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Herbicides and the Aquatic Environment

Rafael Grossi Botelho[1], João Pedro Cury[2],
Valdemar Luiz Tornisielo[1] and José Barbosa dos Santos[2]
[1]*Laboratório de Ecotoxicologia, Centro de Energia Nuclear na Agricultura,*
Universidade de São Paulo – CENA/USP, Piracicaba, SP,
[2]*Universidade Federal dos Vales do Jequitinhonha e Mucuri – UFVJM,*
Diamantina, MG,
Brasil

1. Introduction

The quality of water resources is perhaps currently the most discussed topic when it comes to environmental preservation, since aquatic ecosystems have been suffering changes worldwide in most cases irreversible. Such changes are often associated with human activities such as deforestation, release of industrial and domestic effluents, and even the use of pesticides in agricultural fields, which is one of sources that most contributes to the fall of quality of water resources.

Pesticides are important to the agricultural system. However, it is crucial that they be used with responsibility in order to preserve the quality of the final product and the natural resources that support the production, especially soil and water (Oliveira Junior & Regitano, 2009).

Pesticides are products whose function is to eliminate organisms causing damage to agricultural crops thus ensuring high productivity. Their classification is made according to target species (insecticides, herbicides, fungicides, acaricides, nematicides, etc..) (Alves-Silva & Oliveira, 2003, Sanches et al., 2003), patter of use (defoliants, repellents, and others) (Alves-Silva & Oliveira, 2003; Laws, 1993; Sanches et al., 2003), mechamisns of action (acetylcholinesterase inhibitor, anticoagulants, etc) (Alves-Silva & Oliveira, 2003) or chemical structure (pyretroids, organophosphates, carbamates, etc) (Alves-Silva & Oliveira, 2003; Laws, 1993).

Although these molecules, when applied, have target organisms as their final destination, according to Macedo (2002) 99% of applied pesticides go into the air, water and soil, ie, only 1% reaches its target. This finding is quite disturbing as the world population grows; it means that the use of pesticides will increase (thus increasing food productivity) and natural resources will remain under intense threat from these molecules.

2. Pesticides market in Brazil

Pesticides started to become popular in the middle of the Second World War, when the world discovered the DDT. The ease of accesses of this product and its low cost made it to be extremely used before the discovery of its negative effects. The great successes of this compound in pest control made new products being produced strengthening the agrochemical industry today (Bull & Hathaway, 1986).

Currently, according to the data from National Health Surveillance Agency Anvisa (2010), Brazil is the largest consumer of pesticides in the world and has the largest market for these products with 107 companies authorized to register this compounds, responding for 16% of the world market. According to the sales in Brasil, only in 2010, the industry negotied 342,590 tons of active ingredients and its clear that this number is increasing in recent years (Figure 1).

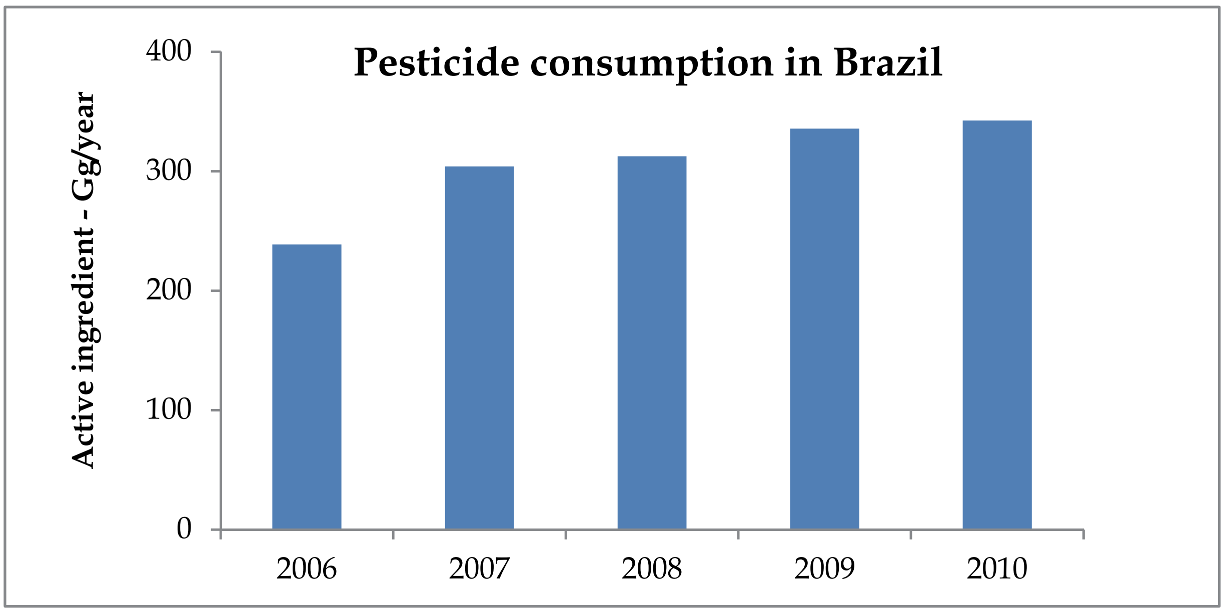

Fig. 1. Pesticide consumption in Brazil, in gig grams of active ingredient, in the period of 2006 to 2010.

Among the classes of pesticides, herbicides are those that make up most marketed worldwide (Moura et al., 2008). These molecules are chemical substances that act by killing or suppressing the development of weeds that impair the productivity of crops of commercial interest (Roman et al., 2007). According to the National Association of Products Industry for Agricultural Defense, only Brazil, one of the leading countries in agriculture with the use of pesticides, 725 000 tons of formulated products were sold in 2009 and herbicides are the main class with 59% (429,693 tons), followed by insecticides and acaricides with 21% (150,189 tons), fungicides 12% (89,889 tons) and others 8% (55,806 tons) (Figure 2) (Sindag, 2010). The problem is that many of these substances are likely to contaminate water resources due to characteristics such as high shift-potential in the soil profile (leaching), high persistence in soil, low to moderate water solubility and moderate adsorption to organic matter present in soil colloids (Almeida et al., 2006). Once present in aquatic environments, these molecules can be absorbed by organisms, and since they live in continual interaction with each other in a complex system of food chains, contamination can result in a drastic imbalance in the ecosystem.

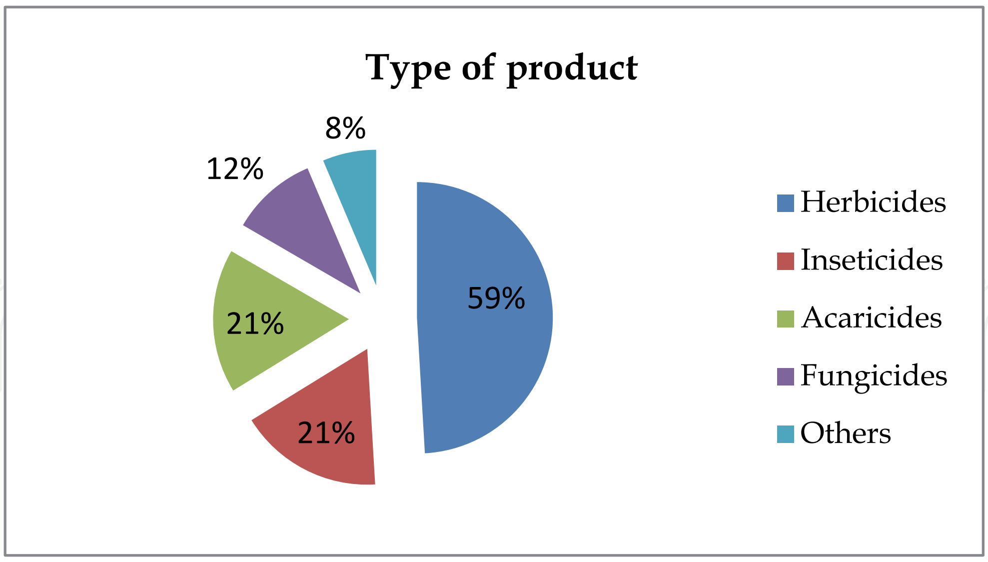

Fig. 2. Pesticide consumption in Brazil by type of product in gig grams of active ingredient, in 2009.

3. Herbicides: leaching and residual effects

Pesticides have an important role in modern agriculture, with new formulations being introduced regularly. Among these, the chlorinated acid-phenoxy herbicides such as 2,4-D and MCPA are commonly used to control weeds in wheat, rice, corn, sugar cane and pasture. The massive use of pesticides has resulted in their presence in the environment in the form of sub-lethal pollution, and problems such as contamination of surface and groundwater have been observed (Legrouri et al., 2005). The concern of environmental protection agencies with the presence of these molecules in soils, water and air has increased greatly in recent times, particularly as it relates to protecting the quality of drinking water (Lagaly, 2001). Due to the commercial importance of agriculture in world and pesticides industry, probably the extensive use of these substances will last for a long period. Therefore, the most feasible would be the rational use of these products through a strict control of its use and handling, aiming, mainly, avoid over dosing, application in undue places and improper washing of packaging and application equipment that many times are held on the banks of rivers (Trovo et al., 2005). Thus, contamination of soils and water due to the extensive use of pesticides over large areas in modern agriculture is a problem that requires research to its remediation (Ignatius et al., 2001).

Considering the transport processes in the environment with which herbicides are related after applied to agricultural areas, leaching and runoff deserve some attention. Surface runoff favors surface water contamination, since the molecule is carried and adsorbed to eroded soil particles or in solution. On the other hand, leaching results in contamination of groundwater, and in this case, chemical substances are carried in solution with the water that feeds the ground water (Spadotto, 2002). Only a low percentage of herbicides in soil are used bioactivity, ie, the remainder is distributed in the environment. This loss of product requires a high amount application, increasing the damage to the environment and consequently to health (Dich et al., 1997).

The knowledge of sorption-desorption processes is of great importance, once determining the amount of product present in the soil is possible to control other processes that can affect the dynamics of these molecules in the soil. If the degree of sorption of a pesticide increases, this compound concentration in water and air decreases. Consequently, the speed of concentration-dependent processes such as volatilization, bioavailability, and vertical movement of pesticides through the soil profile also decrease, thus reducing the risk of contamination of surface and groundwater (Cox et al., 1999).

The aquatic environment has become extremely vulnerable to contamination, Herbicides with high leaching potential, ie, those with low capacity to be retained in the soil are potentially more damaging in this environment by being subject to loading by the underground water flow and deposit with final residual effect on aquatic community. The water pollution is still of concern since often agricultural fields are near lakes, streams and rivers potentiating this environment exposure (Moore et al., 2001) to soluble herbicides. Depend on the physical and chemical characteristics, the residue in water, can bind to the material in suspension, accumulate in the sediment or can be absorbed by aquatic organisms. They can be trasported through the aquatic system by diffusion of water in streans or in the bodies of organisms. Some products may also return to the atmosphere through volatilization. Thus, it is evident that there is a continuous interaction between pesticides, sediment and water, affected by water movement, turbulence and temperature (Nimmo, 1995). This interaction can result in a longer exposure of aquatic organisms to toxic compounds.

Solubility in water is defined as the maximum amount of pure molecule that can be dissolved in water (Lavorenti et al., 2003), being considered the most important physical property related to the transport and fate of organic molecules in aquatic systems, such as herbicides, and also one of determinants of soil sorption coefficient. Thus, herbicides with high solubility have a tendency to be less sorbed to soil colloids (Lavorenti et al., 2003). Therefore, sorption to soil and water solubility becomes important parameters to predict the herbicide trend to move horizontally or vertically in the ground (Extoxnet, 1998).

Another measure of leaching potential for a herbicide is the n-octanol-water partition coefficient (Kow), which measures the hydrophobic or hydrophilic character of a molecule. The Kow is defined as the ratio of the solubility of a compound in octanol (a non-polar solvent) to its solubility in water (a polar solvent). The higher to Kow, the more non-polar the compound (U.S.E.P.A, 2009). In environmental studies, this parameter is also correlated with water solubility, soil sorption coefficient and sediments and bioconcentration in aquatic organisms (Lyman et al., 1990; Sablji et al., 1995; Ran et al., 2002). Herbicides with high log Kow values (> 4.0) or lipophilic tend to accumulate in lipid material, for example, soil organic matter and, consequently, present low mobility (Lavorenti et al., 2003). On the other hand, hydrophilic herbicides (log Kow <1.0) are considered more soluble in water and consequently will present low sorption (Lavorenti et al., 2003) and greater potential for damage to the aquatic community.

Several studies on environmental contamination by pesticides are reported in the literature (Jacomini et al., 2006; Henares et al., 2008). Herbicides of the triazine group, which includes ametryne prometryne, atrazine, simazine, among others, are used worldwide and often detected in samples of soil and water, having high mobility in the environment, lasting years in the ground, water and organisms (Costa Queiroz et al., 1999; Kolpin et al., 2002; Jacomini et al., 2006).

The herbicide atrazine is one of the most widely used herbicides in Brazil, and its use is registred for sorghum, corn, sugarcane and other crops (Rodrigues & Almeida, 2005). Due to its wide use, high persistence and moderate mobility in soil, this herbicide has been detected in several compartments of the environment, especially in surface waters (Buser, 1990) and groundwater (Dörfler et al., 1997). Highest losses of atrazine have been correlated with the first rain or irrigation after its application (Belamie & Gouy, 1992; Patty et al., 1997; Correia et al., 2007). The shorter the time between herbicide application and irrigation or rainfall, the higher the herbicide transport by leaching.

Several authors highlight the problem of contamination of surface and subsurface waters by atrazine (Buser, 1990; Pick et al., 1992; Dörfler et al., 1997; Yassir et al., 1999) so that its use was banned in European Union in 2003 (Sass & Colangelo, 2006). Jablonowski et al. (2009), conducted studies on the persistence of atrazine for more than 20 years after application. Concentrations were detected on average four times higher in the subsurface compared to surface in soil, indicating high risk of contamination of groundwater, even past the experimental period. Armas et al. (2007) found concentrations of atrazine in surface waters of Corumbataí river (São Paulo, Brazil) above the level permitted by Brazilian law. In the United States, atrazine was found at high incidence in surface water and groundwater; research included 178 streams and over 2700 wells (Kolpin et al., 2002). In Australia, atrazine and its metabolites were also detected in low concentrations in groundwater/surface water in several states (Ahmad et al., 2001).

The herbicide clomazone also has high water solubility and persistence in soil and can reach, under aerobic conditions, more than 270 days (California, 2003; Senseman, 2007). After its application on the soil surface, the product may leach into the deeper layers, presenting a potential risk of groundwater contamination and, consequently, watercourses contamination as well (Santos et al., 2008). The clomazone fate and behavior is influenced by organic matter and texture (Loux & Slife, 1989) with edaphic half-life ranging between 5 and 117 days depending on the type of soil and environmental conditions (Curran et al., 1992; Mervosh et al., 1995; Kirksey et al., 1996). Senseman (2007) reports that clomazone persistence is lower in sandy soils than in clay soils.

Monitoring conducted for three years (2000-2003) in two Brazilian rivers (Vacacaí and Vacacaí-Mirim) in Rio Grande do Sul (Brazilian state) detected the presence of the herbicides clomazone, in higher concentration, quinclorac and propanil (Marchesan et al., 2007).

Santos et al. (2008) in a study conducted in shallow waters around the rice-growing areas in Rio Grande do Sul showed that 90% of samples contained clomazone residues. Bortoluzzi et al. (2006) found the presence of this product in surface water adjacent to tobacco crops. The presence of clomazone in water and soil samples have been reported in the literature not only in Brazil but also in other countries such as Spain (Nevado et al., 2007), Italy (Palmisano & Zambonin, 2000), China (Li et al., 2010), Uruguay (Carlomagno et al., 2010) and United States (Gunasekara et al., 2009).

Another herbicide that has concerned researchers is ametryne, whose half-life is between 50 and 120 days in soil and about 200 days in natural water with pH 7.0 and temperature ranging from 5 to 29 °C. This period has been reported as dangerous to the environment by the power of contamination of soils and surface water/groundwater by this product. (Cumming et al., 2002; Laabs et al., 2002; Armas, 2006). Ametryne also has potential to contaminate aquatic environments, once in addition to being transported by runoff, this molecule can undergo leaching. Ametryne residues have also been found in surface waters of Brazil (Armas et al., 2007) although Brazilian law has not yet set a permissible limit in surface waters.

Within the broad class of herbicides, there is no doubt that the most commercialized worldwide is glyphosate. Its occurrence in groundwater was cited only once, in Texas, USA, reported by Hallberg (1989) - under review presented by Amarante Junior et al. (2002) - but the concentration measured was not specified. The direct application as herbicide in surface water to eliminate aquatic plants may be responsible for the presence of glyphosate in surface water.

Due to the rapid adsorption to soil, glyphosate is not readily leached, being unlikely the groundwater contamination. On rare occasions, this herbicide has been detected in water samples, but in general, this occurs due to the difficulty of separating the compounds and also by not being considered a serious water contaminant.

In the case of water pollution, glyphosate can be adsorbed by sediments being carried by them. This interaction is normally fast and occurs within 14 days resulting in much slower natural decay process. The Environmental Protection Agency of the United States (USEPA) sets limits of 700 µg/L glyphosate in drinking water as a “health advisory limit”. However, across Europe is established the limit of 0.1 mg/L as "maximum allowable concentration" for pesticides in drinking water as individual substances, since total concentration does not exceed 0.5 mg/L (IAEAC, 1994). Due to its broad-spectrum herbicide properties, ie being non-selective, systemic and low toxicity to animals, as discussed, it became one of the most used in the world, increasing the need for implementation of monitoring programs.

Various processes for water treatment have been investigated regarding their efficiency in removal of certain herbicides present in fresh water samples. Among them, the anaerobic degradation, electrochemical destruction by photo-Fenton reactions, adsorption on activated carbon, adsorption on clays saturated with inorganic or organic cations and the sorption of anionic molecules in lamellar double hydroxides (HDLs) through the processes of anion exchange or merge, among others might be cited.

Atrazine degradation by anaerobic microorganisms was studied by Ghosh & Philip (2004). Authors demonstrated that the degradation of this molecule is dependent on the amount of product in the effluent and the high organic content in the effluent reduces its rate of degradation.

Based on the properties of clays and clay minerals, several authors studied the removal of herbicides present in water, such as phenoxyacetic acid (Yurdakoc & Akcay, 2000), 2,4-D (Hermosín & Cornejo, 1992), prometrine (Socias-Viciana et al., 1998), dicamba (Carrizosa et al., 2001), linuron, atrazine, acephate, diazinon (Villa et al., 1999).

4. Ecotoxicology

The pesticide toxicity is quite complex and overall the goal is to determine what concentration in a particular product is toxic to an organism. The manifestation of a toxic effect resulting from a chemical substance may occur at a point distant from where was found the entry in the medium, since when it reaches a water environment for example, the pollutant can be transported by droplets or particles in suspension thorugh long distances (Pedrozo & Chasin, 2003).

Pollutant toxicity can be expressed by the effective dose or effective concentration (EC_{50} or ED_{50}) which is the amount of a substance affecting half of one group of organisms. Exposure effects to organisms vary according to the product's physico-chemical properties (solubility, chemical reactivity, stability, particle size, etc.) route of exposure (oral, inhalation, dermal), duration and frequency of exposure , species tested (there are differences in susceptibility among species and the type of effect on each one, differences in the effects on individuals of different sex and age, young and elderly are more sensitive than adults), among others (Chasin & Azevedo, 2003). Considering the possibility of contamination in aquatic environments and the need for using herbicides in order to increase agricultural productivity, scientists around the world are working to learn, alert and minimize the effect of these substances in organisms living in these environments. This concern led to the creation of Ecotoxicology, which according to the french toxicologist René Truhaut is the science that studies the effects of natural or synthetic substances on living organisms, populations and communities, animals or plants, terrestrial or aquatic, that make up the biosphere, thus including the interaction of substances with the environment in which organisms live in an integrated context (Plaa, 1982; Niederlehner & Cairns, 1995).

Toxicity tests are used to know the effects of substances in organisms. These represent an important tool in ecotoxicology enabling to determine the toxic effect or not in a particular substance. In the 80's, environmental agencies around the world especially in the United States and Europe began to develop standardized protocols for toxicity test using aquatic organisms (Usepa, 1996; Oecd, 1984-2004). In 1984, the Usepa established the use of organisms for monitoring water quality (USEPA, 1984). Concomitantly, the Organization for Economic Cooperation and Development (OECD) launched a series of test protocols for toxicity to aquatic organisms, including algae, fish and microcrustaceans in Europe. In Brazil, the first initiative to do a focused approach to the subject was in 1975. After this year, other methodologies using groups of organisms have emerged, highlighting algae (Abnt, 1992; Cetesb, 1994), microcrustaceans (Abnt, 1993, 2005; Cetesb, 1994) and fish (Cetesb, 1990).

These tests are used as mechanisms for understanding the effects of anthropogenic impacts on living organisms which act as representative organisms (Campagna, 2005). Toxicity tests allow assessing the environmental contamination by various pollution sources such as agricultural, industrial and domestic waste, chemical products and medicines in general (Marschner, 1999; Lombardi, 2004) and even also detecting the ability of a toxic agent or a mixture to produce deleterious effects showing the extent to which substances are harmful, how and where effects are manifested (Magalhaes & Filho, 2008). They even provide information about the potential danger of a toxic substance to aquatic organisms such as mortality, carcinogenesis, mutagenesis, teratogenesis, behavioral disorders, cumulative physiological, antagonistic and synergistic effects (Baudo, 1987).

The toxicity depends on the susceptibility of the organisms to a particular chemical compound. Different species have different sensitivities according to their feeding habits, behavior, development, physiology and others (Silva & Santos, 2007). Young individuals are usually more susceptible to chemicals than adults, probably due to the diference in degree of development or detoxification mechanisms (Silva & Santos, 2007). Stressed organisms due to of previous exposure to other toxicants may be more sensitive (Rand & Petrocelli, 1985), a commom scenario in the environment .

Toxicity tests are divided into acute and chronic. The acute test aims to assess the effects on organisms to a short period of exposure, whose goal is determining the concentration of a test substance that produces deleterious effects under controlled conditions. For fish, the observed effect is lethality, from which is determined the toxic agent concentration that causes 50% mortality (LC_{50}). For microcrustaceans there is no mobility from which is calculated the average estimate concentration (EC_{50}) that causes 50% immobility (Rand & Petrocelli, 1985). There are also chronic toxicity tests whose organisms are continually exposed to toxic substances for a significant period of time of their life cycle that can vary from half to two thirds of the cycle (Rand & Petrocelli, 1985). These tests assess sublethal effects such as changes in growth and reproduction, changes in behavior (difficulty in movement, increased frequency of opening of the operculum), physiology, biochemistry and tissue changes (Laws, 1993; Adams, 1995). Chronic toxicity tests directly depend on the results of acute toxicity tests, since sublethal concentrations are calculated from the LC_{50} and EC_{50}. For the choice of test organism are often use the following selection criteria: abundance and availability; significant ecological representation within biocenoses; species cosmopolitanism, knowledge of its biology, physiology and dietary habits; genetic stability and uniformity of its populations; low seasonality index, constant and accurate sensitivity; commercial importance; ease of cultivation in the laboratory and, if possible, species should be native for better representation of ecosystems (Rand & Petrocelli, 1995).

Since Ecotoxicology was created several studies have been made always aiming to evaluate the toxicity of a substance for a particular test organism. For example, Botelho et al. (2009) studied the toxicity of various herbicides for tilapia (*Oreochromis niloticus*) including atrazine, paraquat and some mixtures as alaclhor + atrazine, diuron + MSMA and 2,4D + picloram. The LC_{50} (96 hours) was 5.02 mg.L^{-1} for atrazine. These authors also reported weight loss of organisms at 2.5 and 5.0 mg.L^{-1}. In relation to the other products, after 48 hours of exposure, the misture alaclhor + atrazine was the only one that caused 100% of mortality to the organisms. Other studies involving atrazine showed the following LC_{50} values (96 hours): 18.8 mg.L^{-1} for the fish *Cyprinus carpio* (Neskovic et al., 1993), 10.2 mgL^{-1} for *Rhamdia quelen* (Kreutz et al., 2008) and 42.38 mgL^{-1} to *Channa punctatus* (Nwani et al., 2010). In toxicity studies, sensitivity of organisms can vary even if used the same product, as shown in the aforementioned studies with atrazine.

Several other studies involving atrazine has been performed, highlighting Hayes et al. (2002a), which showed that low atrazine concentration (0.1 ppb) stopped the gonadal development of male frogs, confirming the reports of Parshley (2000). In a laboratory study, Hayes et al. (2002b, 2002c) also reported that this herbicide was related to the feminization of *Rana pipiens*. Palma et al. (2009) found that atrazine concentrations affected the reproduction of *Daphnia magna*.

The herbicide atrazine is classified as a toxic agent, carcinogen and hormone disrupter (Friedmann, 2002) which includes potentially carcinogenic compounds to humans (Biradar & Rayburn, 1995). The presence of this product in the environment presents a risk to wildlife and the ecosystem in general, interfering with hormonal activity in animals and human in low doses. Studies have shown that atrazine can also effect the human reproductive system, decreasing the amount of sperms and increasing the infertility (Pan, 2011).

A research of the University of California analyzed the development of 40 males African frogs since the tadpoles stage to adult phase in water with concentration of atrazine within the limits considered safe by the Environmental Protection Agency (EPA). This group of frogs was compared with another without exposure to contaminated water. Among the frogs developed in the water with the herbicide, 10% became functional females. The others 90% despite having characteristics of males, had low testosterone levels and fertility (Hayes, 2010). Strandberg and Scott-Fordsmand (2002) considering organisms exposed to the herbicide simazine, reported ecological effects, including, bioaccumulation in aquatic organisms.

5. Final remarks

There is a growing choice of weed's chemical management by farmers in many agricultural regions of Brazil and the worldwide. The use of herbicides within technical recommendations offers low risk of contamination of non-target sites; however, when applied intensively and without liability, negative environmental impacts may occur.

The application of leachable products such as atrazine and clomazone concerns researchers. It is necessary to achieve sustainable alternatives to the use of these products: the replacement of non-leachable and less toxic products to the environment or follow the banishment example already performed in some countries.

The adoption of bioremediation techniques to areas already contaminated and investment in pesticide application technology as preventive alternative are some of the possibilities to reduce the waste of these molecules in surface water and groundwater.

The lack of supervision by the authorities in small and large agricultural areas coupled with the lack of knowledge of peoples who apply these products and the facility of acquisition contribute to an intensive use and without responsibility.

If professionals and research groups involved in agribusiness can hardly do for the reduction of environmental impacts from domestic and industrial origin, on the other hand, they have fundamental role in the use and dissemination of Good Agricultural Practice to producers, which will be essential for maintaining the activity in a sustainable way at long term.

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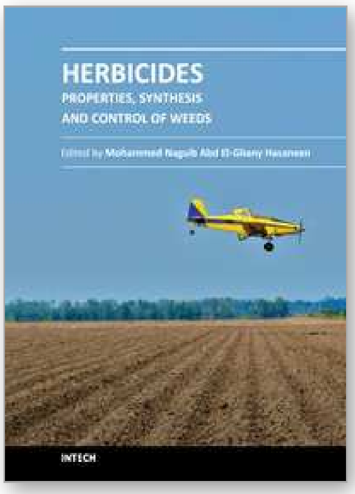

Herbicides - Properties, Synthesis and Control of Weeds
Edited by Dr. Mohammed Nagib Hasaneen

ISBN 978-953-307-803-8
Hard cover, 492 pages
Publisher InTech
Published online 13, January, 2012
Published in print edition January, 2012

This book is divided into two sections namely: synthesis and properties of herbicides and herbicidal control of weeds. Chapters 1 to 11 deal with the study of different synthetic pathways of certain herbicides and the physical and chemical properties of other synthesized herbicides. The other 14 chapters (12-25) discussed the different methods by which each herbicide controls specific weed population. The overall purpose of the book, is to show properties and characterization of herbicides, the physical and chemical properties of selected types of herbicides, and the influence of certain herbicides on soil physical and chemical properties on microflora. In addition, an evaluation of the degree of contamination of either soils and/or crops by herbicides is discussed alongside an investigation into the performance and photochemistry of herbicides and the fate of excess herbicides in soils and field crops.

Rafael Grossi Botelho, João Pedro Cury, Valdemar Luiz Tornisielo and José Barbosa dos Santos (2012). Herbicides and the Aquatic Environment, Herbicides - Properties, Synthesis and Control of Weeds, Dr. Mohammed Nagib Hasaneen (Ed.), ISBN: 978-953-307-803-8, InTech, Available from: http://www.intechopen.com/books/herbicides-properties-synthesis-and-control-of-weeds/herbicides-and-the-aquatic-environment

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University Campus STeP Ri
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51000 Rijeka, Croatia
Phone: +385 (51) 770 447
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中国上海市延安西路65号上海国际贵都大饭店办公楼405单元
Phone: +86-21-62489820
Fax: +86-21-62489821