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Environmental hazards associated with pesticide import into Costa Rica, 1977-2009

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Abstract

Raw pesticide import data from 1977 to 2009 obtained from the Ministry of Agriculture in Costa Rica were processed and analyzed. The quantity of specific active ingredients (a.i.), and chemical groups were calculated by year and presented in ten-year periods. Three sets of environmental hazard indicators were constructed: one for general pesticides exposure to monitor tendencies in time, including total quantities imported divided by significant denominators, such as hectares of protected and wetland areas. The second indicator calculates pesticide use on the Pacific or Caribbean slope. The third one is an assessment of environmental hazards intended to estimate fate and toxicity to aquatic biota. A review of Costa Rican aquatic ecosystems' contamination with pesticides is presented. The annual average import as well as the quantity of pesticides capable of reaching water bodies increased during the analyzed period. The same was observed for harmful a.i., 98% of the pesticides imported were classified as acutely toxic for fish and crustaceans and 73% for amphibians. Approximately 8.4 kg of a.i. were imported per hectare of protected areas and 24.3 kg of a.i. per hectare of wetlands. The contamination of aquatic systems over time by specific pesticides matches quite well the list of imported ones. We recommend using data of pesticide imports as a source of information to evaluate environmental risk exposure and promote changes to reduce impacts on aquatic systems.

Key words

Hazards, Indicators, Pesticides import, Toxicity

Introduction

Pesticides imported for pest control might negatively impact the environment and human health (WHO/UNEP, 1990; Wesseling *et al*., 2001); especially in developing countries such as Costa Rica, where toxic pesticides are used and their negative effects have been observed in the environment (CGR, 2005; Castillo *et al*., 2012). During the last three decades, attention regarding the environmental impact of pesticides has focused on the study of aquatic ecosystems through the implementation of scattered programs for monitoring pesticide concentrations in water, sediment and biota; conducting acute toxicity tests with algae, daphnia and lettuce seeds; the generation of biotic diversity indexes for benthic invertebrates; and the analysis of some episodes of environmental accidents involving death of aquatic fauna (Castillo *et al*., 1997; Castillo *et al*., 2000; de la Cruz *et al*., 2004; Castillo *et al*., 2006; Echeverría-Sáenz *et al*., 2012; Castillo *et al*., 2012; Rizo-Patrón *et al*., 2013). A few studies include chronic and delayed effects, but such effects remain mostly undetermined (Mena *et al*., 2012; Navarro *et al.,* 2013).

Global and local statistics related to production, export, import and use of pesticide to assess trends are scarce, not withstanding the environmental hazards they pose and how valuable they can be in prospective environmental analysis. It has been estimated that the world's pesticide use in 1995 was 2590 million kg (Aspelin, 1997) and that this use dropped almost 9% to reachin 2006 and 2007 a value close to 2363 million kilograms (Grube *et al*., 2011). In countries like the United States (Grube *et al*., 2011) and Sweden (KemI, 2010) a decline in the use of pesticides is also observed. According to data presented by the United States, Environmental Protection Agency in 2011 a reduction of 9.5% in the general pesticides transfer was observed between 1970-1979 and 2000-2007, with annual average of 436.8 and 395.4 million kg respectively. The percentage of pesticides used for agricultural activities increased from 70.9% (309.8 million kg) to 77.8% (307.8 million kg) during the same period, with a total decrease in general pesticide transfer of 0.6% for the whole period (Grube *et al*., 2011). According to the Swedish Chemicals Agency (KemI), in Sweden there has been a 31.8% reduction in sales of pesticides with industrial, agriculture and gardening purposes between 1981-1985 and 2009. The largest reduction was achieved in agriculture and gardening where it fell from an annual average of about 5.3 million kg in 1981-1985 to 1.98 million kg in 2009, a reduction of 62.2% (KemI, 2010).The opposite phenomenon is observed in developing countries such as Brazil and Central America. In Brazil, the Brazilian Institute of Geography and Statistics has reported an increase in pesticide use from 2.7 kg ha^{-1} to 3.2 kg ha^{-1} on major crops between 1997 and 2006 (IBGE, 2002, 2008). In Central America, data from different sources show that pesticide imports to the region have increased from 17.9 million kg (Galvao *et al*., 2002) in 1992 to 36.9 million kg in 2003 (Bravo-Durán *et al*., 2011) to 57.9 million kg in 2010 (CEPAL, 2013).

Total import of pesticides a.i. in Costa Rica has also increased (de la Cruz and Castillo, 2002; Ramírez *et al*., 2009). Eleven to twenty-five percent of the pesticide imported into the country is exported (Ramirez *et al*., 2009; Castillo *et al*., 2012). The import and export activities of these products include in many cases their transportation in technical grade to different parts of the national territory. During this transportation accidents might happen. These accidents, as well as pesticide run off or drift from agricultural lands or other activities, have negatively impacted public health and natural environment (Wesseling *et al*., 2001; CGR, 2005; Castillo *et al*., 2012). Once in the environment, pesticides will be distributed into different environmental compartments according to their physical and chemical characteristics (Miyamoto, 1996). In this way, flora and fauna of different natural ecosystems might be acutely and/or chronically exposed and affected (de la Cruz *et al*., 2004; Mena Torres *et al.,* 2013). Some of the pesticides used in Costa Rica have been reported as acute or chronically toxic to aquatic organisms (BCPC, 2003; Footprint, 2006; de la Cruz *et al*., 2010).These include substances such as paraquat, methamidophos, endosulfan, methomyl and terbufos, which are banned or severely restricted in other countries (Nieto and Henao, 2001).

Physico-chemical and toxicological information, together with proxies of pesticide use data, can be used to construct simple hazard indicators, helpful to environmental authorities as well as researchers for policy making, proposing and evaluating pesticide use and exposure reduction programs and identifying natural populations and ecosystems at risk. In Sweden and UK, a monitoring system based on quantitative indicators of potential risks, to the environment among others, has been implemented (Bergkvist, 2004; Pesticide Forum, 2008). These indicators generally include the impacts as risk score calculations such as concentrations of pesticide pollution in water and of pesticides causing acute or chronic toxicity in biota, and the comparison between them; or the behavior of pesticide users including doses, frequencies, crop extension, biocide action, application equipment, runoff and protection of natural ecosystems. In Costa Rica these indicators have not systematically been constructed and analyzed, even though scattered documentation of undesired impact of pesticides on health and environment have been accumulated through researchers' efforts as well as by some governmental entities (Wesseling, 2003; Humbert *et al*., 2007; Castillo *et al*., 2012). The country has not established yet a consistent monitoring program and farmers' collaboration is not always easy to obtain. The Central American Institute for Studies on Toxic Substances has estimated indicators based on import data to monitor environmental and health hazards from 2003-2004 (Wesseling *et al.,* 2003; de la Cruz *et al.,* 2004; Bravo-Durán *et al.,* 2011, 2013).

In this paper, a sustainable and feasible method for monitoring trends of pesticide imports (1977-2009) and associated environmental hazards through a set of simple general indicators is proposed. It also presents an historical review of the water bodies' contaminated with pesticides, as well as some of the documented damage.

Materials and Methods

Raw import data from 1977 to 2009 were obtained from the Ministry of Agriculture in Costa Rica. The amount in kilograms of specific a.i., chemical groups and primary biocide action were calculated according to the methodology reported by Bravo-Durán *et al.* (2011). Data are presented as averages over ten-year periods. Data before 1990 were difficult to obtain and should be viewed with precaution; though its inclusion here helps to see tendencies.

Three set of general pesticide environmental hazard indicators were constructed: one for general environmental exposure intended to monitor tendencies in time, i.e. comparison of average imports per decade. These include average quantities of pesticide imported divided by significant denominators, such as extension of protected and wetland areas in kg a.i. per protected or wetland ha (SINAC, 1997, 2011; Córdoba-Muñoz, 1998; Ramsar Convention, 2011). The second indicator calculates the pesticide load for the Pacific and Caribbean slope basins. This was calculated as the sum of crop cultured land area in hectares per Caribbean or Pacific slope (INEC, 2007; MAG, 2007; CNP, 2009a, b, c, d, e; 2010, 2011, 2012; SEPSA, 2012) times the pesticides used per crop in kg a.i. ha^{-1} (CGR, 2005; Castillo *et al.,* 2012; Bravo-Durán *et al.,* 2013) and divided by the total cultivated land in the slope in hectares. As data were difficult to obtain, they were approximated with crop areas and pesticide use for the first decade of the 21st century, in the following way:

$$\text{kg a.i. (ha of slope)}^{-1} = \frac{\Sigma_{(i\text{-}n)} (C_{Sha} \times P_{cu})}{\Sigma_{(i\text{-}n)} (C_{sha})}$$

C_{Sha}= Crop cultivated hectares in that slope (ha); P_{cu}= Pesticide use in that crop (kg a.i.ha^{-1}); $\Sigma_{(i\text{-}n)} (C_{Sha})$= Total slope cultivated land (ha); i = each crop

The third set is a group of indicators to assess the environmental hazard, intended to estimate fate and toxicity to aquatic biota. These hazard indicators add a dimension of potential environmental risks related to the quantity and types of pesticides imported. Thereafter, we considered databases and published data (BCPC, 2003; Footprint, 2006; Kegley *et al.*, 2011) and employed scoring procedures to rank pesticides imports by risks associated to their environmental fate, and acute and chronic toxicity to aquatic biota. To evaluate environmental fate, the pesticide a.i. was differentiated with regard to its water solubility, soil mobility, interface water-sediment and soil persistence, as well as bio-accumulation potential. The criteria considered to rank pesticides by increased risk to the aquatic environment were as follows: moderate or high for water solubility >50 mg l^{-1} or >500 mg l^{-1}, soil and water sediment persistence >30 days or >100 days and soil mobility with a koc<150 l kg^{-1} or <500 l kg^{-1}respectively. These criteria were adapted from the IRET (1999); de la Cruz *et al.* (2010); Footprint (2006).These parameters were chosen because it is generally considered that a substance characterized by high water solubility, high soil mobility, long environmental persistence, and high bio-accumulation potential has a good chance of reaching aquatic systems and the biota.

The criteria adopted to evaluate if a pesticide can be harmful for the aquatic biota was its acute or chronic toxicity. For acute toxicity, LC_{50} or EC_{50} for specific representative taxonomic groups were used. The toxicity ranked as high or moderate for amphibians, fish and crustaceans included values <10 or <100 mg l^{-1}. For aquatic plants and algae values of <0.01 mg l^{-1} or <0.1 mg l^{-1} were considered high or moderate (IRET, 1999; Footprint 2006; de la Cruz *et al.,* 2010; Kegley *et al.*, 2011). Values over those mentioned were considered low, special attention was given to a.i. for which no data were available. The criteria adopted for chronic toxicity were based on the European Unión recommendations for pesticides classified as endocrine disruptors (CEC, 2007) and pesticides suspected of causing reproductive and/or endocrine disrupting effects (Colborn *et al.*, 1993; Mnif *et al.*, 2011).

The above criteria were linked to import data. The most imported pesticides, more likely to reach water bodies and contaminate and affect aquatic ecosystems, were prioritized, and the first sixteen are mentioned in this study. An historical matrix of pesticide pollution and impact on natural ecosystems in Costa Rica was constructed from national studies and reports.

Results and Discussion

Imports of pesticide during the past three decades increased more steeply than the area devoted to agriculture in Costa Rica (Fig. 1). It went from an average of 3.8 million kg in the eighties to an average of 11.4 million kg during the first decade of the 21st century (Ramírez *et al.,* 2009; Castillo *et al.,* 2012), while the average area devoted to agriculture increased from about 401 to 444 thousand hectares over the same period (SEPSA, 1983, 1989, 1992, 2001, 2007, 2011). Imported kilograms of pesticides per hectare of cultivated land passed from 7.5 kg i.a. ha^{-1} in 1977-1979 to 25.7 kg i.a. ha^{-1} in 2000-2009, one of the highest in Central America (Bravo-Duran *et al.*, 2011). Export products generally use higher amounts of pesticides per hectare and more land area than the majority of crops for domestic consumption; for example, bananas (49.3 kg a.i. ha^{-1}) pineapple (25.2 kg a.i. ha^{-1}) and melon (60.5 kg a.i. ha^{-1}) (Bravo-Duran *et al.*, 2013) sugar cane (10.1 kg a.i. ha^{-1}), beans (3.0 kg a.i. ha^{-1}), corn (3.1 kg a.i. ha^{-1}), cassava (7.4 kg a.i. ha^{-1}), tomato (37.8 kg a.i. ha^{-1}) and potato (37.3 kg a.i. ha^{-1}) (CGR, 2005; Bravo-Duran *et al*. 2013). The rise of pesticide imports may be a result of the increase in areas cultivated mainly with bananas, pineapple, plantain, palm, melon, rice, oranges and sugar cane occurred in the country during the '90s and the beginning of the 2000s (SEPSA, 2001, 2007). These crops are grown in mono cultures, mainly for export, and they use pesticides intensively. From 2000 to 2009, the most expanding cultures were pineapple, palm, sugar cane and plantain, which have increased their cultivated area from 12500 ha to 40000 ha (pineapple); 39790 to 55000 ha (palm), 47200 to 53030 ha (sugar cane). The area under bananas, rice and coffee cultivation declined slightly during the decade 2000-2009 (SEPSA, 2007, 2011).

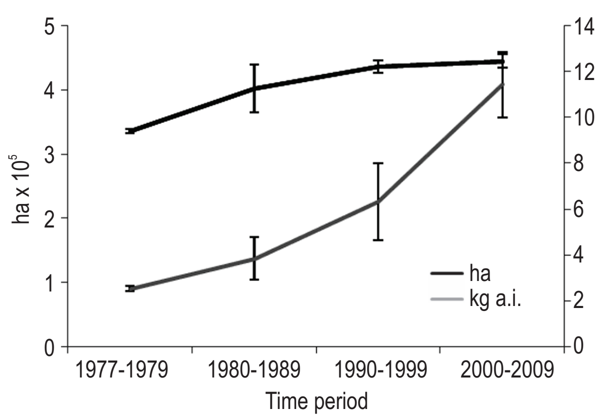

Fig.1 : Pesticide a.i. imported in millions of kilograms of a.i. and hectares of cultivated land in Costa Rica 1977-2009

The most imported group according to biocide action during the seventies corresponded to herbicides (35%) and insecticides-nematicides (35%) followed by fungicides (29%) and fumigants with 1% (de la Cruz and Castillo, 2002); whereas during the following twenty six years (1980-2006), the most imported were the fungicides (54-46%), herbicides (25-30%), insecticides-nematicides (16-15%) and fumigants (1-11%) (Ramírez *et al*., 2009). Crops such as banana, pineapple and coffee used important quantities of fungicides. Banana and pineapple are cultivated at high temperature and humid conditions in lowlands of the country. Coffee is cultivated mainly in highlands, it is especially affected by fungi such as the coffee rust and fungicides are used to control it (INEC, 2007). Herbicides are mostly used in crops such as sugar cane, rice and pasture lands, insecticides are used in rice, sugar cane, coffee, beans, pineapple, vegetables and other fruits; insecticides and nematicides are mainly used in banana and pineapple plantations and fumigants in melon and strawberries (Castillo *et al*., 2000; Castillo and Ruepert, 2005; Bravo-Duran *et al.,* 2013).

Some pesticides belonging to different chemical families are classified as toxic and might cause acute and long-term adverse effects on the organisms living in the aquatic environment near cultivated areas (Mena *et al.,* 2012; Lushchak 2011; Tilton *et al*., 2006). Eighteen chemical groups have dominated the domestic market in Costa Rica during the last 32 years studied (Table 1); namely dithiocarbamates (28-39%), organophosphates (0.6-15%), phenoxiacetic acids (8-35%), phosphones (<11%), benzonitriles (<18%), brominated aliphatics (1-10%), morpholines (<4.8%), bipyridyls (2.5-10%), inorganics (0,1-5%), carbamates (0.2-2,6%), anilides (<7%), triazines, ureas and benzimidazoles (<2%), dinitroanilines (<1%), organochlorines (0.7-14%), chlorinated aliphatics (<0.6%) and conazoles (<1%). In 70's, the dithiocarbamates (38.7%) was followed by phenoxyacetic acids (35.4%), organochlorines (14%) and bipyridyls (9.8%). During the 80's, 90's and first decade of 21st century, dithiocarbamates (28-33%) were followed by organophosphates (11.6-14.8%), phosphones (1.5-10.8%), phenoxyacetic acids (7.8-11.2%), brominated aliphatics (1.1-10.4%) and benzonitriles (4.5-18.2%) respectively.

According to Ramírez *et al.* (2009) more than 70% of the total amount of pesticides imported from 1979-2006 was constituted in descending order by the following a.i.: mancozeb, 2,4-D, chlorothalonil, glyphosate, methyl bromide, terbufos, tridemorph, paraquat, propanil and ethoprophos. Pesticides such as DBCP and DDT were banned in 1988 (de la Cruz and Castillo, 2002) and are no more imported.

Fish are particularly sensitive to pesticides because they are able to absorb and retain dissolved xenobiotics present in the water, via active or passive transport through the gills (Walker, 2001). Dithiocarbamates are among the most important classes of fungicides currently used in the country for agricultural purposes. They have been found to be toxic to algae, crustaceans and fish. Some of them were found to induce embriotoxicity and teratogenicity in rainbow trout, at concentrations below μg l^{-1} (van Leeuwen *et al*., 1986; Tilton *et al*., 2006). Organophosphates and carbamates might inhibit the acetylcholinesterase (AChE) activity in fish brain and muscle. This has been observed in the brains of carps exposed to chlorfenvinphos, chlorpyrifos, diazinon, and carbofuran (Hai *et al*., 1997) as well as in native fish from Costa Rica exposed to ethoprophos, during field and laboratory experiments (Mena *et al.,* 2012). Pesticide may induce oxidative stress via several mechanisms (Lushchak, 2011). It has been observed that organophosphates (dichlorvos, methyl parathion, trichlorfon, fenthion, malathion and ethoprophos) and carbamates might induce oxidative stress on carp and catfish (Dembélé *et al*., 2000; Lushchak, 2011). Triazole fungicides, as well as other related imidazoles used for the protection of crops in the country might accumulate in the fish (Konwick *et al.,* 2006) and cause also oxidative stress (Lushchak, 2011; Toni *et al*., 2011). An increase in the oxidative stress was observed in fish exposed to simazine, a triazine herbicide (Stara *et al*., 2012), and to the insecticide deltamethrin, a pyrethroid (Ensibi *et al.,* 2012). The imidazolinone herbicides imazapic and imazethapyr also caused oxidative stress in carp fish (Moraes *et al*., 2011). Zebra fish embryos exhibited teratogenic effects such as bent spine, uninflected swim bladder and other malformations after exposure to triadimefon (1,2,4-triazole fungicide) (Shaoying *et al.,* 2010) and dichlorophenoxyacetic acid herbicide 2,4-D, and acetanilide herbicide butachlor were found to be genotoxic to catfish (Ateeq *et al*., 2002).

Considerable differences in sensitivity among species of algae to different agricultural pesticides have been found. Twenty-three chemicals from different chemical groups were tested with green algae, diatoms, cyanobacteria and duckweed. Nine of the pesticides tested (5 triazines) were highly toxic to algae; twelve pesticides including triazine inhibited growth of duckweed by more than 50%. Duckweed was the most sensitive organism, being equally affected by all pesticides causing algal toxicity, as well as being acutely affected by sulfonylurea herbicides. Diatoms and cyanobacteria showed sensitivity to glyphosate (Peterson *et al*., 1994).

General exposure indicators : Exposure indicators showed increase in the average amount of pesticides imported in connection with the extension of protected areas (5.9 kg a.i.ha^{-1} in 1980-1989 to 8.4 kg a.i. ha^{-1} in 2000-2009) and wetland areas (10.9 kg a.i. ha^{-1} in 1980-1989 to 24.3 kg a.i. ha^{-1} in 2000-2009) (Table 2). About 62% and 38% of the cultured land of Costa Rica occurs on the Pacific and Caribbean slope basins respectively (CGR, 2005; INEC, 2007; MAG, 2007; CNP, 2009a, b, c, d, e, 2010, 2011, 2012;

Table 1 : Percentage of pesticide a.i. imported by chemical groups. Costa Rica 1977-2009 in tons of a.i. – based on average for the period

	1977-1979	1980-1989	1990-1999	2000-2009
Dithiocarbamate	38.7	27.7	29.4	32.8
Organophosphate	0.64	11.6	14.8	12.4
Phosphone	0	1.5	6.1	10.8
Phenoxyacetic acid	35.4	11.2	7.8	9.4
Morpholine	0	1.4	4.4	4.8
Brominated aliphatic	1	1.1	10.4	4.6
Benzonitrile	0	18.2	4.9	4.5
Bipyridyl	9.8	3.7	2.3	2.5
Carbamate	0.19	2.2	2.6	2.4
Triazine	0	1.5	2.3	2.2
Inorganic	0.06	4.7	2.4	2
Benzimidazole	0	0.8	1.9	1.4
Urea	0	2.2	1.5	1.4
Dinitroanaline	0	1.3	1.2	1.3
Conazole	0	0.21	0.96	0.87
Organochlorine	14	1.2	0.8	0.7
Anilide	0	7	2.3	0.6
Chlorinated aliphatic	0	0	0.016	0.58
Total	99.8	97.5	96.1	95.3
Others[a]	0.2	2.5	3.9	4.7

Symbol %	>9.5		1 a 9.5		0.1 a 1		<0.1		0	

SEPSA, 2012). The pesticide load for the Pacific slope including pasture land was calculated at 4.7 kg a.i. ha^{-1} and for the Caribbean slope at 7.7 kg a.i. ha^{-1} for the first decade of the 21^{st} century. This might be an indication that important and valuable protected and coastal aquatic ecosystems are under chemical stress. Part of this a.i. might reach aquatic systems either dissolved in the water column or associated to suspended particles and expose and affect the fauna inhabiting those ecosystems.

Indicators of environmental fate and toxicity : Pesticides imported into the country from 1977 to 2009 and classified according to their water solubility, soil mobility, water-sediment and soil persistence (IRET, 1999; BCPC, 2003; Footprint, 2006) are shown in Fig. 2. Pesticides with high and low solubility showed an increase in the quantities imported; 54% of those currently imported had low solubility, whereas 38% had high solubility and were more easily carried away by runoff.

Pesticides with low mobility in soil are less susceptible to leaching and runoff. Currently most of the total quantities of pesticides imported into Costa Rica have low mobility; 26% are highly mobile in soil. The risk of impacts on biota increases with pesticide persistence, an increased quantity of pesticides imported is highly persistent in the water/sediment system, approximately 37% of current imports into Costa Rica. On the contrary, proportion of imports of pesticides with high soil persistence has decreased (Fig. 2).

The quantities of pesticides imported according to their toxicity to different biotic groups and for the different decades

Table 2 : Environmental general exposure indicators proposed. Costa Rica 1980-2009

Time period	1980-1989	1990-1999	2000-2009
Protected area (kg a.i. ha^{-1})	5.9	4.9	8.4
Wetland (kg a.i. ha^{-1})	10.9	17.9	24.3
Pacific slope (kg a.i. ha^{-1})	-	-	4.7
Caribbean slope (kg a.i. ha^{-1})	-	-	7.7

analyzed (Fig. 3) show that about 98% of imports are moderately to highly toxic to fish and crustaceans and 42 to 85% are moderately to highly toxic to algae. However, there is a small increase in the importation of pesticides that are moderately toxic to fish during the period 2000-2009. Regarding toxicity to amphibians and aquatic plants, there is no information available for 26% and 61% of the total quantity of pesticide imports. About 49% of the total imported during 2000-2009 is highly toxic to amphibians and 37% is moderately to highly toxic to aquatic plants (Fig. 3).

The 16 most imported pesticides during 2000-2009 (80% of imports) include namely mancozeb, glyphosate, 2,4-D, methyl bromide, tridemorph, chlorothalonil, terbufos, paraquat, metam sodium, ethoprophos, diazinon, diuron, pendimethaline, fosetyl-aluminium, sulphur, and ametryn (Table 3). This coincides with

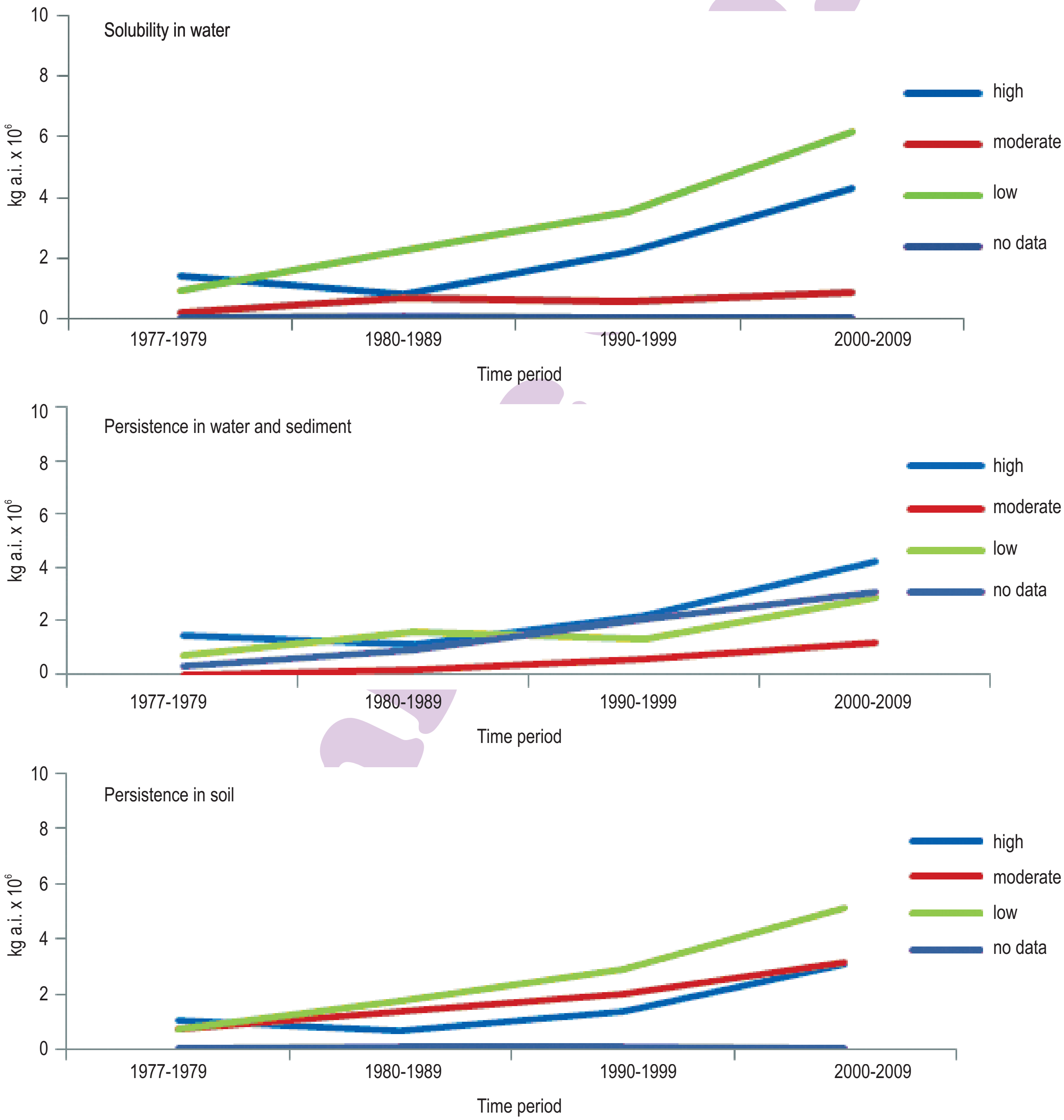

Fig. 2 : Millions of kilograms of pesticides imported with high, moderate and low water solubility, mobility in soil and environmental persistence

Table 3 : Most imported 2000-2009 pesticides a.i. and their toxicity to biota

Active ingredient	Kg a.i. x 10^6	%	Amphibia	Fish	Crustacea	Algae	Aquaticfern
Mancozeb	3.3	29	High	High	High	Moderate	No data
Glyphosate	1.2	40	Moderate	Moderate	Moderate	Low	Low
2,4-d	1	49	Moderate	Moderate	Moderate	Low	Moderate
Methylbromide	0.5	54	No data	High	High	Moderate	No data
Tridemorph	0.51	58	High	High	High	Moderate	No data
Chlorothalonil	0.51	63	High	High	High	Moderate	Low
Terbufos	0.38	66	No data	High	High	Moderate	No data
Paraquat	0.28	68	High	Moderate	High	High	High
Metam sodio	0.26	71	No data	High	High	Moderate	No data
Ethoprophos	0.25	73	No data	High	High	Low	No data
Diazinon	0.18	74	High	High	High	Moderate	No data
Diuron	0.15	76	Moderate	High	High	High	High
Pendimethalin	0.15	77	No data	High	High	High	High
Fosetyl	0.14	78	No data	Low	Low	Low	Low
Sulphur	0.13	80	Moderate	High	High	Moderate	No data
Ametryn	0.11	80	High	High	Moderate	High	High

Symbol:	High	Moderate	Low	No data

those reported for the 1979-2006 period (Ramírez *et al*., 2009). Thirteen of these are highly toxic to aquatic fauna. Paraquat, diuron, pendimethalin and ametryn have high toxicity for aquatic flora. These figures might be evidence that, if necessary precautions to reduce transfer and use of hazardous substances in the country are not taken, these substances will reach natural ecosystems and impair aquatic biota of great economic, aesthetic and recreational value.

Eight a.i. have been imported during all the period analyzed: aldicarb, 2,4-D, dicofol, endosulfan, mancozeb, maneb, metaldehyde, methyl bromide and paraquat. Six of them are classified as endocrine disruptors or as causing effects on reproduction (CEC, 2007; Mnif *et al*., 2011) and most of them are moderately and highly toxic to fishes, crustaceans, algae and amphibians. During 70's and 80's, compounds such as parathion, 2,4,5-T, organochlorines and lead arseniate were in use, most of them with high bioconcentration potential, environmental persistence and high toxicity to fish, crustaceans and amphibians (IRET, 1999; de la Cruz and Castillo, 2002).

The quantity of imported pesticides with endocrine disruption or reproduction effects (EDRE) according to literature (CEC, 2007; Mnif *et al*., 2011) has continuously increased since 1977; although the relative percentage has decreased over time. It goes from 62% in 1977-1779 to 47% in 2000-2009. The exposure of biota in their natural habitat to endocrine disrupting compounds or to compounds with reproduction effects might have long term consequences difficult to evaluate now. Four of the 16 most imported pesticides mentioned in Table 3, are classified as possible EDRE, including 2,4-D, mancozeb, paraquat and pendimethalin. Attention must be given to limit the import of EDRE pesticides into the country.

About 22% (90) of the 411 a.i.of pesticides imported into the country during the study period were analyzed in environmental samples. The country does not have a national monitoring system and the results reported are mainly from the efforts of research activities at the national universities, some in collaboration with governmental institutions. Some of the most imported pesticides have not been analyzed, such as mancozeb, maneb, glyphosate, 2,4-D and methy bromide. The presence of pesticides in aquatic ecosystems reflects both the changes in use (Table 1) and the improvement in the analytical capacity of the country, which has included new a.i. in the chemical analysis (CGR, 2005, 2013; Alfaro Monge, 2011; Castillo *et al.,* 2012).

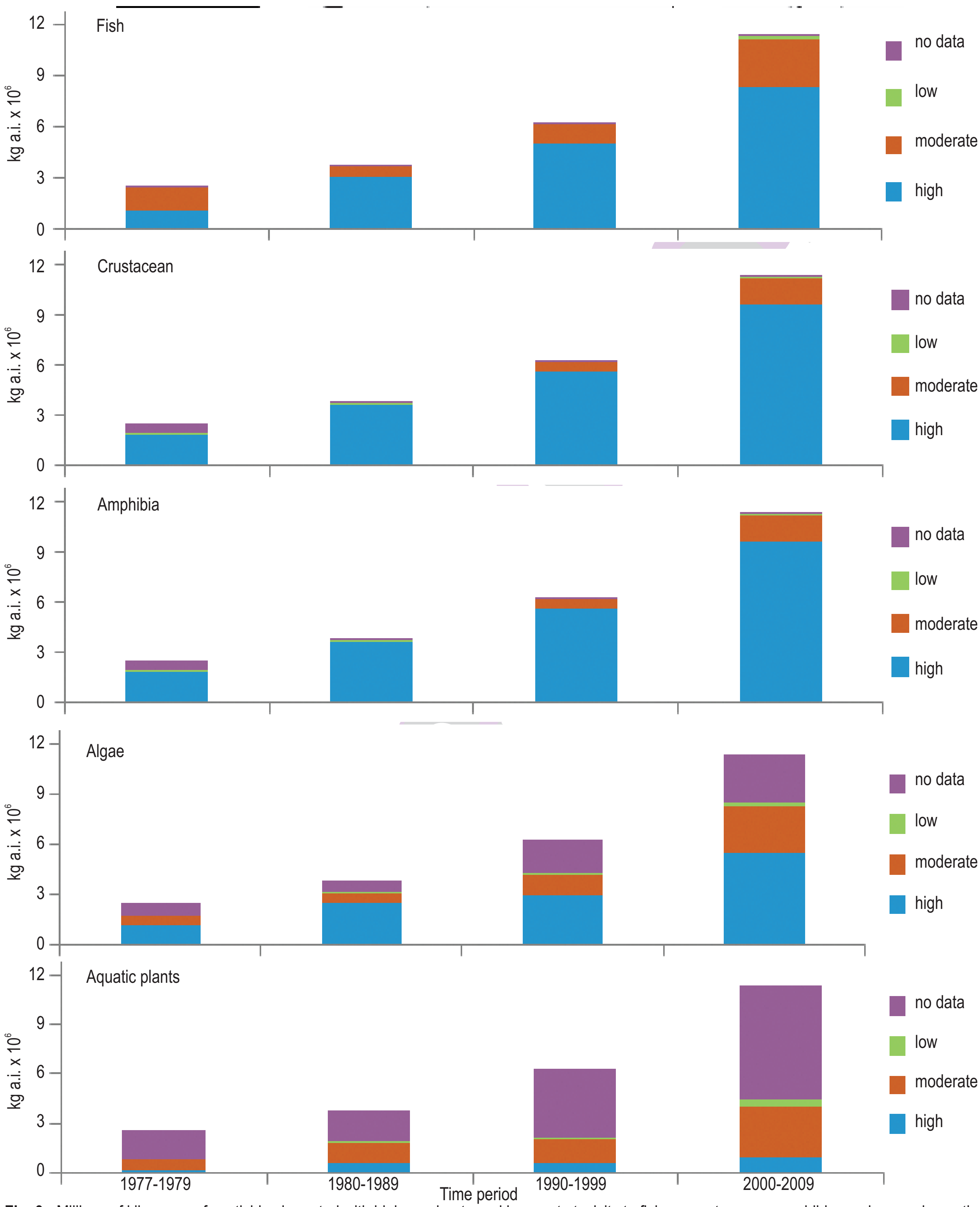

Fig. 3 : Millions of kilograms of pesticides imported with high, moderate and low acute toxicity to fishes, crustaceans. amphibians, algae and aquatic plants. Amphibian EC_{50} (96 hr) for different species, Fish EC_{50} (96 hr) for *Daniorerio, Lepomismacrochirus*, *Oncorhynchus mykiss, Oryzias latipes*. and *Salmotrutta*. Crustaceans EC_{50} (48 hr) for *Ceriodaphnia dubia* and *Daphnia magna*, Algae and aquatic plants EC_{50} (72 hr) for *Pseudokirchneriella subcapitata*; *Selenastrum capricornutum*, *Anabaena flos-aquae*, different species of *Scenedesmus* and *Lemna*. In cases that the mentioned species and experimental time were not found, other species and experimental times are used

Table 4 : Historical pesticide reports in research studies, monitoring programs and environmental accidents. Costa Rica, 1880 - 2009.

Biocide action/ Chemical group	Active ingredient	Decade reported	Reports linked to accidents	Reference
Fungicides				
Anilide	flutolanil[a], metalaxyl	1990-2009	formulation and transport	Fournier *et al.,* 2010; Echeverría-Sáenz *et al.*, 2012
Benzimidazole	carbendazim, thiabendazole[a]	1990-2009	aquatic fauna killings	Castillo *et al.*, 2000; Echeverría-Sáenz *et al.*, 2012
Benzonitrile	chlorothalonil	1980-2009	aquatic fauna killings	Castillo *et al.*, 2000; de la Cruz *et al.*, 2004; Echeverría-Sáenz *et al.*, 2012
Chlorophenyl	tecnazene	2000-2009		Fournier *et al.,* 2010
Conazole	difenoconazole[a,b], epoxiconazole[a], propiconazole[a,b], tebuconazole	1990-2009	aquatic fauna killings; transport	de la Cruz *et al.*, 2004;CGR,2013; Polidoro, 2007; Fournier *et al.,* 2010; de la Cruz *et al.,* 2012; Echeverría-Sáenz *et al.,* 2012
Ditiocarbamate	etylenethiourea[e], mancozeb[c]	2000-2009	aerialspraying; transport	de la Cruz *et al.*, 2004;CGR, 2013
Imidazole	imazalil, prochloraz	1990-2009		Castillo *et al.,* 2000; Alfaro Monge, 2011
Morpholine	tridemorph	2000-2009		de la Cruz *et al.*, 2004
Organochlorine	hexachlorobenzene, quintozene	2000-2009		de la Cruz *et al.*, 2004; Fournier *et al.,* 2010
Organophosphate	edifenphostolclofos-methyl	1990-1999		de la Cruz *et al.*, 2004; CGR, 2005; Fournier *et al.,* 2010
Oxazole	vinclozolin	2000-2009		
Phenylurea	pencycuron	2000-2009		Fournier *et al.,* 2010
Pyrimidine	cyprodinil	2000-2009		Echeverría-Sáenz *et al.*, 2012
Strobilurin	pyraclostrobin	2000-2009		Echeverría-Sáenz *et al.*, 2012
Triazole	bitertanol, myclobutanil, triadimefon, triadimenol	2000-2009		Alfaro Monge, 2011; Echeverría-Sáenz *et al.*, 2012; CGR, 2013
Herbicide				
Anilide	propanil	1990-1999		de la Cruz *et al.*, 2004; CGR, 2005
Benzothiazinone	bentazon	1990-1999		de la Cruz *et al.*, 2004; CGR, 2005
Bipyridylium	paraquat	1980-1989		de la Cruz *et al.*, 2004; CGR, 2005
Chloroacetamide	alachlor, butachlor	2000-2009		de la Cruz *et al.,* 2012; Echeverría-Sáenz *et al.,* 2012
Dinitroaniline	pendimethalin	1990-1999	fish and shrimp killing	de la Cruz *et al.*, 2004; CGR, 2005
Oxidiazole	oxadiazon	1990-1999		de la Cruz *et al.*, 2004; CGR, 2005
Phenylurea	diuron	1990-2009	aquatic fauna killings	CGR, 2005; Alfaro Monge, 2011; Echeverría-Sáenz *et al.*, 2012
Quinoline	quinclorac	1990-1999		de la Cruz *et al.*, 2004
Triazine	ametryn[a], atrazine, terbutryn	1990-2009	aquatic fauna killings	Castillo *et al.,* 2000; Echeverría-Sáenz *et al.*, 2012
Triazinone	hexazinone	1990-2009		CGR, 2005; de la Cruz *et al.,* 2012; Echeverría-Sáenz *et al.*, 2012
Uracil	bromacil	1990-2009	aquatic fauna killings	CGR, 2005, 2013 ; Echeverría-Sáenz *et al.*, 2012
Urea	linuron	2000-2009		Fournier *et al.,* 2010
Insecticides, acaricides, nematicides				
Carbamate	carbaryl, carbofuran[a,c], oxamyl, pirimicarb	1990-2009	leakage vapors from containers; mammals and bees killings	Castillo *et al.,* 2000; CGR, 2005; Alfaro Monge, 2011; Echeverría-Sáenz *et al.*, 2012
Inorganic	aluminiumphosphide[c]		explosion; improper disposal of waste	de la Cruz *et al.*, 2004; CGR, 2005
Organochlorine	aldrin[d], dieldrin[a,d], endrin[d], chlordane[d],DDT[d], heptachlore[d], dechlorane[d], endosulfan, lindane[d]	1980-2009	shrimp killing	de la Cruz *et al.,* 2004; CGR, 2005; Pérez-Maldonado *et al.*, 2010; Alfaro Monge, 2011; Fournier *et al.,* 2010
Organophosphate	acephate, cadusafos, chlorfenvinphos, chlorpyrifos[a,c], diazinon[a,b], dichlorvos[a], dimethoate, ethion, ethoprophos[a,b], fenamiphos[a,b], fenthion, phorate, methamidophos, parathionmethyl, pirimiphos, terbufos[a,b,c], triazophos[a,b]	1980-2009	aquatic fauna killings; mammals and shrimp killing, formulation and transport	Castillo *et al.,* 2000; de la Cruz *et al.*, 2004, CGR, 2005, 2013; Polidoro, 2007; Fournier *et al.,* 2010; Alfaro Monge, 2011; de la Cruz *et al.*, 2012; Echeverría-Sáenz *et al.,* 2012
Phenylpyrazole	fipronil[a]	2000-2009	bees killing	de la Cruz *et al.*, 2004 ; CGR, 2005
Pyrethroid	cypermethrin, permethrin	1990-2009		de la Cruz *et al.,* 2004; Fournier *et al.,* 2010; de la Cruz *et al.,* 2012

[a]reported in episodes of massive fauna killings; [b]more frequently reported in episodes of massive aquatic fauna killings; [c]reported in accidents that do not cause pollution of water bodies; [d]reported in biota and sediment; [e]reported in accidents that cause pollution of water bodies

Before 80's, pesticide pollution of environmental ecosystems was hardly registered besides DDT (Castillo *et al*., 1997). Seventy-three a.i. from thirty-one chemical families and different biocide action have been reported in samples from aquatic systems since 1980 to 2009 (Table 4). The most reported families during the eighties were organochlorines (aldrin, chlordane, DDT, dechlorane, heptachlore and lindane), organophosphates (chlorpyrifos and parathion), bipyridyls (paraquat), and benzonitriles (chlorothalonil), most associated with banana, rice and sugar cane plantation (Castillo *et al*., 1997), although some of the DDT metabolites might be related to the malaria control programs (Pérez-Maldonado *et al*., 2010).

The presence of individual pesticides in the aquatic environment and the quantities of these that were imported agree quite well over time (Table 1 and Table 4); especially at the chemical family level. Fifteen of the 31 chemical families constituting 95.3 to 99.8% of the imports have been reported. Of the 100 most imported pesticides a.i. during this time, 45 a.i. have been detected, 40 of them have high acute toxicity to fish or crustaceans and 12 are suspected EDRE and have high acute toxicity to fish or crustaceans, among them bromacil, carbaryl, carbofuran, endosulfan, mancozeb, and terbutryn (IRET, 1999; Footprint, 2006; CEC, 2007; Mnif *et al*., 2011).The fungicide chlorothalonil and the insecticides chlorpyrifos, and DDT compounds were reported during the three decades period (de la Cruz and Castillo, 1997; CGR, 2005; Perez-Maldonado *et al*., 2010).

From time to time, accidents with pesticides that affect aquatic resources are reported by different communities and governmental offices (CGR, 2005, 2013; de la Cruz *et al.,* 2010; Castillo *et al.,* 2012). Without being exhaustive, of the 29 accidents analyzed (Table 4), five (17%) were related to transportation and/or formulation of pesticides, which included anilides, dithiocarbamates (mancozeb), conazoles (propiconazole), and organophosphates (chlorpyrifos and terbufos). Three (10%) were related to leakage from containers (carbofuran), improper waste disposal and explosions (aluminum-phosphide). Twenty one (72%) of these accidents involved biota mortalities, 3 (10%) include bees (carbofuran and fipronil) and mammals (carbofuran and dichlorvos) and 19 (65%) aquatic fauna, including different species of fish, shrimp and other crustaceans, birds and reptiles. The a.i. detected in water or biota during these environmental accidents were ametryn, bromacil, chlorothalonil, chlorpyrifos, diazinon, dichlorvos, dieldrin, difenoconazole, diuron, epoxiconazole, ethoprophos, fenamiphos, pendimethaline, propiconazole, terbufos, thiabendazole and triazophos. Carbofuran, chlorpyrifos, difenoconazole, fenamiphos, propiconazole, terbufos and triazophos were involved in more than one accident.

Insecticides were the most common biocide group reported in the Northern Pacific (mainly organochlorines) and Caribbean (chlorpyrifos) slopes of the country during 80,s (Castillo *et al.,* 1997). Most die-offs remained unknown at that time. From 1990-1999, evidence of the contamination of aquatic systems increased. Insecticides-nematicides, herbicides and fungicides were associated with aquatic biota deaths in different regions of the country during this decade (de la Cruz *et al.,* 2004). In 2000-2009, the number of reports of contamination of aquatic systems with fungicides increased in the Northern Pacific, the Central Valley and the Southern part of Caribbean lowland of the country. The reason for this might be that aquatic fauna mortalities occurred and there was an increase in the number of studies conducted in areas where vegetables and ornamental plants were produced in the Central Valley (CGR, 2005; Fournier *et al*., 2010). Other reasons might be land expansion in the production of pineapple and plantain, the occurrence of aquatic mortality events and the conducting of more studies on the Northern, Central and Southern part of the Caribbean slope during this decade (Polidoro, 2007; Alfaro Monge, 2011; SEPSA, 2011; Echeverría-Sáenz *et al.,* 2012; CGR, 2013).

Insecticides and nematicides seem to be the group linked to more environmental problems. Compounds from this biocide action group were reported (endosulfan, acephate, chlorpyriphos, dimethoate, fenthion, methamidophos, triazofos, cypermethrin, permethrin) in the Northern and Southern Pacific, in the Northern and Southern Caribbean (carbaryl, carbofuran, DDT´s, chlorpyrifos, diazinon, ethoprophos, fenamiphos, terbufos), and in the Central Valley (carbofuran, endosulfan, dieldrin, acephate, clorpyrifos, diazinon, dimetoate, methyl pyrimiphos). The insecticides and nematicides (dieldrin, diazinon, dichlorvos, ethoprophos, fenamiphos, terbufos, triazophos) which, with the exception of dieldrin have higher toxicity to aquatic organisms and moderated to higher solubility in water and mobility in soil, seem to be linked to more accidents, even though some fungicides (chlorothalonil, difenoconazole, propiconazole), and herbicides (ametryn, bromacil, diuron) were also associated to fauna die-offs, especially in the Caribbean slope (CGR, 2005, 2013; Polidoro, 2007; de la Cruz *et al.,* 2010;Castillo *et al.,* 2012). The geographical occurrence of pesticides is relation to their use in different cultures; herbicides are most used in the production of rice, sugar cane, pineapple and pastures located mainly on the Central and Northern Pacific side of the country. The fungicides, insecticides-nematicides are heavily used in banana production on the Caribbean coast; but also in pineapple and plantains grown in the same region. Pineapple is as well planted at the Northern and Southern Pacific slope. Inseticides-nematicides and fungicides are also used in vegetables, ornamental and flower production, occurring mainly in the Central Valley of the country (Castillo *et al.,* 2000; Castillo and Ruepert, 2005; Fournier *et al.,* 2010; Bravo-Durán *et al.,* 2013).

Import data of pesticides can be used to construct indicators of environmental hazards, such as environmental exposure indicators and environmental hazard indicators. Pesticide imports data seem to be good indicators of environmental pollution occurring in the country; even with limitations, it is clear that the most imported compounds are related to water pollution and die-offs. We recommend using data of pesticide imports, toxicity to different organisms and their use in agriculture as a source of information to document trends in time, evaluate current risk and use as a working tool to create awareness in different stakeholder groups.

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