

Research article

Treatment efficiency of package plants for on-site wastewater treatment in cold climates

Brenda Vidal^{a,*}, Juho Kinnunen^b, Annelie Hedström^a, Elisangela Heiderscheidt^b, Pekka Rossi^b, Inga Herrmann^a^a Department of Civil, Environmental and Natural Resources Engineering, Luleå University of Technology, SE 971 87, Sweden^b Water, Energy and Environmental Engineering Research Facility, Faculty of Technology, 90014, University of Oulu, Finland

ARTICLE INFO

Keywords:

Nitrogen
Phosphorus
Bacteria
Phthalates
Pharmaceuticals
Micropollutants

ABSTRACT

Package plants (PP) are implemented around the world to provide on-site sanitation in areas not connected to a sewage network. The efficiency of PP has not been comprehensively studied at full scale, and the limited number of available studies have shown that their performance varies greatly. Their performance under cold climate conditions and the occurrence of micropollutants in PP effluents have not been sufficiently explored. PP are exposed to environmental factors such as low temperature, especially in cold regions with low winter temperatures and deep frost penetration, that can adversely influence the biochemical processes. The aim of this study was to investigate the treatment efficiency and possible effects of cold temperatures on PP performance, with focus on traditional contaminants (organics, solids, nutrients and indicator bacteria) and an additional assessment of micropollutants on two PP. Eleven PP hosting different treatment processes were monitored. Removal of biological oxygen demand (BOD) was high in all plants (>91%). Six out of the 11 PP provided good phosphorus removal (>71%). Small degrees of nitrification were observed in almost all the facilities, despite the low temperatures, while denitrification was only observed in two plants which achieved the highest nitrification rates (>51%) and had sludge recirculation. No strong correlation between wastewater temperature and BOD, nutrients and indicator bacteria concentration in the effluents was found. The high data variability and the effects of other process parameters as well as snow-melt water infiltration are suggested as possible reasons for the lack of correlation. However, weak negative relations between effluent concentrations and wastewater temperatures were detected in specific plants, indicating that temperature does have effects. When managed adequately, package plants can provide high BOD and phosphorus removal, but nitrogen and bacteria removal remain challenging, especially at low temperatures. Pharmaceutical compounds were detected in the effluents at concentrations within or above ranges reported for large treatment plants while phthalate ester concentrations were below commonly reported effluent concentrations.

1. Introduction

On-site wastewater treatment systems are used for the treatment and disposal of domestic wastewater in areas where households are not connected to a municipal sewage network. Treatment is mostly achieved via the implementation of septic tanks followed by soil-based systems (SBS) such as drain fields or sand filters (Envall et al., 2020; Eveborn et al., 2012; Heinonen-Tanski and Matikka, 2017; Herrmann et al., 2017). Treatment efficiency is often poor due to construction errors, operational deficiencies, or inadequate maintenance (Heinonen-Tanski and Matikka, 2017; Lehtoranta et al., 2022). Consequently, these

systems contribute to the release of e.g., nutrients and pathogens into the environment with adverse effects on water sources (Heinonen-Tanski and Matikka, 2017; Hübinette, 2009; Larsson et al., 2017; Thomasdotter, 2008; Vidal et al., 2018).

As an alternative to SBS, package plants (PP) have been developed. These are prefabricated treatment units based on widely applied biochemical wastewater treatment processes such as coagulation/sedimentation, aerobic/anaerobic biological degradation and filtration. They have become attractive options in areas where space is restricted, or implementation of soil-based systems is limited by the bedrock, soil composition or fluctuating groundwater tables. While the contribution

* Corresponding author.

E-mail address: Brenda.vidal@ltu.se (B. Vidal).<https://doi.org/10.1016/j.jenvman.2023.118214>

Received 25 January 2023; Received in revised form 20 April 2023; Accepted 19 May 2023

Available online 11 June 2023

0301-4797/© 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

of PP to the number of on-site treatment systems operating in Europe today is still small, their implementation has increased in the past five years and this trend is expected to continue. For example, about 5% of the on-site sewage systems currently operating in Sweden are PP (Olshammar, 2021), compared to 2% six years earlier (Olshammar et al., 2015). The proposal for a revised Urban Wastewater Treatment Directive suggests that the scope of the Directive expands to agglomerations above 1000 p.e. and that new standards are developed for decentralized facilities as discharges from small systems were identified as one of the three main sets of problems to be addressed (European Commission, 2022). The collection and treatment of wastewater from agglomerations smaller than 1000 p.e. was also introduced in the Directive (new article 18), as the Member States have the obligation to assess the risks caused by urban wastewater discharges to the environment and human health and take additional measures when necessary (European Commission, 2022).

Although well-established processes are applied in PP, their efficiency has not been comprehensively studied at full scale. The limited number of available studies and monitoring reports have shown that while PP systems can achieve high removal rates their performance can vary greatly (Heinonen-Tanski and Matikka, 2017; Hübner, 2009; Lehtoranta et al., 2014; Thomasdotter, 2008). Heinonen-Tanski and Matikka (2017) studied different types of PP and reported average removal rates of 90% for BOD₇, 62% for tot-P and 40% for tot-N while Vilpas et al. (2005) assessed activated sludge and chemical treatment based PP and reported removal rates ranging from 85% – 99% for BOD₇, 47%–99% for tot-P and 32%–77% for tot-N.

A special requirement for on-site treatment systems operating in cold climate regions (e.g., Canada, Northern Europe, USA, China and Russia) is the ability to sustain treatment performance during periods of low temperatures. Wastewater treatment systems based on biological processes can be especially vulnerable to cold climate conditions as water temperatures strongly influence microbial growth rates, metabolism and substrate affinities (Nedwell, 1999). While it has been shown that treatment in SBS and PP may be disturbed by winter conditions (Heinonen-Tanski and Matikka, 2017; Kauppinen et al., 2014), studies investigating the effect of air temperature on wastewater temperature and any corresponding effects on purification processes are difficult to find. As PP are expected to be more widely implemented and national regulations to become stricter, better understanding of effects of local conditions on the treatment processes is needed. This is not only because of possible impacts on the receiving environment but also for assessment of PP as suitable systems for on-site wastewater treatment.

Additionally, as for large-scale wastewater treatment plants, the occurrence of micropollutants in the inflow and effluent of on-site systems is a cause of concern. While only a few studies have investigated the presence, removal and discharge of micropollutants by on-site wastewater systems, these have reported the discharge of significant quantities of micropollutants into the aquatic environment (Gago-Ferrero et al., 2017; Gros et al., 2017). In general, high removal rates of per- and polyfluoroalkyl substances (PFAs) and phosphorus-containing flame retardants (PFRs) have been observed in PP, whereas SBS have been shown to remove pharmaceuticals more thoroughly (Gros et al., 2017). However, a lot is still unknown regarding the occurrence of micropollutants on-site treatment systems influent and their efficiency in preventing their released into the environment with discharging effluents. More studies have been conducted at larger plants which can provide useful information applicable at small on-site scales. The biological process appears to be the main factor affecting the removal of contaminants of emerging concern including caffeine and pharmaceuticals, with the retention time and the nitrification processes also being suggested as having an influence in larger wastewater treatment plants (Di Marcantonio et al., 2023).

This study addresses some of the knowledge gaps described in the provided background. The main aim was to investigate the treatment efficiency and possible effects of temperature on pollutant removal in

different types of PP treating domestic wastewater. Eleven full-scale PP systems were systematically sampled. The data collected included chemical and biological quality of inflow and outflow water (organics, solids, nutrients and indicator bacteria), wastewater and air temperature. An additional assessment was done on micropollutants on two of the larger PP.

2. Methods

The treatment efficiency of 11 PP was studied in terms of removal of organics, solids, nutrients and indicator bacteria. In addition, the occurrence of selected micropollutants, including pharmaceuticals and phthalate esters (not explored to date), in the influent and effluent of two designated facilities was assessed. The study design was selected to be able to evaluate the variability of influent and effluent wastewater quality over time and effects of seasonal and other operational variations on treatment in full-scale PP.

2.1. Selected package plants

The 11 evaluated on-site facilities included seven types of batch and continuous flow plants (Table 1). Six plants operated in continuous mode: one with a trickling filter (TF), one with a rotating biological contactor (RBC) and four with activated sludge with phosphorus removal by coagulation (ASC) or alkaline filter (ASF1, ASF2, ASF3) systems. Five plants supplied by three different manufacturers operated in batch mode with activated sludge and coagulation for phosphorus removal (SBR1-5). The facilities were located at ~65 °N latitude in a subarctic climate (Dfc classification, Köppen), with about 600 mm precipitation (half falling as snow on average).

All the systems included either a separate or built-in septic tank for sedimentation of coarse particles before the biological treatment. In plants using coagulation, aluminum-based salts were added directly into the process tank before the sedimentation phase (SBR1-5), or a different chamber after the bioreactor at the inlet of the final clarifier (ASC, RBC, TF). For better visualization, photos of some of the facilities can be found in the Supplementary Material.

2.2. Wastewater sampling and analyses

During the period August 2019–April 2021, covering different seasons and temperature ranges, influent samples were collected from the plant's sedimentation tanks before the biological process facility and effluent samples from an outlet pipe, sampling chamber, or the last chamber containing the treated wastewater before discharge. On each sampling occasion the wastewater temperature, total suspended solids (TSS) contents and pH of the samples was measured, and their turbidity on some occasions. In addition, the water temperature was continuously measured in the process tank of three PP with HOB0® Pendant®MX Temp (MX2201) loggers. Data from local weather stations (Swedish Meteorological and Hydrological Institute, n.d.) were used for the air temperature analyses. Levels of BOD, phosphorus (total and dissolved: tot-P and dis-P, respectively), nitrogen (total, nitrite, nitrate and ammonium nitrogen: tot-N, NO₂-N, NO₃-N, NH⁴⁺-N), and the indicator microorganisms *Escherichia coli* and enterococci were measured in approximately 3 L grab samples of influent and effluent water (and chloride in some of these samples for facilities ASC, ASF1-3, SBR1-2 and TF). Portions of samples used for analyses of BOD and nutrients were stored frozen (<-18 °C) until analysis according to the corresponding standards (See supplementary Material), and portions used to determine densities of the bacteria were stored at 5 °C and examined in an accredited laboratory within 24 h. Detailed information about the physicochemical analyses is presented in the Supplementary Material.

Table 1

Specifications of the 11 evaluated package plants, with codes reflecting the main treatment process. S = sedimentation, AS = activated sludge, C = chemical treatment, D = disinfection, F = alkaline filter, R = rotating biological contactor, TF = trickling filter, PF = polishing filter (mineral wool).

Code	Treatment steps	Type	People connected	Age (years) ^a	Number of sampling occasions (n)	Location
ASC	S, AS, C, D	Continuous	32	5	Influent (10) Effluent (10)	Sweden
ASF1	S, AS, F	Continuous	20–30	2	Influent (9) Effluent (10)	Sweden
ASF2	S, AS, F	Continuous	2	2	Influent (10) Effluent (10)	Sweden
ASF3	S, AS, F	Continuous	3	2	Influent (10) Effluent (10)	Sweden
RBC	S, C, R	Continuous	10–30	30	Influent (4) Effluent (4)	Finland
SBR1	S, AS, C	Batch reactor	4	4	Influent (13) Effluent (10)	Sweden
SBR2	S, AS, C	Batch reactor	4	1	Influent (9) Effluent (7)	Sweden
SBR3	S, AS, C,	Batch reactor	2	1	Influent (2) Effluent (5)	Finland
SBR4	S, AS, C,	Batch reactor	1–4	2	Influent (4) Effluent (4)	Finland
SBR5	S, AS, C,	Batch reactor	5	2	Influent (3) Effluent (4)	Finland
TF	S, TF, C, PF	Continuous	12–14	8	Influent (8) Effluent (9)	Sweden

^a Years in operation when the first sample was taken in 2019.

2.3. Micropollutant analyses

Grab samples of the influent and effluent of two facilities (ASC and TF) and blank samples of the sampling equipment (using tap water) were collected on three occasions (March, June and August 2021) for micropollutant analyses, using a stainless-steel sampler, then stored in glass jars before analysis. The investigated micropollutants included 19 pharmaceuticals (Table 2), an artificial sweetener (acesulfame K), caffeine and 15 phthalate: Bis(2-ethylhexyl) phthalate (DEHP), Bis(4-methyl-2-pentyl) phthalate (BMPP), Benzyl butyl phthalate (BBP), Bis(2-ethylhexyl) terephthalate (DEHT), Dibutyl phthalate (DBP), Diethyl phthalate (DEP), Di-n-hexyl phthalate (DHP), Diisobutyl phthalate (DIBP), Diisononyl phthalate (DINP), Diisopentyl phthalate (DISP), Dimethyl phthalate (DMP), Dioctyl phthalate (DNOP), Dipentyl phthalate (DPP), Dicyclohexyl phthalate (DCHP) and Hexyl-2-ethylhexyl phthalate (HEHP). Samples were analysed in an accredited laboratory using LC-MS/MS and GC-MS/MS techniques (detailed information provided in the Supplementary Material).

Table 2

Analysed pharmaceuticals in the influent and effluent of two package plants (ASC and TF).

Compound	Characterization/uses
Diclofenac	Analgesic and anti-inflammatory
Ibuprofen	Analgesic and anti-inflammatory
Bisoprolol	β-blocking agents
Candesartan	ACE inhibitor
Clarithromycin	Antibiotic
Enalapril	ACE inhibitor
Eprosartan	ACE inhibitor
Fenbendazole	Antiparasitic drug
Fluconazole	Antifungal medication
Gabapentin	Anticonvulsant
Ketoprofen	Analgesic and anti-inflammatory
Levetiracetam	Anticonvulsant
Metoprolol	β-blocking agents
Primidone	Anticonvulsant
Ramipril	ACE inhibitor
Sertraline	Antidepressant
Venlafaxine	Antidepressant
Warfarin	Anticoagulant
Xylometazoline	Nasal decongestant

2.4. Removal efficiency calculations and data analyses

The percentage removal efficiency was calculated for each facility using median influent and effluent values (Table 3) on each sampling occasion. The overall removal of each compound was based on the influent concentrations in samples taken from the last chamber of the septic tanks or influent pipe before the process tank, so the estimated removal excluded possible removal in the pre-sedimentation stage. The pre-treatment steps likely differed across PP that had different operational modes, like continuous flow and batch reactors, with the latter providing less time for particles sedimentation based on TSS results. In the few occasions when influent samples were not taken within 24 h of the effluent samples (mainly due to logistics, time constraints and weather e.g., snow cover), the average of all measured influent concentrations was used to calculate the pair-wise removals. Nitrification rates were estimated by subtracting $\text{NH}_4^+\text{-N}_{\text{effluent}}$ from $\text{NH}_4^+\text{-N}_{\text{influent}}$ then dividing by $\text{tot-N} \times 100$, and denitrification rates by subtracting $\text{NH}_4^+\text{-N}_{\text{effluent}}$, $\text{NO}_{2-3}\text{-N}_{\text{influent}}$ and $\text{NO}_{2-3}\text{-N}_{\text{effluent}}$ from $\text{NH}_4^+\text{-N}_{\text{influent}}$ then dividing by $\text{tot-N} \times 100$.

Bacteriological data were \log_{10} -transformed for statistical analyses, half of the lower detection limit was used for left-censored data, and upper detection limits for right-censored data. The significance of differences between influent and effluent concentrations were assessed using the Wilcoxon signed rank test for non-parametric data and correlations between parameters using Spearman rank correlation analysis (with $\alpha = 0.05$ significance level).

3. Results and discussion

3.1. Relationship between air and process temperatures

The possible effect of air temperature on the wastewater temperature in the process tanks was evaluated in three of the PP (Fig. 1C). The air temperature generally influenced the wastewater temperature as seen in two treatment facilities and indicated by linear regression coefficients obtained for ASF3 and SBR1 (Fig. 1A–B) although other factors, such as snow melting also had a considerable impact. The horizontal distribution of the data in Fig. 1A–B indicates that the water temperature in the process tanks (2–20.6 °C in ASF3 and 1.4–17.5 °C in SBR1) can remain stable across a wide range of ambient temperatures, e.g., from –35 to 30

Table 3
Median (in bold) and minimum and maximum concentrations (in parentheses) of selected parameters measured in the influents (Infl) and effluents (Effl) of the studied package plants. Means and standard deviations are also presented in the Supplementary Material (Table S3).

	ASC	ASF1	ASF2	ASF3	RBC	SBR1	SBR2	SBR3	SBR4	SBR5	TF
BOD ₅ (mg/L)	Infl 67.5 (22–226) Effl 38.9 (14–110)	5 (2.2–22.5) 2.1 (<1–5)	28.4 (12.6–78) 1.8 (1.2–5.8)	41.3 (10.4–106) 3.6 (<1–17.5)	91 (32–480) 5.7 (<5–21)	334 (121–545) 13.8 (1.3–124)	266 (111–545) 3.7 (2–34.4)	220 (160–280) 10.9 (3–15)	116.5 (23–210) 4.7 (3–25)	440 (380–500) 36 (21–48)	139.5 (101–548) 13.2 (6.6–175)
P _{tot} (mg/L)	Infl 3.8 (1.9–6.6) Effl 3.3 (2.3–5.6)	0.6 (0.24–1) 0.58 (0.26–3.2)	8.2 (3.3–12) 1.4 (0.3–8.6)	5.8 (2.5–9.9) 3.1 (0.8–6.4)	4.5 (1.6–9.4) 2.2 (1.1–5.2)	9.6 (3.6–15) 2.1 (0.47–4.9)	14 (9.2–36) 2 (1.1–4.2)	4.4 (3.2–5.7) 0.99 (0.4–2.8)	3.8 (1–5.4) 0.2 (0.1–0.5)	25 (23–25) 14.5 (10–21)	6 (4.2–18) 0.3 (0.21–3)
P _{diss} (mg/L)	Infl 2.8 (1.4–4.8) Effl 2.2 (1.3–3.6)	0.4 (0.2–1.1) 0.5 (0.2–1.5)	7.05 (3.2–9.9) 0.7 (0.16–1.5)	4.8 (2.1–8.8) 3 (0.67–6.3)	3.8 (1.4–7.7) 1.8 (1–4.8)	8.9 (2.2–16) 0.2 (0.01–0.8)	11 (8.2–27) 1.1 (0.2–1.7)	3.2 (2.2–4.2) 0.7 (0.2–2.3)	2.9 (0.6–4.2) 0.05 (0.01–0.2)	21 (20–22) 13 (9.5–18)	4.3 (3.5–15) 0.07 (0.02–0.7)
N _{tot} (mg/L)	Infl 40.5 (28–65) Effl 39.5 (30–69)	8.2 (5.5–12) 6.7 (4.6–12)	47 (32–64) 39.5 (22–50)	38.5 (22–84) 26.5 (18–56)	39 (15–73) 27 (12–59)	110 (78–160) 44 (10–83)	140 (70–210) 18 (9–66)	59 (53–65) 36 (4.8–57)	62.5 (28–89) 46 (23–78)	230 (220–240) 190 (87–220)	63 (49–77) 47 (39–84)
NH ₄ (mg/L)	Infl 34 (20–59) Effl 31.5 (22–62)	4.0 (1.4–10) 1.04 (0.01–5.4)	25.5 (18–33) 6.7 (0.9–14)	26 (15–65) 18 (3.4–42)	28 (11–47) 0.7 (0.09–35)	92 (27–130) 14 (0.68–42)	110 (65–170) 3.4 (1.7–22)	49 (46–52) 14 (4.9–23)	34 (22–80) 34 (15–48)	180 (180–210) 175 (52–180)	53 (42–57) 45 (37–62)
E. coli (log ₁₀)	Infl 6.1 (5.2–6.7) Effl 5.9 (4.4–6.7)	4.4 (3.9–5.2) 2.7 (2.5–4.04)	6.4 (4.4–7.7) 4 (2.7–5)	4.6 (4–5.5) 2.8 (1.3–4.3)	5.3 (5–5.7) 2.4 (2–4.2)	5.7 (4.3–6.3) 3.3 (2.4–4.4)	6.4 (2.6–7) 4.2 (3.3–5.7)	1.7 (1.7–1.7) 2.5 (<2–2.8)	4.9 (2.5–5.3) 3.6 (2.4–5.4)	2.8 (2.6–5.1) 4.5 (3–5.6)	6.4 (5.1–6.5) 4.7 (3.8–6.4)
Enterococci (log ₁₀)	Infl 5.2 (4.5–6.3) Effl 4.97 (3.04–5.7)	3.5 (2.7–4.4) 2.1 (<1–2.8)	5.5 (4.5–6.3) 2.9 (2.4–5.2)	2.1 (1–3) 1 (<1–2.7)	5.04 (4.6–5.4) 3.3 (2–3.8)	4.3 (4.1–5.3) 2.7 (1.7–4.3)	4.5 (3.6–4.9) 2.7 (1.8–3.1)	5.5 (5.5–5.5) 1.9 (<1–5.1)	3.03 (1–4.3) 2.1 (1–3)	4.4 (4.3–4.6) 3.6 (3.3–4.4)	4.4 (3.7–5.8) 2.2 (1.7–6.4)

°C, suggesting strong buffering.

Bunce and Graham (2019) observed similar buffering of influent wastewater temperature while monitoring 12 small treatment plants in rural UK (range: 4–19.1 °C) in air temperatures ranging from −1.4 to 24.3 °C. They concluded that seasonal changes were not strong predictors of the reliability and performance of such plants.

The wastewater temperature in the facilities remained positive during the monitoring period, with the lowest values recorded being 1.4, 1.7 and 2 °C in SBR1, ASF2 and ASF3, respectively, during April–May in both years (Fig. 1C). The coincidence of the lowest temperatures with the snowmelt period indicates that cold water from melting snow may have infiltrated the surrounding soil, cooling the process tanks and/or water infiltrated into the sewage pipes before the plants. The influent BOD concentrations in the two samples taken during April–May from SBR1 were lower (140 and 121 mg L^{−1}) than the median (334 mg L^{−1}, Table 3), indicating infiltration of snowmelt. The chloride analysis corroborated the dilution as the influent chloride levels measured in this period were 20.8–26 mg L^{−1}, more than 2-fold lower than those recorded in the summer months (June/August: 55.7–67.7 mg L^{−1}; Table S4). Measures to mitigate snowmelt infiltration into sewers feeding the plants could reduce the cooling effect. Moreover, heat provided by the influent domestic wastewater may significantly help to keep treatment plants at operable temperatures (Viraraghavan, 1985). Biological activity and insulation provided by the snow cover and the systems' components (e.g., an insulating lid) may also contribute to the stability of the wastewater temperature.

3.2. Treatment processes and temperature effects

3.2.1. Organic matter removal

BOD concentrations were significantly lower in effluents than influents at seven of the 11 facilities, with removal rates of 70% at ASF1 and 91–99% at ASF2, ASF3, RBC, SBR1, SBR2 and TF. At the facilities SBR3, SBR4 and SBR5, BOD concentrations were also substantially lower in the effluents than in the influents, but the low number of data points (n = 4–5) limited statistical analysis of the BOD treatment. Among the systems with suitable number of sampling events, the BOD removal levels set by the Swedish authorities (70–90%) (SwAM, 2016) were met by seven PP and those set by Finnish authorities (80–90%) (Finnish Ministry of the Environment, 2017) were met by six. The facility ASC achieved only 42% BOD removal and 22% TSS removal, with effluent presenting high suspended solids (TSS_{Eff} = 51.2 ± 16.9 mg L^{−1}), high turbidity (median value: 115 Nephelometric Turbidity Units) and large variations in the effluent BOD concentrations (54.6 ± 35.9 mg L^{−1}) indicating that its treatment process was performing poorly. In addition, very low median influent concentrations of BOD, phosphorus, nitrogen and indicator bacteria presumably contributed to low removal rates at facility ASF1 (Table 3), where chloride measurements on two occasions (13.1 ± 2.5 mg L^{−1}; Table S4) indicated that dilution of influent wastewater had occurred. As the BOD in the influent was low, the removal of BOD was below regulations in Sweden and Finland, and also in the US where a 30-d average of <30 mg L^{−1} BOD and <30 mg L^{−1} TSS are required (EPA, 2004).

An inverse relationship trend between effluent BOD concentrations and effluent wastewater temperature was observed at ASF2, ASF3, SBR1, SBR2, SBR4, SBR5 and TF facilities (some of them shown in Fig. 2), although no significant correlations were found between the two parameters ($p = 0.823$). In addition, no correlation was found in the pairwise comparison between BOD removal (calculated from influent and effluent concentrations recorded on each sampling occasion) and effluent temperature ($p = 0.092$). The food-to-microbe ratio in some of these systems may not be optimal, which can have a major impact on treatment performance and could explain some of the variability within the data.

The removal of BOD and TSS in activated sludge-based plants is expected to improve with increasing temperatures due to enhancements

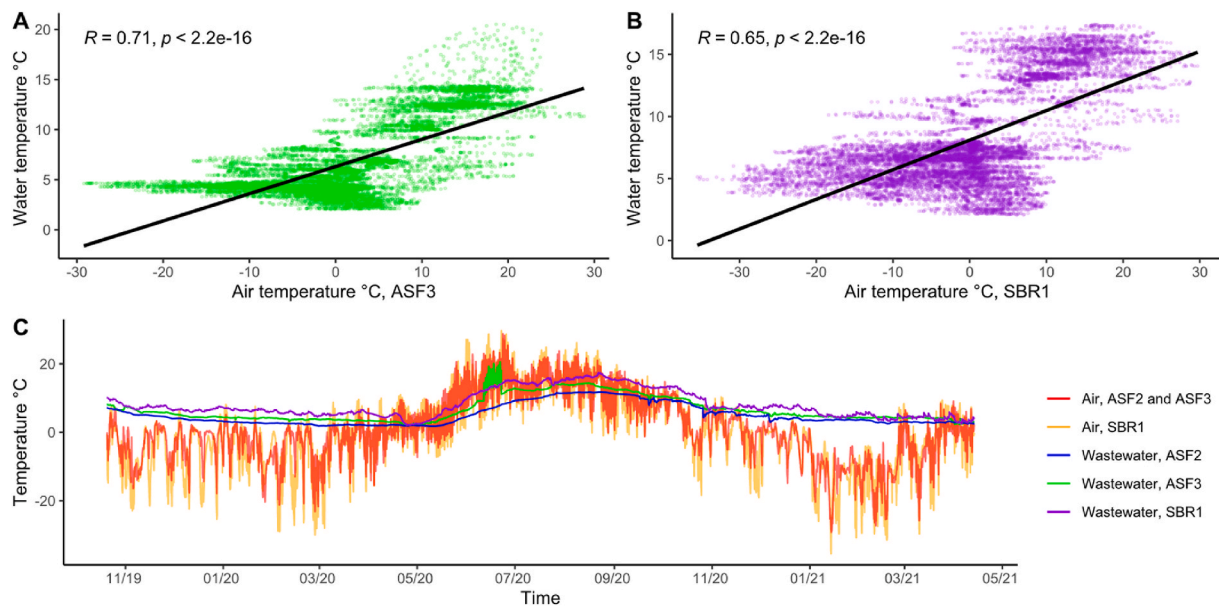


Fig. 1. A and B: Relationships between air and wastewater temperatures in process tanks of two selected facilities, ASF3 and SBR1, respectively. “R” refers to the correlation coefficient and “p” to the p-value indicating the statistical significance. C: Time series of the continuous water temperature measurements in the process tanks of three facilities (ASF2, ASF3 and SBR1) and corresponding air temperatures.

of microbial activity and floc sedimentation (Keefer, 1962; Niku and Schroeder, 1981). However, studies on BOD removal have reported both strong and weak correlations between water temperature and BOD effluent concentrations (Niku and Schroeder, 1981). In this study, in the PP SBR1 e.g., BOD removal was lower during cold months (74% removal

at $T_{Inf} = 7.5\text{ }^{\circ}\text{C}$ and $T_{Eff} = 6.5\text{ }^{\circ}\text{C}$) compared to warm months (>96% removal at $T_{Inf} > 11.5\text{ }^{\circ}\text{C}$). However, it was even lower (<30%) during the snowmelt period, which had below average influent concentrations ($BOD_{Inf} = 140\text{ mg L}^{-1}$) and elevated effluent concentrations ($BOD_{Eff} = 100\text{ mg L}^{-1}$). Therefore, the effect of low wastewater temperatures on

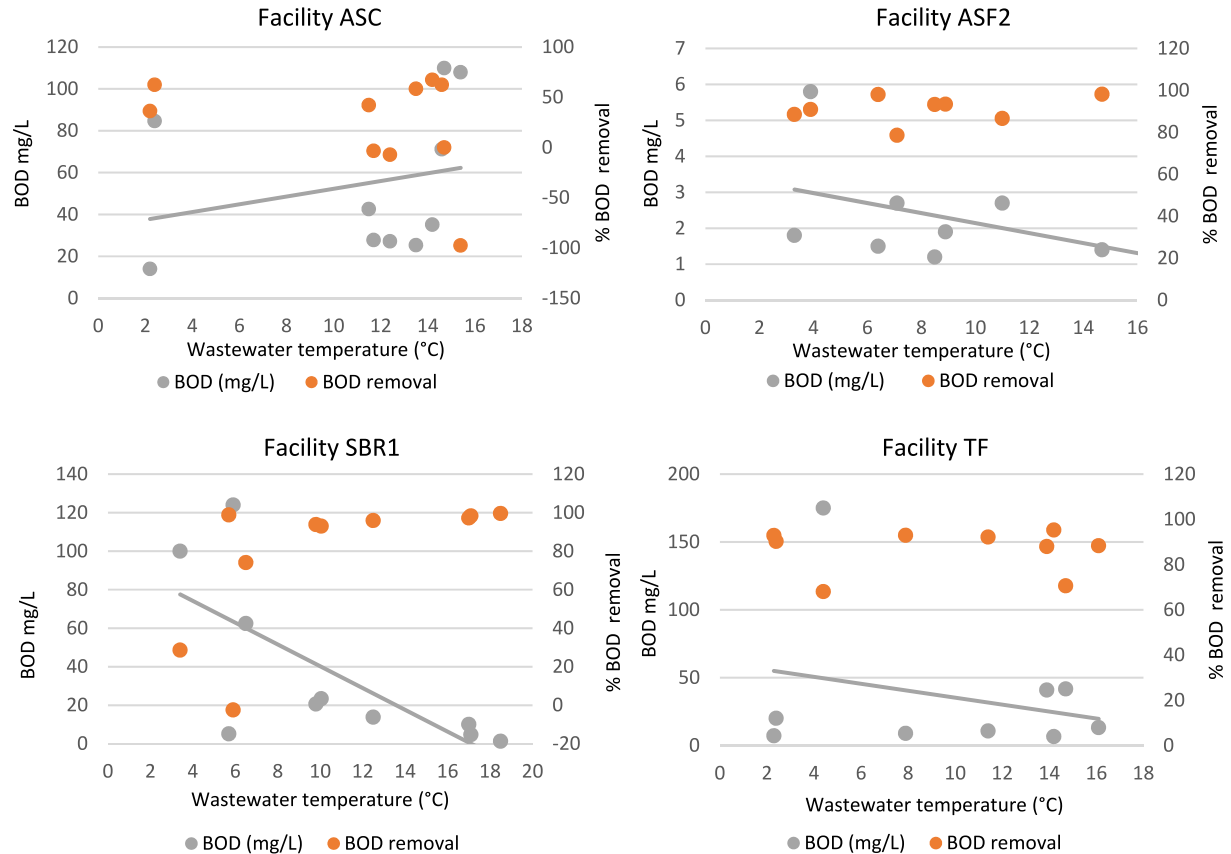


Fig. 2. Effluent BOD, BOD removal rates estimated per sampling event and temperature at four selected facilities with sample sizes $n \geq 9$. The lines show data trends and do not indicate significance.

BOD removal were also exacerbated by the dilution of inflow water which reduced influent BOD concentration and wastewater retention time in the system (Niku and Schroeder, 1981).

3.2.2. Nitrogen treatment

Most of the influent nitrogen was in the form of ammonia in all facilities (Table 3). Tot N removal (the difference between influent and effluent tot N concentrations) was only significant (Wilcoxon signed rank test, p -value < 0.01) at five of the plants: ASF2 (21%), ASF3 (23%), SBR1 (55%), SBR2 (79%) and TF (17%). Most of the nitrogen removed was organic, likely through particle sedimentation or filtration (in the P filters). Nitrification occurred in most studied PP, to different degrees. For example, up to 51 and 78% of the NH_4^+ -N was nitrified in facilities SBR1 and SBR2, respectively, while in ASF2 and ASF3 less than 44 and 27% of the NH_4^+ -N was nitrified, respectively. In facilities SBR1 and SBR2, denitrification of NO_{2-3} also contributed to tot N removal (42% and 64% of the nitrified N was denitrified). Nine of the 11 plants did not meet N removal levels set by the Swedish authorities (50% (SwAM, 2016),) and Finnish authorities (30–40%, (Finnish Ministry of the Environment, 2017)). In SBR1 and SBR2, effluent tot-N concentrations (18.9 ± 13.3 and $7.2 \pm 7.4 \text{ mg L}^{-1}$, respectively) were within ranges of concentrations measured in effluents of other facilities due to the high influent concentrations (Table 3), likely due to partial recirculation of the sludge, despite good removal rates.

Package plants' configurations strongly influence their nitrogen removal capacity, as the treatment mechanisms depend on variables such as substrate quality and quantity, pH ranges, and the presence of aerobic and anaerobic niches for microorganisms capable of transforming nitrogenous compounds (Sharma and Ahlert, 1977). Nitrogen removal by denitrification can be fostered by recirculating the water or sludge from the aerated process tank into a tank with sufficient substrate such as the primary sedimentation or septic tank with anaerobic

conditions (Shaw and Dorea, 2021). This was only possible for the batch reactors, so PP which operated in that mode, i.e., SBR1 and SBR2, efficiently removed N. Certain degrees of denitrification have also been observed by Johannessen et al. (2012) in PP that provide organic-rich substrate in anaerobic environments through wastewater recirculation and contact between the nitrate and influent wastewater. Moreover, anaerobic conditions could be unintentionally provided at the bottom of tanks where chemical precipitation occurs, e.g., in facility TF. However, the alkaline effluent environment in facilities with reactive filter material (e.g., ASF2 median $\text{pH}_{\text{eff}} = 10$ and ASF3 median $\text{pH}_{\text{eff}} = 9.3$) could inhibit denitrification due to high pH (above 10 as discussed by Renman et al. (2008)).

No significant correlation was found between the wastewater temperature and effluent total N concentrations ($p = 0.301$) or NO_{2-3} -N ($p = 0.619$; Fig. 3) in the whole dataset and individual facilities. The only exception was facility SBR1 where strong correlations between the water temperature and the effluent tot-N ($r = -0.9$) and NH_4 -N ($r = -0.75$), but not ΔNO_{2-3} -N ($p = 0.364$), were found, likely indicating the removal of organic particulate nitrogen. These results are in line with previous studies of on-site treatment systems including PP e.g., Vilpas and Santala (2007) where no correlation between the air or wastewater temperature and nitrogen removal was found.

Temperature reportedly affects the ammonia-oxidizing bacterial community, together with other variables such as pH, alkalinity, oxygen concentration, retention time and organic load. Bacterial growth and activities can be retarded or inhibited by low temperatures (Rodríguez-Caballero et al., 2012), so nitrification rates are expected to be higher during warmer periods. Temperatures below 8.3°C reportedly limit nitrification, and little or no growth of nitrifying bacteria occurs at temperatures below 4°C (Sharma and Ahlert, 1977; Taylor Eighmy and Bishop, 1989).

In this study, despite the lack of strong correlations between

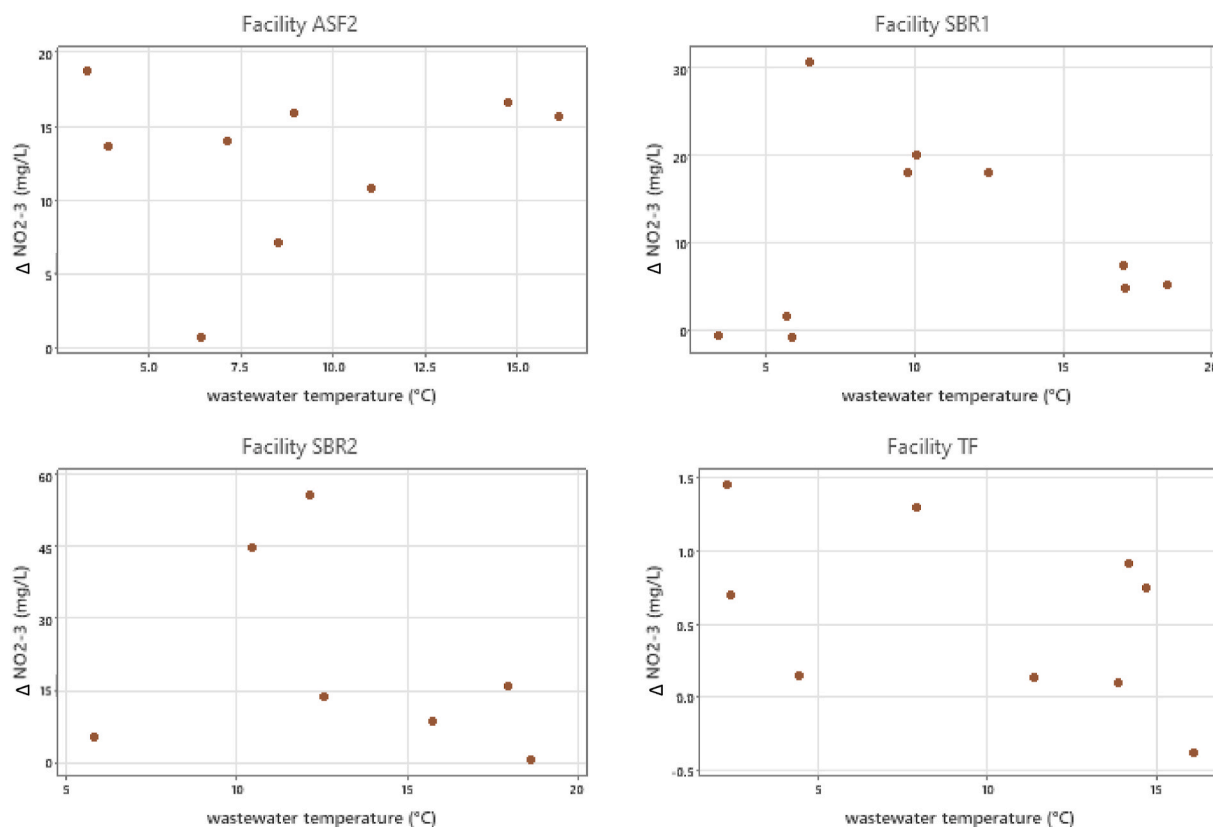


Fig. 3. Effluent temperatures and effluent minus influent NO_{2-3} -N concentrations in four selected facilities where nitrification occurred to some extent and the sample size was $n > 7$.

wastewater temperature and nitrogen removal, the trends observed in facilities with efficient BOD reduction ($n > 7$) indicated a generally inverse relationship between effluent nitrogen (tot-N and $\text{NH}_4\text{-N}$) and wastewater temperature. Measured temperatures in the studied facilities were lower than the range considered optimal for growth of nitrifying bacteria (28–36 °C) and often below the limit for nitrification and growth of nitrifiers (Sharma and Ahlert, 1977), which likely explains the low nitrification rates in most facilities. For example, nitrification rates in SBR1 and SBR2 were highest during the warmest periods (<18.6 °C) and lowest in cold periods (e.g., nitrification rates at 3.4, 5.6 and 17 °C at SBR1 were negative, 36% and 87%, respectively). Difficulties in establishing a thriving community of nitrifying bacteria and maintaining it stably during cold periods in most plants probably exacerbated disruptions of nitrogen removal. Furthermore, the hydraulic residence time in small PP is likely too short for high nitrification rates to be reached (Dincer and Kargi, 2000).

Another important factor for nitrification is pH. The pH of wastewater treated in SBR1 and SBR2 (plants with high tot N removal) decreased substantially from 6.9 to 7.9 and 8.1–8.9 to 4.9 and 6, respectively. Nitrification consumes alkalinity and is normally inhibited by low pH (Sharma and Ahlert, 1977). The decrease in wastewater pH values observed in SBR1 and SBR2 indicated both consumption of alkalinity due to nitrification and low buffering capacity. Adjustment of pH in plants where N removal is targeted might be necessary to sustain nitrification.

3.2.3. Phosphorous removal in different P-targeting facilities

In eight of the 11 studied plants chemical coagulation and sedimentation were the main phosphorus-removal processes (Table 1). The effluent tot-P concentrations were significantly lower (Wilcoxon signed rank test, p -value <0.02) than the influent concentrations in three of the eight facilities that included chemical precipitation (SBR1, SBR2 and TF). The tot-P median concentrations in RBC, SBR3, SBR4, SBR5 were lower in the effluent than in the influent (Table 3), but the limited data did not allow further statistical analysis.

The proportion of dissolved phosphorus was higher in influent than in effluent samples (Table 4) from most facilities (all except RBC, SBR3 and SBR5), indicating that some of the dissolved phosphorus precipitated and the particulate form predominated after the treatment.

Coagulation/flocculation of phosphorus is generally affected by temperature, because effects of reducing the temperature on the solubility of aluminum hydroxide species raise the optimal coagulation pH, and change floc characteristics such as size (Bratby, 2016). However, we detected no significant correlation between the wastewater temperature and total phosphorus removal rates ($p = 0.277$), or effluent concentrations ($p = 0.08$). Neither the measured effluent concentrations nor calculated removal rates had a clear monotonic relationship with the wastewater temperature. The lack of correlation between temperature and total phosphorus concentration was likely due to the high variability

of the data, and effects of other parameters, such as type and dosage of coagulant (assumed to be similar between the systems), pH, alkalinity and ionic strength (Clark and Stephenson, 1999; Zhou et al., 2018).

In the facilities that included chemical precipitation treatment the effluent TSS concentration was strongly correlated with effluent concentrations of tot-P ($r = 0.69$, $p = 0.000$) and particle-bound P, estimated from differences between tot-P and diss-P ($r = 0.78$, $p = 0.000$). Thus, the low phosphorus removal observed in some plants could be due to insufficient sedimentation and subsequent release of particles. Furthermore, the pH strongly influences phosphorus precipitation due to its impact on chemical charges and competition between metal hydrolysis products and natural organic compounds (Bratby, 2016). The optimum pH reported for phosphorus removal (using aluminium coagulants) is 5.5–6.0 (Bratby, 2016), so the chemical precipitation of phosphorus may have been impaired in facilities with median pH lower than that range, such as SBR1 which had $\text{pH} < 5$ in more than half of the effluent samples. The P treatment was also probably compromised in the investigated PP by the lack of thorough mixing (high turbulence) at the dosing points, which is required for good precipitation of dissolved P (Kroiss et al., 2011). In summary, suboptimal pH, low chemical dosage, insufficient mixing of coagulant, insufficient flocculation and sedimentation, equipment malfunction or lack of coagulant are commonly reported reasons for low P removal in PP (Johannessen et al., 2012; Kroiss et al., 2011) and may explain the low removals observed in some of the plants we studied.

Three of the facilities studied had alkaline filters for P removal (Table 1): ASF1, ASF2 and ASF3, with estimated tot-P removal rates of 3%, 83% and 46%, respectively. Influent wastewater into ASF1 was clearly affected by dilution, as previously discussed, and was not considered in this analysis. Estimated pair-wise removal rates varied considerably with time at each facility. For example, ASF2 removed between 28% of tot P in February 2020, when there was an unusually high effluent tot-P concentration (8.6 mg L^{-1}) and 94% in June 2020, and on both occasions the effluent pH was high (10.1 and 9.7, respectively). ASF3 removed tot P less efficiently, between 23% (in June 2020) and 83% (in October 2019), with effluent pH values (7.8 and 9.7, respectively) lower than those measured in ASF2.

The phosphorus removal capacity of alkaline filters is affected by the residence time, influent phosphorus concentrations, pH and temperature. The removal mechanism mainly involves precipitation reactions after dissolution of calcium ions from the filter material (Eveborn et al., 2009; Jucherski et al., 2021; Vohla et al., 2011). Precipitation of calcium phosphates is favored by high pH (Feenstra and De Bruyn, 1979), which gradually decreases with time during operation (Eveborn et al., 2009; Renman and Renman, 2010). As pH falls below 9 due to exhaustion of the alkaline filter media, previously precipitated calcium phosphates may dissolve (Eveborn et al., 2009). In this study, the effluent pH measured in facility ASF3 was below 9.9 at the beginning of the sampling campaign (Fig. 4b) and decreased over time, indicating that the

Table 4

Average influent and the effluent diss-P/tot-P ratios, effluent mean concentrations and removal rates for all investigated facilities. C = chemical treatment; F = alkaline P-filter.

Facilities	P-removal method	Influent diss-P/tot-P ratios		Effluent diss-P/tot-P ratios		Effluent concentration (mg L^{-1})		P removal (%)
		Mean	StDev	Mean	StDev	Diss-P	Tot-P	
ASC	C	0.75	0.03	0.65	0.09	2.5 ± 0.8	3.8 ± 1.2	13
ASF1	F	0.78	0.06	0.83	0.16	0.7 ± 0.4	0.9 ± 0.9	3
ASF2	F	0.89	0.04	0.55	0.30	0.7 ± 0.4	1.98 ± 2.4	83
ASF3	F	0.90	0.05	0.89	0.24	2.9 ± 1.8	3.2 ± 1.6	46
RBC	C	0.84	0.02	0.88	0.07	2.4 ± 1.7	2.7 ± 1.9	50
SBR1	C	0.86	0.07	0.27	0.35	0.3 ± 0.3	2.4 ± 1.5	78
SBR2	C	0.84	0.03	0.44	0.24	0.98 ± 0.6	2.3 ± 0.98	86
SBR3	C	0.71	0.02	0.77	0.16	0.98 ± 0.8	1.2 ± 0.99	71
SBR4	C	0.73	0.03	0.33	0.29	0.07 ± 0.07	0.2 ± 0.2	95
SBR5	C	0.86	0.03	0.90	0.05	13.4 ± 3.9	15 ± 4.7	42
TF	C	0.81	0.03	0.26	0.16	0.1 ± 0.2	0.8 ± 0.9	95

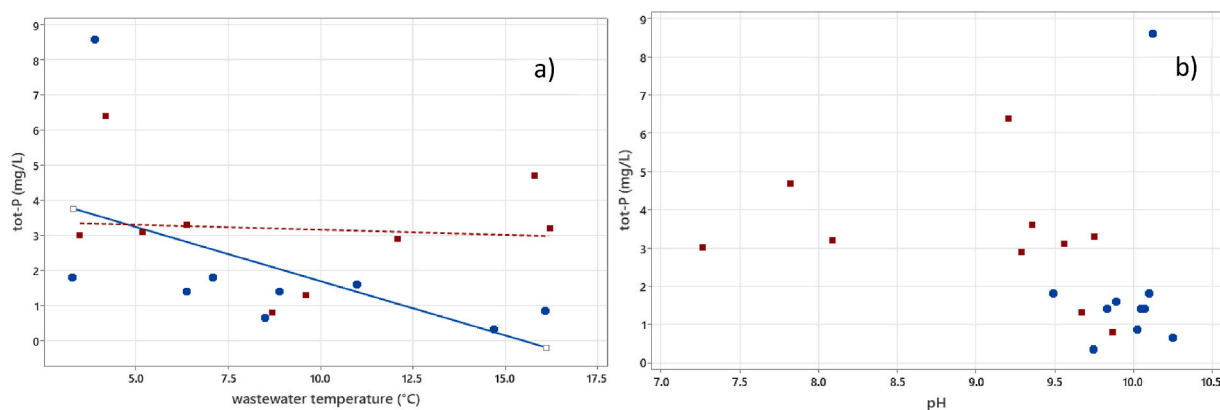


Fig. 4. Relations between the water temperature (a) and effluent pH (b) with effluent phosphorus concentrations in facilities ASF2 (blue) and ASF3 (red) featuring alkaline P-filters (facility ASF1 is not shown due to the dilution issues). Sequential regression coefficients (R-Sq) values for the ASF2 and ASF3 data are 30 and 0.7%, respectively.

filter media was reaching exhaustion. The low pH at the end of the sampling campaign (pH = 7.3), and higher tot-P concentrations in the effluent than influent indicate that some dissolution of previously formed calcium phosphates may have occurred. Moderately negative correlations (based on combined data from ASF2 and ASF3) were found between the effluent pH and effluent tot-P concentrations ($r = -0.47$, $p = 0.04$) and diss-P ($r = -0.60$, $p = 0.006$), confirming the inverse relation between these parameters.

The ratios of dissolved phosphorus to total phosphorus (diss-P/tot-P) were higher in the influent than the effluent of ASF2 (0.89 ± 0.04 and 0.55 ± 0.3 , respectively), but the same (0.9) in the influent and effluent of ASF3, with high effluent diss-P concentrations (Table 3), indicating that the soluble phosphorus had not been successfully precipitated, while the measured removal was mainly due to particulate phosphorus.

A strong correlation between the effluent wastewater temperature and effluent phosphorus concentrations was found at facility ASF2 ($r = -0.71$, $p = 0.03$), but not at ASF3 ($p = 0.86$) (Fig. 4a). Previous laboratory studies have shown that the temperature significantly affects the binding capacity of phosphorus in alkaline filter materials, as precipitation of calcium phosphates is an endothermal reaction, favored by high temperatures (Herrmann et al., 2014; Jucherski et al., 2021). Herrmann et al. (2014) found that total P binding capacities of two tested filter materials increased 1.2- and 1.5-fold when the temperature was increased from 4.3 to 16.5 °C. Because the filter material in ASF3 was only exhausted towards the end of the sampling campaign, changes in its chemistry may have masked temperature's negative impact on the treatment process. Nevertheless, the highest effluent tot-P concentrations we recorded (6.4 and 8.6 mg L⁻¹) were at ASF2 and ASF3 during cold months (4.2 and 3.9 °C, in February 2020).

3.2.4. Inactivation of indicator bacteria

High concentrations of indicator bacteria were found in effluents of most plants, <2.3 – 5.9 (log₁₀) *E. coli* and <1 – 4.9 (log₁₀) enterococci per 100 mL, exceeding in most cases European Union limits for acceptable bathing water quality—200 cfu/100 mL for enterococci and 500 cfu/100 mL for *E. coli* (EU, 2006)—in line with similar studies of on-site systems (Hübinette, 2009; Vilpas and Santala, 2007). Similar inactivation rates were obtained by plants with different configurations e.g., ASF2 operating in continuous flow and with an alkaline P-filter had an average removal of 2.5 log₁₀ for *E. coli* whereas SBR1 operating in batches had 2.1 log₁₀ removal. The removal rates were generally low, <2.5 (log₁₀), suggesting that extra polishing treatment steps (e.g., chlorination and UV radiation (EPA, 2003)) would be needed to obtain more hygienic effluents. Likely, the short retention times typically found in on-site PP did not allow enough sedimentation, encapsulation in the flocs, natural decay or predation mechanisms to occur (Henze et al.,

2008). Methods like chlorination or UV radiation can be implemented as tertiary treatment and their disinfection success is related to the concentration of colloidal and particulate constituents in the wastewater. While low dosages of UV radiation may not inactivate some viruses, spores and cysts, the method has no residual effect that could harm humans or aquatic life, as compared to chlorine (EPA, 2003).

No correlation was found between the effluent temperature and densities of either *E. coli* ($p = 0.154$) or enterococci ($p = 0.834$), in line with previous studies of conventional activated sludge systems operating at 9–15 °C (Barrios-Hernández et al., 2020). However, regression analyses of variables at specific facilities revealed strong inverse correlations at SBR1 between wastewater temperature and effluent densities of both enterococci and *E. coli* (R-Sq = 70.9 and 48.1%, respectively, $n = 9$). Moreover, at SBR1, the lowest *E. coli* effluent densities (log₁₀ 2.4) were recorded in August, when the water temperature was one of the highest measured (about 17 °C), and highest (log₁₀ 4.4) in April, when the water temperature was coldest ($T_{\text{influent}} = 3.7$ °C, $T_{\text{effluent}} = 5.9$ °C).

3.3. Occurrence and removal of micropollutants in selected facilities

Twelve of the 19 analysed pharmaceutical compounds were detected in at least one of the influent samples taken from facilities TF and ASC (Table S1). Of these 12 compounds, nine were detected in effluents of the TF and nine (not the same) on effluents of ASC facilities (Table 5). The pharmaceuticals detected included anti-inflammatory drugs (diclofenac, ibuprofen and ketoprofen), β -blocker drugs (bisoprolol and metoprolol), an ACE inhibitor (enalapril), anticonvulsants (gabapentin and levetiracetam) and the antidepressant venlafaxine. Effluent concentrations of bisoprolol, anticonvulsant levetiracetam and ketoprofen were within ranges of previously reported effluent concentrations from large WWTPs and on-site systems, but those of venlafaxine and metoprolol were slightly higher (Table 5). Effluent concentrations of diclofenac were generally higher than reported concentrations in effluents of conventional wastewater treatment plants (WWTPs), but within ranges of those reported in effluents of on-site sewage systems. In contrast, effluent concentrations of ibuprofen and gabapentin were much higher than those recorded in previous studies. Low concentrations of the ACE inhibitor, enalapril were found, while previously reported concentrations have typically been sub-limit of quantification (Leiviskä and Risteelä, 2022).

The stimulant caffeine and sweetener acesulfame K were detected in all the influent and effluent samples of the two studied facilities. Acesulfame K is a commonly used artificial sweetener with suggested suitability as an indicator of wastewater contamination in the environment due to its persistence (Doummar and Aoun, 2018; Luo et al., 2014). Its insensitivity to treatment was observed in this study, as the influent and

Table 5

Summary of concentrations of the selected pharmaceuticals (except enalapril), caffeine and Acesulfame K ($\mu\text{g L}^{-1}$) detected in influents and effluents in this study and effluents of previous studies. LOQ = Limit of quantification; ND= Not detected. All the measured concentrations are shown in Table S1.

Compound	This study		Other studies
	Range (influent)	Range (effluent)	Range (effluent)
Diclofenac	TF: 0.43–13 ASC: 0.15–2.4	TF: 0.54–14 ASC: 0.85–1.2	0.2 ^a (max.), <0.001–0.69 ^b , 1.4 ^c (max.), 0.03–0.14 ^d , 2.4–3.9 ^e
Venlafaxine	TF: 1.1–1.5 ASC: <LOQ	TF: 1.3–1.4 ASC: <LOQ	0.548 ^a (max.), 0.5–1.3 ^e ,
Bisoprolol	TF: <LOQ–0.06 ASC: 0.1–0.8	TF: <LOQ–0.06 ASC: 0.2–0.5	0.423 ^a (max.), 0.04–1.1 ^e
Ibuprofen	TF: 9.6–35 ASC: 17–69	TF: 6.7–15 ASC: 22–67	2.1 ^a (max.), ND–55 ^b , 17 ^c (max.), 0.1–4.2 ^d
Ketoprofen	TF: 0.6–8 ASC: <LOQ	TF: 0.006–1.5 ASC: <LOQ	1.6 ^a (max.), 0.003–3.92 ^b , 2.1 ^c (max.), <0.003–0.03 ^d , 0.1–0.5 ^e
Metoprolol	TF: 2.1–3.1 ASC: 0.7–1.7	TF: 1.6–2.9 ASC: 0.5–1.5	0.003–0.25 ^b , 0.03–0.1 ^d , 0.6–1 ^e
Gabapentin	TF: <LOQ–0.01 ASC: 43–62	TF: 0.008–0.08 ASC: 29–78	3–42.6 ^d , <0.9–6.5 ^e
Levetiracetam	TF: 0.7–1.5 ASC: 0.2–0.7	TF: <LOQ–0.8 ASC: 0.3–0.5	0.1–1 ^e
Caffeine	TF: 180–303 ASC: 46–187	TF: 73–192 ASC: 99–149	3 ^a (max.), ND–43.5 ^b , 70.8 ^c (max.), 0.2 ^e (max.)
Acesulfame K	TF: 40–140 ASC: 1.8–11	TF: 37–123 ASC: 7.1–19	2500 ^a (max.)

^a Loos et al. (2013). Effluents from 90 WWTPs.

^b Luo et al. (2014). Review of effluents from WWTPs in various countries.

^c Matamoros et al. (2009). Effluents from 13 on-site wastewater treatment systems.

^d Kasprzyk-Hordern et al. (2009). Effluents of a WWTP treating mainly communal wastewater using trickling filter beds.

^e Leiviskä and Risteelä (2022). Effluents of one WWTP with conventional activated sludge.

effluent concentrations did not vary greatly (Table 5). Caffeine is excreted unchanged in the urine but, in contrast to Acesulfame K, it is readily degradable in conventional WWTPs and conventional on-site sewage systems, with high reported removals (>70%) (Deblonde et al., 2011; Luo et al., 2014; Matamoros et al., 2009). In this study, concentrations in facility TF were clearly lower in the effluent than the influent (Table 5), but not in facility ASC, corroborating the poor performance of ASC's biological treatment.

Diclofenac and metoprolol are considered to be poorly removed (<40%), ketoprofen moderately removed (40–70%), and ibuprofen and caffeine highly removed (>70%) in conventional WWTPs according to a simple classification scheme (Luo et al., 2014). Our results are consistent with these general patterns, particularly for diclofenac and metoprolol, whereas ibuprofen was removed to a lesser extent (<45%) than previously reported and only in facility TF. Caffeine was also removed to a lower degree (about 50% in TF, negligible in ASC) than reported in the literature. In contrast to Kasprzyk-Hordern et al. (2009), we did not detect high removal (about 84% was reported) of the anticonvulsant gabapentin by activated sludge treatment. The antidepressant venlafaxine was found in all the influent and effluent samples of facility TF, in similar ranges, confirming its persistence in aerobic processes and ability to resist wastewater treatment, as previously reported (Falås et al., 2016).

Caffeine, ibuprofen, metoprolol and venlafaxine may be environmentally relevant micropollutants, based on their removal efficiency, estimated persistency, bioconcentration factor, toxicity potential, concentration in wastewater effluents and frequency of detection in samples (Gros et al., 2017). Use of ibuprofen is considered to result in moderate environmental risk and the predicted non-effect concentration (PNEC) is $1 \mu\text{g L}^{-1}$ according to the Swedish pharmaceutical database (FASS, n.d.). The lowest PNEC for diclofenac is $0.05 \mu\text{g L}^{-1}$ according to the Norman Ecotoxicology Database (Norman, n.d.). A suggested environmental quality standard (EQS) of $0.040 \mu\text{g L}^{-1}$ for diclofenac is under consideration for inclusion in the EU water legislation (watchlist) (Leverett et al., 2021). All the effluent diclofenac concentrations recorded in this study exceeded $0.5 \mu\text{g L}^{-1}$ (Table 5 and Table S1), implying environmental hazards because of its toxicity to aquatic organisms.

Seven of the 15 analysed phthalates (DEHP, BBP, DEHT, DBP, DEP, DIBP, DMP) were detected on at least one occasion in effluents of the two studied plants (Tables S2a–b, Fig. 5). The highest concentrations measured in the effluents were of the phthalates DEP and DIBP (380 and 270 ng L^{-1} in TF effluents, 240 and 310 ng L^{-1} in ASC effluents, respectively). Concentrations of DBP and DEHT were also high in effluents of facility ASC (408 and 380 ng L^{-1} , respectively). The influent and effluent concentrations of the mentioned phthalates were below concentrations reported in effluents of conventional WWTPs (Deblonde et al., 2011; Gao and Wen, 2016; Luo et al., 2014), likely due to their smaller scale (e.g., excluding industrial wastewater). The concentrations of phthalates in facility TF were always lower in the effluents than the influents, indicating some removal, but concentrations were higher in some effluents from the plant ASC than the influents (Fig. 5, Table S2a).

DEHP is included in the European Union's list of priority substances (EC, 2013). The highest DEHP effluent concentration measured in the present study was $0.27 \mu\text{g L}^{-1}$ (Table S2a), which is lower than the EQS (annual average value) for inland and other surface waters of $1.3 \mu\text{g L}^{-1}$ (EC, 2013) and not shown in the graph due to the high concentrations found in two of the three blanks taken.

Biodegradation by bacteria and fungi, or adsorption, are considered important mechanisms in the degradation and transformation of phthalate esters (Gao and Wen, 2016). The degradation is much slower in anaerobic than in aerobic conditions because of the lack of syntrophic microbial communities, together with sub-optimal temperatures, pH, initial phthalates concentrations and carbon sources (Gao and Wen, 2016). The removal rates in conventional WWTPs are reportedly high for most phthalates, including DEP, DBP, BBP, DEHP and DMP, e.g., >90% according to Deblonde et al. (2011) and 73–87% according to Gao and Wen (2016). We found stronger removal of phthalates in facility TF than in ASC (Fig. 5.), in accordance with the other measured parameters and indicating that removal of phthalate esters will be sub-optimal in PP with suboptimal biological treatment.

Micropollutants' fate and likelihood of escape in WWTP processes are affected by various characteristics, such as their sorption capacity (affected by their hydrophobicity) and biodegradability (affected by their bioavailability), so non-volatile and polar compounds are most

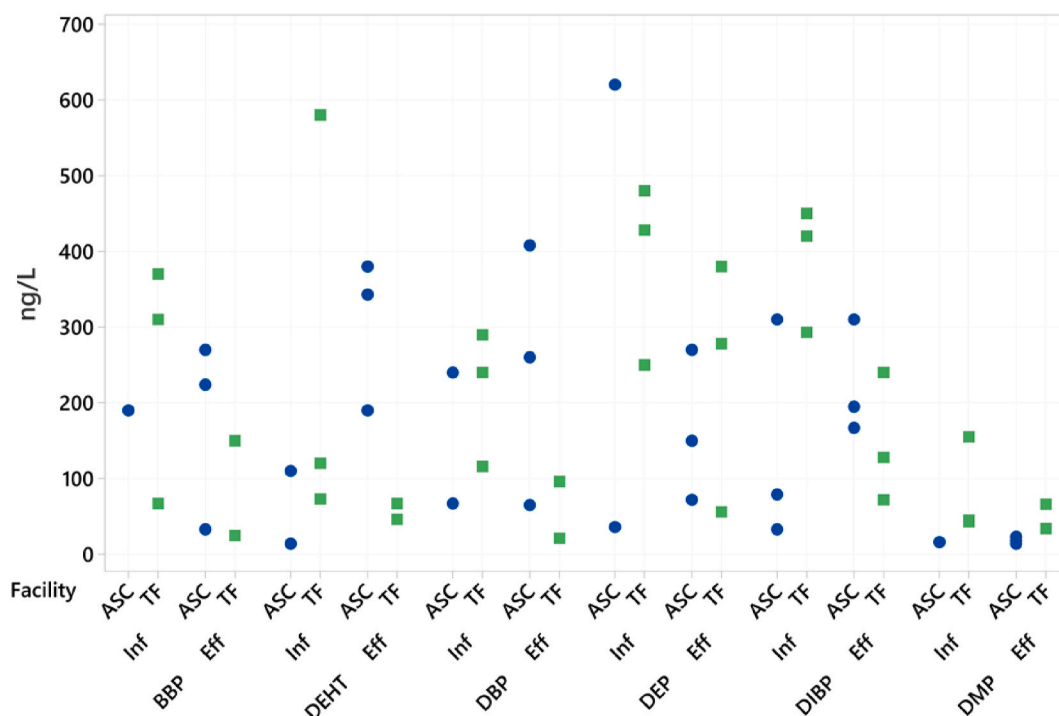


Fig. 5. Influent and effluent concentrations of six selected phthalate esters detected in the two studied facilities, ASC (blue markers) and TF (green markers). All the measured concentrations, including blanks, are shown in Table S2a-b.

likely to remain untreated (Luo et al., 2014). Parameters of the treatment plants, such as sludge and hydraulic retention times, likely influence micropollutants' removal as they affect key components of the microbial community (e.g. nitrifying bacteria), and time available for biodegradation and sorption processes (Fernandez-Fontaina et al., 2012). The PP investigated in this study generally had shorter retention times (and thus presumably the contact time for biodegradation and sorption processes) than full scale WWTPs.

Gros et al. (2017) found no clear differences between on-site PP and medium and large scale WWTPs in concentrations and removals of micropollutants (e.g., pharmaceuticals), and concluded that pharmaceuticals removals in both types of facilities seem to be rather low. However, there are treatment-related variations, e.g., use of trickling filters reportedly results in lower removal of pharmaceuticals (<70%) than activated sludge systems (85%) in full-scale WWTPs (Kasprzyk-Hordern et al., 2009). We found that the facility with a trickling filter (TF) seemed to provide higher removal than the activated sludge plant (ASC), which was underperforming, so the results should not be extrapolated. Most PP do not discharge directly into receiving water bodies, but into ditches or infiltration beds where the effluent is in contact with soil and vegetation, so further treatment processes may occur before the substances reach aquatic environments. Nevertheless, the results indicate that many on-site sewage plants may function poorly and discharge significant amounts of micropollutants into the surrounding environment and receiving waters, in contrast to general results of studies limited to relatively well-functioning sewage systems, as discussed by Blum et al. (2017).

4. Conclusions

The treatment efficiency of eleven PP was studied in terms of removal of organics, solids, nutrients and indicator bacteria. In addition, the occurrence of selected micropollutants, including pharmaceuticals and phthalate esters (not explored to date), in the influent and effluent of two designated facilities was assessed. The conclusions can be summarized as follows:

- Wastewater temperatures remained above freezing ($>1.4^{\circ}\text{C}$) even during periods with the lowest outside temperatures (about -30°C). Snow melting appeared to have a stronger cooling effect (due to water infiltration into the systems) on the wastewater temperatures than air temperatures.
- No strong correlations were found between PP effluent BOD or nutrient concentrations and the wastewater temperature, although e.g., the BOD removal appeared to be the lower in some facilities (e.g., SBR1, SBR2) during the coldest months.
- Nitrogen removal was generally low, however, both nitrification and denitrification were observed. Nitrification rates were high ($>51\%$) in SBR1 and SBR2, especially during the warmest periods. Denitrification was also observed, to a limited extent, in these facilities which included water/sludge recirculation of the nitrified N to the septic tank.
- Phosphorus removal rates were good ($>71\%$) in six of the 11 plants, and highest in plants with coagulation. However, no clear relationships between temperature and total phosphorus concentration or removal were established.
- An effect of temperature on the adsorption/precipitation processes in alkaline P filters was confirmed in one facility (ASF2) where the effluent pH remained high (>9.5), but not in the plant where the filter media was already likely exhausted (ASF3, effluent pH <7.3).
- Large densities of *E. coli* and enterococci indicator bacteria were found in effluents of most plants as the removal rates were low, <2.6 (\log_{10}), suggesting that additional treatment steps are needed to improve sanitation.
- Thirteen of the 19 analysed pharmaceutical compounds were detected in at least one of the effluent samples taken from the two monitored facilities. Seven of 15 analysed phthalates were detected on at least one occasion in effluents of the two studied plants. Therefore, PP for domestic wastewater treatment may be sources of micropollutants for the receiving environment.

Credit author statement

Brenda Vidal: Methodology, Formal analysis, Investigation, Data curation, Writing – Original draft, Writing – Reviewing and editing, Visualization. **Juho Kinnunen:** Methodology, Formal analysis, Investigation, Data curation, Writing – Reviewing and editing, Visualization. **Annelie Hedström:** Conceptualization, Writing – Reviewing and editing, Supervision. **Elisangela Heiderscheidt:** Conceptualization, Writing – Reviewing and editing, Supervision, Project administration, Funding acquisition. **Pekka Rossi:** Conceptualization, Writing – Reviewing and editing, Funding acquisition. **Inga Herrmann:** Conceptualization, Methodology, Writing – Reviewing and editing, Supervision, Project administration, Funding acquisition. All authors approved the final article.

Funding sources

This work was supported by the Interreg Nord European Regional Development Fund (grant no. NYPS, 20201833), Region Norrbotten (grant no. NYPS, 20201991), the Swedish Agency for Marine and Water Management (1:11-anslag för havs-och vattenmiljö, grant nos. 1634/20 and 00929-2021) and J. Gust. Richert foundation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The raw data is available in the Swedish National Data Service (SND) with the following DOI number <https://doi.org/10.5878/48t5-qm50>.

Acknowledgements

The authors thank Kerstin Nordqvist for the laboratory analyses of nutrients, Luleå, Älvsbyn and Haparanda municipalities in Sweden, as well as Tornio and Oulu in Finland, for their support during the inventory work. We also thank the operators and owners of the studied treatment plants for their collaboration and enthusiasm during the sampling campaign, and colleagues who helped with the field work (Lina Büngener, Haoyu Wei, Rasmus Klapp, Ivan Milovanovic, Sarah Lindfors and Emmanuel Okwori).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2023.118214>.

References

- Barrios-Hernández, M.L., Pronk, M., García, H., Boersma, A., Brdjanovic, D., van Loosdrecht, M.C.M., Hooijmans, C.M., 2020. Removal of bacterial and viral indicator organisms in full-scale aerobic granular sludge and conventional activated sludge systems. *Water Res.* X 6, 100040. <https://doi.org/10.1016/j.wroa.2019.100040>.
- Blum, K.M., Andersson, P.L., Renman, G., Ahrens, L., Gros, M., Wiberg, K., Haglund, P., 2017. Non-target screening and prioritization of potentially persistent, bioaccumulating and toxic domestic wastewater contaminants and their removal in on-site and large-scale sewage treatment plants. *Sci. Total Environ.* 575, 265–275. <https://doi.org/10.1016/j.scitotenv.2016.09.135>.
- Bratby, J., 2016. *Coagulation and Flocculation in Water and Wastewater Treatment*, third ed. IWA Publishing, London.
- Bunce, J.T., Graham, D.W., 2019. A simple approach to predicting the reliability of small waste water treatment plants. *Water (Switzerland)* 11, 2397. <https://doi.org/10.3390/w11112397>.
- Clark, T., Stephenson, T., 1999. Development of a jar testing protocol for chemical phosphorus removal in activated sludge using statistical experimental design. *Water Res.* 33, 1730–1734. [https://doi.org/10.1016/S0043-1354\(98\)00372-8](https://doi.org/10.1016/S0043-1354(98)00372-8).

- Deblonde, T., Cossu-Leguille, C., Hartemann, P., 2011. Emerging pollutants in wastewater: a review of the literature. *Int. J. Hyg Environ. Health* 214, 442–448. <https://doi.org/10.1016/j.ijheh.2011.08.002>.
- Di Marcantonio, C., Chiavola, A., Gioia, V., Leoni, S., Cecchini, G., Frugis, A., Ceci, C., Spizzirri, M., Boni, M.R., 2023. A step forward on site-specific environmental risk assessment and insight into the main influencing factors of CECs removal from wastewater. *J. Environ. Manag.* 325 <https://doi.org/10.1016/j.jenvman.2022.116541>.
- Dinçer, A.R., Kargı, F., 2000. Effects of operating parameters on performances of nitrification and denitrification processes. *Bioprocess Eng.* 23, 75–80. <https://doi.org/10.1007/s004499900126>.
- Doummar, J., Aoun, M., 2018. Assessment of the origin and transport of four selected emerging micropollutants sucralose, Acesulfame-K, gemfibrozil, and iohexol in a karst spring during a multi-event spring response. *J. Contam. Hydrol.* 215, 11–20. <https://doi.org/10.1016/j.jconhyd.2018.06.003>.
- EC (European Commission), 2013. Directive 2013/39/EU Amending the Water Framework Directive and the Environmental Quality Standards Directive.
- Envall, I., Fagerlund, F., Westholm, L.J., Åberg, C., Bring, A., Land, M., Gustafsson, J.P., 2020. What evidence exists related to soil retention of phosphorus from on-site wastewater treatment systems in boreal and temperate climate zones? A systematic map protocol. *Environ. Evid.* 9, 1–11. <https://doi.org/10.1186/s13750-020-00205-9>.
- EPA, 2004. *Local Limits Development Guidance*.
- EPA, 2003. *Wastewater Technology Fact Sheet - Disinfection for Samll Systems*.
- EU, 2006. Directive 2006/7/EC of the European Parliament and of the Council of 15 February 2006 Concerning the Management of Bathing Water Quality and Repealing Directive.
- European Commission, 2022. Proposal for a revised Urban Wastewater Treatment Directive [WWW Document]. https://environment.ec.europa.eu/publications/proposal-revised-urban-wastewater-treatment-directive_en, 4.17.23.
- Eveborn, D., Gustafsson, J.P., Hesterberg, D., Hillier, S., 2009. XANES speciation of P in environmental samples: an assessment of filter media for on-site wastewater treatment. *Environ. Sci. Technol.* 43, 6515–6521. <https://doi.org/10.1021/es901084z>.
- Eveborn, D., Kong, D., Gustafsson, J.P., 2012. Wastewater treatment by soil infiltration: long-term phosphorus removal. *J. Contam. Hydrol.* 140, 24–33. <https://doi.org/10.1016/j.jconhyd.2012.08.003>.
- Falås, P., Wick, A., Castronovo, S., Habermacher, J., Ternes, T.A., Joss, A., 2016. Tracing the limits of organic micropollutant removal in biological wastewater treatment. *Water Res.* 95, 240–249. <https://doi.org/10.1016/j.watres.2016.03.009>.
- Feenstra, T.P., De Bruyn, P.L., 1979. Formation of calcium phosphates in moderately supersaturated solutions. *J. Phys. Chem.* 83, 475–479. <https://doi.org/10.1021/j100467a010>.
- Fernandez-Fontaina, E., Omil, F., Lema, J.M., Carballa, M., 2012. Influence of nitrifying conditions on the biodegradation and sorption of emerging micropollutants. *Water Res.* 46, 5434–5444. <https://doi.org/10.1016/j.watres.2012.07.037>.
- Finnish Ministry of the Environment, 2017. Finnish Environmental Protection Act, Ch. 16 on the management and transport of wastewater in areas outside the sewage network. (In Finnish and Swedish). <https://www.finlex.fi/sv/laki/alkup/2017/20170157>, 1.18.22.
- Gago-Ferrero, P., Gros, M., Ahrens, L., Wiberg, K., 2017. Impact of on-site, small and large scale wastewater treatment facilities on levels and fate of pharmaceuticals, personal care products, artificial sweeteners, pesticides, and perfluoroalkyl substances in recipient waters. *Sci. Total Environ.* 601–602, 1289–1297. <https://doi.org/10.1016/j.scitotenv.2017.05.258>.
- Gao, D.W., Wen, Z.D., 2016. Phthalate esters in the environment: a critical review of their occurrence, biodegradation, and removal during wastewater treatment processes. *Sci. Total Environ.* <https://doi.org/10.1016/j.scitotenv.2015.09.148>.
- Gros, M., Blum, K.M., Jernstedt, H., Renman, G., Rodríguez-Mozaz, S., Haglund, P., Andersson, P.L., Wiberg, K., Ahrens, L., 2017. Screening and prioritization of micropollutants in wastewaters from on-site sewage treatment facilities. *J. Hazard Mater.* 328, 37–45. <https://doi.org/10.1016/j.jhazmat.2016.12.055>.
- Heinonen-Tanski, H., Matikka, V., 2017. Chemical and microbiological quality of effluents from different on-site wastewater treatment systems across Finland and Sweden. *Water* 9, 47. <https://doi.org/10.3390/w9010047>.
- Henze, M., van Loosdrecht, M., Ekama, G., Brdjanovic, D. (Eds.), 2008. *Biological Wastewater Treatment*. IWA Publishing, London.
- Herrmann, I., Nordqvist, K., Hedström, A., Viklander, M., 2014. Effect of temperature on the performance of laboratory-scale phosphorus-removing filter beds in on-site wastewater treatment. *Chemosphere* 117, 360–366. <https://doi.org/10.1016/j.chemosphere.2014.07.069>.
- Herrmann, I., Vidal, B., Hedström, A., 2017. Discharge of indicator bacteria from on-site wastewater treatment systems. *Desalination Water Treat.* 91, 365–373. <https://doi.org/10.5004/dwt.2017.21416>.
- Hübinette, M., 2009. Tillsyn på minireningsverk inklusive mätning av funktion [Inspection of package plants including measurement of functioning]. Report 2009: 07. Västra Götalands county (In Swedish).
- Johannessen, E., Eikun, A.S., Ek, M., Krogstad, T., Junested, C., 2012. Performance of prefabricated package plants for on-site wastewater treatment in the Vansjø- and Hobøl watershed (Morsa), Norway. *VATTEN – J. Water Manag. Res.* 68, 107–114.
- Jucherski, A., Walczowski, A., Bugajski, P., Józwiakowski, K., Rodziejewicz, J., Janczukowicz, W., Wu, S., Kasprzyk, M., Gajewska, M., Mielcarek, A., 2021. Long-term operating conditions for different sorption materials to capture phosphate from domestic wastewater. *Sustain. Mater. Technol.* 31, e00385 <https://doi.org/10.1016/j.susmat.2021.e00385>.

- Kasprzyk-Hordern, B., Dinsdale, R.M., Guwy, A.J., 2009. 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, 363–380. <https://doi.org/10.1016/j.watres.2008.10.047>.
- Kauppinen, A., Martikainen, K., Matikka, V., Veijalainen, A.-M., Pitkänen, T., Heinonen-Tanski, H., Miettinen, I.T., 2014. Sand filters for removal of microbes and nutrients from wastewater during a one-year pilot study in a cold temperate climate. *J. Environ. Manag.* 133, 206–213. <https://doi.org/10.1016/j.jenvman.2013.12.008>.
- Keefer, C.E., 1962. Temperature and efficiency of the activated sludge process. *J. Water Pollut. Control Fed.* 34, 1186–1196.
- Kroiss, H., Rechberger, H., Egle, L., 2011. Phosphorus in water quality and waste management. In: *Integrated Waste Management - Volume II*. IntechOpen. <https://doi.org/10.5772/18482>.
- Larsson, C., Forsberg, B., Engström, T., 2017. Uppföljande Kontroll Av Nya Små Avloppsanläggningar [Follow up Control of New Small Wastewater Facilities. Kungälv municipality (In Swedish)].
- Lehtoranta, S., Laukka, V., Vidal, B., Heiderscheidt, E., Postila, H., Nilivaara, R., Herrmann, I., 2022. Circular economy in wastewater management—the potential of source-separating sanitation in rural and peri-urban areas of northern Finland and Sweden. *Front. Environ. Sci.* 10, 1. <https://doi.org/10.3389/fenvs.2022.804718>.
- Lehtoranta, S., Vilpas, R., Mattila, T., 2014. Comparison of carbon footprints and eutrophication impacts of rural on-site wastewater treatment plants in Finland. *J. Clean. Prod.* 65, 439–446. <https://doi.org/10.1016/j.jclepro.2013.08.024>.
- Leiviskä, T., Risteelä, S., 2022. Analysis of pharmaceuticals, hormones and bacterial communities in a municipal wastewater treatment plant – comparison of parallel full-scale membrane bioreactor and activated sludge systems. *Environ. Pollut.* 292 <https://doi.org/10.1016/j.envpol.2021.118433>.
- Leverett, D., Merrington, G., Crane, M., Ryan, J., Wilson, I., 2021. Environmental quality standards for diclofenac derived under the European Water Framework Directive: 1. Aquatic organisms. *Environ. Sci. Eur.* 33, 1–11. <https://doi.org/10.1186/S12302-021-00574-Z/FIGURES/4>.
- Loos, R., Carvalho, R., António, D.C., Comero, S., Locoro, G., Tavazzi, S., Paracchini, B., Ghiani, M., Lettieri, T., Blaha, L., Jarosova, B., Voorspoels, S., Servaes, K., Haglund, P., Fick, J., Lindberg, R.H., Schwesig, D., Gawlik, B.M., 2013. EU-wide monitoring survey on emerging polar organic contaminants in wastewater treatment plant effluents. *Water Res.* 47, 6475–6487. <https://doi.org/10.1016/j.watres.2013.08.024>.
- Luo, Y., Guo, W., Ngo, H.H., Nghiem, L.D., Hai, F.I., Zhang, J., Liang, S., Wang, X.C., 2014. A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment. *Sci. Total Environ.* <https://doi.org/10.1016/j.scitotenv.2013.12.065>.
- Matamoros, V., Arias, C., Brix, H., Bayona, J.M., 2009. Preliminary screening of small-scale domestic wastewater treatment systems for removal of pharmaceutical and personal care products. *Water Res.* 43, 55–62. <https://doi.org/10.1016/j.watres.2008.10.005>.
- Nedwell, D.B., 1999. Effect of low temperature on microbial growth: lowered affinity for substrates limits growth at low temperature. *FEMS Microbiol. Ecol.* 30, 101–111. <https://doi.org/10.1111/j.1574-6941.1999.tb00639.x>.
- Niku, S., Schroeder, E.D., 1981. Factors affecting effluent variability from activated sludge processes. *J. Water Pollut. Control Fed.* 53, 546–559.
- Olshammar, M., 2021. Datinsamling om teknikuppgifter för små avlopp [Collection of technical data for small sewage plants. In: Report 28. Norrköping (In Swedish)].
- Olshammar, M., Ek, M., Rosenquist, L., Ejhed, H., Sidvall, A., Svanström, S., 2015. Uppdatering av kunskapsläget och statistik för små avloppsanläggningar [Update in the state of knowledge and statistics for small sewage plants. In: Report 166. Norrköping (In Swedish)].
- Renman, A., Hylander, L.D., Renman, G., 2008. Transformation and removal of nitrogen in reactive bed filter materials designed for on-site wastewater treatment. *Ecol. Eng.* 34, 207–214. <https://doi.org/10.1016/j.ecoleng.2008.08.006>.
- Renman, A., Renman, G., 2010. Long-term phosphate removal by the calcium-silicate material Polonite in wastewater filtration systems. *Chemosphere* 79, 659–664. <https://doi.org/10.1016/j.chemosphere.2010.02.035>.
- Rodriguez-Caballero, A., Hallin, S., Pålsson, C., Odlaire, M., Dahlquist, E., 2012. Ammonia oxidizing bacterial community composition and process performance in wastewater treatment plants under low temperature conditions. *Water Sci. Technol.* 65, 197–204. <https://doi.org/10.2166/wst.2012.643>.
- Sharma, B., Ahlert, R.C., 1977. Nitrification and nitrogen removal. *Water Res.* 11, 897–925. [https://doi.org/10.1016/0043-1354\(77\)90078-1](https://doi.org/10.1016/0043-1354(77)90078-1).
- Shaw, K., Dorea, C.C., 2021. Biodegradation mechanisms and functional microbiology in conventional septic tanks: a systematic review and meta-analysis. *Environ. Sci. Water Res. Technol.* 7, 144–155. <https://doi.org/10.1039/d0ew00795a>.
- SwAM, 2016. Havs- Och Vattenmyndighetens Allmänna Råd Om Små Avloppsanordningar För Hushållsspillvatten HVMFS 2016:17 [Swedish Agency for Marine and Water Management's General Advice on Small Sewer Systems for Domestic Wastewater]. Swedish Agency for Marine and Water Management (In Swedish).
- Swedish Meteorological and Hydrological Institute, n.d. SMHI [WWW Document]. URL www.smhi.se.
- Taylor Eighmy, T., Bishop, P.L., 1989. Distribution and role of bacterial nitrifying populations in nitrogen removal in aquatic treatment systems. *Water Res.* 23, 947–955. [https://doi.org/10.1016/0043-1354\(89\)90167-X](https://doi.org/10.1016/0043-1354(89)90167-X).
- Thomasdotter, M., 2008. En Undersökning Av Funktionen Hos Minireningsverk I Marks Kommun [An Evaluation of the Function of Package Plants in the Municipality of Marks]. Göteborg University (In Swedish).
- Vidal, B., Hedström, A., Herrmann, I., 2018. Phosphorus reduction in filters for on-site wastewater treatment. *J. Water Process Eng.* 22, 210–217. <https://doi.org/10.1016/j.jwpe.2018.02.005>.
- Vilpas, R., Kujala-Räty, K., Laaksonen, T., Santala, E., 2005. Haja-asutuksen Ravinnekkuorimituksen Vähentäminen – Ravinnesampo [Decreasing Nutrient Loading in Sparsely Populated Regions]. Vammala, Finland.
- Vilpas, R., Santala, E., 2007. Comparison of the nutrient removal efficiency of onsite wastewater treatments systems: applications of conventional sand filters and sequencing batch reactors (SBR). *Water Sci. Technol.* 55, 109–117. <https://doi.org/10.2166/wst.2007.134>.
- Viraraghavan, T., 1985. Temperature effects on onsite wastewater treatment and disposal systems. *J. Environ. Health* 48, 10–13.
- Vohla, C., Köiv, M., Bavor, H.J., Chazarenc, F., Mander, Ü., 2011. Filter materials for phosphorus removal from wastewater in treatment wetlands—a review. *Ecol. Eng.* 37, 70–89. <https://doi.org/10.1016/j.ecoleng.2009.08.003>.
- Zhou, H., Li, X., Xu, G., Yu, H., 2018. Overview of strategies for enhanced treatment of municipal/domestic wastewater at low temperature. *Sci. Total Environ.* <https://doi.org/10.1016/j.scitotenv.2018.06.100>.