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Occurrence of micropollutants in the wastewater streams of cruise ships

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ABSTRACT

Nowadays the protection of the marine environment raises increasing academic and public attention. The issue of organic micropollutants is of equally high importance for the marine ecosystems. Maritime vessels are considered to significant sources of micropollutants especially if the ship carries many passengers, which is often true for cruise ships which frequent attractive and sensitive sea areas. The emission pathways for micropollutants include wastewater discharges and sewage sludge disposal. The findings of the German research and development project NAUTEK contribute to bridging the knowledge gap about micropollutant emissions from cruise ships. As expected, micropollutants were detected in both the blackwater and greywater on board, emitted from either the passengers or certain ship operations. In total, 16 out of 21 target substances were detected. Peak concentrations of pharmaceuticals could be found mainly in blackwater (peak conc. Carbamazepine 3.9 μg/L, Ibuprofen 29 μg/L, Diclofenac 0.04 μg/L), while greywater is mainly characterized by substances such as ointment residues, UV-filters and flame retardants (peak conc. Diclofenac 0.65 μg/L, Bisphenol A 8 μg/L, Tris(1-chloro-2-propyl) phosphate 136 μg/L). Further analyses suggest a gradual removal of the micropollutants by the onboard MBR plant (MBR effluent peak conc. Carbamazepine 0.47 μg/L, Ibuprofen 6.8 μg/L, Diclofenac 0.3 μg/L). Findings of this research provide a critical stepstone for shaping technical solutions for onboard micropollutants removal and water resource recycling.

1. Introduction

It is borne out by cruise operators' figures that every year more than 20 million passengers embark on a cruise trip. For many years, the cruise ship industry has been one of the fastest growing tourism sectors worldwide and its passenger count has been increasing rapidly. Similarly, the ongoing construction of many new cruise ships shows how optimistic the cruise industry is about the future. However, the cruise industry should be held accountable for numerous environmental problems, including critical emissions such as exhaust gases and wastewater. In view of the latter, the creation and maintenance of luxurious conditions aboard results in high water and resource consumption and hence high wastewater discharge. In addition, further wastewater streams are derived from laundries, galleys and from other activities like ship cleaning. In view of the existing legal regulations (primarily Annex IV of MARPOL Convention which contains regulations for the prevention of pollution by sewage from ships), most of the operating cruise ships are equipped with wastewater treatment systems. Specific statements regarding treatment performance cannot be made due to missing administrative plant monitoring.

In the maritime context only blackwater is officially regarded as wastewater. In most cases, greywater is nonetheless also treated on board of cruise ships, which seems appropriate in view of the actual pollution loads from greywater.

There is still significant room for treatment system improvements. The latest technical developments are aimed at integrating nutrient removal mainly into market available treatment systems. Since it was proven that micropollutants harm aquatic life [1] the issue of micropollutants has become an important topic on the world's task list for wastewater treatment improvement. Micropollutants encompass substances such as pharmaceuticals,

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Table 1
Specification of target substances.

Compound	Subordinated group	CAS number	Method for analysis	Reference substance[a]	Measurement uncertainty [%]
Pharmaceuticals					
Carbamazepine	Anticonvulsant	298-46-4	HPLC/MS-MS	Sigma	60–80
Dimenhydrinate	Antiemetic	523-87-5	GC/MS	Sigma	70–100
Ibuprofen	Analgesic and anti-inflammatory	15687-27-1	HPLC/MS-MS	Fluka	50–90
Diclofenac	Analgesic and anti-inflammatory	15307-86-5	HPLC/MS-MS	Sigma	55–65
Naproxen	Analgesic and anti-inflammatory	22204-53-1	HPLC/MS-MS	Sigma	70
Propyphenazone	Analgesic and anti-inflammatory	479-92-5	GC/MS	Fluka	94
Metoprolol	Beta blocker	37350-58-6	HPLC/MS-MS	Sigma	85–120
Atenolol	Beta blocker	29122-68-7	HPLC/MS-MS	Sigma	10
Bezafibrate	Cholesterol-lowering drug	41859-67-0	HPLC/MS-MS	Sigma	65–100
Clofibric acid	Cholesterol-lowering drug	882-09-7	HPLC/MS-MS	Fluka	70–80
Clarithromycin	Antibiotic	81103-11-9	HPLC/MS-MS	Sigma	80–85
Sulfamethoxazole	Antibiotic	723-46-6	HPLC/MS-MS	Fluka	80–90
Trimethoprim	Antibiotic	738-70-5	HPLC/MS-MS	Sigma	75
Ethinyl estradiol	Estrogen	57-63-6	GC/MS	Sigma	80
Verapamil	Antiarrhythmic	52-53-9	HPLC/MS-MS	Sigma	60
Caffeine	Analeptic	58-08-2	GC/MS		20–45
Personal Care Products					
Benzophenone	UV filter	131-57-7	GC/MS	Fluka	75–95
Methylbenzyli-dene camphor	UV filter	36861-47-9	GC/MS	Fluka	62
Tonalide	Fragrance	21145-77-7	GC/MS	SAFC	50–60
Chemicals					
Bisphenol A	Plastic Softener	80-05-7	GC/MS	Fluka	85–110
Tris(1-chloro-2-propyl) phosphate (TCPP)	Flame retardant	13674-84-5	GC/MS	Fluka	100

[a] Standard/pure substance of each substance to establish the analytical method for this substance.

personal care products, endocrine disruptors, anti-flame retardants and many more. Because of a lack of specific legal requirements, reducing the micropollutants emissions from (cruise) ships are apparently not yet on the ship owners' agenda.

For the first time, the cooperative R&D project “Sustainable Solutions for Wastewater Treatment and Reuse on Cruise Liners (NAUTEK)” places the micropollutants issue in the context of wastewater management aboard cruise ships. Why investigate the occurrence of micropollutants aboard? First, the ashore discharge standards - after some time – are expected to become relevant for the offshore regions. Second, in line with continuous efforts for energy saving, waste or greywater reuse solutions will potentially play an important role. While reflecting on reuse solutions the micropollutants issue can be a critical bottleneck. To address all these open questions, the project NAUTEK focused on the development of a “future-proof” modular wastewater treatment scheme.

The present article aims to provide a comprehensive overview about the occurrence of selected micropollutants in different wastewater streams aboard cruise ships. In detail, black and greywater streams on cruise ships were subjected to in-depth investigations for the first time. The sampling methods and analyses were carried out by Hamburg University of Technology (TUHH), Germany, within the scope of the cooperative R&D project “Sustainable Solutions for Wastewater Treatment and Reuse on Cruise Liners (NAUTEK)” and in cooperation with a large cruise operator. The findings presented in this article provide essential criteria for the concrete design of techniques to be used for onboard micropollutants removal.

2. Methods

After establishing sampling points, accurate sampling, and sample processing as well as using high-end analytical methods for micropollutants detection, as illustrated in details below.

2.1. Selection of target substances

The determination of target substances was based on their likelihood of appearance onboard cruise ships, either originated from passengers or certain cruise ship operations. For example popular painkillers, beta blockers, and also typical compounds of sun protection products and ship cleaning agents were worth considering. Further conversations with the pharmacy staff and doctors onboard a cruise ship as well as in-depth literature review - particularly dealing with the occurrence and fate of micropollutants in the aquatic environment - were conducted. Finally, 21 micropollutants were selected as target substances. The selected compounds belong to the following groups: pharmaceuticals (16 substances), personal care products (3 substances) and chemicals (2 substances). Table 1 specifies the tested substances. It comprises the CAS Number, the parameter classification, the specific method for analysis, the distributor of the reference substance and the specific measurement uncertainty.

2.2. Sampling

In total 12 sampling episodes took place on four different medium-sized cruise ships (total capacity 2600–3300 persons) during calls at Hamburg Port, Germany. The grab sampling was carried out during passenger disembarkation and embarkation. It is worth noting that all cruise ships subject to the investigations were equipped with nearly similar membrane bioreactor systems as shown in Fig. 1. Generally, the plant operation was not geared to support nitrification and the denitrification tank was bypassed. Only one cruise ship had a denitrification system in operation.

Appropriate sampling points were identified and implemented with the assistance of the ship crews (also displayed in Fig. 1). In total four wastewater streams were sampled: the blackwater vacuum tank, the mixed greywater stream, the laundry greywater and the final effluent (MBR-permeate). Table 2 provides an overview of the different sampling episodes indicating number of samples taken from each cruise ship. The sampling frequency depended on ships calling at Hamburg Port within the investigation period. There was no sampling after mixing black- and greywater due to the absence of collecting tank or mixing tank upstream from the treatment plant.

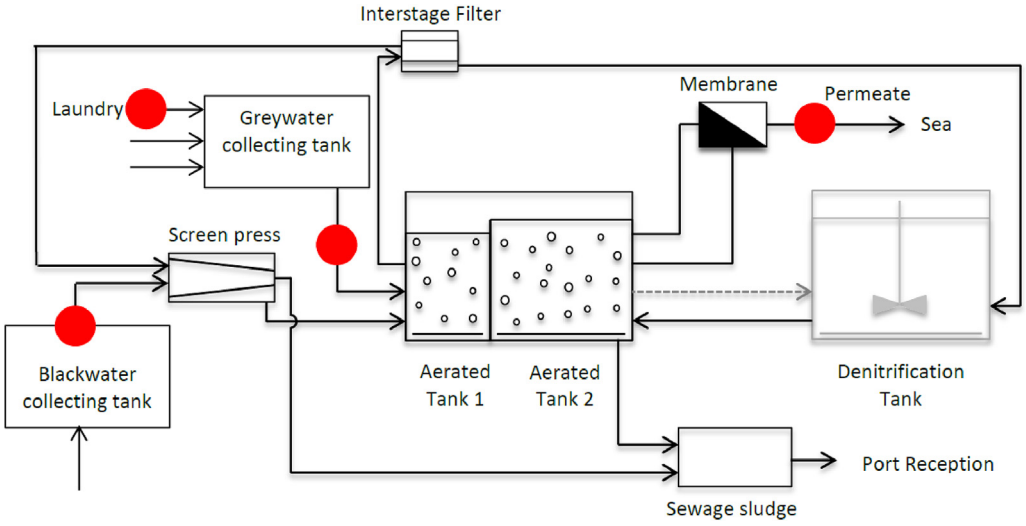


Fig. 1. Schematic set-up of wastewater treatment systems and distribution of sampling points (red spots).

2.3. Sample processing and analysis

For the grab sampling, glass bottles - previously rinsed with acetone - were used to avoid any contamination. For the analysis of the target compounds, membrane-filtered (pore width 0.45 μm) samples (1 L of greywater or 300–800 mL of blackwater, pH 6.9–7.2) were filtered through ABS ELUT-NEXUS solid phase extraction cartridges (Agilent Bond ELUT), first at pH 7 (500 mg/12 mL) and subsequently at pH 3 (200 mg/12 mL). Subsequently, the cartridges were dried in a mild nitrogen flow. The analytes were eluted from the dried cartridges with methanol containing 0.1% acetic acid and with unadulterated methanol. The combined eluates were concentrated by means of a rotational vacuum evaporator to a volume of 2 mL. These concentrates were either analyzed by gas chromatography coupled to mass spectrometry (GC/MS) or by liquid chromatography/mass spectrometry (HPLC/MS-MS). The applied analytical methods were established for each substance by either GC-MS parameters (retention time, target and qualifier masses, temperature program) or LC-MS/MS parameters (tuning each single compound, gradient LC chromatography). The analysis of each compound was verified by standard addition methods. Detection limits were determined from the calibration curves (according to the German Standard DIN 32645) and estimated with enrichment and dilution factor. The concrete framework for the chromatographic analyses is described in Table 3. The detection limit of each analysis varied widely from 0.02 up to 0.5 μg/L depending on the consistency of the single samples. Possible reasons for this include measuring inaccuracies of the analytical equipment, the quality of sample preparation and interferences triggered by matrix effects, the enrichment factor and the dilution factor for analyzing. The two applied analytical methods (GC/MS and HPCL/MS-MS) led to different limits of detection. Due to high solids content, the blackwater filtration (as part of the sample preparation) required an especially long time (mostly two days). Thus, the analyses were carried out with a reduced sample volume of 300–800 ml. This led to a lower enrichment factor and a higher value for the limit of detection. Another reason for varying limit of detection was the heterogeneous wastewater matrix. Organic compounds such as surfactants aggravated the evaluation of peaks in the mass spectra. In some cases the matrix required a higher dilution to achieve chromatographic sorting which significantly influenced the limit of detection.

Table 2
Details of sampling episodes.

Cruise ship	No. of samples			
	Blackwater	Greywater	Laundry greyw.	MBR-permeate
1	8	8	3	8
2	0	1	0	1
3	1	1	0	1
4	2	2	2	2
Total	11	12	5	12

2.4. Theory and calculation

Some practical restrictions onboard made certain samplings and detections substantially difficult or even unmanageable so that rough calculations became necessary in order to derive some conclusions that might be of interest. This applies to 1) the conclusions with regard to the mixed wastewater, 2) the information on micropollutant loads and 3) the estimation in terms of micropollutant removal performance of the existing MBR-plants, as specified in the following:

1) Due to the fact that there was no access to a sampling point for mixed wastewater, any specifications with regard to micropollutants concentrations in the mixed waste water are based on the assumed blackwater and greywater ratio of 1:7 (compare [2]).
2) Application of a simple equation based on the assumption that one person generates 31 and 220 L of blackwater and greywater per day, respectively. This valuation is derived from a comprehensive review of data made available by scientists, ship owners, ship yards and professional organizations – as compiled in Ref. [2].
3) As there were no possibilities to take time-corresponding samples, any interpretations in terms of the micropollutant removal performance are subject to substantial uncertainty. Removal performance was estimated by comparing maximum effluent concentrations with maximum influent concentrations (compare item 1). This leads to a highly hypothetical reduction rate in micropollutants which must be understood as not more than a first orientation.

Table 3
Instrumentation and conditions for analysis of the selected trace organics in blackwater and greywater.

Method	GC/MS	HPLC/MS
Analytes	caffeine, dimenhydrinate, 17α-ethinyl estradiol, bisphenol A, propyphenazone, tris(1-chloro-2-propyl) phosphate, methylbenzylidene camphor, benzophenone, tonalide	atenolol, carbamazepine, clarithromycin, metoprolol, tonalide, trimethoprim, verapamil, bezafibrate, clofibric acid, diclofenac, ibuprofen, naproxen, sulfamethoxazole
Chromatograph	Agilent 6090N, autosampler 7683B, cold injection system Gerstel KAS 4	Agilent system with 1200 binary pump, 1200 Autosampler and 1260 column oven
Mobile phase	helium 5.0, 1 mL/min	A:demineralized water with 0.1% acetic acid B: methanol with 0.1% acetic acid, 0.3 ml/min gradient elution A 90%–10% / B 10%–90%
Column	DB-5MS, 30 m × 0.25 mm i.d., 0.25 μm film thickness	Phenomenex Synergi Fusion 4u RP 80A, 150 × 3 mm
Injection	2 μL	10 μL
Mass spectrometer	Agilent MSD 5975B with ChemStation G1701DA	AB Sciex API2000 with Analyst Version 1.5.1
Ion source	EI 70 eV, 230 °C	turbo spray; polarity: positive/negative
Quadrupole temperature	150 °C	–

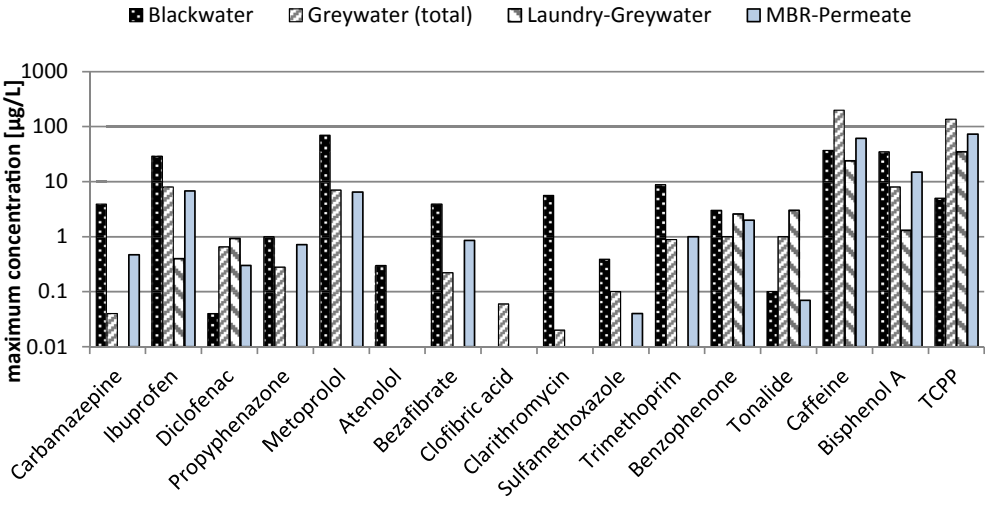


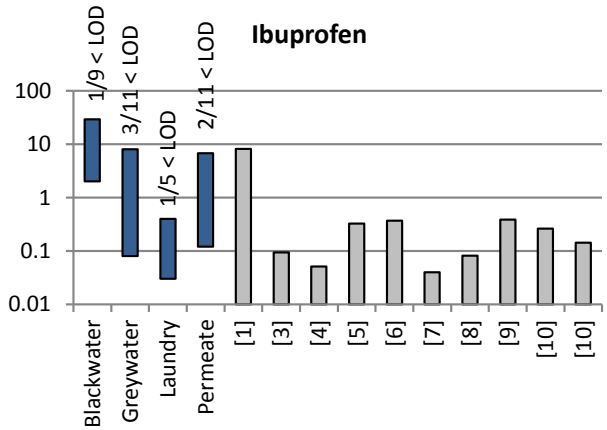
Fig. 2. Maximum concentrations in blackwater and relevant greywater streams on cruise ships.

3. Results and discussion

As a first step the maximum concentrations are given for all substances concerned and for all examined wastewater streams. Some of the target substances were measured at significantly high concentrations while others were not detected during the entire sampling phases. In most cases obvious differences in the micropollutants load could be observed between blackwater and greywater. Residues from orally-administered pharmaceuticals were especially common in blackwater. In contrast, non-pharmaceutical residues and food residues such as the flame retardant TCPP and caffeine were more prevalent in greywater. As a first step, Fig. 2 shows a compilation of the measured maximum concentrations of micropollutants in blackwater, greywater,

Table 4
Specification of number of samples (n_{total}) and number of samples below limit of detection ($n_{belowLOD}$).

Substance	Blackwater		Greywater (total)		Laundry-greywater		MBR-permeate	
	n_{total}	$n_{belowLOD}$	n_{total}	$n_{belowLOD}$	n_{total}	$n_{belowLOD}$	n_{total}	$n_{belowLOD}$
Carbamazepine	11	3	12	11	5	5	12	7
Ibuprofen	9	1	11	3	5	1	11	2
Diclofenac	11	9	12	6	5	2	12	5
Propyphenazone	9	9	9	7	5	5	9	7
Metoprolol	11	1	12	5	5	5	12	2
Atenolol	9	8	9	9	5	5	9	9
Bezafibrate	11	8	12	11	5	5	12	6
Clofibrate	11	11	12	11	5	5	12	12
Clarithromycin	11	8	12	11	5	5	12	12
Sulfamethoxazole	11	9	12	9	5	5	12	11
Trimethoprim	9	3	9	4	5	5	9	2
Benzophenone	11	6	12	0	5	0	12	0
Tonalide	11	10	12	8	5	0	12	11
Caffeine	10	0	11	0	4	2	11	0
Bisphenol A	11	1	12	3	4	1	12	0
TCPP	9	3	9	0	3	0	9	0



n indicates number of samples, [_] indicates reference
[1] 1-year sampling episode at 4 WWTPs
[3] n = 20
[4] 5 measurement campaigns at 1 WWTP
[5] n = 11
[6] n = 49
[7] n= 5
[8] EU-wide monitoring survey at 90 WWTPs
[9] n=39
[10] n = 10
[10] n=10

Fig. 3. Ibuprofen concentration in blackwater and greywater streams on 4 cruise ships compared with selected average values for effluents of land-based wastewater treatment plants [1,3–10].

greywater from laundries and the MBR-plant effluent (permeate). The compounds Dimenhydrinate, Naproxen, Ethinyl estradiol, Verapamil and Methylbenzylidene camphor were not detected in any of the samples. Table 4 contributes to a better understanding of Fig. 2 by providing detailed information about the total amount of samples (n_{total}) in relation to the amount of samples below limit of detection ($n_{below\ LOD}$).

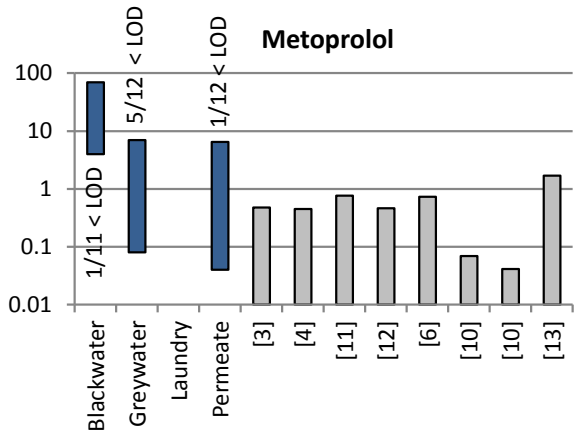
In addition, four micropollutants of high relevance were selected to be examined in depth using the entire array of analyses. Hence, the results for Carbamazepine, Diclofenac, Ibuprofen and Metoprolol are specifically represented and supplemented by an itemized comparison with land-based treatment facilities. Figs. 3–6 contain information about maximum and minimum concentrations as well as the total amount of samples in relation to the amount of samples below LOD (indicated as $n_{below\ LOD}/n_{total}$). Furthermore, the figures also provide a comparison between the ship's final effluent (permeate) and individual average values from land-based treatment facilities which were primarily taken from peer-reviewed publications.

In view of the presented analytical results, consistent conclusions cannot be drawn from Figs. 3–6 for

- Metoprolol concentration in greywater from laundries
- Diclofenac concentration in blackwater
- Carbamazepine concentration in greywater and greywater from laundries

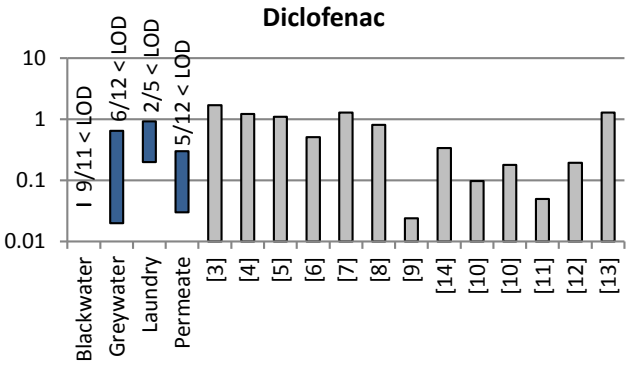
4. Discussion

Overall, all results are consistent and equally plausible. The succeeding discussion follows the wastewater categories introduced above and culminates in a first appraisal of reduction in micropollutants while treating wastewater in onboard systems.



n indicates number of samples, [_] indicates reference
[3] n=30
[4] five measurement campaigns at 1 WWTP
[11] n = 3
[12] n = 4
[6] n = 29
[10] n = 10
[10] n = 10
[13] n = 6

Fig. 4. Metoprolol concentration in blackwater and greywater streams on 4 cruise ships compared with selected average values for effluents of land-based wastewater treatment plants [3,4,6,10–13].



n indicates number of samples, [_] indicates reference
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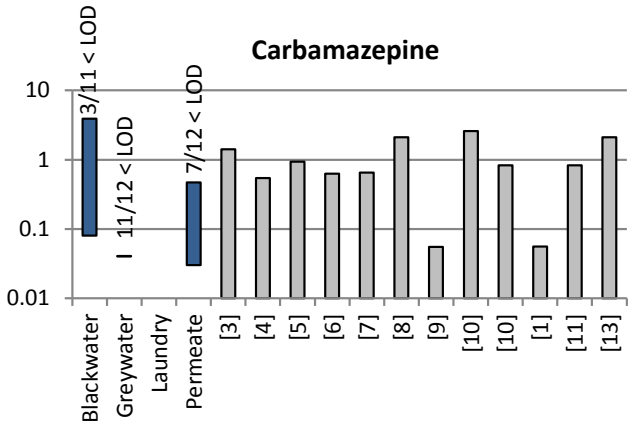
[4] five measurement campaigns at 1 WWTP
[5] n = 3
[6] n = 11
[7] n = 4,
[8] n = 49
[9] n= 5
[14] n = 42
[10] n = 10
[10] n = 10
[11] EU-wide monitoring survey at 90 WWTPs
[12] n=39
[13] n = 6

Fig. 5. Diclofenac concentration in blackwater and greywater streams on 4 cruise ships compared with selected average values for effluents of land-based wastewater treatment plants [3–14].



n indicates number of samples, [_] indicates reference
[3] n = 38
[4] five measurement campaigns at 1 WWTP
[5] n = 3
[6] n = 11
[7] n = 4
[8] n = 30
[9] n= 5
[10] n = 10
[10] n = 10
[1] 1-year sampling episode at 4 WWTPs
[11] EU-wide monitoring survey at 90 WWTPs
[13] n = 6

Fig. 6. Carbamazepine concentration in blackwater and greywater streams on 4 cruise ships compared with selected average values for effluents of land-based wastewater treatment plants [1,3–11,13].

4.1. Blackwater

11 of the 16 traceable compounds showed higher peak concentrations in the blackwater compared to greywater. Unsurprisingly, most of the micropollutants detected in blackwater were oral pharmaceutical residues. However, non-drugs such as Benzophenone, Tonalide, Bisphenol A and TCPP were also detected in blackwater. There is concrete reason to believe that the human body absorbs these compounds after exposure (compare [14–18]).

4.2. Greywater

Diclofenac, Caffeine, Tonalide and TCPP showed higher concentrations in greywater than in blackwater. Besides, Clofibric Acid was detected only once in the entire greywater stream. Compared with blackwater, greywater was more likely to contain Diclofenac and at significantly higher measured maximum concentrations. This observation is possibly due to the fact that Diclofenac is broadly applied in salve form and is washed off easily. The Caffeine detected most likely originates from the galleys, where coffee grounds and coffee residues from buffets are discharged into the greywater drainage system. Tonalide, a fragrance belonging to the group of synthetic musks, is widely used to provide aromas to laundry detergents and in personal care products such as shampoos and body lotion. Thus, Tonalide can easily reach the greywater systems in various ways. The flame retardant TCPP easily diffuses from host materials and finally reaches the greywater system through laundries, hand-washing basins or showers.

Table 7
Comparison of max concentrations in MBR-permeate and max concentration in the receiving marine environment (nanograms per litre).

Compound	Max. conc. in permeate (ng/L)	Max. conc. marine environment [19] (ng/L)
Carbamazepine	470	3.1–157
Diclofenac	300	4.1–9.7
Ibuprofen	9000	12–109
Metoprolol	6800	6–158

4.3. Greywater from laundries

Seven target compounds were detected in the greywater from laundries, particularly, Diclofenac and Ibuprofen. As both drugs are painkillers commonly applied in salves, they are easily rubbed off by towels, clothes and bed linen. Furthermore, Benzophenone (a widely applied UV filter in sunscreens), Bisphenol A (ubiquitous softener), Tonalide, Caffeine and TCPP could be detected in greywater as well. The latter two reached maximum concentrations exceeding 10 μg/L.

4.4. MBR-permeate

Out of the total set of 16 micropollutants detected in all wastewater streams, 13 compounds were found in the plant permeate. The higher concentration micropollutants in the raw wastewater also ended up in higher concentrations in the permeate, although only non-time-corresponding data were considered. At this point, the huge share of greywater in the total wastewater stream becomes noticeable as well. Eleven compounds

Table 5
Hypothetical reduction in micropollutant concentrations in the course of non-targeted treatment (μg/L).

Compound	Max BW	Max GW	Max. mixed wastewater[a]	Max. in permeate	Reduction[b]
Carbamazepine	3.9	0.04	0.52	0.47	9.6%
Diclofenac	0.04	0.65	0.57	0.3	47.4%
Ibuprofen	29	8	10.6	9	15.1%
Metoprolol	70	7	14.9	6.8	54.4%

[a] Calculation.
[b] Purely hypothetical as first approach.

Table 6
Rough estimation of micropollutants loads from an average cruise ships (assumptions: wastewater generation is 251 L per person and day, 4000 persons on board).

Compound	Max. conc. in permeate (μg/L)	LOAD μ (person day)$^{-1}$	LOAD mg (ship day)$^{-1}$	Annual LOAD per ship (kg)
Carbamazepine	0.47	117.97	471.88	0.172
Diclofenac	0.3	75.3	301.2	0.109
Ibuprofen	9	2259	9036	3.298
Metoprolol	6.8	1706.8	6827.2	2.491

were detected at concentrations exceeding 0.1 μg/L and 6 compounds including two pharmaceuticals exceeding concentrations of 1 μg/L (Ibuprofen, Metoprolol, Benzophenone, caffeine, Bisphenol A and TCPP).

4.5. Further assessment

As mentioned above any conclusions with regard to the performance of micropollutant removal are subject to high uncertainties. As a first approach non-time-corresponding data for Carbamazepine, Diclofenac, Ibuprofen and Metoprolol are presented in Table 5. In view of the "reduction rates", a certain removal performance may be assumed, although the MBR treatment system was not targeted on the removal of micropollutants.

As supplement, Table 6 provides a specification of the emitted loads both per person and day as well as per ship and day under the assumption of an average cruise ship with a capacity of 4000 persons. As mentioned above, this model case is based on a wastewater generation of 251 L per person and day (see Methods). Particular mention should be made of the calculated annual loads, also indicated in Table 6. For instance, in the case of Ibuprofen the emitted load reaches 3.30 kg/a per ship.

In addition, Table 7 compares the maximum micropollutant concentrations in the MBR-permeate with the maximum concentrations in the receiving marine environment. For the compounds considered in Table 5, the permeate shows significant higher concentrations when compared to the receiving marine environments - as compiled by Ref. [19]. In view of this comparison, the micropollutant emissions originating from cruise ships are a potential concern, particularly on popular touristic sea routes located in sensitive sea areas.

5. Conclusions

The issue related to organic micropollutants such as pharmaceuticals, personal care products and other trace chemicals in wastewater is not settled finally but is still subject to ongoing intensive research and a discourse on how to remove these compounds from wastewater (and why such an approach is even necessary). The present study on micropollutants in wastewater from cruise ships is a novel one that is unique from all other studies on land-based treatment plants. It gives a first impression about to which extent micropollutants are detected in different wastewater streams produced on cruise ships. As anticipated, numerous micropollutants were identified in blackwater and all greywater streams. Thus, this study also served to determine which micropollutants occur and what are the significant differences in concentration and loads. In this manner the differences between blackwater and greywater became obvious. In blackwater the oral pharmaceutical residues prevail, while in greywater the non-pharmaceutical residues such as TCPP are more common. However, pharmaceuticals such as painkillers - very likely deriving from salves - may also be present in greywater at significant concentrations.

Although there is apparently a slight trend towards higher Metoprolol and Ibuprofen concentrations in the onboard MBR permeate the detailed discussion made in this article for Ibuprofen, Metoprolol, Carbamazepine and Diclofenac suggests no substantial differences in micropollutant concentrations between the onboard MBR-permeate and the land-based treatment plant effluents. This does not mean, however, that the issue of micropollutant emissions from cruise ships is negligible. The onboard MBR plant effluent may release some micropollutants at concentrations up to almost 100 μg/L. Hence, it is necessary to address this issue in the development of a sustainable cruise ship industry. More and more members of the cruise industry have committed themselves to sustainability. Thus, it is time for the ship owners to pay attention to the issue of micropollutant emissions and even to consider such aspects while designing a new onboard treatment system. In general, overcoming the micropollutant issue will contribute much more than an intermediate progress for achieving a closed-loop recycling onboard.

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References

[1] L.H. Santos, A.N. Araújo, A. Fachini, A. Pena, C. Delerue-Matos, M. Montenegro, Ecotoxicological aspects related to the presence of pharmaceuticals in the aquatic environment, J. Hazard. Mater. 175 (2010) S. 45–S. 95.
[2] S. Köster, L. Westhof, L. Keller, Stand der Technik der Abwasserreinigung an Bord von Kreuzfahrtschiffen, gwf 157 (2016) S. 528–S. 537.
[3] BLAC, in: Arzneimittel in der Umwelt. Auswertung der Untersuchungsergebnisse. Bericht an die 61, Umweltministerkonferenz (UMK), 2003.
[4] C. Abegglen, B. Escher, J. Hollender, S. Koepke, C. Ort, A. Peter, H. Siegrist, U. Gunten, S. von Zimmermann, Ozonung von gereinigtem Abwasser. Schlussbericht Pilotversuch Regensdorf. Dübendorf, 2009.
[5] H. Herbst, C. Maus, Spurenstoffelimination auf der Kläranlage Warburg. Machbarkeitsstudie, September 2013.
[6] S. Scharf, O. Gans, R. Sattelberger, Arzneimittelwirkstoffe im Zu- und Ablauf von Kläranlagen, 2002.
[7] M. Clara, C. Scheffknecht, S. Weiß, Eintrag von Arzneimittelwirkstoffen in die Umwelt. Bregenz, 2013.
[8] T.A. Ternes, Occurrence of drugs in German sewage treatment plants and rivers, Water Res. 32 (1998) S. 3245–S. 3260.
[9] S.K. Behera, H.W. Kim, J.-E. Oh, H.-S. Park, Occurrence and removal of antibiotics, hormones and several other pharmaceuticals in wastewater treatment plants of the largest industrial city of Korea, Sci. Total Environ. 409 (2011) S. 4351–S. 4360.
[10] B. Kasprzyk-Hordern, R.M. Dinsdale, A.J. Guwy, The removal of pharmaceuticals, personal care products, endocrine disruptors and illicit drugs during wastewater treatment and its impact on the quality of receiving waters, Water Res. 43 (2009) S. 363–S. 380.
[11] R. Loos, R. Carvalho, D.C. António, S. Comero, G. Locoro, S. Tavazzi, B. Paracchini, M. Ghiani, T. Lettieri, L. Blaha, B. Jarosova, S. Voorspoels, K. Servaes, P. Haglund, J. Fick, R.H. Lindberg, D. Schwesig, B.M. Gawlik, EU-wide monitoring survey on emerging polar organic contaminants in wastewater treatment plant effluents, Water Res. 47 (2013) S. 6475–S. 6487.
[12] L. Lishman, S.A. Smyth, K. Sarafin, S. Kleywegt, J. Toito, T. Peart, B. Lee, M. Servos, M. Beland, P. Seto, Occurrence and reductions of pharmaceuticals and personal care products and estrogens by municipal wastewater treatment plants in Ontario, Canada, Sci. Total Environ. 367 (2006) S. 544–S. 558.
[13] T.A. Ternes, J. Stüber, N. Herrmann, D. McDowell, A. Ried, M. Kampmann, B. Teiser, Ozonation: a tool for removal of pharmaceuticals, contrast media and musk fragrances from wastewater? Water Res. 37 (2003) S. 1976–S. 1982.
[14] A.C. Watkinson, K.R. Brain, K.A. Walters, J. Hadgraft, Prediction of the percutaneous penetration of ultra-violet filters used in sunscreen formulations, Int. J. Cosmet. Sci. 14 (1992) S. 265–S. 275.
[15] D.L. Giokas, A. Salvador, A. Chisvert, UV filters: from sunscreens to human body and the environment, TrAC Trends Anal. Chem. 26 (2007) S. 360–S. 374.
[16] L. Roosens, A. Covaci, H. Neels, Concentrations of synthetic musk compounds in personal care and sanitation products and human exposure profiles through dermal application, Chemosphere 69 (2007) S. 1540–S. 1547.
[17] A.M. Hormann, S. vom Saal, Frederick, S.C. Nagel, R.W. Stahlhut, C.L. Moyer, M.R. Ellersieck, W.V. Welshons, P.-L. Toutain, J.A. Taylor, D.O. Carpenter, Holding thermal receipt paper and eating food after using hand sanitizer results in high serum bioactive and urine total levels of Bisphenol a (BPA), PLoS One 9 (2014) S. e110509.
[18] H.M. Stapleton, S. Klosterhaus, S. Eagle, J. Fuh, J.D. Meeker, A. Blum, T.F. Webster, Detection of organophosphate flame retardants in furniture foam and U.S. House dust, Environ. Sci. Technol. 43 (2009) S. 7490–S. 7495.
[19] K. Nödler, D. Voutsa, T. Licha, Polar organic micropollutants in the coastal environment of different marine systems, Mar. Pollut. Bull. 85 (1) (2014) 50–59, 15 August 2014.