Available at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/watres

Exposure of the marine deposit feeder *Hydrobia ulvae* to sediment spiked with LAS congeners

A. Mauffret [a,*], A. Temara [b], J. Blasco [a]

[a] The Marine Sciences Institute of Andalusia (ICMAN, CSIC), Puerto Real, Spain
[b] The Procter & Gamble Company, Brussels, Belgium

ARTICLE INFO

Article history:
Received 25 September 2009
Received in revised form
10 February 2010
Accepted 16 February 2010
Available online 19 February 2010

Keywords:
LAS
Homologues
Mixture toxicity
Sediment
Deposit feeder
Acclimation

ABSTRACT

The lethal and sub-lethal toxicity of LAS congeners to the mollusc gastropod *Hydrobia ulvae* were assessed in spiked sediment bioassays. This complements the little knowledge available to date on mixture effects in the sediment compartment. The LAS homologues joint effect was concentration additive ($\sum TU_i = 0.8–1$). As opposed to the 10-d LC10 based on the sediment associated LAS concentration (91–330 mg/kg) which was independent of the homologue chain length, the LC10 based on the dissolved LAS fraction (0.804–0.068 mg/L) decreased as the homologue chain length increased from 10 to 13 carbons. The quantitative structure-activity relationship (QSAR) derived from these data was log (1/LC10 (mol/L)) = 0.64 log K_{ow} + 4.40 ($n = 5$; $r^2 = 0.76$; $s = 0.24$). It showed an apparent higher toxicity compared to the typical QSAR for polar narcosis in water-only systems probably due to the simultaneous exposure of the snail to LAS through the dissolved and the sediment associated fractions. The egestion rate of the surviving snails recovered after few days' exposure (1-d NOEC: 40–107 mg/kg, 9-d NOEC: 65–190 mg/kg) which suggests that the organisms were able to acclimate to LAS during the exposure.

1. Introduction

Joint effect of compounds in a mixture may be similar (additive), stronger (synergistic) or weaker (antagonistic) than expected from the observed effects of separate exposures (Altenburger et al., 2003). Joint effect depends on the interactions between the chemicals in the mixture and their mode(s) of action with respect to the biological response. The interactions depend on the influence of one chemical on the biological responses of the other chemicals in the mixture. Furthermore, the mode of action depends on the sites of primary action of the chemicals, which can be specific or unspecific (Escher and Hermens, 2002). Specific modes of action encompass reactive and receptor mediated mechanisms. An unspecific mode of action is defined as baseline toxicity or narcosis and is the reference case because it is the minimal toxicity of any given chemical (e.g. Boeije et al., 2006; Hodges et al., 2006a; Hwang et al., 2003). It is displayed by passive diffusion of the contaminant in biomembranes and therefore directly correlated with environmental concentration and compound hydrophobicity. Consequently, quantitative structure-activity relationship (QSAR) approaches can be developed with a limited number of chemicals exerting baseline toxicity and used to predict the toxicity of a wider set of chemicals with a similar hydrophobicity and mode of action. Baseline toxicity mainly includes two modes of action: non-polar and polar narcosis (Table 1). Polar narcosis is slightly more toxic than predicted by non-polar narcosis. Indeed, in fish, the lethal body residues of chemicals acting by non-polar narcosis have been found to range between 2 and 8 mmol/kg body weight; which is greater than the lethal body residues of polar narcotics (0.6–1.9 mmol/kg body weight)

* *Corresponding author*. Tel.: +34 956832612; fax: +34 956834701.
E-mail address: aourell.mauffret@icman.csic.es (A. Mauffret).
0043-1354/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.watres.2010.02.020

Table 1 – Selected QSAR developed in aquatic systems for non-polar and polar narcosis including equations developed specifically with LAS congeners.

Species	Endpoints	Equation	Reference
1. Non-polar narcosis in aquatic system (mol/L)			
Fish *Pimephales promelas*	96h-LC50	$\log(1/LC50) = 0.85 \log K_{ow} + 1.39$ $n = 58$, $r^2 = 0.94$, $s = 0.36$	(Verhaar et al., 1995)
Fish *Brachydanio rerio*	28-32-d NOEC ELS	$\log(1/NOEC) = 0.90 \log K_{ow} + 2.30$ $n = 27$, $r^2 = 0.92$, $s = 0.33$	(Verhaar et al., 1995)
Crustacean *Daphnia magna*	48-h LC50	$\log(1/EC50) = 0.95 \log K_{ow} + 1.32$ $n = 49$, $r^2 = 0.95$, $s = 0.34$	(Verhaar et al., 1995)
Crustacean *Daphnia magna*	16-d NOEC, growth, reprod.	$\log(1/NOEC) = 1.05 \log K_{ow} + 1.85$ $n = 10$, $r^2 = 0.97$, $s = 0.39$	(Verhaar et al., 1995)
Algae *Selenastrum capricornutum*	72-96-h EC50 growth	$\log(1/EC50) = 1.00 \log K_{ow} + 1.23$ $n = 10$, $r^2 = 0.93$, $s = 0.17$	(Van Leeuwen et al., 1992)
2. Polar narcosis			
2. 1. Typical polar narcosis in aquatic system (mol/L)			
Fish *Pimephales promelas*	96-h LC50	$\log(1/LC50) = 0.73 \log K_{ow} + 2.16$ $n = 86$, $r^2 = 0.90$, $s = 0.33$	(Verhaar et al., 1995)
Crustacean *Daphnia magna*	48-h EC50 immobilis	$\log(1/EC50) = 0.56 \log K_{ow} + 2.79$ $n = 37$, $r^2 = 0.77$, $s = 0.37$	(Verhaar et al., 1995)
2. 2. LAS in aquatic system (mol/L)			
Crustacean *Daphnia magna*	LC50	$\log(1/LC50) = 0.64 \log K_{ow} + 2.44$ $n = 12$, $r^2 = 0.96$, $s = 0.15$	(Roberts, 1991)
Crustacean *Daphnia magna*	48-h LC50	$\log(1/LC50) = 0.77 \log K_{ow} + 2.47$ $n = 17$; $r^2 = 0.96$; $s = 0.16$	(Hodges et al., 2006b)
2. 3. LAS in sediment system (mol/L or mol/kg)			
Mollusc *Hydrobia ulvae*	10-d LC10 mol/L	$\log(1/LC10) = 0.64 \log K_{ow} + 4.40$ $n = 5$; $r^2 = 0.76$; $s = 0.24$	Present study, Fig. 2
Mollusc *H. ulvae*	10-d LC10 mol/kg	$\log(1/LC10) = -0.22 \log K_{ow} + 4.01$ $n = 5$; $r^2 = 0.43$; $s = 0.17$	Present study, Fig. 2

ELS: early life-stage, n: number of data, r^2: correlation coefficient, s: standard error of estimate.

(McCarty and Mackay, 1993). Roberts and Costello (2003) suggested that the mechanistic difference between non-polar and polar narcosis is due to different water – membrane partitioning processes that depend on the contaminant polarity. In polar narcosis partitioning, part of the narcotic molecule associates with the head groups of the membrane lipids, whereas in non-polar narcosis, the narcotic molecule moves freely in all directions in the membrane.

To our best knowledge, the application of mixture assessment in sediment systems is currently based on extrapolation from data obtained in aquatic systems with equilibrium partitioning (EqP) calculations, and assuming thus that the porewater is the main exposure route (Di Toro et al., 2000; Van Leeuwen et al., 1992). However, though the EqP model is applicable in numerous cases (e.g. Rico–Rico et al., 2009), departures have been reported for several benthic deposit feeders, mainly with oligochaete or amphipod (e.g. Simpson and King, 2005) and also with the gastropod *Hydrobia ulvae* (Mauffret et al., 2010). *H. ulvae* is a deposit feeder widely distributed along the Atlantic coast, usually occurring at extremely high densities (up to 100,000 ind./m^2) (Sola, 1996). The hydrobiidae family constitutes a primary food source for larger gastropods, crustaceans, fishes and birds.

The anionic surfactant Linear Alkylbenzene Sulfonate (LAS) is a major high production volume chemical with 340 kt consumed in Europe in 2005, mainly in detergents (HERA, 2009). Worldwide growth of LAS is expected to average about 1.7% per year during 2008–2013 (Modler et al., 2009). LAS concentrations in estuarine and coastal sediments are typically below 2 mg/kg where sewage treatment systems are installed (Lara-Martín et al., 2006). However, close to urban sites not equipped with an adequate WWTP, concentrations can reach up to 750 mg/kg (Gonzalez-Mazo et al., 1999). The commercial material consists in a complex mixture of homologues and isomers identified by an alkyl chain length normally ranging from 10 to 13 carbons and by the position of attachment of the benzenesulfonate group on the alkyl chain from the central to the external carbon (6–2 phenyl). LAS can thus be modelled as a multicomponent mixture. In aquatic systems, the LAS mixture has been suggested to exert its toxicity by polar narcosis mechanisms because of the similarity with polar chemical characteristics of the body burden measured in midge (0.2–0.7 mmol/kg body weight) (Hwang et al., 2003) and of the QSARs developed with individual LAS homologues in water-only systems (Roberts, 1991) (Table 1). In the present work, LAS lethal and sub-lethal effects to *H. ulvae* were determined to assess the mixture effects in sediment systems.

2. Materials and methods

2.1. Chemicals

The LAS mixture and individual homologues were kindly provided by the Petresa Company (Spain). The total activity of the LAS mixture (CAS Nr. 68411-30-3, MW 341.5) was 46.8%