



Occurrence of pesticide residues in fish from south American rainfed agroecosystems



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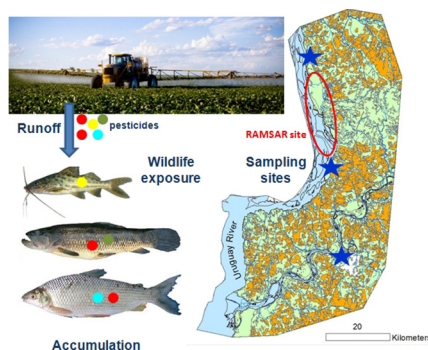
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HIGHLIGHTS

- Multiple substances approved for agriculture detected at sublethal concentrations.
- Spatial differences are observed for non-migratory fishes depending on land use.
- Bioaccumulation positively associated to log Kow and trendy active ingredients.
- Trifloxystrobin exhibited higher occurrence in fish in spite of its reduced persistence.

GRAPHICAL ABSTRACT



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ABSTRACT

Environmental sustainability of South American rainfed agroecosystems is of current concern. In this work, we evaluate the occurrence of multiple pesticide residues in muscle tissue of wild fish species from two large rivers in South America (Uruguay and Negro Rivers). Two sampling campaigns (representing summer and winter crops) were performed during 2015 targeting a wide biodiversity of fish species used for human consumption (ranging from migratory to non-migratory and from detritivorous to top-predators). Three different localities associated to rainfed agriculture were assessed, two of them enclosed to a RAMSAR site (National Park “Esteros de Farrapos e Islas del Rio Uruguay”). Pesticide residues occurred in muscle tissue of 143 from 149 sampled fishes (96%). Thirty different pesticides were detected at concentrations from <1 to $194 \mu\text{g kg}^{-1}$. Incidence of pesticides in fish were tightly related to: i) features of the contaminant: (Kow, environmental persistence and mobility) and ii) intensity of use of particular pesticides and land dedicated to rainfed agriculture. Trifloxystrobin, metolachlor and pyraclostrobin showed the highest rates of occurrence. Of great concern is that strobirulins have highest toxicity to fish from those detected compounds. From the pattern of pesticides occurring for non-migratory fish species it was possible to trend important spatial differences related to the intensity of rainfed agriculture. Results suggest a regular exposition of aquatic wild biota to sublethal concentrations of multiple semi-polar pesticides.

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1. Introduction

The intensification of agriculture in Southern South America has caused remarkable changes in the use of land by highly technified agricultural techniques. Particularly in Argentina, Brazil, Paraguay and Uruguay, an intensive and specialized cropping system under rainfed conditions has been adopted. Nowadays, soybean itself represents 13% of the total income of Uruguayan exportations (Uruguay XXI, 2015). Agricultural practices shifted from a crop-pasture rotation (CPR) technique to a continuous annual cropping under no-till (CCNT) (Franzluebbers et al., 2014; Wingeyer et al., 2015). More than a decade of increasing crop area has been accompanied by changes in land tenancy, tillage practices, reduced crop diversity and greater overall productivity. For more than a decade CCNT became the predominant technique in rainfed agriculture (García-Préchac et al., 2004; Franzluebbers et al., 2014; Wingeyer et al., 2015). Uruguayan rainfed crops increased from 278,000 ha in 2004 to 1334 million ha in 2015 (DIEA, 2015; Harriet et al., 2017).

In Uruguay, typical rainfed summer crop under CCNT include the sowing of soybean (*Glicine max*, rounding 50% of land area), sorghum (*Sorghum spp.*) and maize (*Zea mays*) during October – November period. Harvest is typically performed during March – April. Generally, after 1 month of fallow, a rainfed winter crop of wheat (*Triticum aestivum*) or barley (*Hordeum vulgare*) is sown in June – July to harvest prior to the next summer crop. In such scenario, a rapid increase in the amount of pesticides applied was particularly registered for Uruguay as well as in other South American countries (Schreinemachers and Tipraqsa, 2012). These new, highly technified procedures are based in the massive use of pesticides to maximize yields (Novelli et al., 2017; Wingeyer et al., 2015). Demand of herbicides, fungicides and insecticides has been continuously growing as higher portions of land are dedicated to rainfed agriculture (DGSA, 2017).

However, although Good Agricultural Practices (GAPs) are generally followed, it is well known that pesticide residues may diffuse over different environmental compartments and reach non-target biota (Andreu and Picó, 2012; Belenguer et al., 2014; Ccancapa et al., 2015; Ginebreda et al., 2016; Masiá et al., 2015; Masiá et al., 2013; Niell et al., 2015; Niell et al., 2017). Fish muscle tissue has been habitually selected for monitoring several non-polar organic contaminants (Kalachova et al., 2011; Molina-Ruiz et al., 2015; Munaretto et al., 2013; Rose et al., 2015; Sapozhnikova and Lehotay, 2013; Sapozhnikova and Lehotay, 2015).

Despite the wide range of substances currently approved for use in agriculture, their effects and circulation among environmental compartments and particularly on wildlife are not studied or clearly recorded (Köhler and Triebkorn, 2013). The study of the diffuse contamination due to pesticides is a difficult and broad task as the agrochemicals range from low to relatively high solubility, toxicity and persistence. Therefore, wildlife organisms living in streams or rivers whose basin is subjected to rainfed CCNT practices are potentially exposed to pesticides. Their occurrence in local species would be able to depict spatial variation of exposure to pollutants. Aquatic biomonitoring strategy is useful in the identification of risk and prioritization approaches (Chalar et al., 2013).

Moreover, fish are important components structuring aquatic communities, with a great range of body sizes, habitats and trophic positions (Daufresne and Boët, 2007). They are consumed by humans and provide a source of protein to artisanal fishermen and local people. In order to understand the consequences of particular agricultural practices, wild fish species with different and similar feeding habits and migratory behavior should be examined.

The aim of this study is to show main findings of pesticide residues occurring in fish muscle tissue of selected South American fish species collected in fishing campaigns accomplished in 2015 at different locations of rainfed agroecosystems in Uruguay.

This study particularly focuses on different fish species living into the RAMSAR convention site National Park “Esteros de Farrapos e Islas

del Río Uruguay” (RAMSAR, 2017). Moreover, we show spatial distribution of occurring pesticides and its positive relationship with land dedicated to rainfed agriculture. Finally, we discuss the association with physicochemical properties, soil degradation and amount of pesticides that foster accumulation in fish tissues.

2. Materials and methods

2.1. Site description

Uruguay River and Negro River are the most important freshwater courses in Uruguay. Uruguay River divides Uruguay and Argentina whereas Negro River divides southern to northern territories of Uruguay. This study was carried out with fish samples from 4 sampling sites. San Javier (SJ) (32°41'00"S 58°08'00"O) and Nuevo Berlin (NB) towns (32°58'45"S 58°03'10"O) are two sites located at margins of Uruguay River. Both locations are adjacent to the National Park “Esteros de Farrapos e Islas del Río Uruguay” – a RAMSAR Convention site. The Uruguayan National System of Protected Areas (SNAP) administrates this Park. This area constitutes a system of river wetlands, islands and islets that are permanently or temporarily flooded as a result of floods of the River. Wetlands, natural field and scrubland ecosystem predominates. However, neighboring areas of NB are particularly dedicated to intensive CCNT plantations of soybean, maize, barley, wheat, sorghum. On the other hand, SJ neighboring areas range from intensive CCNT crop to natural pasture. In this area, secondary water streams that cross the rainfed fields fall into the main rivers (Uruguay River or Negro River). Forestry of exotic species (mainly *Eucalyptus spp.*) for cellulose pulp production is located between both localities. Fig. 1 shows the sites under study and land use.

The city of Mercedes (MC) (33°15'00"S 58°01'55"O) lies on the margin of the Negro River, 50 km upstream where the Negro River falls into the Uruguay River (Fig. 1C). Particularly, the surrounding areas of MC and NB developed the most intensive CCNT agriculture. Rainfed agriculture is performed at southern and northern margins of the Negro River. Fig. 1B shows land use in NB, SJ and MC. In MC land use is predominantly dedicated to soybean – wheat annual CCNT.

San Gregorio de Polanco (SGP) (32°37'00"S 55°55'00"O) is also located at margin of Negro River but >200 km upstream from MC and separated by two hydroelectric dams. Most land use is particularly associated to grassland and forestry although there is incipient rainfed agriculture. Natural herbaceous field dominates in this area as seen on Fig. 1D. SGP was selected as a CCNT negative site.

Intensity of land dedicated to rainfed agriculture is mostly associated to MC and NB. On the other hand, SJ has less land use dedicated to rainfed agriculture and some amount of protected area surrounding. Finally, SGP was considered as a negative control site of rainfed agriculture.

2.2. Sampling

Two sampling campaigns were developed in SJ, NB and MC at the beginning of April 2015 (fall season, after summer crop) and at the end of September 2015 (spring season, after winter crop). Sampling dates were selected in order to have two contrasting scenarios of agricultural production at the end of each crop. For SGP sampling campaign was performed at September 2015.

In all sites, fish samples were obtained from local fishermen. Individuals were size measured and taxonomically identified according to morphometric characters and visual inspection (Teixeira de Mello et al., 2011).

A total of eight fish species were acquired from these four sampling sites. Five non-migratory species (*Hoplias malabaricus*, *Rhamdia quelen*, *Pimelodus maculatus*, *Paraloricaria vetula*, *Hypostomus commersonni*) and three migratory ones (*Salminus brasiliensis*, *Megaleporinus obtusidens*, *Prochilodus lineatus*). Feeding habits of these species range

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