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Risk assessment for drugs of abuse in the Dutch watercycle

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ABSTRACT

A screening campaign of drugs of abuse (DOA) and their relevant metabolites in the aqueous environment was performed in the Netherlands. The presence of DOA, together with the potential risks for the environment and the possible human exposure to these compounds through consumption of drinking water was investigated. Sewage water (influent and effluent), surface water of the rivers Rhine and Meuse, and drinking water (raw and finished) were analysed by four different laboratories using fully in-house validated methods for a total number of 34 DOA and metabolites. In this way, data reported for several compounds could also be confirmed by other laboratories, giving extra confidence to the results obtained in this study. In total 17 and 22 DOA were detected and quantified in influent and effluent sewage samples, respectively. The tranquilizers oxazepam and temazepam, and cocaine and its metabolite benzoylecgonine were found in high concentrations in sewage water. Nine compounds were possibly not efficiently removed during treatment and were detected in surface waters. The results indicated that substantial fractions of the total load of DOA and metabolites in the rivers Rhine and Meuse enter the Netherlands from abroad. For some compounds, loads appear to increase going downstream, which is caused by a contribution from Dutch sewage water effluents. As far as data are available, no environmental effects are expected of the measured DOA in surface waters.

In raw water, three DOA were detected, whereas in only one finished drinking water out of the 17 tested, benzoylecgonine was identified, albeit at a concentration below the limit of quantification (<1 ng/L). Concentrations were well below the general signal value of 1 μg/L, which is specified for organic compounds of anthropogenic origin in the Dutch Drinking Water Act.

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1. Introduction

Drugs of abuse (DOA) and their metabolites have recently been recognised as a novel group of environmental contaminants (Zuccato et al., 2008a). Owing to the increased sensitivity of analytical methods and the high level of world-wide consumption of DOA, they are among the growing number of emerging compounds that are detected at trace concentrations in the aqueous environment, including sewage water and surface waters.

DOA refers to both illegal drugs and misused prescription drugs, such as tranquilizers. They have received special attention recently since a novel approach allowed to study DOA consumption patterns of a population through sewage water analysis (Daughton, 2001; Zuccato et al., 2008b; van Nuijs et al., 2010; Thomas et al., 2012). Following consumption and excretion, some DOA and their metabolites are continuously released into the aquatic environment due to their insufficient elimination in sewage treatment plants (STPs) (Huerta-Fontela et al., 2008; Kasprzyk-Hordern et al., 2009; van Nuijs et al., 2009a; Postigo et al., 2010). Recent studies have shown the presence of DOA and their metabolites in STP effluents and river water in Australia (Irvine et al., 2011), Europe (Boleda et al., 2009; van Nuijs et al., 2009a; Postigo et al., 2010; Baker and Kasprzyk-Hordern, 2011; Hernandez et al., 2011) and North America (Jones-Lepp et al., 2004; Bartelt-Hunt et al., 2009).

Although the reported concentrations in surface waters are in general low, possible toxicological effects on animals, plants and humans may occur as a result of their presence in the aquatic environment. Especially, long-term effects on organisms and the effects of combined exposure to multiple compounds are of potential concern. However, so far, little ecotoxicological information for DOA is available and a well-founded scientific risk assessment is not yet possible. Although some information is available on DOA removal and transformation products formed during (drinking) water treatment processes (Huerta-Fontela et al., 2008), much more research is required for a better knowledge and understanding of these processes. In the Netherlands, where approximately 40% of the drinking water is produced from surface water, little is known about the occurrence of DOA and their metabolites in the Dutch water cycle. Exploratory studies conducted in the period 2007–2010 have revealed the presence of benzoylecgonine, methadone, codeine and three tranquilizers (nordazepam, temazepam and oxazepam) in Dutch surface waters and sewage effluents (de Voogt et al., 2011; Hogenboom et al., 2009). The results from this study implied a clear need for a more detailed monitoring campaign in the Netherlands.

This work presents the results of a large monitoring exercise on the occurrence of DOA and metabolites in the Dutch watercycle. To the best of our knowledge, this study is one of the largest of this kind in Europe, both in terms of number of analytes investigated and types of water studied. In addition, samples were individually analysed by four different laboratories, using their own validated analytical methodology. Five DOA were determined by all four laboratories and additional seven by at least two laboratories. The fact that three DOA (amphetamine, MDMA and benzoylecgonine) were found in several water samples by all laboratories allowed the performance of an interlaboratory exercise.

Beforehand, a selection of compounds was made, applying the following criteria: the results of the aforementioned preliminary inventory studies; international occurrence data on DOA and metabolites in the aqueous environment (Baker and Kasprzyk-Hordern, 2011; Bartelt-Hunt et al., 2009; Boleda et al., 2009; Hernandez et al., 2011; Irvine et al., 2011; Jones-Lepp et al., 2004; Postigo et al., 2010; van Nuijs et al., 2009a); the estimated DOA consumption in the Netherlands, which was based on criteria such as (il)legal import volumes and anonymous surveys (van Laar et al., 2007), the availability of reference standards and internal isotope-labelled standards, and the scope of the methods applied by the different laboratories participating.

The main objectives pursued within this study were (1) to evaluate the occurrence of DOA and metabolites in the Dutch watercycle (sewage influents and effluents, surface water and drinking water); (2) to perform an ecotoxicological risk assessment of the levels of DOA observed in surface waters.

2. Methods and materials

2.1. Sampling sites and sample collection

The sampling campaign in this study was performed by the Dutch National Institute for Public Health and the Environment (RIVM). All water samples were analysed by three laboratories: RIVM, KWR Watercycle Research Institute and University Jaume I (UJI). In addition, sewage water samples from four STPs (Utrecht, Apeldoorn, Amsterdam, Eindhoven) were also analysed by the University of Antwerp (UA).

Figure S1 of the Supplementary Information (SI) presents an overview of the sampling locations. Samples were collected from 65 sites and corresponded to three different types of water:

(1) Surface water: samples were collected at all nine surface water intake points for drinking water production in the Netherlands. Eight of these locations were part of the Meuse and Rhine river basins, and one was part of the Ems river basin. In addition, samples were taken at five locations along the rivers Rhine and Meuse.
(2) Raw water and finished drinking water: samples were taken at ten production sites where drinking water is produced from surface water and another seven drinking water production sites where drinking water is produced from river bank filtration. Raw water refers to the source water that enters the drinking water production facility. At some production sites this raw water has undergone pre-treatment, e.g., direct filtration, subsoil passage in the dune areas or storage in a reservoir, before it enters the drinking water treatment plant. Finished drinking water refers to water that is distributed as tap water. Drinking water treatment mostly consists of a combination of coagulation/flocculation and filtration/flotation, UV/H_2O_2 treatment or ozonation followed by activated carbon filtration.
(3) Sewage water: influent and effluent water samples were collected from eight STPs. The size of these conventional