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Environmental impact of pesticides after sewage treatment plants removal in four Spanish Mediterranean rivers

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The re-use of sewage treatment plant (STP) effluents is currently one of the most employed strategies in several countries to deal with the water shortage problem. Some pesticides are bio-accumulative and due to their toxicity they can affect non-target organisms, especially in the aquatic ecosystems, threating their ecological status. Despite these facts, and to our knowledge, there are few peer-reviewed articles that report concentrations of pesticides in Spanish STPs.

This work presents the results of an extensive survey that was carried out in October of 2010 in 15 of the STPs of Ebro, Guadalquivir, Jucar and Llobregat rivers in Spain. Forty-three currently used pesticides, belonging to anilide, neonicotinoid, thiocarbamate, acaricide, juvenile hormone mimic, insect growth regulator, urea, azole, carbamate, chloroacetanilide, triazine and organophosphorus, have been monitored. Integrated samples of influent and effluent, and dehydrated, lyophilized sludge from 15 STPs located along the rivers were analyzed for pesticide residues. With these data, removal efficiencies are also calculated.

Extraction of water samples was performed through Solid Phase Extraction (SPE) and sludge samples were extracted using the QuEchERS method. Pesticide determination was carried out using Liquid Chromatograph – tandem Mass Spectrometry (LC-MS/MS). Recoveries ranged from 48% to 70%, in water samples, and from 40 to 105 %, in sludge samples. The limits of quantification were 0.01-5 ng L-1 for the former, and 0.1-5.0 ng g-1 for the latter.

In terms of frequency of detection, 31 analytes were detected in influent, 29 in effluent and 11 in sludge samples. Organophosphorus pesticides were the most frequently detected in all wastewater samples, but azole, urea, triazine, neonicotinoid and the insect growth regulator were also commonly found. Imazalil revealed the maximum concentration in wastewater samples from all rivers except the Guadalquivir, in which diuron presented the maximum one. Eleven pesticides including five organophosphorus, two azoles, one triazine, one chloroacetanilide, one juvenile hormone mimic and one acaricide were detected in the sludge samples. Accordingly, organophosphorus were the most frequently detected pesticides in the sludge samples, but the highest concentration was observed for imazalil. The higher concentration of this azole in the influent and their possible stronger adsorption may be the reason for their higher concentration in the sludge samples. The removal efficiency of pesticides was calculated from the analyte concentration in influent (Cin) and effluent (Cef): [(Cin-Cef)/Cin] x 100%. The removal of organophosporus ranged from -810,47 to 93,11%, meanwhile azoles and ureas were not removed in the STPs.

The poor elimination of pesticides by sewage treatment plants presented in this study could be related to the treatment process used, hydraulic and solid retention times, besides the dilution and temperature of the raw sewage and the plant's configuration. These poor efficiencies are responsible of the high pesticides concentration (e.g.diuron) found in some effluents, which may endanger water quality of the ecosystem when they are re-used or directly discharged into the river. In fact, with respect to the Maximum Allowable Concentrations (MAC) stipulated by the Directive 2008/105/EC for pesticides in inland and other surface waters (Council of the European Communities, 2008), diuron exceeded these limits. Nevertheless, it is important to emphasize that, even though, the pesticides concentrations measured were relatively low (according to directives); this study analysed just some of them. A wide variety of other compounds, including other pesticides and pesticides transformation products, may contribute to the bad quality of the water ecosystems.

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