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Review

Long lasting perfume — A review of synthetic musks in WWTPs

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ABSTRACT

Synthetic musks have been used for a long time in personal care and household products. In recent years, this continuous input has increased considerably, to the point that they were recognized as emerging pollutants by the scientific community, due to their persistence in the environment, and hazardous potential to ecosystems even at low concentrations. The number of studies in literature describing their worldwide presence in several environmental matrices is growing, and many of them indicate that the techniques employed for their safe removal tend to be ineffective. This is the case of conventional activated sludge treatment plants (WWTPs), where considerable loads of synthetic musks enter mainly through domestic sewage.

This review paper compiles and discusses the occurrence of these compounds in the sewage, effluents and sludge, main concentration levels and phase distributions, as well as the efficiency of the different methodologies of removal applied in these treatment facilities.

To the present day, it has been demonstrated that WWTPs lack the ability to remove musks completely. This shows a clear need to develop new effective and cost-efficient remediation approaches and foresees potential for further improvements in this field.

1. Introduction

The expansion of industrial activities has promoted an increase of environmental pollution, mainly due to the generation of considerable amounts of waste. Therefore, quality control became an extremely important issue for the scientific community, particularly directed to the safety of our surrounding environment. In the last years, however, the focus of environmental research has been broadened from conventional priority pollutants such as polycyclic aromatic hydrocarbons (PAHs), pesticides or polychlorinated biphenyls (PCBs) to a continuous surge of emerging micropollutants (Bu et al., 2013). Among these compounds, personal care products (PCPs) are one of the most important groups (Liu et al., 2010; Polo et al., 2007), which includes antimicrobial agents, insect repellents, preservatives, UV filters and fragrances (Liu and Wong, 2013; Richardson et al., 2005).

Synthetic musk fragrances are PCPs with widespread use, incorporated in several personal care and household products (e.g. lotions, perfumes, shampoos, washing powders, softeners, air fresheners) as fragrance additives and fixative elements (Ramírez et al., 2011; Zhang et al., 2008). They are usually divided into 4 main groups according to their chemical structure: nitro, polycyclic, macrocyclic and alicyclic musks. Due to their potential toxicity, namely phototoxic, neurotoxic, carcinogenic and oestrogenic activity (Hu et al., 2011; Polo et al., 2007), most nitromusks were phased out from the market. In fact, in Europe, musk ambrette (MA), musk moskene (MM) and musk tibetene (MT) were banned from cosmetic products, while the use of musk xylene (MX) and musk ketone (MK) is restricted (European Parliament, 2009) due to suspected carcinogenic effects at high concentration levels (Polo et al., 2007). Recently, the European Commission under the new chemical regulation REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals), considered MX a very persistent and very bioaccumulative (vPvB) substance and therefore decided to ban it as well (European Commission, 2011).

Abbreviations: 2-AMK, 2-amino musk ketone; 2-AMX, 2-amino musk xylene; 4-AMX, 4-amino musk xylene; ADBI, celestolide; AETT, versalide; AHMI, phantolide; AHTN, tonalide; ATII, traseolide; DPMI, cashmeran; HHCB, galaxolide; HHCB-lactone, galaxolidone; HRT, hydraulic retention time; K_{OW}, octanol-water partition coefficient; MA, musk ambrette; MC4, muskonate; MK, musk ketone; MM, musk moskene; MT, musk tibetene; Musk-NN, ethylene brassylate; MX, musk xylene; OTNE, orbitone; PAHs, polycyclic aromatic hydrocarbons; PCBs, polychlorinated biphenyls; PCPs, personal care products; REACH, Registration, Evaluation, Authorisation and Restriction of Chemicals; SRT, solids retention time; STRB, sludge treatment reed bed; UV, ultraviolet; vPvB, very persistent and very bioaccumulative; WWTP, wastewater treatment plant.

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With the decreasing use of nitromusks, polycyclic musks became the most important commercial synthetic musks and currently dominate the global market. The most representative compounds of this class are galaxolide (HHCB), tonalide (AHTN), celestolide (ADBI), phantolide (AHMI), traseolide (ATTI) and cashmeran (DPMI). Nevertheless, recent studies reported their potential oestrogenic and anti-oestrogenic effects (Hu et al., 2011; Toivanen et al., 2008; Yamauchi et al., 2008) and presence in different environmental compartments. For this reason, their use in cosmetic products is currently under discussion. In contrast, macrocyclic musks (e.g. ethylene brassylate, exaltolide) represent only a small fraction of the market (3–4%) and are almost exclusively used in perfumes, given their relatively high production costs. Moreover, having a more intense odour, smaller quantities are needed to obtain the same performances as other synthetic musks. These compounds show a similar chemical structure to the musks of natural origin, and hence seem to be more environmental-friendly, with greater degradability in the environment than the previous classes (Vallecillos et al., 2012a). Alicyclic musks are the 4th generation of musk odourants and are known as the linear musks (e.g. cyclomusk, helvetolide; Arbulu et al., 2011). Their use in personal care products is still very limited. As mentioned previously, synthetic musks are incorporated in products used in our everyday life (Correia et al., 2013; Homem et al., 2013). Following their application, most of these compounds are released via household effluents, reaching the WWTPs (García-Jares et al., 2002). Due to their physicochemical properties, they are considered bio-accumulative, lipophilic and only partially biodegradable. For this reason, some of them, namely polycyclic musks, are not completely removed during wastewater treatment (Lv et al., 2010; Posada-Ureta et al., 2012; Zeng et al., 2005; Zhang et al., 2008) and are therefore frequently found in surface waters (e.g. Lv et al., 2009; Posada-Ureta et al., 2012; Yang and Ding, 2012). In fact, the application of sludge and/or biosolids to agricultural fields is a direct input of musks into the soil (e.g. Chase et al., 2012), and thus into the food chain (Hu et al., 2011), whereas the discharges of effluents are the major route for water and aquatic biota contamination (e.g. Duedahl-Olesen et al., 2005; Nakata et al., 2012; Reiner and Kannan, 2010). Apart from persistent or pseudo-persistent (due to continuous emission), musks are also semi-volatile, which may explain their detection in remote areas (long-range atmospheric transport; Arbulu et al., 2011; Ramírez et al., 2011). Therefore, this phenomenon also plays a role in the dispersion of musks in the environment.

With the increase of material published on these subjects, there is a lack of systematic investigation that can provide a good overview of the incidence and behaviour of musks in their main contamination route, the WWTPs. Bearing in mind the aforementioned information, the aim of this work is to present levels, describe the fate and trends of musks detected in the WWTPs, as well as to discuss the efficiency of the removal methodologies applied in these treatment facilities.

2. Methodology

Scientific publications regarding the occurrence and fate of musks in WWTPs between 1996 and 2014 were searched and presented in this work. This literature review was done using several available electronic databases: Scopus®, Elsevier®, Taylor & Francis®, ACS Publications®, Springer® and Google® Scholar.

3. Discussion

The WWTPs represent the main potential source of environmental contamination for musks, but at the same time the crucial point for remediation actions through the development and application of removal processes. For that reason, an increasing number of studies about musks in WWTPs have emerged in recent years (Fig. 1). These studies focused on the assessment of diverse procedural steps of WWTPs, with sampling strategies covering different matrices (sludge, wastewater and even the surrounding air). The analysis of these matrices can be challenging given their complexity and, for that reason, several analytical approaches have been developed.

3.1. Overview of WWTPs

Wastewater treatment plants were initially designed in the 20th century to remove pathogens, organic matter and nutrients from the final effluents (Amy et al., 2008), which should not be a new source of contamination. However, the demands on wastewater treatment systems have since increased dramatically, and nowadays the attention is focused on emergent pollutants (i.e. pharmaceuticals, personal care products, perfluorinated compounds, etc). These compounds have been detected in ng L^{-1} to mg L^{-1} concentration levels in WWTPs and to ensure that they will not affect the environment or pose a threat to human health, the implementation of new technologies and control measures is required (Amy et al., 2008; Weiner and Matthews, 2003). Conventional wastewater treatment generally involves four main stages: preliminary, primary, secondary and tertiary treatment.

In the preliminary treatment, elements on the influent flow prone to cause maintenance and operational problems in the system are removed. Afterwards, wastewater is conducted to a primary treatment that usually involves a physical process (Tchobanoglous et al., 2003; Weiner and Matthews, 2003), but chemicals (flocculants and coagulants) are sometimes added to enhance the removal of suspended solids and, to a lesser extent, dissolved solids. In the secondary treatment, biological processes are used to remove most of the biodegradable organic matter. At this stage, different methods can be employed, but trickling filters (percolated fixed beds) and activated sludge systems are the most common. In some WWTPs, this is the final stage of the process and the effluent is ready to be released into the environment. There are, however, cases where additional processes are applied – tertiary treatment – to further improve its quality. The objective is to remove residual suspended solids and some specific contaminants, such as nutrients and toxic substances (e.g. nitrogen, phosphorous, heavy metals), by filtration over activated carbon or reverse osmosis, chlorination, ozonation, degradation with UV-light and biological nutrient removal (Tchobanoglous et al., 2003; Weiner and Matthews, 2003).

Within the whole WWTP processes, sludge is produced from nearly all treatment phases, and must be treated before disposal. In order to reduce their volume, a thickening and/or dewatering treatment is applied, and a stabilization process to reduce problems

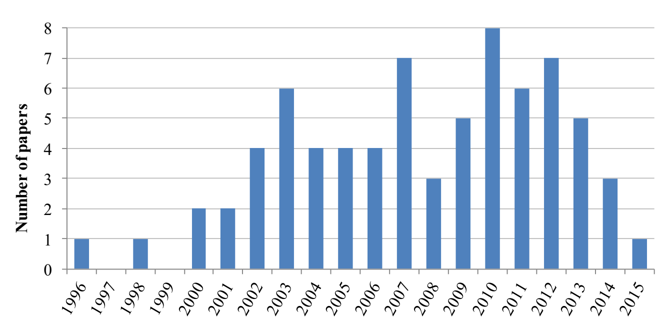

Fig. 1. Number of publications per year studying synthetic musks in WWTPs.