Environ Sci Pollut Res (2018) 25:33001–33013
DOI 10.1007/s11356-017-9726-6

Pharmaceuticals and personal care products (PPCPs) in Arctic environments: indicator contaminants for assessing local and remote anthropogenic sources in a pristine ecosystem in change

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Received: 16 January 2017 / Accepted: 6 July 2017 / Published online: 31 July 2017

Abstract A first review on occurrence and distribution of pharmaceuticals and personal care products (PPCPs) is presented. The literature survey conducted here was initiated by the current Assessment of the Arctic Monitoring and Assessment Programme (AMAP). This first review on the occurrence and environmental profile of PPCPs in the Arctic identified the presence of 110 related substances in the Arctic environment based on the reports from scientific publications, national and regional assessments and surveys, as well as academic research studies (i.e., PhD theses). PPCP residues were reported in virtually all environmental compartments from coastal seawater to high trophic level biota. For Arctic environments, domestic and municipal wastes as well as sewage are identified as primary release sources. However, the absence of modern waste water treatment plants (WWTPs), even in larger settlements in the Arctic, is resulting in relatively high release rates for selected PPCPs into the receiving Arctic (mainly) aquatic environment. Pharmaceuticals are designed with specific biochemical functions as a part of an integrated therapeutically procedure. This biochemical effect may cause unwanted environmental toxicological effects on non-target organisms when the compound is released into the environment. In the Arctic environments, pharmaceutical residues are released into low to very low ambient temperatures mainly into aqueous environments. Low biodegradability and, thus, prolonged residence time must be expected for the majority of the pharmaceuticals entering the aquatic system. The environmental toxicological consequence of the continuous PPCP release is, thus, expected to be different in the Arctic compared to the temperate regions of the globe. Exposure risks for Arctic human populations due to consumption of contaminated local fish and invertebrates or through exposure to resistant microbial communities cannot be excluded. However, the scientific results reported and summarized here, published in 23 relevant papers and reports (see Table S1 and following references), must still be considered as indication only. Comprehensive environmental studies on the fate, environmental toxicology, and distribution profiles of pharmaceuticals applied in high volumes and released into the Nordic environment under cold Northern climate conditions should be given high priority by national and international authorities.

Keywords Pharmaceuticals · Personal care products · PPCP · Arctic · AMAP · Arctic Monitoring and Assessment Programme · Environmental pollution · Regulation · Risk assessment

Responsible editor: Philippe Garrigues

Electronic supplementary material The online version of this article (doi:10.1007/s11356-017-9726-6) contains supplementary material, which is available to authorized users.

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Introduction

The number of priority pollutants identified in the Arctic environment is continuously increasing due to the rapid

development in modern trace-analytical technology for the ultra-trace level quantification of anthropogenic pollutants at ultra-trace levels. The list of relevant Arctic pollutants is comprehensively documented in the newly published reports of the Arctic Monitoring and Assessment Programme (AMAP: www.amap.no). AMAP is regularly reporting on Arctic pollution issues since the early 1990s (Hansen 1998, Hansen 2000, Hansen et al. 2002, Heidam et al. 2004, Bakke et al. 2008, Bonefeld-Jorgensen 2010, Donaldson et al. 2010, Hung et al. 2010, Muir and De Wit 2010, Kallenborn et al. 2012, Hung et al. 2016). However, pharmaceuticals and personal care products (PPCPs) have not been considered as a priority pollutant group in Arctic environments until today. Most PPCPs are expected to be found close to the sources and the primary sources. Furthermore, all major contamination sources like sewage and domestic waste facilities are usually associated with densely populated regions (Daughton 2003a, Kanda et al. 2003, Xia et al. 2005, Lishman et al. 2006, Carballa et al. 2007, Moldovan et al. 2007). Recent studies, however, indicate that the decentralized character of local PPCP sources (i.e., sewage treatment) in combination with low-temperature Arctic climate and low technological standards for waste treatment facilities in Arctic settlements extends the environmental stability of this residues considerably compared to conditions found in lower latitude regions (Gunnarsdottir et al. 2013a, Weigel et al. 2004a, Bergheim et al. 2010). Therefore, national and regional authorities are now looking into the occurrence, distribution pathways and environmental fate of PPCPs in Arctic environments (Huber et al. 2016).

As a first attempt to summarize the current scientific knowledge on the occurrence and fate of PPCP in the Arctic, AMAP has conducted a comprehensive survey. A detailed report is published for regulators and authorities (AMAP, 2016). The major findings and conclusions are published here as scientific review as a scientific foundation for highly needed discussions on the expected implications of PPCP residues in the Arctic.

General considerations

In 2007, the Norwegian State Pollution Control Authorities (now Norwegian Environmental Agency), published a priority list for PPCP environmental monitoring based upon a comprehensive literature survey (Grung et al. 2007). Along these recommendations, the Nordic screening program (a pollutant screening program of the Nordic Council of Ministers (NCM)) initiated and funded several annual pollutant screening programs focusing on PPCPs in a variety of environmental matrices. Thus, several circum-Arctic nations, the Arctic Council, as well as NCM acknowledge today the imminent risk associated with the uncontrolled release of pharmaceutical agents and personal care products in the Arctic environment. In addition to research studies published in peer-reviewed scientific journals, several regional screening initiatives have been completed and reported (Carlson et al. 2013, Kleywegt et al. 2011, Lishman et al. 2006, Servos et al. 2005, Vieno et al. 2007, Vasskog et al. 2008, Vasskog et al. 2006, Huber et al. 2013a, Dye et al. 2007, Kaj et al. 2014, Lee et al. 2003, Lindqvist et al. 2005, Metcalfe et al. 2003, Weigel et al. 2004a, Kallenborn et al. 2008a, Mutter 2014, Kankaanpää et al. 2014, Thomas et al. 2014). Based on this literature information, the here reported first assessment was conducted. This review is based on references reporting PPCP contamination from locations within the entire AMAP monitoring area. For our review, we also included adjacent sub-Arctic regions (below 66° N latitude) like the entire Hudson Bay, Ontario, Manitoba, Alaska, the northwest coast of Norway, Southern and Western Greenland, Finland, Faroe Islands and Iceland, Northern Norway, Sweden, and Finland (Fig. 1). However, no information from the Eastern Arctic regions was available for our comparison.

Based upon this first comprehensive literature survey, it is safe to conclude that PPCPs have been identified and confirmed as relevant pollutants also in the Arctic environment. Their presence is reported in a variety of environmental compartments (Fig. 1). The identified compound groups are listed in Table 1.

Environmental implications

Pharmaceuticals applied in human medical and/or veterinary therapy are usually excreted unchanged and released directly in the municipal or hospital sewer systems. Also, uncontrolled discharge into municipal waste by patients or private household cannot be excluded as important source for environmental contamination in the Arctic. PPCP load is retained and/or eventually degraded into transformation products. Therefore, not only the target parent compounds but also the properties of the major transformation products must be included into an appropriate and comprehensive risk assessment of PPCPs in the Arctic environment (Daughton 2003b).

Usually pharmaceuticals are characterized by their therapeutic effects during medical treatment. A selection of pharmaceutical active substance groups as well as personal care products found in Arctic environments is listed in Table 2.

A total of 110 different PPCP-related substances have been identified in Arctic samples in research and screening studies published as scientific articles and reports (see references above). This already quite large number of PPCPs identified, however, reflects only the capability of today's analytical methods available for the quantification of these substances and not necessarily an exhaustive list of PPCPs present in the Arctic environment. The progress of technological

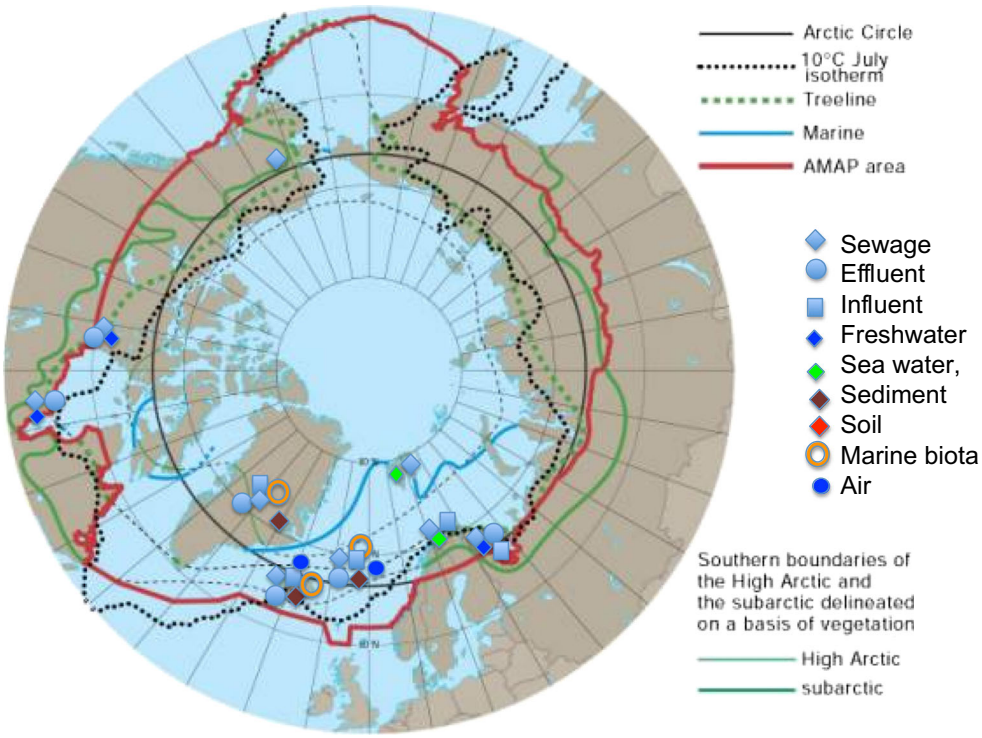

Fig. 1 Arctic/sub-Arctic locations and regions where PPCPs were found in environmental samples. The respective investigated environmental compartments are listed (figure modified with courtesy of AMAP)

development will, in the future, inevitably lead to the identification of new, hitherto unidentified PPCPs.

The majority of the compounds have been identified in raw sewage or related matrices (i.e., effluent, recipients). However, a substantial number of compounds were also found in freshwater, seawater, and biota, illustrating the inherent environmental risk posed by these environmental contaminants. Due to the expected high concentrations of target PPCPs in WWTP related samples, the majority of the here reported studies were conducted in these type of sample materials (sewage and effluent). The low trace concentration level for PPCPs expected in the recipient or in seawater is still a challenge for today's analytical methods.

Levels and pathways

PPCPs are produced as active agents for supporting general health and hygiene as well as disease treatment and therapeutic applications in medicine and veterinary medicine. In general, the human community structure in the Arctic is characterized by a decentralized, scattered distribution of minor settlements with a few cities as cultural and social centers. In addition, no production of PPCPs is reported from Western Arctic environments. Thus, potential release from production plants as potential pollutant source must be excluded. This is a different feature compared to middle latitude compared to middle latitude regions. These characteristic differences have considerable consequences for the release and fate of pharmaceutical residues in the Arctic environment. Installation of state-of-the-science small-scale/medium-scale WWTPs is usually economically not affordable for the small communities of the Arctic (Gunnarsdottir et al. 2013b). Therefore, sewage is often released more or less untreated from single households directly or via collecting processes into the aquatic environment. Due to their generally low environmental mobility, the presence of PPCP-related substances in the Arctic environment is restricted to the vicinity of defined local anthropogenic sources. However, as indicated in a recent investigation, some PPCP-related substances might even be transport over long distances (atmosphere, ocean currents), and thereby reaching Arctic environments from remote release sources. Such behavior is, i.e., reported for volatile siloxanes (Vorkamp and Rigét 2014). However, the majority of the PPCP residues identified as pollutants in Arctic environments are directly associated with human activities in the region (usage and application). Environmental transport over medium or long distances into Arctic environments is less relevant for PPCPs. Accordingly, the direct release from WWTPs, as well as release from households and diffusive seeping from disposal sites is considered today the predominant source for the presence of PPCPs release in the Arctic (Kummerer 2009c, Gunnarsdottir et al. 2013b). Consequently, most published studies on fate and distribution of selected PPCPs in the Arctic have focused on the compound-specific distribution originating from the direct release via sewage or uncontrolled disposal into receiving waters (Weigel et al. 2004a, Weigel et al. 2004b, Vieno et al. 2007, Vasskog et al.

Table 1 Compound groups and the respective environmental compartments PPCP are reported in

Compound groups (number of substances)	Atmosphere		Terrestrial		Freshwater			Marine			Waste
	Air	Snow	Soil	Biota	Water	Sediment	Biota	Water	Sediment	Biota	Sewage
NSAIDs (11)					X	X		X			X
Antidepressants/SSRI (10)					X	X		X			X
β (beta)-blockers (5)					X	X					X
Calcium channel blockers (1)						X					X
ACE blockers (3)						X					X
Angiotensin receptor antagonists (3)						X					X
Antiepileptics (1)					X						X
Antimicrobials (4)					X			X			X
Antibiotics (14)					X			X			X
Additives (5)	X		X		X					X	X
Fragrances (5)					X						X
Steroid hormones (5)					X	X					X
Lipid regulators (3)					X						X
Illicit drugs (4)					X						X
Stimulants (2)					X			X			X
Anti-cancer (2)											X
Artificial sweeteners (2)					X						X
Surface tension suppressors (6)					X	X	X				X
Hypnotics (1)											X
Antidiabetics (1)											X
Anticoagulants (2)						X					X
Diuretics (4)						X					X
Chelating agents (1)						X					X
UV-filters (3)											X
Bisphenol monomers (5)											X

A detailed compound list is provided in Table 2. The number of individual PPCPs for each class is given in parentheses

NSAID non-steroidal anti-inflammatory and antipyretic analgesic drugs, *SSRI* selective serotonin re-uptake inhibitors, *ACE* acetylcholine esterase

2008, Servos et al. 2005, Mutter 2014, Metcalfe et al. 2003, Lishman et al. 2006, Lindqvist et al. 2005). However, a few reports confirm the presence of selected PPCPs also in biota, sediment, soil, and air, thus identifying the potential for contaminant transfer across environmental compartments (Vorkamp and Rigét 2014, Bakke et al. 2008, Huber et al. 2013b). The preferred environmental compartment in which the target substances are likely to be found is, thus, mainly dependent on the environmental conditions and the substance-specific physical and chemical properties (volatility, reactivity, solubility, etc.)

In a first survey for 2001–2003 contacted in the medium-size Norwegian Arctic city of Tromsø (2002: 45,000 inhabitants), sewage sludge, sewage effluents, and receiving seawater samples were analyzed for selected PPCPs for the first time in the vicinity of the municipal several sewage treatment facilities (Weigel et al. 2004a). Already 15 years ago, the levels of ibuprofen, its metabolites, and caffeine in the detected surface seawater were high (medium ng/L range) despite strong sea currents in the receiving seawaters (Tromsø Sound). The tidal currents were originally expected to lead to effective dilution and dispersion of the effluent water which, i.e., contains PPCPs (Weigel et al. 2004a). However, the year-round low ambient water temperature and the consequently low degradation rates in combination with high emission volumes lead to the continuous presence of the target PPCPs in the surrounding seawater in the vicinity of the WWTP locations (Weigel et al. 2004a). Since hospital sewage is physically separated from household sewage by different sewage systems, source-specific emission differences were identified (Weigel et al. 2004a).

This first study confirmed that the cold Northern ambient conditions in the aqueous environment may have considerable influence on the environmental properties including slow microbial transformation of environmental pollutants such as PPCPs. In Tromsø, sewage samples from three WTTP facilities were collected. In all Tromsø WWTPs, sewage is only processed with primary

Table 2 PPCP concentrations in samples from the Pharmafate pilot study in 2007 (Kallenborn et al. 2008a)

Conc. (ng/L)	Oslo (VEAS)		Tromsø		Longyearbyen	
Target PPCPs	Effluent (n = 1)	Seawater (n = 2)	Effluent (n = 8)	Seawater (n = 8)	Effluent (n = 5)	Seawater (n = 2)
Ibuprofen	10	n.d.–52	448	n.a.	30–403	0.4–1
Hydroxy-ibuprofen	126	188–243	3614	n.a.	8–1398	2–34
Carboxy-ibuprofen	42	109–213	70,170	n.a.	411–34,028	6–26
Diclofenac	25	n.d.–48	78	n.a.	30–1074	1–4
Triclosan	11	n.d.	350	n.a.	28–803	2–2.3
Caffeine	23	5–96	n.a.	n.a.	501–50,704	24–41
Citalopram	238	n.a.	63–102	<LOQ	<LOQ	n.d.
Desmethyl-citalopram	310	n.d.	118–215	<LOQ	<LOQ	n.d.
Didesmetyl-citalopram	10	n.a.	6–10	n.d.	n.d.	n.d.
Fluoxetine	8	n.a.	1–5	n.d.	n.d.	n.d.
Norfluoxetine	2	n.a.	0.7–2.5	n.d.	n.d.	n.d.
Fluvoxamine	1	n.a.	0.8–1.7	n.d.	n.d.	0.5–0.8
Sertraline	8	n.a.	8–90	n.d.	n.d.	<LOQ
Desmethylsertraline	6	n.a.	n.d.	n.d.	n.d.	n.d.
Paroxetine	4	<LOQ	3–13	n.d.	n.d.	0.6–1.4
Tetracycline	n.d.	n.d.	n.a.	n.a.	0.6–1.1	n.d.
Trimethoprim	0.8–0.9	n.d.	n.a.	n.a.	0.07–0.15	n.d.
Sulfamethoxazole	0.2–0.3	n.d.	n.a.	n.a.	n.d.	n.d.

LOQ limit of quantification, *n.a.* not analyzed, *n.d.* not detected

treatment (filtration). The concentration of caffeine in the Tromsø sewage effluent (Weigel et al. 2004a) was considerably higher than reported for sewage effluent and river water from a major German city at that time (Hamburg, 900,000 inhabitants (Weigel et al. 2004a).

In a follow-up study, PPCP levels were determined in effluent water samples and receiving seawater from three typical WWTPs along a longitudinal gradient in Norway, from Oslo (60° N) Tromsø (70° N) and Longyearbyen, Svalbard (78° N) (Kallenborn et al. 2008a). The average levels for all target PPCPs from this study are summarized in Table 2. Local specific differences were found for PPCP distribution patterns and samples analyzed.

The high levels reported in the receiving seawater samples from Longyearbyen surpassed in some cases the PPCP levels found in Tromsø. As a follow-up of these above-described pilot studies, the Norwegian Pollution Control Authorities conducted a first literature survey on the occurrence of selected PPCPs in the environment. The survey provided a first overview of the current state of knowledge regarding usage, distribution patterns, and occurrence of these compounds in the Norwegian environment (Grung et al. 2007). Recommendations for compound prioritization and implementation into future monitoring programs were made. This also included recommendations for appropriate sampling and monitoring strategies. Information on sales volumes for the selected compounds in Norway and the other Nordic countries were included (Grung et al. 2007).

Due to these first findings, the NCM Nordic screening program initiated a series of screening studies on the environmental levels and distribution profiles of PPCPs (Kaj et al. 2014, Huber et al. 2013b, Grung et al. 2007, Dye et al. 2007). These studies covered the following matrices: waste water treatment plants (WWTPs)—influent, effluent water, and sludge; surface water, including seawater and freshwater; and freshwater and marine sediment samples (see Supplementary Material, Table S2). For WWTP recipients, samples included water samples from up- and downstream from fish farms, other fresh or seawaters, groundwater, in particular in the proximity of hospitals and farms (NCM 2012). However, when considering the number of investigated samples and PPCP compounds, the available data sets are still quite small and scarce in order to provide the needed scientific conclusions for sound regulation strategies.

A comprehensive survey on PPCPs in the Nordic environment is reported in a recent NCM report (NCM 2012). The focus of that study was on WWTPs, including sewage, sewage influent, sewage effluent water, as well as sediment and water samples and biota associated with the receiving environment of the WWTPs. Of all target PPCPs selected, a few compounds, such as diclofenac and ibuprofen, were detected in all samples in significant levels. Estrone (both a synthetic and naturally occurring estrogen) was detected in most of the analyzed samples.

Transformation and degradation

The transformation of PPCP compounds is mainly governed by their physical-chemical properties and the underlying environmental conditions. As previously discussed, cold seawater temperatures seem to delay degradation of PPCPs. The comparison of ibuprofen patterns in Tromsø with published data from Germany (Weigel et al. 2004) indicated that the carboxylated transformation product (Ibu-CX) was more stable in the cold seawater environment around Tromsø (annual average temperature 4–6 °C) compared to mid-latitude environments. The transformation of environmental pollutants, including PPCPs, is strongly influenced by ambient conditions including temperature, microbiological environment, as well as light conditions. These influences were also found to be important for the PPCPs identified during a subsequent study in Tromsø (Vasskog et al. 2006) where selected SSRI antidepressants including their transformation products were found in considerable concentrations in sewage effluent samples. Similar levels were also found in untreated sewage effluent collected in Longyearbyen (Svalbard; ca. 2200 inhabitants, 2008) for the same study, where comparable levels of antidepressants were measured (Vasskog et al. 2008). Thus, physical-chemical properties, consumption profiles (waste water profiles), the waste water treatment technologies applied including controlled and uncontrolled degradation/transformation, as well as the environmental conditions in the marine environment are important influencing factors for the final PPCP profiles identified in Arctic sewage effluent samples (Kallenborn et al. 2008a).

Based upon the above assumptions, a controlled laboratory experiment was performed focusing on the temperature-dependent microbiological transformation of benzylpenicillin applying the Zahn-Wellens test (ZWT) for microbiological degradation in raw sewage sludge (Bergheim et al. 2010). The temperature-dependent degradation of benzylpenicillin (Pen-G) was studied at 5, 12.5, and 20 °C during an experimental period extending from the OECD recommended standard 28 days testing period to a final duration of 42 days. In addition to the dissolved organic carbon content (DOC), the concentration levels of Pen-G as well as the major transformation products identified were monitored continuously by chromatography coupled to mass spectrometric detection during the entire experimental period. The comparison of DOC loss at different ambient temperatures revealed considerable temperature dependence for the Pen-G degradation rate. The maximum DOC loss was slowest at 5 °C and the total DOC degradation (considered as a measure of the available total microbial carbon source in the experiment) was not completely reached even after 42 days of the experiment (Bergheim et al. 2010). This finding is considered a clear indication for the temperature dependence of the biodegradation pathways for Pen-G.

In particular, direct excretion of antibacterial and antiviral agents, in combination with incomplete removal during the passage through the WWTP will lead to continuous high concentration release into the cold Arctic aqueous environment. This has the potential to lead to bacterial and viral resistance with severe consequences for human populations and the environment at concentrations already observed in ng/L level range (see Table S2). Many antimicrobial agents, including antibiotics, are semi-synthetic or synthetic derivatives of naturally occurring biologically active substances (i.e., antibiotics, hormones) and are observed to be persistent over time in the environment. The environmental persistence of these anthropogenic synthetic substances is easily explained by the fact that they have been specifically developed to retain activity and resist degradation by the biochemical activities of the organism until they reached the target location within the treated organism.

A large proportion of antibiotics consumed during medical treatment is usually excreted chemically unchanged (Hirsch et al. 1999, Bergheim et al. 2015, Kummerer 2009a, Kummerer 2009b, Kummerer 2003, Braschi et al. 2013, Milic et al. 2013, Ding and He 2010). The potential impact of the continual environmental release of antimicrobial substances in Arctic aqueous and marine coastal environmental is, thus, a shift in bacterial populations from being predominantly sensitive to pharmaceutically produced antimicrobials to predominantly resistant phenotypes. Such a shift would consequently lead to the potential transfer of genetic information (resistance determinants) into clinically relevant microbial pathogens (Kallenborn et al. 2008b). A currently published study explored and confirmed these mechanisms by using functional metagenomics to investigate the resistomes of bacterial communities isolated from different layers of the Canadian high Arctic permafrost (Perron et al. 2015). From selected bacteria strains sampled from ancient layers, eight genes conferring clinical levels of resistance against aminoglycoside, beta-lactam, and tetracycline antibiotics were isolated. In bacteria sampled from the overlaying active layer, another ten different genes were isolated and characterized conferring resistance to all six antibiotics tested in this study. The study concluded that the development of antibiotic resistance genes is functionally diverse and identified as important microbial defense mechanisms even prior to the anthropogenic use of antibiotics, contributing to the evolution of natural reservoirs of resistance genes (Perron et al. 2015).

Even triclosan, a commercially used chlorinated antimicrobial agent, is associated with the development of microbial multidrug resistance when present in the aqueous environment (Carey and Mcnamara 2015). However, reliable research is still lacking for assessing appropriately the impacts and consequences of triclosan release into the aqueous environment. The presence of triclosan clearly induces multidrug resistance in environmental microbial communities (Carey and

Mcnamara 2015). Direct physiological risks for Arctic higher terrestrial organisms associated with the currently observed triclosan levels in the Arctic are not expected under the currently observed conditions (Reiss et al. 2009).

Levels and distributions

In total, 110 different substances from 25 different PPCP groups were identified in atmospheric, terrestrial, freshwater, and marine samples. The compound groups and concentration distributions together with the relevant references are listed as Table S1 in the supplementary material of this study. The freshwater environment is considered the major recipient for PPCPs. The majority of all PPCPs identified have been identified in sewage influent, sewage effluent, sewage, and sludge samples (Fig. 3).

Sewage influent Fifty-five of the total of 110 (50%) reported PPCPs were identified in sewage influent samples. The majority of the reported data are from several Nordic reports (see Supplementary material, Table S1). PPCP concentration data from sewage influent is reported from nine Arctic regions (Northern Norway, Finland, Faroe Islands, Iceland, Greenland, Svalbard, Ontario, Manitoba, and Alaska). The highest measured concentration was reported for prescription-free, "over the counter" NSAIDs including salicylic acid (874 μg/L in Ontario, Canada), acetaminophen/paracetamol (506 μg/L in Faroe Island), naproxen (109 μg/L in Iceland), and the anticoagulants dipyridamole (166 μg/L in Faroe Islands). Most of the PPCP compounds were found in middle ng/L to low μg/L concentration range in the sewage effluent samples analyzed (Fig. 2).

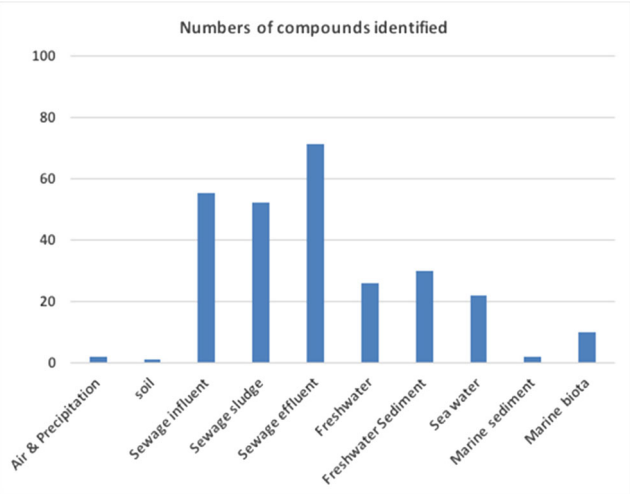

Fig. 2 Sample type-specific distribution profile of the PPCPs identified in Arctic environmental samples. *Y-axis*: compound numbers identified in the respective matrix, *x-axis*: sample type

Sewage and sludge Fifty-two of the 110 (47%) reported PPCPs were identified in sewage and sludge samples. The majority of the reported data are from several Nordic reports and peer-reviewed research publications (see Supplementary material, Table S1). PPCP concentration data from sewage and sludge is reported from nine Arctic regions (Northern Norway, Finland, Faroe Islands, Iceland, Greenland, Svalbard, Ontario, Manitoba, and Alaska). In general, water-soluble residues are enriched in sewage, whereas less polar compounds are found mainly in sludge samples. Consumption volumes obviously reflect directly the amount of active PPCP substances found in sewage related samples. The average levels of all compounds identified were found in the middle ng/L range for sewage and ng/g range for sewage sludge. In sewage sludge, SSRI antidepressants were found in the highest concentrations (metoprolol = 549 μg/g and propranolol = 680 μg/g in Iceland). For sewage, the highest levels were found for additives and surfactants (siloxane D5 = 4.3 μg/g in Faroe Islands and ATAC type = 680 μg/g in Iceland).

Sewage effluent Seventy-one of the 110 (65%) reported PPCPs were identified in Arctic sewage effluent samples. The majority of the PPCP-related compounds are confirmed in sewage effluent. This again supports the importance of sewage-related matrices as primary source for PPCPs. In addition, many PPCP compounds are immobilized in the exposed organism through phase II metabolization such as glutathione-*S*-transferase-induced conjugation. These conjugates are usually not easily quantified in standard analytical procedures without direct cleavage during the preparation method as additional sample treatment step. However, during passage of the WWTP procedure the conjugates are often cleaved, the original substance is available again and thus the respective PPCP levels identified in the target effluent samples are consequently considerable higher compared with inflow or sewage. All identified PPCP substances were found in concentration from medium ng/L to low μg/L concentration range. The predominant compounds were quantified in medium μg/L range. The ibuprofen metabolite carboxy-ibuprofen (CX-Ibu) was found in levels up to 70 μg/L in Tromsø (Norway) and 38 μg/L in Longyearbyen (Svalbard). Acetaminophen/paracetamol was found in levels up to 71 μg/L in the Faroe Island effluents. The surfactant BAC was found up to 60 μg/L in effluent samples from Greenland.

Freshwater Twenty-six of the total of 110 (24%) reported PPCPs were identified in freshwater samples. Usually all detected PPCP concentrations were found in the low ng/L range. The highest concentrations, however, were identified in the high ng/L to low μg/L range. Naproxen was quantified in max. 199 ng/L in Ontario (Canada) surface freshwater water samples, and carbamazepine was found in maxim concentrations of 749 ng/L in

Onario (Canada) surface water samples. However, the antibiotics lincomycin (max. 1413 ng/L) and monensin A (max. 810 ng/L) dominated Ontario surface water samples. The levels are only slightly lower compared to the respective effluent samples demonstrating again the insignificant retention properties for standard WWTPs concerning this type of contamination.

Freshwater Thirty of the total of 110 (27%) reported PPCPs were identified in freshwater sediment samples. Usually all detected PPCP concentrations were found in the low ng/g range. The highest concentrations, however, were identified in the high ng/g to low μg/g range. Sertraline was quantified in max. 1 μg/g in sediment from the Faroe Islands. The angiotensin II receptor antagonist losartan was found in 392 ng/g in Island sediment. 17β Ethinyl estradiol was measured in 302 ng/g in Greenland sediment. However, surfactants like BAC (1.3 μg/g) in Faroe Island sediment and ATAC type surfactants (1 μg/g) in Iceland sediment were the dominating PPCP substances in freshwater sediments. Mainly surface-active substances and less polar compounds were associated with sediment as carrier matrix.

Marine environment Especially for the Arctic, the marine (coastal) environment is considered an important recipient for PPCPs. Most settlements are located in the coastal zone and, thus, emit sewage related waste directly into the receiving marine environments.

Seawater Twenty-two of the total of 110 (20%) reported PPCPs were identified in seawater samples. In many cases, the sewage is released directly into coastal seawater recipients after minimal of treatment. Therefore, effluent sample characterization should be applied when interpreting these data if not mentioned otherwise. Usually all detected PPCP concentrations were found in the low ng/L range. The highest levels were found in the high ng/L range. Citalopram (SSRI antidepressant) was determined in max. 612 ng/L concentration in surface seawater in Tromsø (influenced by direct hospital effluent). Carbamazepine was detected with maximum concentrations of 47.7 ng/L in Manitoba seawater (Hudson Bay, Canada). The stimulant caffeine was quantified in concentrations up to 126 ng/L in receiving seawater samples from Tromsø (Norway).

Marine sediment Only one screening studies confirmed the presence of PPCPs in marine sediments from the Barents Sea. Bisphenol A (max. 11 ng/g) and the artificial fragrance musk xylene (max. 4.1 ng/g) were quantified in these sediment samples.

Marine biota Eleven of the total of 110 (10%) reported PPCPs were identified in seawater samples. Usually all detected PPCP concentrations were found in the high pg/g to low ng/g range. The highest levels were found in the low ng/g range. The siloxanes D5 (max. 10 ng/g in Icelandic marine mammals) and D6 (max. 5.2 ng/g in Faroe Island fish) were quantified in a comprehensive Nordic study. ATAC type of surfactants was found up to 6.7 ng/g in marine fish from Greenland. The synthetic musks phantolide (2.5 ng/g Ontario), Cashmeran (2.5 ng/g in Greenland), and musk tibetene (max. 6 ng/g in Greenland) where found in polar bear tissue.

Distribution profiles

The highest levels identified in the available studies was reported for metoprolol for Manitoba sewage sludge (Table S2, Fig. 3) is only as a single value and, thus, does not reflect a general trend. However, this level reported here should rather be considered a strong signal for the high degree of variation in the release patterns of this type of compounds reflecting the demographic profile and the treatment technologies of the respective sewage treatment facilities (Kallenborn et al. 2008b).

However, these results confirm again that sewage effluent and sewage sludge must be considered as the major source of PPCPs also for the Arctic environment. The here provided review supports the conclusion that for PPCPs consumed in high volumes like ibuprofen, acetaminophen/paracetamol, and acetyl salicylic acid (all over the counter NSAID PPCPs without general prescription regulations), the population density, and the STP technology are considered the main determining factor for the release levels and the overall concentration profiles and not primarily the environmental stability and mobility.

For those compounds identified in sewage effluent samples from more than four Arctic locations, the maximum levels

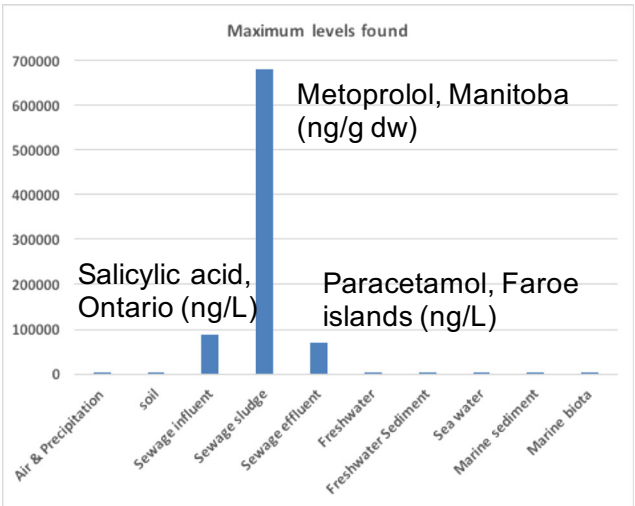

Fig. 3 Maximum concentration levels identified in the respective Arctic environmental matrices

reported are compared (Table 4). In accordance with previous assumptions, the highest concentrations in this comparison were found for prescription-free agents (ibuprofen and acetaminophen/paracetamol). However, for compounds where controlled application is advised by national prescription regulations (i.e., diclofenac, paroxetine, citalopram), the therapeutic application scheme is also contributing significantly to the release profile (Table 3).

For diclofenac (usually prescribed anti-inflammatory NSAID agent in Northern Europe), the highest concentrations were reported in Longyearbyen (Svalbard, around 2200 inhabitants) and found considerably higher compared with Tromsø (North Norway) an Arctic city with more than 60,000 inhabitants. Similar profiles were also found for prescribed Antidepressants (i.e., SSRI agents). For these compounds, national prescription regulations contribute to the respective release profiles and thus the concentration variability. Sertraline dominates in Iceland whereas citalopram is the predominant SSRI residue in effluent samples from the Faroe Islands (Table 3).

Comparison with other regions

A first comparison of level and distribution profiles for selected pharmaceuticals identified in the here performed survey revealed that rather compound-specific physical-chemical properties (like (environmental half-life times), environmental conditions, as well as release levels are the governing factors for the overall concentration levels found than the population density and human population characteristics (Table 4).

Selected compounds, like siloxanes which are reaching the Arctic via atmospheric long-range transport (in addition to local sources), are showing significantly lower concentrations in Arctic air compared with densely populated regions (urban atmosphere in Chicago, IL). However, the very high Arctic levels found for ibuprofen, metoprolol, and paracetamol (two to three orders of magnitude higher compared with the levels found in western urban environments) indicate significant higher environmental stability in the immediate vicinity of the releasing source with obvious hazardous consequences for the local ecosystems.

This comparison again illustrates impressively the situation the Arctic is currently facing with respect to the introduction of this type of new emerging pollution issues where no regulatory or restricting measures are taken yet. Substances, identified and acknowledged as highly important for maintaining good living and health standards of the human population on our globe, may exhibit unintended negative hazardous effects in the pristine Arctic environment when directly exposed to uncontrolled release with potential consequences both for the receiving ecosystems and the human populations still largely dependent on harvesting from the wealth of the Arctic ecosystems.

Concluding remarks

Although the overall volume pharmaceuticals applied for medical and veterinary treatment is low compared to the densely populated regions of the globe, the lack of modern WWTP installations also in larger settlements/cities in the North in addition to low-temperature ambient conditions (slow microbiological transformation and no photochemical degradation during winter) results in surprisingly high release rates of selected pharmaceuticals into the Arctic environment (Vasskog et al. 2008, Vasskog et al. 2006, Weigel et al. 2004a, Gunnarsdottir et al. 2013b, Weigel et al. 2004b, Huber et al. 2013a, Green et al. 2008, Kallenborn et al. 2008b, Kallenborn et al. 2008a).

During the past decade, a number of national and international research and screening studies have confirmed the presence of PPCPs in the Arctic environment. Many of these studies are currently described in the new assessment reports published and updated by the Arctic Monitoring and Assessment Programme (AMAP). As already outlined earlier, the absence of modern WWTP installations also in larger settlements/cities in the Arctic is obviously resulting in relatively high release rates of selected PPCPs. As a direct consequence, the combination of lower elimination/degradation rates and

Table 3 Maximum concentration levels for regional comparison of selected PPCPs in Arctic sewage effluent samples (ng/L). All compounds listed here were found in sewage effluent samples from >4 locations

Location/ region	Ibuprofen	Diclofenac	Acetaminophen/ paracetamol	Sertraline	Paroxetine	Citalopram
Ontario (Can)	4000		740			
Greenland	2800		25,800	2	20.8	192
Iceland	5800	390	8540	299	89.3	69.2
Faroe Islands	4500	597	71,500	23	149	540
Longyearbyen (N, Svalbard)	403	1074				
Tromsø (N)	448	48		90	13	102

Table 4 Comparison of selected PPCP levels with non-Arctic reports

Conc. unit	Compounds	Sample matrix	Levels Arctic (maximum)	Other regions (maximum)	Non-Arctic sampling location	References
ng/m^3	Siloxanes (D4, D5)	Air	4	1100	Chicago (IL, USA)	Yucuis et al. 2013
ng/L	Ibuprofen	Sewage influent	87,400	3600	Källby (SWE)	Bendz et al. 2005)
ng/g	Metoprolol	Sewage sludge	680,000	500	WWTP Terrassa, Spain	Radjenovic et al. 2009
ng/L	Paracetamol	Sewage effluent	71,000	150	Källby (SWE)	Bendz et al. 2005
ng/g	BAC	Freshwater sediment	1300	1500	Long Island (NY)	Li and Brownawell 2010
ng/L	Citalopram	Seawater	612	27	San Francisco Bay	Nodler et al. 2014
ng/g	Bisphenol A	Marine sediment	11	10,500	Taiwan	Huang et al. 2012

reduced retention in WWTP treatment is contributing to surprisingly high release concentration into the receiving Arctic (mainly) aquatic environment.

Pharmaceuticals are designed to express a specific biochemical function at low levels as a part of an integrated therapeutically procedure. This biochemical effect, desirable during therapy, may cause unwanted environmental toxicological effects on non-target organisms when the compound is released into the environment. In the Arctic environments, pharmaceutical residues are released into low to very low ambient temperatures in receiving aqueous environments. Low biodegradability and, thus, prolonged residence time must be expected for the majority of the pharmaceuticals entering the aquatic system. This is especially critical when significant amounts of antibiotic/antimicrobial agents are released in a low-temperature environment, thus enhancing the potential for resistance against these substances in the local microbial communities.

The environmental toxicological consequence of the continuous release is, thus, expected to be different compared to temperate regions of the globe. The impact on the human populations due to consumption of contaminated local fish and invertebrates or through exposure to resistant microbial communities cannot be excluded.

However, the scientific results so far available through published papers and reports must still be considered as indication only. Comprehensive environmental studies on the fate, environmental toxicology, and distribution profiles of pharmaceuticals applied in high volumes and released into the Nordic environment under cold Northern climate conditions should be given high priority by national and international authorities. This is also necessary to ensure that local food sources can also be harvested by the future generations of indigenous populations without any concern for health and well-being.

The following information is missing for and comprehensive risk assessment in Arctic environments when considering the environmental fate of PPCPs:

- Medium- and long-term monitoring data are not yet available from national or regional monitoring activities.
- A future research priority should be laid upon time and spatial trend investigations for PPCPs in Arctic environments.
- Comprehensive information on source apportionment and assessment of source strength is missing for a reliable risk assessment in the Arctic.
- Scientific emphasis should be focused on environmental fate assessment (i.e., DIPSR approach) including uptake into species exploited for human consumption.
- The elucidation of transformation processes and the risk evaluation of major transformation products as an integrated part of fate assessment are not available.
- Reliable scientific information on environmental toxicology and effects studies in the Arctic designed for PPCPs including cocktail effects and information on non-target effect mechanisms are missing

Recommended research and monitoring priorities

The available scientific information confirms that PPCPs were found in high concentrations in source near Arctic locations. Some of the reported substances exhibit even considerable mobility and are found in samples in considerable distance from the primary sources as demonstrate above. However, only sparse science-based information is available in order to provide a reliable foundation for science-based regulations and recommendations for mitigation. We therefore recommend a future stronger research focus on fate and distribution properties for relevant PPCPs in the Arctic environment.

We recommend therefore to

- Identify and implement relevant PPCPs into already established long-term national and regional monitoring programs

- Validate the available analytical methods for the purpose of continuous and regular monitoring. This includes harmonization method, joint quality control criteria, as well as continuous method inter-comparison
- Develop suitable fate and transport models designed for Arctic conditions
- Include aspects on PPCP fate and distribution in Arctic environments in already available research programs
- Investigate the combined environmental effects of PPCP in combination with non-chemical stressors such as water and soil quality as well as on-going Arctic climate change
- Support priority research on PPCP residues with respect to sources, effects, and consequences for human health exposure

For more detailed information on research priorities for PPCPs as environmental pollutants, we refer to a recent strategic study (Rudd et al. 2014).

Acknowledgements The authors appreciate to open-minded discussions with several anonymous reviewers during the work on the current AMAP assessment report. The Arctic Monitoring and Assessment Programme (AMAP) supported this review both intellectually as well as economically. Roland Kallenborn thanks the Research Council of Norway (RCN), the Norwegian Environmental Agency (Miljødirektoratet), and the Screening Program of the Nordic Council of Ministers (NCM) for the financial support of several screening and research project on PPCP-related topics. We appreciate the financial support of ARTEK 2016 (DTU Copenhagen) allowing the presentation of the here given overview during the ARTEK Conference in Sisimiut (Greenland).

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