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## Ecotoxicological risk assessment of chemical pollution in four Iberian river basins and its relationship with the aquatic macroinvertebrate community status

Maja Kuzmanović<sup>a,\*</sup>, Julio C. López-Doval<sup>b</sup>, Núria De Castro-Català<sup>c</sup>, Helena Guasch<sup>d</sup>, Mira Petrović<sup>e,f</sup>, Isabel Muñoz<sup>c</sup>, Antoni Ginebreda<sup>a</sup>, Damià Barceló<sup>a,e</sup>

<sup>a</sup> Water and Soil Quality Research Group, Dep. of Environmental Chemistry, IDAEA-CSIC, Jordi Girona 18-26, 08034 Barcelona, Spain

<sup>b</sup> Institute of Biosciences, Department of Ecology, University of São Paulo, R. do Matão, Travessa 14, 321 Butantã 05508-090 São Paulo, Brazil

<sup>c</sup> Department of Ecology, Universitat de Barcelona, Av. Diagonal, 643, 08028 Barcelona, Spain

<sup>d</sup> Institute of Aquatic Ecology and Department of Environmental Sciences, University of Girona, Campus de Montilivi, 17071 Girona, Spain

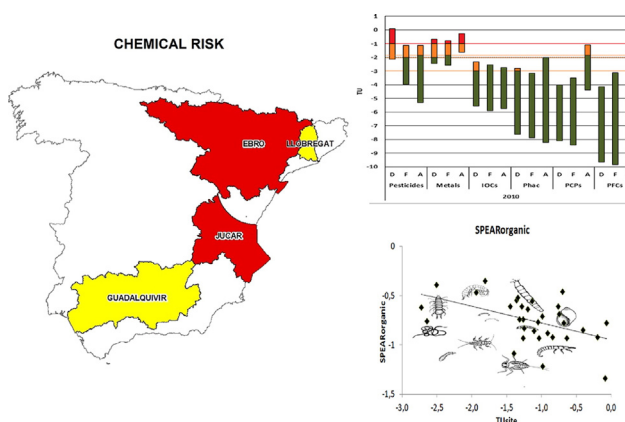
<sup>e</sup> Catalan Institute for Water Research (ICRA), H2O Building, Scientific and Technological Park of the University of Girona, Emili Grahit 101, 17003 Girona, Spain

<sup>f</sup> ICREA, Passeig Lluís Companys 23, 08010 Barcelona, Spain

### HIGHLIGHTS

- Ecological risk assessment of chemical pollution was performed in four Iberian rivers.
- Relationships between chemical risk and biological responses in situ were examined.
- Acute risk effects at more than 40% and chronic at all sites were present.
- Both regulated and emerging compounds are responsible for chronic risk.
- Decrease of sensitive species was correlated with the increase of the risk.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Ecotoxicological risk assessment of chemical pollution in four Iberian river basins (Llobregat, Ebro, Júcar and Guadalquivir) was performed. The data set included more than 200 emerging and priority compounds measured at 77 sampling sites along four river basins studied. The toxic units (TU) approach was used to assess the risk of individual compounds and the concentration addition model (CA) to assess the site specific risk. Link between chemical pollution and aquatic macroinvertebrate communities in situ was examined by using four biological indexes: SPEAR (“Species at Risk Index”) as the indicator of decline of sensitive species in relation to general organic (SPEAR<sub>organic</sub>) and pesticides (SPEAR<sub>pesticides</sub>) pollution; and Shannon and Margalef biodiversity indexes. The results of the study suggested that organic chemicals posed the risk of acute effects at 42% of the sampling sites and the risk of chronic effects at all the sites. Metals posed the acute risk at 44% of the sites. The main drivers of the risk were mainly pesticides and metals. However, several emerging contaminants (e.g. the antidepressant drug sertraline and the disinfectant triclosan) were contributing to the chronic effects risk. When risk associated

\* Corresponding author.

River basins  
Emerging contaminants  
Macroinvertebrate communities

with metals and organic chemicals was compared, the latter dominated in 2010, mainly due to the presence of highly toxic pesticides, while metals did in 2011. Compounds that are not regulated on the European level were posing the risk of chronic effects at 23% of the sites. The decline of sensitive macroinvertebrate taxa expressed in terms of SPEAR index was correlated with the increase of toxic stress related to organic compounds. Biodiversity indexes were negatively correlated with the metals and the urban land use type in the catchment.

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## 1. Introduction

Aquatic ecosystems are impacted by a variety of stressors, including organic and inorganic stressors, excess input of nutrients, geomorphological alterations, land use changes, hydrological stress, invasive species and pathogens (Vörösmarty et al., 2010). As a consequence, the biodiversity decline is one of the greatest ecological problems threatening aquatic ecosystems (Beketov et al., 2013). However, little is known beyond the described effects of single stressors on specific ecological endpoints (Navarro-Ortega et al., 2015) and our understanding of the main causes for the losses of biodiversity still remains vague (Beketov et al., 2013). Rivers are receiving numerous chemical compounds originated from anthropogenic activities on a daily basis. As a result, complex mixtures of potentially dangerous compounds are present in the aquatic environment. However, site-specific exposures can vary a lot and some sites are likely to be affected more than others due to local conditions and specific vulnerability characteristics (Brack et al., 2015). Thus, the characterization of the constituents of these mixtures and the identification of the compounds of the highest concern in different spatial frameworks is one of the key issues for the protection of natural ecosystems (Vörösmarty et al., 2010).

Besides a number of regulated pollutants which are known to exhibit adverse effects, there is a large number of chemicals currently in use that are not taken into account in the routine water quality monitoring (Barceló and Petrovic, 2007). These compounds are commonly referred to as emerging contaminants. They encompass a variety of substances used both in industry and households; such as pharmaceuticals, personal care products, hormones, industrial chemicals or their byproducts and the transformation products, all together having in common that their environmental allowed levels are not regulated. In the European Union, the Water Framework Directive (WFD) Directive 2013/39/EU (2013) is the legislation concerning the chemical pollution which aims to achieve good chemical status of water bodies by meeting the Environmental Quality Standards (EQS) for the 45 so-called priority substances (PS) and priority hazardous substances (PHS). In addition, under the WFD, the EU member states are obliged to set quality standards for river basin specific pollutants discharged in each water body and to take action to meet these quality standards as a part of ecological status. A question that remains open is to what extent priority pollutants represent chemical status in comparison with unregulated chemicals. Here we address this issue from the perspective of their associated ecotoxicological risk.

Another challenge for the scientist dealing with aquatic risk assessment is revealing the link between water pollution and biological community responses. Due to the presence of multiple stressors, their unknown joint effects and the complexity of the biological responses, it is very difficult to distinguish the influence of particular stressors on affected ecosystems. Moreover, in recent years, studies in ecology are increasingly emphasizing that biodiversity loss implies more than the mere loss of species (i. e. taxonomic diversity) (Feld et al., 2014). Hence, the functional component of biodiversity should rather be addressed by using the concept of biological traits (e.g. generation time, body size) (Beketov and Liess, 2008; Feld et al., 2014). Commonly used taxonomic richness and diversity metrics (e.g. Shannon or Margalef diversity indexes) are dependent on both anthropogenic influences and natural longitudinal gradient of environmental factors in rivers as altitude, temperature, stream width, nutrition status and velocity (Minshall et al., 1985; Beketov and Liess, 2008; Paller et al.,

2006) so they might not be able to characterize the toxicant specific influence of ecosystems. To cope with this problem stressor specific, traits based metric SPEAR index was developed for pesticides (Liess and Von Der Ohe, 2005), general organic toxicants (e.g. petrochemicals, synthetic surfactants) (Beketov and Liess, 2008) and salinity (Schäfer et al., 2011a) which is poorly dependent on the natural longitudinal factors (Beketov and Liess, 2008).

In this context our study is addressing the following objectives. First, to assess the area specific levels of the risk posed to aquatic ecosystems on the river basin level for more than 200 emerging and priority pollutants in four Iberian river basins using the toxic unit concept. Second, to evaluate whether the current list of WFD priority pollutants is enough to estimate the ecotoxicological risk in these basins or there are other compounds present that could be more or equally important in terms of risk. And third, to determine the potential relationship between the ecotoxicity associated with local mixtures of pollutants and aquatic macroinvertebrate biological community responses using four different metrics: Shannon and Margalef biodiversity indexes and SPEAR<sub>pesticides</sub> and SPEAR<sub>organic</sub>.

To tackle these questions we used as case study four rivers of the Iberian Peninsula for which both biological and chemical data were previously gathered (Navarro-Ortega et al., 2012).

## 2. Materials and methods

### 2.1. Study area

Four Iberian river basins (Fig. 1) were studied as the representatives of Mediterranean rivers. Detailed description of the study area can be found elsewhere (Kuzmanović et al., 2015).

The Llobregat is the river situated in the North East of Iberian Peninsula. The lower part of the basin is subjected to strong anthropogenic pressures due to high proportion of the urban and industrial land use types in that area. In the middle part of the basin most of the agricultural lands are situated. As a typical Mediterranean river, Llobregat is subjected to decreased flow in the summer periods as a consequence of Mediterranean climate (Gasith and Resh, 1999). The Ebro is the large river situated in North of the Peninsula. The main pressures for water quality are coming from agriculture developed along the river basin. The urban and industrial centers are scattered in the basin, mostly in the North East and central part of the basin. The Júcar basin is situated in the East of Iberian Peninsula characterized by semi-arid climate. The most of the agricultural and urban areas are located in the medium and lower parts. Thus, these areas are receiving the most of the combined pressures together. The Guadalquivir basin, situated in the South of the Peninsula as a consequence of the high population, is subjected to strong anthropogenic pressures that may cause deterioration of water quality. A large portion of the basin is devoted to agricultural use which might result in water quality deterioration due to input of pesticides and fertilizers.

### 2.2. Sampling

The data used for this study were gathered within the Spanish research SCARCE-CONSOLIDER project (Navarro-Ortega et al., 2012). Extensive monitoring of water, sediment and biota from the four Iberian river basins was carried out in two monitoring campaigns (autumn 2010 and 2011). The autumn of 2010 was characterized by intense

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