Science of the Total Environment 562 (2016) 13–25

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

A first screening and risk assessment of pharmaceuticals and additives in personal care products in waste water, sludge, recipient water and sediment from Faroe Islands, Iceland and Greenland

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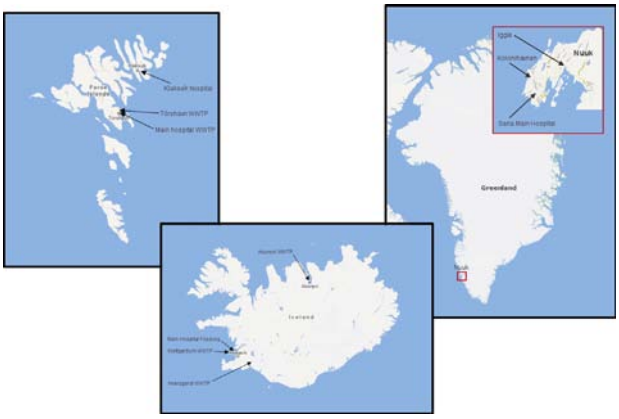
HIGHLIGHTS

- Analysis of a wide spectrum of selected PPCPs for the first time in the sub-arctic
- Identification of a high number of PPCPs in WWTPs and recipients from FO, IS and GL
- Highest ecotoxicological risk was found stemming from discharge of surfactants.
- High ecotoxicological risk from discharge of additives in personal care products

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 8 January 2016
Received in revised form 1 March 2016
Accepted 2 March 2016
Available online xxxx

Editor: Jay Gan

ABSTRACT

A screening of a broad range of pharmaceuticals and additives in personal care products (PPCPs) in sub-arctic locations of the Faroe Islands (FO), Iceland (IS) and Greenland (GL) was conducted. In total 36 pharmaceuticals including some metabolites, and seven additives in personal care products were investigated in influent and effluent waters as well as sludge of waste water treatment plants (WWTPs) and in water and sediment of recipients. Concentrations and distribution patterns for PPCPs discharged via sewage lines (SLs) to the marine environment were assessed. Of the 36 pharmaceuticals or metabolites analysed 33 were found close to or above the limit of detection (LOD) in all or a part of the samples. All of the seven investigated additives in personal

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http://dx.doi.org/10.1016/j.scitotenv.2016.03.063
0048-9697/

Keywords:
PPCPs
Hormones
WWTP
Recipient
Waste water
Ecotoxicological risk assessment

care products were detected above the LOD. Some of the analysed PPCPs occurred in every or almost every sample. Among these were diclofenac, ibuprofen, lidocaine, naproxen, metformin, citalopram, venlafaxine, amiloride, furosemide, metoprolol, sodium dodecyl sulphate (SDS) and cetrimonium salt (ATAC-C16). Additionally, the study encompasses ecotoxicological risk assessment of 2/3 of the analysed PPCPs in recipient and diluted effluent waters. For candesartan only a small margin to levels with inacceptable risks was observed in diluted effluent waters at two locations (FO). Chronical risks for aquatic organisms staying and/or living around WWTP effluent pipe-outlets were indicated for 17β-estradiol and estriol in the three countries. Additives in PCPs were found to pose the largest risk to the aquatic environment. The surfactants CAPB and ATAC-C16 were found in concentrations resulting in risk factors up to 375 for CAPB and 165 for ATAC-C16 in recipients for diluted effluents from Iggia, Nuuk (GL) and Torshavn (FO) respectively. These results demonstrates a potentially high ecological risk stemming from discharge of surfactants as used in household and industrial detergents as well as additives in personal care products.

1. Introduction

In recent years, pharmaceuticals and additives in personal care products (PPCPs) and their metabolites are reason for concern due to their occurrence and fate in the aquatic environment (Kümmerer, 2004; Vieno et al., 2007; Kallenborn et al., 2008; Azzouz and Ballesteros, 2013) as many of them have adverse ecological health effects at low concentrations (Halling-Sørensen et al., 1998; Jørgensen and Halling-Sørensen, 2000; Heberer, 2002; Kümmerer, 2004). Limited information is available about synergistic, additive and antagonistic effects of complex compound mixtures and their in vivo effects in the ecosystems (Cleuvers, 2003). Fate, transport and, in a certain degree, also emission patterns of a broad range of PPCPs in natural aquatic environments is poorly understood. The main route of PPCPs and hormones into the aquatic ecosystems is via discharge of effluents from wastewater treatment plants (WWTPs) to adjacent recipients as rivers, lakes or the sea. WWTPs were initially not designed to remove specifically these above mentioned compounds, whereas their main purpose was to remove organic matter, both solid and dissolved (i.e. mainly fats and nutrients), from WWTP influent waters. As a result, PPCPs and hormones may only be partly eliminated by sorption to particles and biodegradation (Vieno et al., 2007; Azzouz and Ballesteros, 2013); thus, compounds that are not removed are being discharged to the recipient ecosystem where they may impact non-target organisms. However, incomplete removal of several PPCPs and their metabolites from waste waters has been observed in a range of WWTPs with different methods of waste water treatment (Vieno et al., 2007; Tauxe-Wuersch et al., 2005; Kanda et al., 2003; Ternes, 1998). In addition to the properties of the investigated compound itself, other factors that will affect the elimination rate of the PPCPs in sewage treatment (Vieno et al., 2007) are for example type of the treatment process (Kanda et al., 2003; Joss et al., 2006), degree of dilution of raw sewage (Ternes, 1998; Tauxe-Wuersch et al., 2005; Joss et al., 2006), temperature (Vieno et al., 2005, Castiglioni et al., 2006; Azzouz and Ballesteros, 2013), solids retention time (Kreuzinger et al., 2004; Clara et al., 2005) and hydraulic retention time (Tauxe-Wuersch et al., 2005). Also, seasonal variations together with individual geographical climate conditions are influencing the removal efficiency with biological and chemical treatment processes, bio- and photo-degradation and sorption to particles (Vieno et al., 2005; Kallenborn et al., 2008; Azzouz and Ballesteros, 2013). In the Nordic countries, and especially in the Arctic and sub-Artic, the cold and harsh climatic conditions impose challenges regarding design and operation of WWTPs. Pharmaceutical residues degrade significantly slower in the high latitude of the Nordic aquatic environment than in similar facilities in lower latitudes. Removal by photodegradation is retarded during the winter when sunlight in the Arctic is limited, and the low temperatures in the Arctic slow down the rate of degradation in the environment (Kallenborn et al., 2008).

In some areas of the Arctic and the sub-Arctic, treatment of waste water is inadequate or completely lacking, as for example in Greenland where no treatment facility for industrial or domestic waste water exists (Gunnarsdóttir et al., 2013). Discarded and excreted PPCPs and hormones are discharged without any clearance. In Iceland, the most significant development has occurred in the metropolitan area of the capital Reykjavik, where almost all of the inhabitants have acceptable sewage treatment (Environment Agency of Iceland, 2013). Some municipalities around the coastline still discharge untreated sewage into the fresh water and marine environment. Sewage treatment is mostly conducted as primary treatment including mechanical means to lower the content of suspended particles, which is permitted in less sensitive coastal areas. Secondary treatment with biological treatment processes is required inland and by the seaside in areas which have not been classified as less sensitive (Environment Agency of Iceland, 2013). In the Faroe Islands, the sewage treatment is based on primary treatment in sedimentation tanks, which may be privately installed for each household or a few households shared or installed by the municipality for groups of houses in a neighbourhood. The effluents from these sedimentation tanks are discharged to the nearshore marine area.

During the preparation of a review on available information of presence, concentrations and distribution patterns of PPCPs in the Nordic Countries (Nordic Council of Ministers, 2012) it became evident that such data were either not or only very sparsely available for the Faroe Islands, Iceland and Greenland. The island societies are similar in that they are surrounded by vast expanses of ocean with long distances to heavily industrialised areas in Europe and North America. Traditionally, the economies were heavily based on marine resources. The dependence on income from fish export is expected to increase with increasing aquaculture. Further, tourism is growing and important income source in parts of this area whereas in other parts it represents a sector whose growth potential is assumed to be substantial (Sméradóttir et al., 2014). With these two main economic sectors, the quality of the marine environment is of utmost importance for these societies. At present, the enormous dilution factor of the recipients of the WWTPs compared to the relative low number of inhabitants, and in many instances also the low population density, has provided a setting and an assumption, where the discharge of organic waste (i.e. waste water) would likely not pose harm to the natural environment; even when applying none or only a first tier purification prior to discharge. However, a main underlying presumption for utilising the purification capacity of the natural environment is that the waste consists mainly of potential nutrients. The validity of this assumption decreases when the waste contains traces of persistent and/or biologically active and potentially toxic compounds as PPCPs and hormones. With an ageing population and increased pressure on the public health care system, the application of pharmaceuticals, whether prescription drugs or over-the counter medicine, as well as the application of personal care products may be assumed to increase rather than decrease as the economic situation improves. The discharge of pharmaceuticals which may exert pharmaceutical activity to non-target organisms together with a long lifetime of several substances in aquatic environments raises the need to investigate the possible occurrence of PPCPs in these coastal areas.

The main aim of the present study was to assess concentrations and distribution patterns for PPCPs and hormones discharged via sewage lines to the marine environment in Faroe Islands, Iceland and