



The occurrence and spatial distribution of pesticides in sea water of the Agadir bay (South of Morocco)

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Abstract

The Agadir Bay is one of the most important economic centers of Morocco, but it is the site of several pollutions due to the various anthropic activities (agricultural, industrial, tourism), the bay opens onto the plain of Souss which is a highly developed agricultural region. The intensification of modern agriculture in Souss Massa region, especially for vegetable crops destined mainly for export, has been accompanied by the extensive use of pesticide in order to increase yields. In order to assess the contamination by pesticides in sea water of Agadir bay, the water samples are collected, seasonally, between winter 2009 and autumn 2010. The concentrations of pesticides in water, were analysed by gas chromatography using electron capture detector (GC/ECD). Among 32 investigated pesticides, only seven molecules (vinchlozolin, endosulfans, lindane, dichlofluanide, bupirimate, aldrin and tetradifon) have been detected. The total concentration of pesticides residues detected in the different sites shows that the waters of the two estuaries and Anza beach are more polluted compared to the beaches of Aghroud and Cap Ghir. The level of contamination of water by total pesticides presented a seasonal variation for all sites, the highest concentrations are obtained in spring and the lowest in the autumn, also the results shows an increases contamination from north to south of Agadir bay. The results obtained in this study show a presence of the pesticides residues during all sampling season, this contamination could be related to the most intense use of this product in agriculture of Souss Massa region.

1. Introduction

The socio-economic life of the Agadir Region is based on fishing, agriculture and tourism. This requires good knowledge and management of the environment in the Region (resources and ecosystems). Several studies have focused on the study of coastal ecosystems: ecotoxicological study [1-2-3], environmental characterization and stand structure [4], species biology [5-6-7], evaluation of chemical contamination [8-9-10], evolution of the ecosystem environment [4-11], impact of port activities on the coastal environment [12], impact of wastewater on the marine environment [4-11].

However, no research project has been undertaken to study the impact of pesticides used in agriculture on marine ecosystems. Whereas, the region knows an intense agricultural activity using many products. Indeed, considered as one of the most important agricultural poles of Morocco and practicing a modern and very productive agriculture, the region records the use of considerable quantities of phytosanitary products as fertilizer and pesticides to protect crops in order to increase yields. These products are not without impact on agricultural land, on the water table and certainly on the coast. These products are drained to the coast ecosystems [13-14].

The aim of the study is to evaluate the levels of some pesticides residues in waters collected from five sites along the coast of Agadir bay and to provide information for further research and inform decision-makers to take effective measures to gradually reduce the use of these products. in agriculture and, indeed, their impact on soil, plants, water table and on coastal ecosystems.

2. Material and Methods

2.1. Study sites

Five sites of Agadir Bay located in the north and the south have been studied:

a- Massa river estuary

The location site (30 ° 02.491 N ; 9 ° 48.641 W) is situated at 45 kilometres to the south of Agadir city, in a mainly agricultural area, particularly the lucerne (*Medicago sativa*) and is widely used for cattle breeding. The Massa river estuary is Wetlands of international importance for migratory birds, it was classified as a Ramsar site in January 2005. It is also part of Souss Massa National Reserve and contains a lot of endemic flora and fauna. The Massa river estuary is one of the marshy brackish freshwater environment which is home to more than 30 species of birds breeding, migrating or wintering, some of which are among the most remarkable. This ecosystem is made up of fine sand and coastal strip separating the lagoon from the Atlantic Ocean. This cord is composed of dunes constantly displaced by the wind. A cordon of sand is formed there and disappears due to wave dynamics, water-current dynamics and sediment of the beach, while the complete bar closure refers to the importance and the frequency of water exchange between the ocean and the river at the level of the estuary [15].

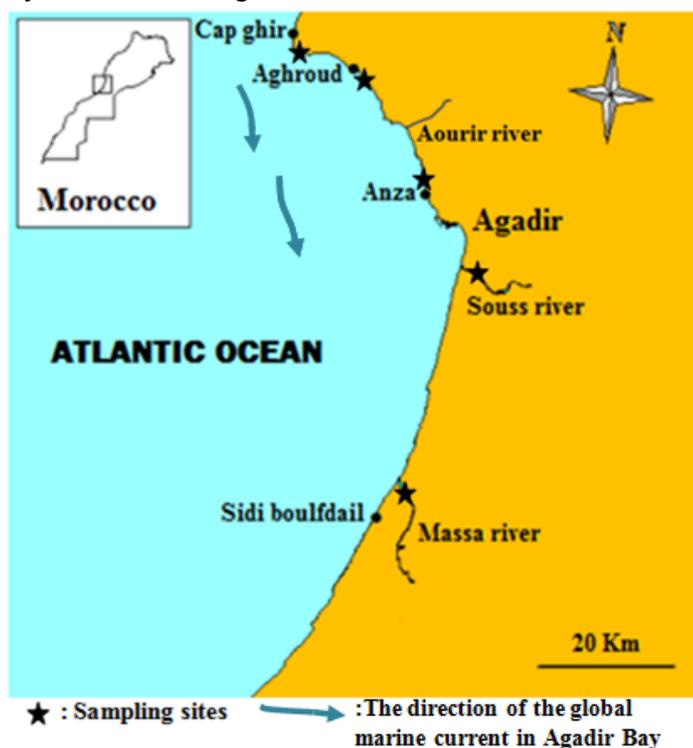


Figure 1 : Location of Agadir bay in Morocco and the sampling sites.

b- Souss river estuary

The location site (30°21,858'N ; 009°35,303'W) it is the natural border to the north of the Souss-Massa National Park. The Souss river estuary constitutes an important biotope for avifauna, particularly migratory birds and is included in the list of wetlands protected under the Ramsar Convention. It is also a part of Souss Massa National Reserve and contains a lot of endemic flora and fauna [16]. However, Souss river crosses one of the most modern and productive agriculture areas in Morocco, whose products are destined to a large market [16- 17].

c) Anza Beach

The location site (30 ° 26 'N ; 9 ° 38' W) is situated at 10 Km in the North of Agadir and has a sandy beach and a rocky coast. This site is subject to industrial waste and effluents (liquids, solids, dust) from various small industrial factories, located in the Anza region, and domestic effluents directly pouring into the sea and without any prior treatment. It is also near to the Aourir river estuary, which most of the edges of this river are covered with banana crops requires the intensive use of pesticides. All the studies carried out on this site have shown that it is a polluted site with a relatively high degree [1-5-6-8-18].

d –Aghroud Beach

The location site (30 ° 36'N ; 9 ° 47' W) is situated at 35 Km north of Agadir city on the road leading to Essaouira city. It is a sandy beach whose most dominant sands have a size between 0.125 and 0.2 mm with a median particle size of 0.180mm, the classification index [19] varies between 1.04 and 1.19 indicating a well-

classified sediment. The various studies carried out at the laboratories [6] have shown that this site is not very polluted because it is far from any industrial or agricultural activity, but there are some fishing activities and tourism.

e- Cap Ghir

The location site (30 ° 38 'N ; 9 ° 53' W) is situated at 50 km north of Agadir city , on the road leading to Essaouira city. It has a rocky coastline open to the sea and exposed to hydro-dynamism. It corresponds to the northern limit of the up welling high concentrations of nutrients especially in summer [20]. This site is far from any type of pollution; it contains a high density of mussels.

2.2 Water sampling

Each season, 500 ml of water is taken, put in glass bottles with Teflon stoppers. The bottles used are washed with detergent, rinsed with distilled water and then with acetone. After that, they are dried in the oven. The sampling consists of mixing and shaking the water bottle well and immersing the sample bottle with the cap under the surface of the water and removing the cap under water. Collected samples are put a cooler box with ice and immediately transported to the laboratory where they are stored at 4 C° until extraction, the storage period does not exceed 24 hours.

2.3. Extraction and cleanup

For each sample of water withdrawn, 250 ml is used for the determination of pesticides residues. To this quantity is added 30 ml of saturated solution in sodium chloride and 50 ml of hexane and then gently stirred to remove the gases formed and allow it to settle for phase separation by avoiding emulsions. Thereafter, the organic phase is recovered in a round bottom flask topped with a glass funnel, lined with a glass cotton pad and containing 20 g of sodium sulphate (Na₂SO₄) (extraction 3 times). The filter is then washed with 20 ml of hexane. the organic phases are evaporated on a rotary evaporator under vacuum at a temperature less than 50°C; and the extract is taken up with 3 ml of hexane solution. The concentrated hexane residues are analysed using gas chromatography (GC) analysis.

2.4. Compounds Identification

An HP7890 gas chromatograph equipped with two Ni electrons captures detectors (GC-ECD) has been used for detecting the pesticides levels in water samples. 1µL of each sample and a blank extract are injected, separately, into the GC-ECD. For every five injections made, the GC-ECD programmed conditions are calibrated by injecting 1µL of hexane. Quantification is achieved by comparing the peak areas of sample injections with those of the 17 standards analysed under the same conditions.

3. Results and discussion

3.1 . Total Pesticide residues in seawater of Agadir bay : Spatial and seasonal variations.

The figure 2 shows the total pesticide residues in seawater of different sites studied in Agadir bay and in different seasons.

a. Spatial distribution

To compare pesticide contamination in different sites studied in Agadir Bay, we chose the samples taken in the spring because the concentrations recorded maximum values in all sites. Figure 2 shows the concentrations recorded in different sites.

The data show clearly that the level of total pesticide contamination increases from north to south of Agadir bay and that areas facing the estuaries are more contaminated by total pesticides compared to the rest of the bay sites. Indeed, the total concentration of pesticides residues detected in the different sites shows that the waters of the two estuaries (Massa and Sous) and Anza beach (not far from Aourir estuary) are more polluted compared to the beaches of Aghroud and Cap Ghir. The Massa estuary shows the highest concentrations (312.74 ng / l) followed by the Souss estuary (291.09 ng / l), and by Anza site (291.09 ng / l) ; while the beaches of Aghroud and Cap Ghir are less polluted by pesticides ; probably because these two last sites are far from any agricultural activity.

b. Seasonal variation

The level of contamination of water by total pesticides presented a seasonal variation (figure 3). For all sites the highest concentrations are obtained in spring and the lowest in the autumn for two years studies. Massa estuary presents the highest concentration (312.74 ng/l) observed in spring 2010, followed by the Souss estuary with a value of (291.09 ng/l), then the Anza beach with a concentration of (122.54 ng/l), also recorded in spring 2010. In this last season, the concentration in beaches of Aghroud and Cap Ghir is respectively 11.44 and 14.72 ng/l.

The results also show that the concentrations of total registered pesticides in the waters of the different sites during year 2010 were higher than those recorded in year 2009.

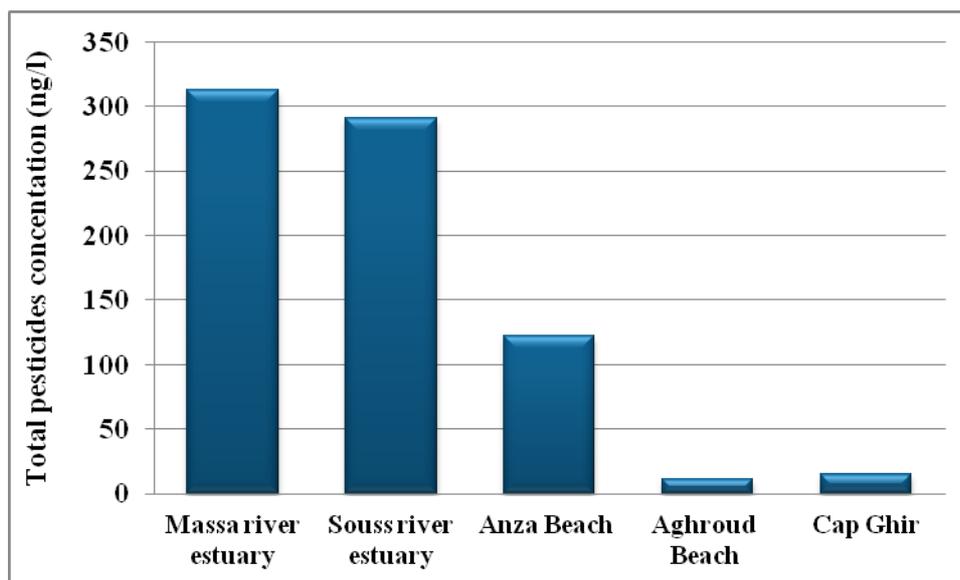


Figure 2 : Total pesticides detected in seawater sampled in spring in 2010 different sites of Agadir bay.

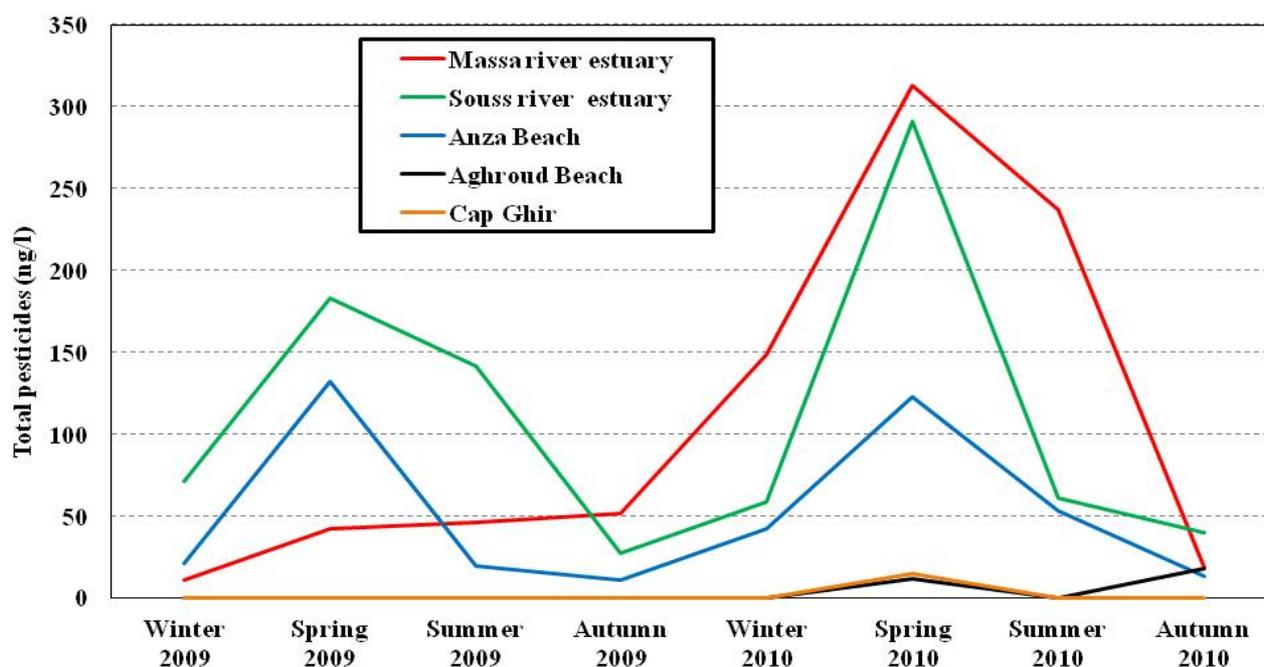


Figure 3 : Total pesticides detected in seawater sampled in different sites of Agadir bay.

3.2. Individual pesticide residues in water of Agadir bay

a. Massa estuary

The results of the analysis of pesticides residue in water samples collected in the estuary of Massa are shown in Figure 4. Among 32 investigated pesticides, only five (Dichlofluanide, lindane, Endosulfan S, Bipurimate and Tetradifon) have been detected. The level of concentrations depends on the nature of the pesticide and the sampling period. The highest concentration is Tetradifon 160,404 ng/l recorded at spring 2010. The concentrations of Endosulfan oscillate between ND (undetected) and 96,119 ng/l ; those of Bupirimate, Dichlofluanide and lindane vary between ND and respectively 62,644 ng/l, 56,219 ng/l and 46,134 ng/l.

The seasonal variation of the individual pesticides residues in water of the Massa estuary shows that the highest concentrations are obtained in spring and the lowest.

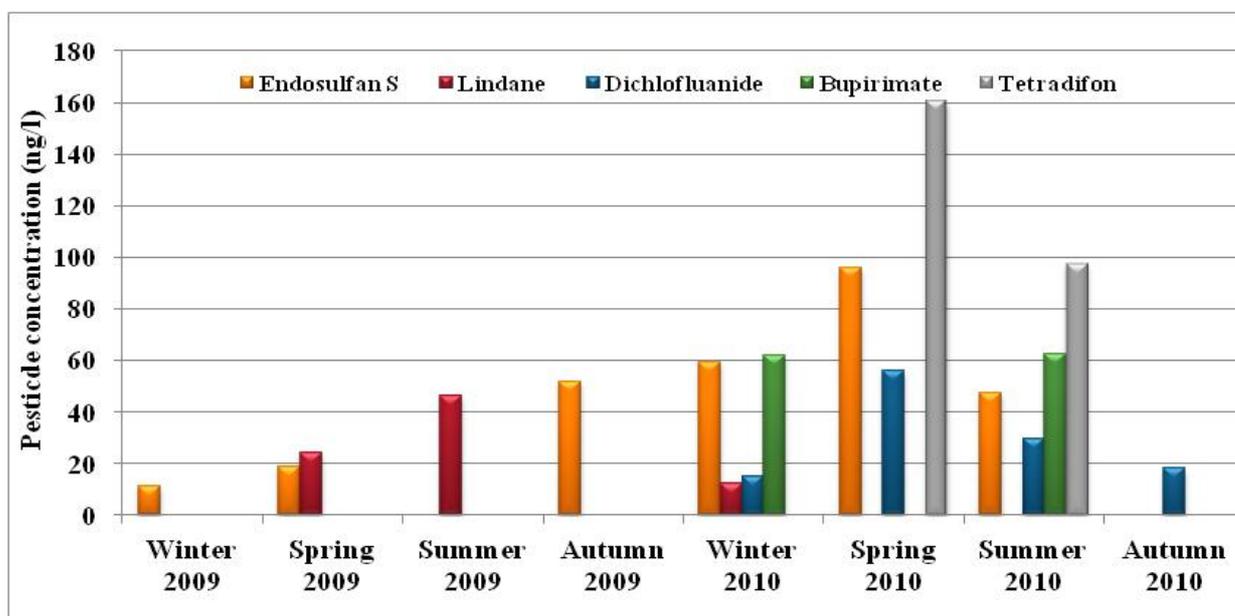


Figure 4: Seasonal variation of individual pesticides in water of Massa river estuary

b. Souss river estuary

The study showed that water samples collected between winter 2009 and autumn 2010 in Souss estuary are contaminated by the following compounds : Endosulfan S, vinchlosolin, lindane, aldrin, terradifon, dichlofuanide and bipurimate (figure 5). Endosulfans, lindane and dichlofuanide are the most abundant residues in the estuary, with concentrations ranging between ND and 69,304 ng/l for lindane ; between ND and 68,800 ng/l for dichlofuanide and between ND and 68,553 ng/l for endosulfans. The concentrations of aldrin vary between ND and 33,957ng/l, those of bipurimate oscillate between ND and 21,428 ng/l. Those of tetradifon vary between ND and 151,969 ng/l, while those of vinchlozolin are detected only in autumn 2010 with a concentration of 24.3 ng/l.

The seasonal variation of the individual pesticides *residues in water of the Souss estuary* shows high values in the spring and low concentrations in autumn (Figure 5).

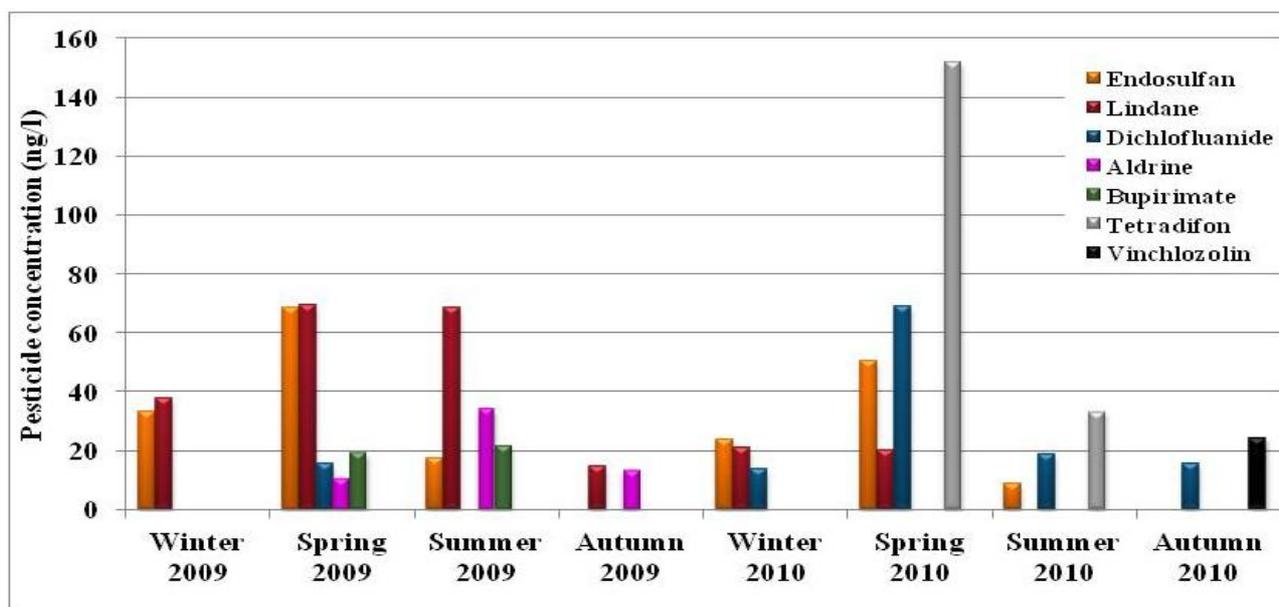


Figure 5: Seasonal variation of individual pesticides in water of Souss estuary

c. Anza beach

Variations of pesticide residues in the waters of the Anza beach during two years of study are presented in Figure 6. The analysis shows that in water of this ecosystem, the level of contamination vary with the seasons and the nature of the pesticides studied. Among the desired compounds alone, Endosulfans, Lindane, Aldrin,

Bupirimate and dichlofuanide have been detected. Concentrations of Endosulfan S range from ND and 35.523 ng/l. Those of lindane oscillate between ND and 53.372 ng/l. Concentrations of aldrin oscillate between ND and 22.506 ng/l, and those of Bipurimate vary between ND and 84.161 ng/l, whereas dichlofuanide has concentrations varying between ND and 18.31 ng/l.

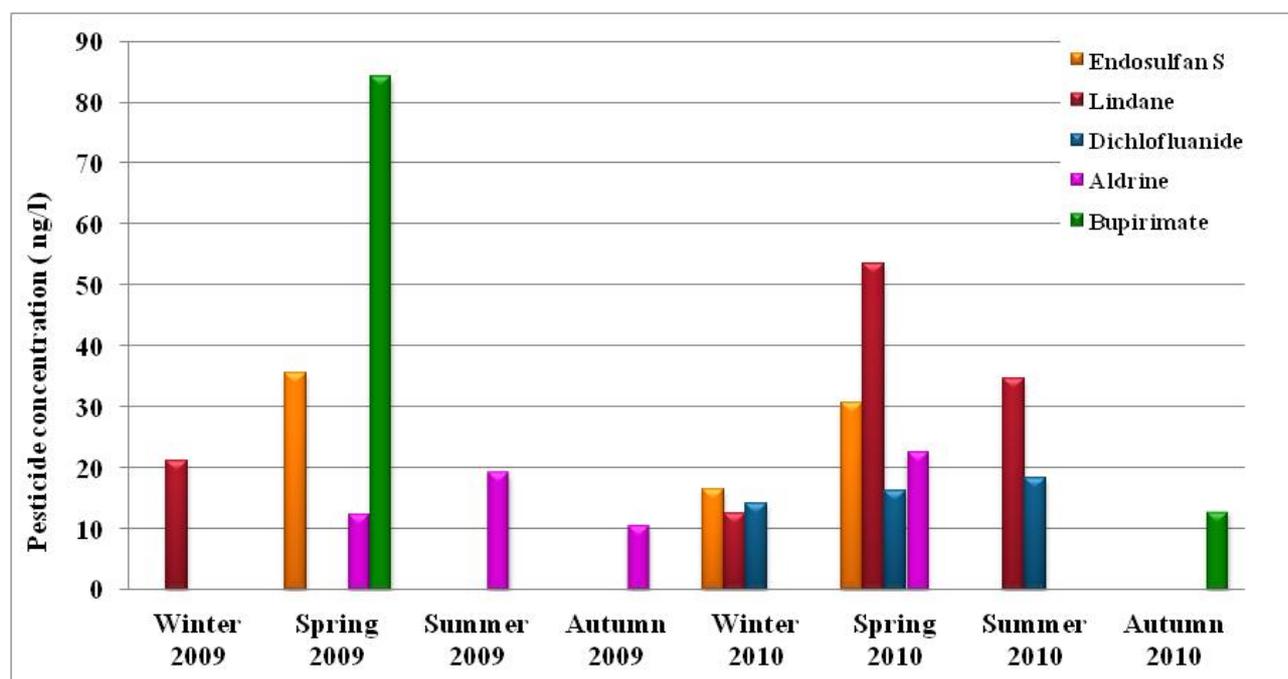


Figure 6 : Seasonal variation of individual pesticides in water of Anza beach

The seasonal variation of the individual pesticides residues in water of the Anza beach follows the same profile than the seasonal variation of total pesticides with the high concentrations in spring and the low ones in autumn.

d. Aghroud and Cap Ghir sites.

The results of residue analyses of pesticides in water samples collected in Aghroud beach during the study period show that, among 32 investigated compounds in this study, only two molecules have been detected: Dichlofuanide in autumn 2010 (18.127 ng/l) and tetradifon in spring 2010 (11,448 ng/l). The analyses of water samples taken from Cap Ghir beach during study period show that, among the 32 investigated molecules pesticides, just tetradifon was detected in this ecosystem in spring 2010 with a concentration of 14.729 ng/l.

3.3. Discussion

The results of the analyses of the water samples taken at the various sites show the presence of pesticide residues in the waters of Agadir Bay at concentrations varying according to the nature of pesticides and water samples taken in different periods of time. The waters of the estuaries of Souss and Massa present the highest concentrations of the total pesticides compared to the waters of Aghroud and Cap Ghir beaches. The high levels of contamination of waters of these two estuaries (Massa and Souss) are mainly due to the contributions of the pollutants by Souss and Massa rivers (that cross large agricultural areas). The contamination of the Anza site can also be explained by its proximity to the Oued Aourir estuary. The latter is located 3 km north of Anza site. Comparaison of pesticides level in Massa, Souss and Anza shows that the contamination increases from north to south (Anza, Souss and Massa). This fact could be explain by the presence of the current North South in Agadir Bay [21]

Thus, comparaison of pesticides level in différent seasons shows that the contamination is higher in spring in all sites studied. Significant peaks are observed during this season, coincides with the period of pesticides use in agriculture, usually taking place in spring. This explains the high levels of the bay seawaters by these pollutants in spring.

Of the 32 compounds investigated, only vinchlozolin, endosulfans, lindane, dichlofuanide, bupirimate, aldrin and tetradifon have been detected. Endosulfan S is present whereas the endosulfans A and B are rarely present. This could explain the fact that endosulfans A and B are less persistent in the environment, and are rapidly transformed to endosulfan S, whose persistence is higher than the first ones. Indeed, the half-life of the

degradation of endosulfan A is about one to three months then that of endosulfan B and endosulfan sulfate could range from two to six years [22].

About the level of contamination in Agadir Bay contamination, it is important to point out that the values obtained do not exceed the Maximum Residue Limit (MRL) (0.1 µg / L) from the European Union. Our results are comparable to those recorded in Togo in Adta water wells (0.07 µg/l) and lower than those detected in the Anié River (0.57 µg / L), the Mono River 0.69 µg / L [23] and Senegal where Cissé et al [24] obtained levels of 1.26 µg/l for endosulfan A and 1.84µg/l for endosulfan B in the water wells in the Niayes area. On the other hand, our concentrations are higher than in Ghana where [25] endosulfan is found at a concentration of 0.036 ng/L in the waters of Volta Lake.

The aldrin level in water of Agadir Bay varies between 0.01 µg/l and 0,03µg/l. These concentrations do not exceed the value of the (MRL) of the European Union (0.03 µg/l), and are less than those detected in other regions of the world, in Senegal the aldrin was detected at a value of 1.70 µg/l [24]. As for lindane, the levels in the water of the Souss estuary (ND-0.069 µg / l), of Massa estuary (ND - 0.046 µg/l) and of Anza Beach (ND - 0.053 µg/l) are comparable to those detected in surface water of many regions in China (1.81-61.95 µg/l) in water of the Qiangtang River [26]; (0.052-0.515µg/l) in the Mingjiang River; (0.031-0.129.8 ng/l) in the river of Jiulong; (0.070-0.992 µg/l) in the Tonghui River and (0.004-0.005 µg/l) in the river of Yangtse [27-28-29]. These two compounds, lindane and aldrin, are among the compounds which have been banned in Morocco since 1984. Their presence in the waters of the estuaries of Souss and Massa rivers could be explained either by the fact that these molecules are still use illegally or by a release of these pesticides from old sediments.

Dichlofluanid is frequently detected at all sampling sites except Cap Ghir, with high concentrations in the waters of Souss and Massa estuaries especially during spring 2010. This can be attributed to the use of Euparenne (commercial product of dichlofluanid) as an anti-powdery-mildew fungicides in Souss region[30] and its solubility in water (1.3 mg/l at 20°C).

This compound is easily degradable in water by photolysis, giving a more toxic by-product (dichlorofluoromethane)[31]. The same research team has shown that the presence of organic matter in water decreases photodegradation of dichlofluanid, which is often the case, especially in estuaries.

Bupirimate (AAKO: commercial product), used as a systemic fungicide against the powdery mildew of several vegetable and fruit crops, is rarely detected in water of Agadir Bay. This could be explained by its rapid hydrolysis at pH 7 and its half-life of 63 days, and high solubility in water (22 mg/l). (Index of phytosanitary products, Version: 04.07.2013).

Tetradifon is an acaricide with a non-systemic action. Its presence in the water of Agadir Bay can be attributed to its use in the fight against a broad spectrum of larvae of insect pests on citrus, and its persistence in water due to its low solubility in water 0.078 mg/l and its non-volatility.

Vinchlozolin (Ronilan DF: commercial product) is rarely observed in the waters of Agadir bay. It is used as a preventive contact fungicide to protect crops vegetables and vegetables against gray mold (*Botrytis cinerea*) [32]. The absence of this molecule can be linked to its rapid hydrolysis, as well as to a temperature the half-life of vinchlozolin is 2.8 hours at 35°C[33] and at photo-degradation, about 10% of vinchlozolin is degraded after exposure to UV ($\lambda \geq 290$ nm) for 8 h [34].

Conclusion

The results reported in this study made it possible for the first time to provide an overview of levels of pesticide contamination in Agadir region ecosystems one of the most important agricultural areas of the Kingdom of Morocco, in particular for crops destined for export. This study reveals contamination of the surface waters, in the bay ecosystems by pesticides used in agriculture (Endosulfans (A, B and S), Vinchlozolin, Lambda - cyhalothrin, Lindane, Aldrin, Bupirimate, Bifenthrin, fenarimol, deltamethrin, Tetradifon and dichlofluanid). The noticed contamination is close or even below that known and listed in various regions of the world. This contamination of water, by pesticides observed in this work, may be harmful to aquatic life especially for detritivores and filtering organisms that are capable of accumulating large quantities of these pesticides in their tissues from water and subsequently transfer these pollutants through the food chain.

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