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Research article

Personal care products in surface, ground and wastewater of a complex aquifer system, a potential planning tool for contemporary urban settings

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ABSTRACT

The use and discharge of personal care products (PCPs) result in their presence in the aquatic environment. This study investigates the occurrence and fate of some PCPs in wastewater, surface and groundwater in an urbanized area in the North of Italy. We investigated four UV filters: phenylbenzimidazole sulfonic acid (PBSA), benzophenone-3 (BP3), benzophenone-4 (BP4) and 4 methylbenzilidine-camphor (4-MBC), and two antibacterial agents: triclosan (TCS) and triclocarban (TCC). BP3, BP4 and PBSA were detected in all WWTPs and concentrations ranged 27–822 ng/L (BP4 > PBSA > BP3). TCS was the only disinfectant detected in wastewater and ranged from <0.2 to 1690 ng/L. Removal efficiencies in WWTPs were good for BP3 and TCS (80–100%), but were quite low for PBSA and BP4 (0–40%). Consequently, PBSA and BP4 were the most abundant substances in surface water, detected up to 560.4 ng/L. TCS was also found in surface water (<0.2–161.0 ng/L). Only PBSA and TCS were found in untreated groundwater, and levels were higher in wells close to rivers, suggesting the contribution of surface water to this contamination, but not from the catchment and the sewer networks. These PCPs were confirmed to be ubiquitous in all the aquifers sampled, being reliable descriptors of human presence. The use of these data as direct indicators of pollutant's loads for the aquifers deriving from human presence could provide early warnings on chemicals that are continuously introduced into surface waters, identifying dynamic urban trends and suggesting paths for the planning in urban regions and for appropriate investment and rehabilitation strategies of infrastructure.

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1. Introduction

The growing environmental stress from modern cities and territories is an increasing alarming topic, starting to affects the way how urban planners consider ecology during the planning process (Forman, 2008). Currently 40% of the cities have an adjacent water supply and urban expansion has direct effects on the aquifer. There is a potential risk of overwhelm sewage wastewater systems, reducing their effectiveness, posing a pollution threat to the receiving water bodies, groundwater pollution and soil contamination. Wastewater, in fact, contains not only microorganisms, organic matter, and excess nutrients, but a wide range of contaminants, including a variety of pharmaceuticals and hormones, pesticides, drugs of abuse, toxic trace elements and metals, total suspended solids (TSS) and many more classes of substances. Urban regions, with their heavy anthropic pressure on the territory increase the discharge and accumulation of pollutants in the environment.

Personal care products (PCPs) are one of the classes of contaminants found ubiquitously in environmental waters as a result of the anthropic impact, with the potential for endocrine disruption and developmental toxicity (Nohynek et al., 2010) (Brausch and Rand, 2011) (Liu and Wong, 2013) (Richardson and Ternes, 2014). This group of emerging contaminants includes different organic substances such as antimicrobial agents/disinfectants, synthetic musk/fragrances, insect repellants, preservatives and sunscreens/

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UV filters.

Among PCPs, UV filters are considered very important because of their widespread use (Liu and Wong, 2013) to protect the skin from sun exposure absorbing ultraviolet radiation during exposure to the sun (Latha et al., 2013). Growing concern about the UV consequences for human health (e.g melanoma or other skin cancers) has boosted the use of sunscreen products with high sun protection factors, making them important for their extensive use (Latha et al., 2013). Consequently, the amounts discharged in the environment have increased too and several studies investigated their potential impact on environmental media (Gago-Ferrero et al., 2012) (Tsui et al., 2014b) and possible effects on human health (Gilbert et al., 2013). They enter the environment indirectly from wastewater treatment plant (WWTPs), which collect wastewater after daily personal hygiene (bathing, shaving, toilet use), and directly from people swimming and bathing in river and lakes. These compounds also bioaccumulate in fish due to their lipophilicity and stability (Poiger et al., 2004) and some of them have potential estrogenic activity (e.g 4-MBC) (Kunz and Fent, 2006) or may pose a risk for aquatic species (Fent et al., 2010).

The presence of these substances in aquifers and in WWTPs influents and effluents is amply documented in recent reviews (Ramos et al., 2015) (Ramos et al., 2016) (Wilkinson et al., 2017) (Montes-Grajales et al., 2017) (Yang et al., 2017) (Tanwar et al., 2014) and in a book dedicated to PCPs (Díaz-Cruz and Barceló, 2015) as is their removal during wastewater treatment processes (Balmer et al., 2005) (Kasprzyk-Hordern et al., 2009) (Sun et al., 2014) (Tsui et al., 2014a). UV filters concentrations in WWTPs influents range from few ng/L to μg/L and show seasonal variations with higher concentrations in warmer seasons (Balmer et al., 2005). Their incomplete removal in WWTPs seems an important discharge pathway to the aquatic environment (Kasprzyk-Hordern et al., 2009) where they can be found in surface water (e.g. rivers and lakes) (Kasprzyk-Hordern et al., 2008) and can ultimately reach groundwater (Jurado et al., 2014). UV filters and their distribution have been reported in the marine environment (Tsui et al., 2014a).

Antimicrobial agents are another major class of PCPs that are used in a number of household and PCPs products, including cosmetics, liquid hand soap, deodorant bar soap, sponges, toothpaste and cutting boards, as well as shoes, towels and clothes. Triclosan (TCS) and triclocarban (TCC) are the antimicrobial agents most frequently detected in WWTPs influents and effluents (Halden and Paull, 2005). Their incomplete removal leads to their distribution in surface water at concentration ranging from the detection limits (low ng/L) up to the μg/L levels (Brausch and Rand, 2011). The environmental concentrations of antimicrobial agents in some cases exceed toxic threshold values for certain aquatic organisms such as algae and crustacea (Chalew and Halden, 2009). Both TCS and TCC have been shown to bioaccumulate in algae (Coogan et al., 2007) and to persist in the environment, particularly under anaerobic conditions (Ying et al., 2007). There is evidence that TCS has weakly estrogenic properties (Ishibashi et al., 2004). It was listed in 1986 in the European Community Cosmetics Directive (76/768/EEC) to regulate its use as preservative in cosmetic products at a maximum concentrations of 0.3% and its safety in use is constantly evaluated by the EU Scientific Committee on Consumer Products (European Commission and Directorate General for Health and Consumers, 2009) (European Commission and Directorate General for Health and Consumers, 2011). Since March 2010 TCS can no longer be used as surface biocide in food contact material in Europe and recent studies showed a possible influence of this compound on the spreading of antimicrobial resistance (European Commission and Directorate General for Health and Consumers, 2010).

UV filters and antimicrobial agents are ubiquitous emerging pollutants and very little data are currently available in Italy on the occurrence of these compounds in different water matrices and are limited to the south area of the country (Celano et al., 2014) and there is a clear need to characterize the sources and environmental behavior of these substances.

The study investigated the occurrence and fate of organic UV filters and antibacterial agents in wastewater, surface and groundwater in order to provide information on their sources, the mass loads flowing into the territory and the aquifer vulnerability to pollutants migration. This the most urbanized area in Italy, comprising all the provinces around, which border the city, and functioning in an integrated manner with large geographical and functional networks, like the water system. Here the complex cooperation between administrative organizations adds difficulties in managing resources and in strategic planning for the goods that are in common, like the aquifer system.

We collected the receiving surface water before and after the main point of discharge of WWTPs. Groundwater was collected during two comprehensive measurement campaigns of three different aquifer levels, partially separated by aquitards, in the urban territory. These results might potentially be a useful tool for large-scale, long-term, urban planning, if employed as direct indicators of pollutant's loads for the aquifers deriving from human presence.

2. Experimental

2.1. Selection of the analytes

In this study the selection of the analytes was made among the most relevant PCPs taking into account their presence and persistence in the aquatic environment documented in previous studies (Brausch and Rand, 2011) and their ecological and toxicological relevance. Benzophenone-3 (BP3), benzophenone-4 (BP4) and 4-methylbenzylidene camphor (4 MBC) are ubiquitous and frequently detected in WWTP effluents and surface waters worldwide (Brausch and Rand, 2011) (Jurado et al., 2014). BP3 is responsible of endocrine alteration in fish also at low concentration (Blüthgen et al., 2012). 4-MBC can accumulate in fish tissues (Balmer et al., 2005), can affect the benthic invertebrates reproduction (Schmitt et al., 2008) and seems to have estrogenic activity (Kunz and Fent, 2006). Phenylbenzimidazole sulfonic acid (PBSA) was selected because it is not totally removed during wastewater treatments and can therefore be discharged in the environment (Rodil et al., 2012). A recent study reported that the exposure to this compound can alter biochemical parameters and enzyme activities in fish plasma (Grabicova et al., 2013). TCS (TCS) and TCC (TCC) were selected because they are frequently detected in wastewater and TCS has potential implication with the spread of antibiotic resistance (European Commission and Directorate General for Health and Consumers, 2009).

2.2. Sampling sites

2.2.1. Wastewater

Wastewater samples were collected from three WWTPs located in the city of Milan (Fig. 1). They receive urban wastewaters from the whole city and from part of its metropolitan area. WWTP A collects wastewater from the central and eastern part of Milan, WWTP B receives wastewater from the western part of the city and WWTP C collects 10% of wastewater from the eastern area of the city and additional wastewater from the surrounding area. The population equivalents served by these plants ranged from 1,100,000 to 250,000 (estimated from biological oxygen demand) and the volume of treated wastewater ranged on average between

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