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ECOTOXICOLOGICAL EFFECTS OF ORGANOTIN COMPOUNDS ON OYSTER CULTURE

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ABSTRACT

The toxic effect of trialkyltin compounds is used in antifouling paints to protect vessels against biofouling organisms. Tributyltin oxide (TBTO) or fluoride (TBTF) leaches by diffusion from the paint layer into the seawater. Trialkyltin compounds in aqueous solutions are resistant to hydrolysis and biodegradation, but are rapidly converted into diakyl, monoalkyl, and inorganic tin by UV irradiation.

As inhibiting agents of the oxidative phosphorylation, organotin compounds are extremely dangerous for aquatic organisms such as molluscs. The following estimated toxicity levels for Crassostrea gigas oyster larvae have been put forward : at 100 $\mu g.l^{-1}$ the fecondation is inhibited, at 10 $\mu g.l^{-1}$ the cleavage of 20 % of the eggs is affected, larval development is inhibited at 1 $\mu g.l^{-1}$ and already affected at 0.1 $\mu g.l^{-1}$.

Even at a minimal leaching rate of approximately 1 $\mu g.cm^{-2}.day^{-1}$ guaranteeing an antifouling effect, the organotin input is very high in areas where important pleasure-craft activity takes place. The studies on

the effects of antifouling paints on oysters in French coastal zones, where both oyster grounds and pleasure boating areas are located, have demonstrated that :

- the level of contamination with TBT (tributyltin) of the natural seawater is of the same order than the levels causing a toxic effect on oyster larvae in laboratory experiments ;
- the total tin content in oyster tissues increases in the vicinity of ports and areas where a great number of boats are moored ;
- there is an obvious correlation between shell malformations of Crassostrea gigas and input areas of organotin ;
- in laboratory experiments TBTO can produce shell malformations similar to those observed in marine areas.

Taking the deleterious effects of TBTO into account, French authorities have forbidden the use of organotin in antifouling paints (decree of September 14, 1982.)

KEYWORDS

Marine ecotoxicology, Hazard assessment, Bioassays, Antifouling paint, Organotin, Oysters, Crassostrea gigas, Calcification anomalies.

INTRODUCTION

Due to their toxic effect on marine fouling organisms, the application of organotin compounds in antifouling paint formulations has largely increased in the last 10 years. During the same period pleasure boating in coastal areas has considerably increased as well. In fact, in many regions pleasure boating and oyster culture go side by side. For example, during July and August 15 000 boats cruise the Basin of Arcachon, one of the most important French zones for oyster culture, located at the Atlantic coast to the south of the Gironde.

Since 1975 culture of the Pacific oyster, Crassostrea gigas in this bay encounters difficulties in those areas where little water exchange with the ocean exists, namely :

Panels coated on one side with an antifouling paint containing tributyltin fluoride (TBTF) were used as organotin input. For the tanks receiving the oyster samples IV and V, the panel surface was respectively 50 and 500 cm^2. In this test procedure samples II and III were used as references for the ecotoxicological tests in the natural environment (I) and in the laboratory (IV and V).

3. Survey of the tin levels in oyster tissue in different areas of the French Atlantic coast. The chemical analysis was performed by spectrophotometric atomic absorption according to the method of Thibaud (1980).

RESULTS AND DISCUSSION

INFLUENCE OF MARINA INPUTS ON THE OYSTER SHELL GROWTH

Fig. 1 and 2 show the variation of the ratio Lv/Ev in 18-months-old oysters, along the east coast of Oléron Island and in the vicinity of La Rochelle, both located to the north of the River Gironde.

Oléron Island

The main source of organotin is located in the Perrotine channel where approximately 300 boats stay during the whole summer. Tidal currents transport the polluted water from station I to station X. Fig. 1 clearly shows that the ratio Lv/Ev increases from station I to station X, indicating that the shell thickness decreases towards a normal value reached at station X. The malformations are most pronounced within a radius of 1 km from the mouth of the Perrotine channel. In this area the ratio Lv/Ev is lower than 6 and the Japanese oysters are ball shaped.

La Rochelle coast

Numerous sources of organotin originating from treatments with antifouling paints are present along the coast, In Fig. 2 the main sources are plotted (from north to south) :

- Port du Plomb : capacity of 70 pleasure crafts ;
- La Pallice (commercial harbour) : 15 dock sites ;
- Port des Minimes : capacity of 2 700 pleasure crafts ;
- Port du Loiron : 60 anchorages during the summer.

- high mortality rates of the larvae in the natural environment not related to unfavourable hydroclimatic conditions (water temperature, salinity, dissolved oxygen);
- malformations of the oyster shell due to a disturbance of the calcification mechanisms ; the shells become thickened with cavities containing a gelatineous substance.

Shell malformations have also been observed in other oyster growing regions such as Marennes-Oléron and La Rochelle, but the phenomenon was limited to the vicinity of the marinas. Consequently, the pollutants input through marinas, especially of organotin compounds has been suspected to be responsable for the malformations of the oyster shells (Alzieu et al., 1980). The present paper gives the general conclusions of the studies conducted at the laboratories of the "Institut Scientifique et Technique des Pêches Maritimes" (ISTPM) concerning the impact of organotin on oyster-shell calcification.

MATERIALS AND METHODS

Three kinds of studies were performed :

1. Biometry of oysters living in the vicinity of marinas or areas of organotin input. The degree of malformation was determined by the ratio Lv/Ev, where Lv represents the upper-valve length and Ev the upper-valve thickness. The objective was to determine if any correlation existed between the distance from the input site and the degree of shell malformation.
2. Laboratory and field experiments to investigate if oysters exposed to water polluted by organotin compounds, can develop malformations similar to those observed in the natural environment. One stock of adult oysters without any shell malformations was divided in five samples dispatched as follows :
   I in the marina of Boyardville (capacity of 300 boats) ;
   II in the River Seudre near the water intake of the laboratory ;
   III, IV, and V, each in a 150 l tank at the laboratory. Seawater was provided by tidal movement (tanks were successively filled and drained according to the tidal periode).

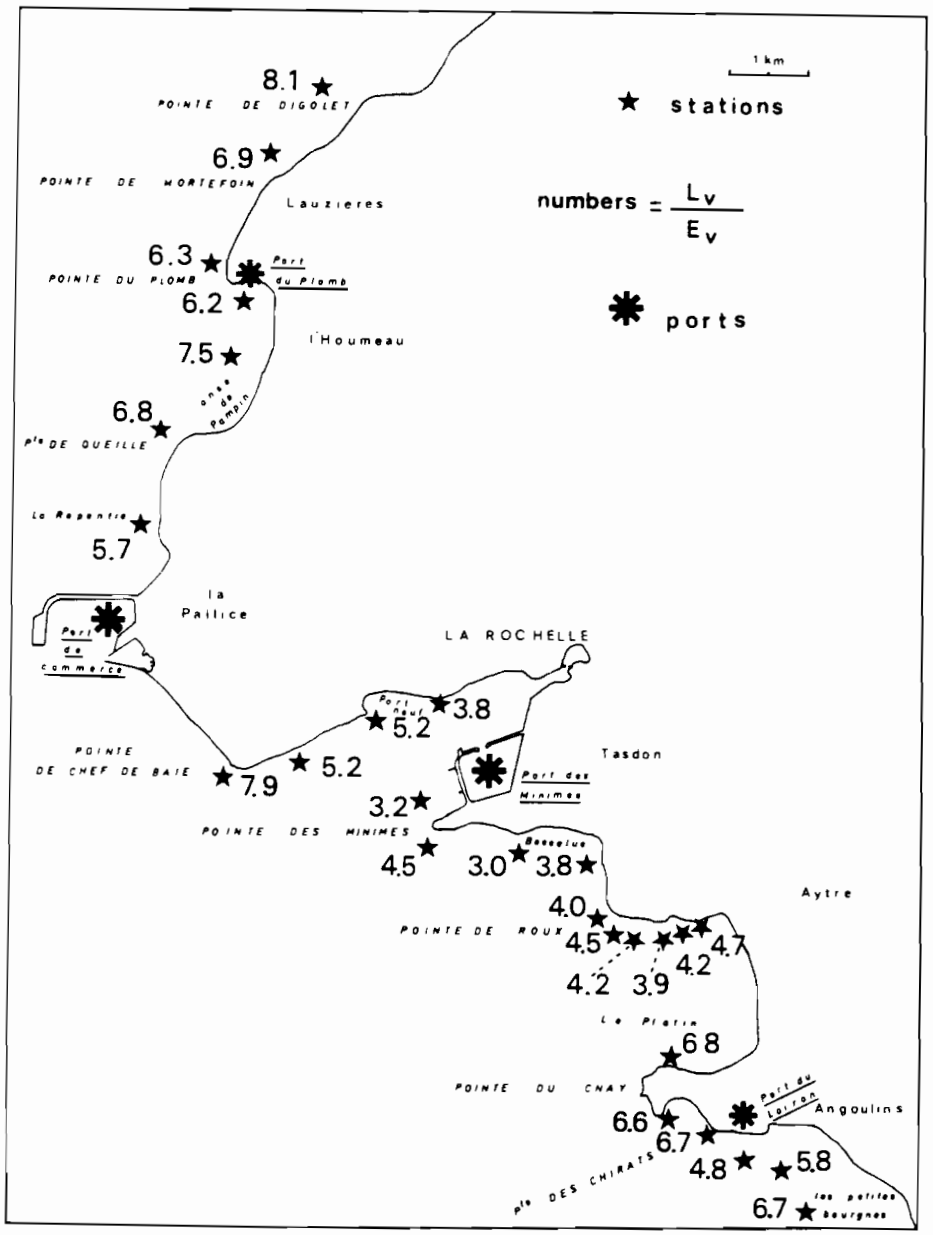

Fig. 2. Correlation between shell malformation in Japanese oysters and various organotin inputs along the French coasts (Alzieu et al., 1981 (1982)).

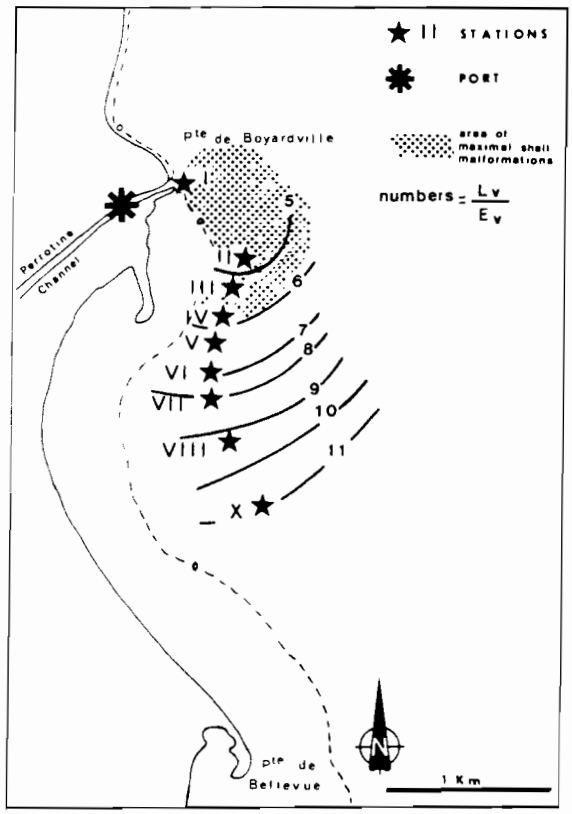

Fig. 1. Correlation between shell anomalies and organotin input from the Perrotine channel (Cléron Island). Lv = upper-valve length, Ev = upper-valve thickness (Alzieu et al., 1981 (1982)).

The Lv/Ev values range between 3.0 (Besselue station) and 8.1 (Pointe de Rigolet). The shells are in poor condition (ball shape) in all areas south of Pointe de Chef de Baie. The local inputs at Port du Plomb and Port du Loiron have a supplementary adverse effect on the Lv/Ev ratio.

In conclusion, it can be said that the vicinity of pleasure-craft ports and a commercial harbour can heavily affect the quality of the oyster shells. However, numerous chemicals pollutants, other than organotin, are present in marina waters. Therefore the possible adverse effects of organotin on shell calcification have been evaluated by means of laboratory experiments.

Fig. 3. Anomalies in the calcifications of the upper valve in the Japanese oyster. a = sample IV in tank with 50 cm^2 panel coated with antifouling paint, b = sample I in Port de Boyardville, c = sample III reference tank.

ENVIRONMENTAL LEVEL OF TRIBUTYLTIN

The actual levels of organotins in the natural environment are difficult to assess because the chemical analysis of alkylated tin compound at very low concentration is difficult. Measurements of the total tin content of the tissues of oysters living in the Basin d'Arcachon have demonstrated that tin is preferably accumulated in the digestive gland and thegills. The values range between < 0.5 to 17.4 $mg.kg^{-1}$ on a dry weight

EFFECTS OF TRIBUTYLTIN ON CALCIFICATION MECHANISMS

During these experiments from June through December, observations were made on the lethality rate, shell morphology, and metal content of the tissue (tin, copper, zinc, cadmium, lead).

Lethality

High mortality rates were noted in all samples in contact with tributyltin. The control organisms in samples II and III were alive at the end of the experiment, while the lethality reached 100 % in sample V (tank with panel surface of 500 cm^2) after 30 days, and in sample I(Port de Boyardville) after 170 days. In sample IV (tank with panel surface of 50 cm^2), a mortality rate of 30 % was observed after 110 days.

Metal content

In unpolluted waters, values for the total tin content in oyster tissue were always lower than 1 $mg.kg^{-1}$ on dry weight basis. In sample I (Port de Boyardville), on the contrary, the total tin content had increased up to 110 $mg.kg^{-1}$ after 80 days. In sample IV the maximal level attained was 25 $mg.kg^{-1}$. The lead and cadmium levels were not affected by the test and remained constant during the experiment. The copper and zinc content, on the contrary, showed some disturbances as compared to the reference samples.

Calcification anomalies

The oysters in samples I (Port de Boyardville) and IV (panel surface of 50 cm^2) developed gel-filled cavities after respectively 100 and 110 days of experimentation during the shell-growth period. Sampling occurred on September 15. Fig. 3 shows the aspect of the oyster shells in samples II and IV as compared to the reference. The oysters in sample V died before shell malformation occurred.

This experiment proves that organotin compounds present in natural environments (marinas) are toxic for adult oysters and induce malformations of the oyster shells. Tin can be heavily accumulated in the oyster tissue as a result of exposure to tributyltin released by antifouling paints.

basis (Alzieu et al., 1980). Preliminary results obtained from water sampling in the same area, during the 1982 summer give concentrations of total organic tin between 0.1 and 0.3 $\mu g.l^{-1}$ tin.

CONCLUSIONS

Organotins released in the seawater by antifouling paints, represent a great danger for the oyster culture, since trialkyltin appears to be :

- toxic for adult oysters : oysters do not survive in polluted marinas; recently Heral et al. (1983) determined that 0.5 $\mu g.l^{-1}$ of tributyltin fluoride resulted in 100 % mortality after 35 days ;
- toxic for oysters larvae : different toxic levels for larval development have been estimated (His and Robert, 1980) :
  - 100 $\mu g.l^{-1}$ inhibition of the fecondation ;
  - 10 $\mu g.l^{-1}$ adverse effects on the egg cleavage (20 %) ;
  - 1 $\mu g.l^{-1}$ inhibition of the larval growth ;
  - 0.1 $\mu g.l^{-1}$ affects the larval development ;
- responsible of shell malformations due to adverse effects on the calcification mechanisms, inducing the formation of gel-filled cavities,

As a result of these findings the French authorities have forbidden the use of organotin antifouling paints for boats less than 25 m long (decree of September 14, 1982).

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