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Review

Effect of polycyclic musk compounds on aquatic organisms: A critical literature review supplemented by own data

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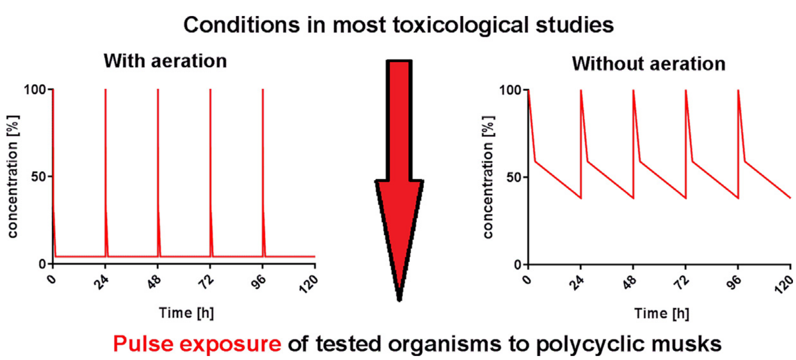
HIGHLIGHTS

- Polycyclic musks are aromatic substances extensively used in personal care products.
- Ubiquitous occurrence, pseudo-persistence, tendency for bioconcentration
- They tend to volatilize from experimental baths under laboratory conditions.
- The tested organisms are mostly exposed to concentration pulses.
- Toxicity of polycyclic musks for aquatic organisms may be underestimated.

GRAPHICAL ABSTRACT



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ABSTRACT

Synthetic musk compounds are extensively used in personal care and cosmetic products around the world. Because they are not completely removed in sewage treatment plants, they eventually end up in aquatic environments. The aim of this review was to summarize published information on effects of polycyclic musks on aquatic organisms and to discuss whether the experimental design of toxicological studies involving these substances could influence the results obtained. With the exception of one study run in a flow-through system, all published toxicological studies on synthetic polycyclic musks have been conducted in semi-static or even static systems. Based upon data in the literature and our own results, we conclude that in toxicological tests with semi-static set-ups, concentrations of polycyclic musks decrease with time between bath exchanges, and, as a result, tested organisms are not being exposed to stable concentrations but rather to concentration pulses. The duration and character of these pulses are influenced mainly by aeration of experimental baths, as polycyclic musks have a tendency to volatilize from water baths. Under semi-static conditions, tested organisms may be subjected to lower concentration of the tested substance for relatively long periods. Those levels may even fall below the limits of quantification. During these periods, some level of detoxification and/or elimination (depuration) of the toxicant may reduce toxic effect of the previous exposures. Consequently, toxicity of polycyclic musk substances for aquatic organisms obtained under these conditions may be underestimated. Based upon existing data in the literature, therefore, it is very difficult to correctly estimate risk of polycyclic musks to aquatic organisms.

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1. Introduction

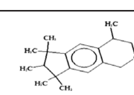
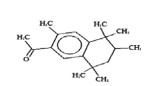
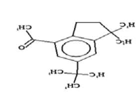
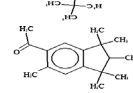
In recent decades, attention has been directed to contamination of aquatic environments by various pollutants of anthropogenic origin, including synthetic musk compounds. To utilize their typically musky scent, these compounds have been synthesized to replace expensive natural fragrances. Synthetic musks include a broad range of compounds having similar properties that can be divided into four main groups according to their chemical structures: nitro, polycyclic, macrocyclic, and alicyclic musk fragrances. Polycyclic synthetic musks are the most widely used today (Vallecillos et al., 2015), mainly as fragrance ingredients in numerous products in daily use, such as perfumes, cosmetics, body lotions, soaps, shampoos, toothpaste, antiperspirants, laundry detergents, fabric softeners, household cleaners, air fresheners, and other scented household products (Gautschi et al., 2001; Nakata et al., 2015). Although the proportion of musk in products is usually below 2% (Martínez-Girón et al., 2010), their discharging through wastewater could give rise to persistent pollution problems in natural waters because the consumption of fragrant products is huge all over the world (Fang et al., 2017). Worldwide, polycyclic musks production increased from 4300 tons in 1987 (61% of the market) to 5600 tons in 1997 (71% of the market; Rimkus, 1999), and to 6000 tons in 2004 (1000 tons of galaxolide and 5000 tons of tonalide Lange et al., 2015). Tonalide and galaxolide are the dominant types produced. In the first decade of the 21st century, they comprised *ca* 95% of the European market and *ca* 90% of the North American market for all polycyclic musks (Reiner et al., 2007). For this reason, both compounds have been included on the United States Environmental Protection Agency's high production list, which includes chemicals with annual production >4500 tons (USEPA, 2006). Because the polycyclic musk versalide was banned in the 1980s for reasons of its adverse neurotoxic effects (Heberer, 2002), it has not been included in this review.

2. Classification and physicochemical properties of polycyclic musks

Polycyclic musks are compounds with molecular formulas of $C_{17}H_{24}O$ (celestolide, phantolide) or $C_{18}H_{26}O$ (galaxolide, tonalide, traseolide) containing one central aromatic benzene ring adjacent to at least one pentane (in the case of indane derivatives) or hexane (isochroman and tetralin derivatives) ring, highly substituted mainly by methyl groups (Table 1) (Rimkus, 1999). All polycyclic musks exert low water solubility. Logarithms of the *n* octanol water partition coefficient (log K_{OW}) are high, ranging from 5.7 (tonalide) to 6.31 (traseolide), and thus show the lipophilic nature of these substances (Table 2). Therefore, they are presumed to bioconcentrate in organisms (Table 2). Bioaccumulation is made more intensive by their low rates of biological and chemical degradation in the environment, which causes their high relative persistence (Heberer et al., 1999). In-river half-life of the two most used polycyclic musks, galaxolide and tonalide, was estimated at 15 and 67 days (Bester, 2005).

The majority of scents and odours are volatile organic compounds (VOCs). The VOCs Solvents Emissions Directive of the European Union (EC, 1999) classifies VOCs as organic compounds with vapour pressure higher than or equal to 0.01 kPa measured at 20 °C. Although polycyclic

Table 1
Basic characteristics of synthetic polycyclic musks.

Musk	Abbreviation	Derivative	Commonly used chemical name	Chemical (IUPAC) name	CAS number	EC number	Molecular formula	Structure
Galaxolide (Abbalide; Pearlide; Chromanolide)	HHCB	Isochroman	1,3,4,6,7,8 Hexahydro 4,6,6,7,8,8 hexamethyl cyclopenta[*g*] benzopyran	4,6,6,7,8,8 Hexamethyl 1,3,4,7 tetrahydrocyclopenta[*g*] isochromene	1222-05-5; 214-946-9	214-946-9	$C_{18}H_{26}O$	
Tonalide (Fixolide; Tetralide; Musk Plus; Tentarome; Astralide)	AHTN	Tetralin	7 Acetyl 1,1,3,4,4,6 hexamethyl 1,2,3,4 tetrahydronaphthalene	1 (3,5,5,6,8,8 Hexamethyl 6,7 dihydronaphthalen 2 yl) ethanone	21145-77-7; 1506-02-1	216-133-4; 244-240-6	$C_{18}H_{26}O$	
Celestolide (Crysolide)	ADBI (DTI)	Indane	4 Acetyl 1,1 dimethyl 6 *tert*-butyldihydroindene	1 (6 *tert* butyl 1,1 dimethyl 2,3 dihydroinden 4 yl)ethanone	13171-00-1	236-114-4	$C_{17}H_{24}O$	
Phantolide	AHDI (AHMI)	Indane	6 Acetyl 1,1,2,3,3,5 hexamethyl dihydroindene	1 (1,1,2,3,3,6 Hexamethyl 2H inden 5 yl)ethanone	15323-35-0	239-360-0	$C_{17}H_{24}O$	
Traseolide	ATII	Indane	5 Acetyl 1,1,2,6 tetramethyl 3 isopropyl dihydroindene	1 (1,1,2,6 Tetramethyl 3 propan 2 yl 2,3 dihydroinden 5 yl) ethanone	68140-48-7	268-799-0	$C_{18}H_{26}O$