



Caffeine and paraxanthine in aquatic systems: Global exposure distributions and probabilistic risk assessment



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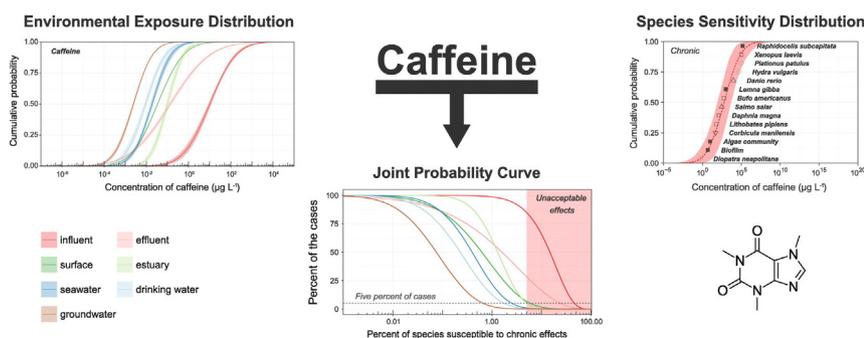
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HIGHLIGHTS

- Caffeine and Paraxanthine present similar environmental exposure distributions.
- Unacceptable environmental risk from chronic exposure to caffeine in 3 matrices.
- Negligible environmental risk expected from acute aquatic exposure to caffeine.
- Unacceptable environmental risk from paraxanthine exposure in all matrices but two.
- Negligible Human health risk from exposure to caffeine via drinking or groundwater.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 2 June 2017

Received in revised form 8 August 2017

Accepted 8 August 2017

Available online xxx

Editor: D. Barcelo

Keywords:

Caffeine
Paraxanthine
Aquatic systems
Exposure distribution
Probabilistic risk assessment

ABSTRACT

This study presents one of the most complete applications of probabilistic methodologies to the risk assessment of emerging contaminants. Perhaps the most data-rich of these compounds, caffeine, as well as its main metabolite (paraxanthine), were selected for this study. Information for a total of 29,132 individual caffeine and 7442 paraxanthine samples was compiled, including samples where the compounds were not detected. The inclusion of non-detect samples (as censored data) in the estimation of environmental exposure distributions (EEDs) allowed for a realistic characterization of the global presence of these compounds in aquatic systems. EEDs were compared to species sensitivity distributions (SSDs), when possible, in order to calculate joint probability curves (JPCs) to describe the risk to aquatic organisms. This way, it was determined that unacceptable environmental risk (defined as 5% of the species being potentially exposed to concentrations able to cause effects in > 5% of the cases) could be expected from chronic exposure to caffeine from effluent (28.4% of the cases), surface water (6.7% of the cases) and estuary water (5.4% of the cases). Probability of exceedance of acute predicted no-effect concentrations (PNECs) for paraxanthine were higher than 5% for all assessed matrices except for drinking water and groundwater, however no experimental effects data was available for paraxanthine, resulting in a precautionary deterministic hazard assessment for this compound. Given the chemical similarities between both compounds, real effect thresholds, and thus risk, for paraxanthine, would be expected to be close to those

Abbreviations: CAF, Caffeine; PXT, Paraxanthine; WWTP, Waste water treatment plant; DWTP, Drinking water treatment plant; HQ, Hazard Quotient; MEC, Measured Environmental Concentration; WWTP, Waste Water Treatment Plant; PNEC, Predicted no effect concentration; LOD, Limit of detection; LOQ, Limit of quantification; MDL, Method detection limit; QSAR, Quantitative Structure-Activity Relationships; AF, Assessment Factor; AC, Acceptable Concentration.

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observed for caffeine. Negligible Human health risk from exposure to caffeine via drinking or groundwater is expected from the compiled data.

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1. Introduction

It has been almost twenty years since the iconic study by Halling-Sørensen et al. (1998), and others (Daughton and Ternes, 1999; Heberer, 2002), focused the attention of the scientific community in the field of “emerging” contaminants. During those two decades an increasing amount of data, both on the presence of these compounds in the environment, as well as their potential environmental effects, has been collected. While the societal and regulatory interest on these compounds has increased greatly since then (BIO Intelligence Service, 2013; EC, 2013; EC, 2015), most attempts to assess the potential environmental risks of these compounds have failed to make full use of the whole extent of the available data; with quick, deterministic, hazard assessments still being the default approach. These assessments are often limited to the calculation of Hazard Quotients (HQ) based on limited toxicological data and direct comparison to point concentrations, measured on a particular study. Attempts to make full use of the existing exposure data, and fully characterize the exposure distributions of these emerging contaminants have been very limited, with only a small number of authors attempting such an approach (Boxall et al., 2014; Christensen et al., 2009; Corrales et al., 2015; Kristofco and Brooks, 2017; Straub, 2008; Straub, 2013; Straub, 2016; Straub and Stewart, 2007).

While it is true that, for most compounds, the extent of the available effects data is insufficient for the development of environmental risk assessments at a higher tiers (Tier-2 and up), the refinement of the Tier-1 exposure characterization stage could benefit from the inclusion of probabilistic techniques, even within individual studies, by estimating the probability of exceeding a particular effect threshold (e.g. a predicted no-effect concentration, PNEC). Furthermore, for some common compounds (such as caffeine) there is, in fact, sufficient exposure and effects data available for the application of full probabilistic Tier-2 assessments.

These approaches, however, do present some limitations. These commonly result from the nature of environmental monitoring data, which is rich in censored data (e.g. samples below the detection limit), as well as from the heterogeneity of formats in which this data is available in the literature, where data is commonly presented just as summary tables (with max-min, median, or mean concentrations) or even in graphical form. On the few instances in which emerging contaminants have been studied via the use of environmental exposure distributions (EEDs), these kind of issues have been dealt with either by limiting the distribution to that of maximum measured concentrations (Corrales et al., 2015; Kristofco and Brooks, 2017), or by back-calculation of the full original dataset from summary data, whenever possible, prior to the fit of to the EED (Straub, 2008; Straub, 2013; Straub, 2016; Straub and Stewart, 2007). Current computational power and statistical tools allow for the combined inclusion of both censored data, as well as summary data (max-mean-min) directly into the analysis, by directly including this interval data in the calculations.

Additionally, while some of the mentioned studies (Straub, 2008; Straub, 2013; Straub, 2016; Straub and Stewart, 2007), implicitly compare EEDs to probabilistic representations of effects data (e.g. species sensitivity distributions, SSDs), none of the studies found, made complete application of the probabilistic risk assessment methodologies by deriving joint probability curves, which can serve as useful tools to provide a quantifiable estimate of the risk (Solomon et al., 2000).

Caffeine, a natural alkaloid produced by many plant species, is a highly used stimulant contained in both recreational products (coffee, tea, caffeinated beverages) as well as many pharmaceutical products

(Buerge et al., 2003). After consumption, caffeine is extensively metabolized, with <5% being excreted unchanged in urine (Magkos and Kavouras, 2005). Paraxanthine (1,7-dimethylxanthine), is the main metabolite in humans, accounting for 80% of the total caffeine excretion in humans (Magkos and Kavouras, 2005). The presence of caffeine (CAF) and paraxanthine (PXT), in the environment has been extensively studied, and its environmental ubiquity has made it a commonly used anthropogenic marker (Buerge et al., 2003; Cantwell et al., 2016; Ferreira et al., 2005). Even before the attention of the scientific community focussed on emerging contaminants in the late 90s/early 2000s, a number of studies had already reported the presence of caffeine in environmental matrices as early as 1978 (Cain et al., 1980; Rogers et al., 1986), making it one of the most data-rich compounds among those considered as “emerging contaminants”.

As such, the main aims of this study were: 1) to characterize, in the most extensive way to date, the global presence of caffeine (as well as its main metabolite, paraxanthine) in aquatic environments, and 2) to use such an ubiquitous, and commonly studied, emerging contaminant – caffeine –, as a case study on how to make effective use of the whole set of publicly available exposure (including non-detect data) and effects data and the kind of approaches that can be applied for the characterization of the risks posed by emerging contaminants in aquatic systems.

2. Materials and methods

2.1. Data collection

2.1.1. Exposure data

Data on the presence of caffeine and paraxanthine (1,7-dimethylxanthine) in aquatic systems was collected from the primary scientific literature as well as public monitoring data repositories.

For the collection of CAF and PXT data from the primary literature, queries were performed in the main scientific repositories (e.g. Web of Knowledge, Pubmed, Scopus), which included the name of the searched compound as well as keywords such as “water”, “aquatic”, “wastewater”, “river”, and their flexible versions (e.g. aqua*). A total of 522 studies published prior to January 2017, were collected from this search. The compiled studies were reviewed, and the following information (when available) was extracted: water matrix evaluated, country/geographic region where samples were collected, year, season, number of samples collected, limit of detection (i.e. LOD, LOQ, MDL), number of detected samples, number of quantified samples, as well as the specific measured concentrations of caffeine and paraxanthine for each quantified sample.

The following matrices were considered: fresh surface water, seawater, estuary water, groundwater, wastewater influent/sewage, wastewater effluent and rain water. Seawater samples include those collected in coastal and open sea locations. Estuary waters include those collected on estuary habitats, i.e. the tide-affected, brackish water environment representing the confluence between a river and the sea. All surface water bodies not considered as sea or estuary were considered surface water, independently of their lentic or lotic nature. This includes, lakes, rivers, reservoirs and wetlands among others. Samples were considered wastewater influent when the samples were collected at the entrance of a wastewater treatment plant (including samples collected after initial screen-filtering and grit removal) or when collected directly on a sewer. In a number of occasions (considered via expert judgement) samples collected directly from septic tanks were considered as influent. The aim of this study was not to

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