Science of the Total Environment 725 (2020) 138420

Contents lists available at ScienceDirect

Science of the Total Environment

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

Monitoring and modelling of influent patterns, phase distribution and removal of 20 elements in two primary wastewater treatment plants in Norway

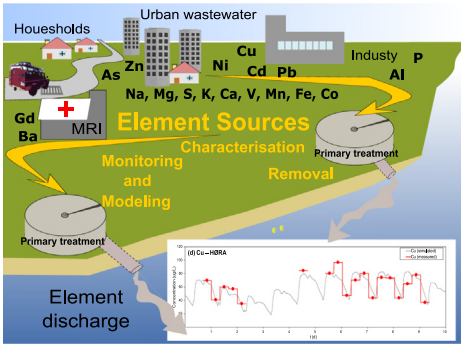
Julia Farkas [a,*,1], Fabio Polesel [b,1,2], Marianne Kjos [c], Patricia Almeida Carvalho [c], Tomasz Ciesielski [d], Xavier Flores-Alsina [e], Steffen Foss Hansen [b], Andy M. Booth [a]

[a] SINTEF Ocean, Brattørkaia 17C, 7010 Trondheim, Norway
[b] DTU Environment, Technical University of Denmark, Bygningstorvet 115, 2800 Kongens Lyngby, Denmark
[c] SINTEF Industry, 7465 Trondheim, Norway
[d] Department of Biology, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway
[e] Process and Systems Engineering Center (PROSYS), Department of Chemical and Biochemical Engineering, Technical University of Denmark, Søltofts Plads, Building 229, 2800 Kongens Lyngby, Denmark

HIGHLIGHTS

- 20 elements were measured in WWTPs with primary sewage treatment.
- Removal efficiencies correlated with element's association with particles
- Diurnal release patterns indicate anthropogenic contribution to element discharge.
- Enrichment in sludge indicates anthropogenic sources for P, Ni, Cu, Zn, As, and Cd.
- Modelling can be used to predict the release patterns of selected elements.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 17 March 2020
Received in revised form 1 April 2020
Accepted 1 April 2020
Available online 6 April 2020

Editor: Paola Verlicchi

Keywords:
Elements
Wastewater treatment
Removal
Characterization

ABSTRACT

Many small- or medium-sized communities in Northern Europe employ only primary wastewater treatment plants (WWTPs) and effluent discharges can be a relevant source of pollution. The current study combines monitoring and modelling approaches to investigate concentrations, influent patterns, size distribution and removal of 20 elements for the two primary WWTPs (Ladehammeren, LARA; Høvringen, HØRA) serving Trondheim, the third largest city in Norway. Element concentrations were determined in raw influent wastewater, effluents and biosolids, and diurnal inflow patterns were assessed. The elemental distribution in particulate, colloidal and dissolved fractions of untreated wastewater was characterized using filtration separation and electron microscopy. An influent generator model and multivariate statistical analyses were used to determine release patterns and to predict the (co-)occurrence of selected elements. Raw influent wastewater concentrations for most elements were similar in the two WWTPs, with only Ca, Mn, Fe, Co and Ba being significantly higher ($p < 0.05$) in HØRA (which receives more household and hospital discharges). Removal efficiencies varied between elements, but

* Corresponding author.
E-mail address: julia.farkas@sintef.no (J. Farkas).
[1] The authors contributed equally to this work
[2] Present address: DHI A/S, Agern Allé 5, 2970 Hørsholm, Denmark.

https://doi.org/10.1016/j.scitotenv.2020.138420

Principal component analysis
Modelling

in most cases reflected their association with particulates. Nanosized particles of several elements were detected, with Cu/Zn being most common. Measured concentrations of most elements followed typical diurnal wastewater discharge patterns and enrichment factors calculated for biosolids confirmed the importance of anthropogenic sources for P, Cu, Zn, Cd, As, Cr, Ni, Pb, V, Co and Fe. Elemental concentrations generally correlated well with total suspended solid (TSS) concentrations at HØRA, while this was less pronounced in LARA (possibly due to higher industrial contributions). In one of its first applications for WWTP influent pattern examination, principal component analysis was found to be instrumental for source identification of target elements, showing significant differences between LARA and HØRA influents. The combined experimental, statistical and modelling approaches used herein allowed for improved understanding of element sources, patterns of discharge and fate in primary WWTPs.

1. Introduction

Municipal wastewater treatment plants (WWTPs) receive highly heterogeneous household and industrial discharges in terms of inorganic element composition. These include potentially toxic elements and metals that, if not adequately removed, can be released to recipient surface waters, sediments and soils, posing a threat to organisms (Deycard et al., 2014; Park et al., 2020). Arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), lead (Pb), nickel (Ni) and zinc (Zn) have been classified as potentially toxic and priority substances in surface water, biosolids and agricultural soils under existing regulations (e.g., Council of the European Union, 1986). Furthermore, the presence of large quantities of phosphorus (P) in wastewater is one the major causes for eutrophication in receiving freshwater and marine environments (de-Bashan and Bashan, 2004). Elements such as sulfur (S), iron (Fe), sodium (Na), calcium (Ca), potassium (K) and magnesium (Mg) have been the object of increasing attention due to their impact on WWTP operating conditions (e.g., pH) and their relevance in the context of resource recovery (Batstone, 2009; Flores-Alsina et al., 2015).

In several small- and medium-sized communities in Northern regions (e.g., Norway, Iceland, Canada) and South-East Europe, WWTPs with only preliminary and primary treatment are in use (Government of Canada, 2017; European Environmental Agency, 2017; Berge and Sæther, 2018). Such limited treatment can result in a reduced removal of conventional pollutants (suspended solids, organics, nutrients) and emerging pollutants compared to WWTPs with biological and advanced treatment steps (Vogelsang et al., 2006; Polesel et al., 2018). Within the past decades, several studies and monitoring programs have investigated the fate of inorganic elements (including metals and toxic elements) in WWTPs, largely focusing on full-scale facilities with secondary and tertiary treatment steps (Buzier et al., 2006; Cantinho et al., 2016; Goldstone et al., 1990a, 1990b, 1990c) (Shafer et al., 1998; Yoshida et al., 2015). These studies revealed (i) the limited contribution of primary treatment to the overall removal of elements; (ii) considerable variability in removal efficiencies for the same element.

The presence of many elements in WWTPs is primarily linked to domestic sewage, commercial and industrial discharges, stormwater runoff and a possible contribution from infiltrating groundwater (Sörme and Lagerkvist, 2002; Ziolko et al., 2011; Drozdova et al., 2019). Atmospheric deposition and stormwater runoff, which contains traffic-related emissions from tire wear, asphalt wear and exhaust particles, contribute to discharges mostly under wet weather conditions (Becouze-Lareure et al., 2016; Sabin et al., 2005; Shafer et al., 1998; Sorme and Lagerkvist, 2002). Chemically-enhanced treatment (e.g., flocculants) may also lead to a considerable additional input to WWTPs (Buzier et al., 2006; Polesel et al., 2018). Recent advances in nanotechnology have led to an increase in the use of metal and metalloid engineered nanomaterials (ENMs) in consumer products such as paints, fabrics and washing machines (Benn and Westerhoff, 2008; Farkas et al., 2011; Kaegi et al., 2010, 2008). Release of ENMs from products can lead to increased metal loads in household discharges during dry weather periods. For example, Ti and Ag ENMs originating from clothing, washing machines, personal care products and food are frequently detected in WWTP influents (Kiser et al., 2009; Li et al., 2013; Polesel et al., 2018; Westerhoff et al., 2011).

Identifying sources of elements and understanding their behavior and fate in WWTP systems is crucial for the development of predictive tools, which can help inform regulatory actions and improve treatment processes and requirements (Cantinho et al., 2016; Snip et al., 2014). Specifically, deterministic modelling and statistical tools to derive pollutant concentration in WWTPs influents have proven useful to optimize the design and control of WWTPs and improve their performance (for review see Martin and Vanrolleghem, 2014). However, these tools have typically focused on conventional pollutants, with the few applications to priority or other trace pollutants (e.g., De Keyser et al., 2010) being hampered by a lack of monitoring data.

In the current study, we combined sampling and characterization in full-scale WWTPs with the use of statistical and predictive modelling tools to study the occurrence and fate of 20 elements in two Norwegian WWTPs employing primary treatment. Analytes included (i) elements listed in priority substance lists, such as those from the Agency for Toxic Substances and Disease Registry (ATSDR) or the EU sewage sludge directive (Al, V, Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Ba, and Pb); (ii) elements regarded as conventional pollutants and/or of relevance for WWTP operation (Na, Mg, K, Ca, P, S and Fe); and (iii) Gd, which has recently emerged as potentially hazardous contaminant in aquatic compartments (Rabiet et al., 2009; Rogowska et al., 2018). A novel methodology was applied to characterize the occurrence of this large and heterogeneous set of elements associated with particulate, colloidal (nanosized) and dissolved fractions in influents, while concentrations in biosolids were used to determine enrichment due to e.g., anthropogenic sources. The combination of detailed particle size fraction characterization and standard chemical analyses was instrumental in the development and application of statistical (principal component analysis) and modelling tools (influent generation). The results improve our understanding of release patterns and anthropogenic input of elements into urban WWTPs and subsequent downstream environments.

2. Materials and methods

2.1. WWTP description

Trondheim is the third largest city in Norway. It is served by two WWTPs, Ladehammeren (LARA) and Høvringen (HØRA), which have a capacity of 120,000 and 170,000 population equivalents (PE), respectively. Both WWTPs receive substantial industrial loading contributions (up to 40% in the case of LARA). The treatment train has previously been described in detail (Polesel et al., 2018). In brief, treatment in LARA and HØRA includes fine screening, sand and fat removal, chemically-aided flocculation-coagulation ($ClFeO_4S$ and polyamine in LARA, polyacrylamide in HØRA) and primary sedimentation. Primary sludge is thickened and pasteurized before anaerobic digestion, followed by

centrifugal dewatering. The effluents of both WWTP are discharged directly into Trondheimsfjord at depths of 40 and 65 m, respectively.

2.2. Sampling

Samples were taken during a 7-day dry weather period in October 2016, in parallel in LARA and HØRA. To determine influx and removal efficiencies of the selected elements 24 h composite samples of raw influent wastewater and treated effluent were collected using refrigerated automatic samplers. These were volume-proportional (VP) composite samples of influent and effluent in LARA ($n = 6$) and effluent only in HØRA ($n = 5$), while daily raw influent wastewater samples in HØRA were composited flow-proportionally from hourly composite samples. Grab samples of biosolids (LARA: $n = 3$; HØRA: $n = 2$) were collected at the end of the sludge treatment line.

To study the diurnal influent patterns of selected elements and conventional pollutant indicators, 8 h composite flow-proportional (FP) samples of raw influent wastewater were taken by compositing of 1 h time proportional samples (Teledyne ISCO®, Lincoln NE, US; 5 min frequency) based on flow data. Samples covered morning (M), evening (E) and night (N) discharges. Sample containers and equipment were subject to a multiple step cleaning procedure, including an acid cleaning (HNO_3) step before each use. The conventional WWTP pollutant endpoints analyzed in the 8 h samples were total chemical oxygen demand (COD_{tot}), soluble COD (COD_{sol}), total nitrogen (N_{tot}), ammonium nitrogen (NH_4-N), total phosphorus (P_{tot}) and TSS. The concentrations of these conventional pollutants have been presented previously (Polesel et al., 2018) and the results are used in this study only for statistical and modelling assessment.

2.3. Sample preparation and fractionation

Influent and effluent 24 h composite samples were concentrated by evaporation and then stored at 4 °C until analysis for total elemental concentrations. Additionally, the target elements were quantified in 8 h FP raw influent wastewater samples and in three different size fractions in the 8 h samples: (i) particulate fraction (>0.7 μm), (ii) colloidal fraction (0.7 μm to 3 kDa), and (iii) dissolved fraction (<3 kDa). To achieve this, samples were sequentially filtered through 2.7 μm and 0.7 μm pore size glass fibre filters (Whatman, USA). The dissolved fraction in the filtrate was subsequently separated from the colloidal fraction using ultrafiltration (3 kDa filter, Amicon®, Merck Millipore, Ireland).

2.4. Elemental analysis

The elements Na, Mg, Al, P, S, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Ba, Pb and Gd were quantified in the composite 8 h raw influent wastewater samples, in each size fraction (particulate, colloidal and dissolved) of the 8 h influent samples, in daily 24 h composite samples (IN and OUT) and biosolids using a inductively coupled plasma triple quadrupole mass spectrometry (ICPQQQ, Agilent 8800; Agilent Technologies, USA) using ^{115}In and ^{89}Y as internal standards (Inorganic Ventures, USA). The sample digestion is described in detail elsewhere (Polesel et al., 2018).

2.5. Sample imaging

To further characterize the elements present in particulate form and to study their association with other particulate matter, samples were imaged using scanning transmission electron microscopy (STEM) coupled with energy-dispersive X-ray spectroscopy (EDS). Samples were applied to copper TEM grids (Agar Scientific, UK) and imaged with a FEI Titan G2 60–300 microscope equipped with a DCOR probe Cs-aberration corrector operating at 300 kV. Elemental analyses were conducted with Energy-dispersive X-ray spectroscopy (EDS) SUPERX detector.

2.6. Data presentation and analyses

The relative removal efficiency (%) of each element was calculated based on the corresponding influent and effluent concentrations from the same sampling period. The relative distribution of each element in the three size fractions (particulate, colloidal, dissolved) was calculated from concentrations determined in the corresponding non-fractionated 8 h samples (assumed as 100%), allowing to verify the closing of mass balances. Enrichment factors (EF) for elements in sludge were calculated relative to their concentration in the upper continental crust (UCC) using Al as reference element (Taylor and McLennan, 1995; Westerhoff et al., 2015).

Data were statistically analyzed using GraphPad Prism 7 (GraphPad Software Inc., USA). To determine differences between groups, the data sets were analyzed for normality (Shapiro-Wilk normality test) and significant differences evaluated using either ANOVA (for multiple comparisons) or *t*-test (for comparison of 2 groups). For statistical analysis and data presentation (Table 1), values below the limit of detection (LOD) were substituted by half the LOD. Groups with 50% or more of the samples featuring values below the LOD were excluded from statistical analyses. To understand the correlation patterns between elements, size fractions and other measured variables, principal component analysis (PCA) was applied using SIMCA® 15 (Sartorius Stedim Data Analytics AB, Umeå, Sweden).

2.7. Diurnal pattern generation

A previously calibrated phenomenological influent generator for both LARA and HØRA was used to derive diurnal profiles of selected elements (Gernaey et al., 2011; Polesel et al., 2018). Diurnal profiles for inorganic elements were obtained by applying a conversion factor to profiles for the conventional pollutant indicators (TSS, COD_{tot}, COD_{sol}, N_{tot}, NH_4-N, P_{tot}), where a significant correlation was observed. Conversion factors indicated the typical content of inorganic elements per unit mass of each conventional pollutant (i.e. mg element per g TSS) and were calculated as the slope of the linear regression. Simulated diurnal profiles were subsequently compared with measured 8 h concentration data.

3. Results and discussion

3.1. Element concentrations in influent, effluent and sludge

Element concentrations determined in the 24 h composite samples of raw influent wastewater (VP for LARA, FP for HØRA), treated effluent (VP for LARA and HØRA) and in grab samples of sludge are presented in Table 1. Concentrations in the raw influent wastewater were highest for Na (117.5 mg L^{-1}, LARA; 119 mg L^{-1}, HØRA) and Ca (45 mg L^{-1}, LARA; 57 mg L^{-1} HØRA), followed by K, S and Mg (15–19 mg L^{-1}; Table 1). Fe, P and Al also reached mg L^{-1} concentration levels in the raw influent wastewater, while other elements occurred in the μg L^{-1} range or below. Influent concentrations of most elements were relatively similar in both WWTP (e.g., Na, Mg, K) or slightly higher in HØRA, compared to LARA (e.g. Al, Cu, Ni, Zn, Gd). However, Ca, Mn, Fe, Co and Ba concentrations were significantly higher ($p < 0.05$) in HØRA compared to LARA. This is potentially related to the slightly higher relative contribution of household discharges (Cu, Fe, Ni, Zn), hospital discharges (Ba, Gd) and groundwater infiltration (estimated to be 37% for HØRA, 35% for LARA; Polesel et al., 2018) (Ca) (Sörme and Lagerkvist, 2002; Bau and Dulski, 1996). Priority elements Al, V, Co, Ni, Cu, Zn, Cd, Ba and Pb were detected in every raw influent wastewater sample, while As and Cr were below the detection limit in 33% and 15% of the samples in

Table 1
Concentrations (μg L^{-1}, if not otherwise stated) of analyzed elements in raw influent wastewater, treated effluent and sludge in LARA and HØRA. The detection frequency (df %) for each element is given. Elemental concentrations in sludge were analyzed in sludge as wet weight (ww). Dry weight (dw) concentrations were recalculated from determined dry weights of the respective samples. Data is presented as mean ± SD.

	IN				OUT				SLUDGE (all in mg kg^{-1})	
	LARA		HØRA		LARA		HØRA		LARA	HØRA
	df %	Mean ± SD (Min–max)	df %	Mean ± SD (Min–max)	df %	Mean ± SD (Min–max)	df %	Mean ± SD (Min–max)	Mean (±SD) ww dw*	Mean (±SD) ww dw*
Na (mg L^{-1})	100	117.5 ± 9.5 (104.2 ± 126.2)	100	119 ± 37.8 (91.4–184.7)	100	510.6 ± 23.9 (477.6–539.1)	100	10.4 ± 20 (95.6–146.8)	1582 ± 43 2534 ± 104	1466 ± 35 2051 ± 162
Mg (mg L^{-1})	100	14.6 ± 0.9 (13.2–15.7)	100	14.8 ± 4.5 (11.5 ± 22.5)	100	669 ± 2.0 (63.6–68.9)	100	12.6 ± 4.5 (10.5 ± 16.7)	328 ± 51 525 ± 82	633 ± 76 882 ± 57
Al	100	1895 ± 590 (1176–2908)	100	2253 ± 574 (1520–2960)	100	223 ± 80.3 (126–332)	100	352.4 ± 36.3 (311–406)	3023 ± 150 4853 ± 444	3385 ± 1 4732 ± 261
P	100	6022 ± 688 (5115–6886)	100	6505 ± 783 (5426–7221)	100	1530 ± 109 (1425–1765)	100	4057 ± 216 (3860–4414)	8403 ± 464 13,457 ± 666	4098 ± 147 5723 ± 108
S (mg L^{-1})	100	16.7 ± 8.7 (14.9–17.4)	100	15.6 ± 2.8 (13.9–20.5)	100	68.2 ± 2.7 (65.8–72.1)	100	13.7 ± 1.35 (12.7–16.1)	5178 ± 530 8322 ± 1162	2383 ± 23 3332 ± 215
K (mg L^{-1})	100	19.2 ± 1.4 (17.5–20.6)	100	19.2 ± 2.1 (17–22)	100	32.2 ± 1.1 (30.4–33.3)	100	17.9 ± 0.7 (17–18.6)	1285 ± 117 2065 ± 274	1607 ± 58 2249 ± 204
Ca (mg L^{-1})	100	45.3 ± 2.3 (41.6–47.4)	100	56.7 ± 2.5* (53.9–60.8)	100	58.9 ± 2.7 (55.5–62.5)	100	52.3 ± 2.6 (48.2–55.5)	5360 ± 201 8584 ± 190	7780 ± 279 10,866 ± 206
V	100	3.63 ± 1.38 (2.13–5.98)	100	3.88 ± 0.96 (2.50–4.94)	100	1.38 ± 0.11 (1.29–1.59)	100	0.48 ± 0.08 (0.38–0.59)	21.1 ± 1.3 34 ± 3.5	15.8 ± 1.2 22 ± 2.9
Cr	83	2.30 ± 1.6 (<LOD–4.88)	100	3.62 ± 1.58 (1.23–4.85)	0	<LOD	0	<LOD	20.3 ± 1 32.5 ± 3.5	21.9 ± 2 30.7 ± 4.2
Mn	100	91.1 ± 9.9 (77.8–107.4)	100	138.5 ± 11.5* (125.7–151.8)	100	135.6 ± 3 (131.1–139.8)	100	93.6 ± 4.9 (88.1–99.4)	79 ± 3 127 ± 11	125 ± 1 175 ± 11
Fe	100	1586 ± 199 (1022–2425)	100	2981 ± 636* (2305–3688)	100	8424 ± 758 (7544–9673)	100	406 ± 33.7 (360–442)	45,903 ± 2283 73,586 ± 5036	7641 ± 245 10,692 ± 929
Co	100	0.45 ± 0.29 (0.16–0.91)	100	0.89 ± 0.29* (0.49–1.12)	100	6.05 ± 0.33 (5.46–6.39)	50	0.15 ± 0.04 (<LOD-0.198)	3.05 ± 0.24 4.9 ± 0.6	2.9 ± 0.13 4.1 ± 0.4
Ni	100	4.84 ± 0.95 (3.51–6.25)	100	5.65 ± 0.72 (4.46–6.18)	100	8.74 ± 0.53 (8.06–9.43)	100	2.71 ± 0.371 (2.28–3.15)	11.1 ± 0.97 17.9 ± 2.3	11.5 ± 0.9 16.1 ± 2.1
Cu	100	80.4 ± 13.0 (66.1–103)	100	94.6 ± 18.6 (73.9–125)	100	33.5 ± 6.91 (31.9–51.6)	100	59.4 ± 21.6 (37.5–92.8)	90 ± 7.3 145 ± 16	105 ± 0.32 147 ± 8
Zn	100	107 ± 15.1 (82.9–123)	100	131.3 ± 26.1 (96.2–163)	100	52.9 ± 10.7 (34.9–66.4)	100	44.2 ± 5.5 (36.2–51.8)	147 ± 11.2 236 ± 27	227 ± 1.8 318 ± 16
As	67	1.60 ± 0.55 (<LOD–2.1)	100	1.69 ± 0.56 (<LOD–2.28)	83	1.55 ± 0.51 (1.51–2.16)	83	1.47 ± 0.45 (<LOD–1.99)	1.86 ± 0.14 3 ± 0.4	1.19 ± 0.01 1.7 ± 0.1
Cd	100	0.12 ± 0.05 (0.08–0.18)	100	0.25 ± 0.13 (0.11–0.40)	50	0.07 ± 0.05 (<LOD–0.17)	67	0.11 ± 0.09 (<LOD–0.27)	0.21 ± 0.03 0.33 ± 0.06	0.33 ± 0.02 0.45
Ba	100	29.7 ± 4.7 (21.9–35.4)	100	46.9 ± 4.8* (40.7–51.9)	100	10.9 ± 1.5 (9.1–12.6)	100	17.9 ± 1.3 (15.4–19.1)	55 ± 4 88 ± 10	82 ± 0.2 115 ± 6
Gd	67	0.26 ± 0.2 (<LOD–0.53)	80	0.39 ± 0.23 (<LOD–0.71)	67	0.17 ± 0.11 (<LOD–0.35)	80	0.36 ± 0.16 (<LOD–0.55)	0.29 ± 001 0.46 ± 0.03	0.42 ± 0.1 0.58 ± 0.11
Pb	100	2.89 ± 0.44 (2.19–3.35)	100	3.44 ± 0.55 (2.23–3.98)	100	1.11 ± 0.20 (0.91–1.32)	100	1.38 ± 0.55 (0.72–2.19)	4.86 ± 0.48 7.8 ± 1.1	8.01 ± 0.06 11.2 ± 0.7

* Indicates significant differences ($p < 0.05$; non-parametric *t*-test, Mann-Whitney) between influent concentrations in LARA and HØRA.

LARA. Similarly, Gd was below the detection limit in both LARA and HØRA in several samples (Table 1, shown as detection frequency, df).

The raw influent wastewater concentrations of the metals Cr, Ni, Cu, Zn, As, Cd and Pb are generally comparable to those in previous studies (Buzier et al., 2006; Cantinho et al., 2016; Carletti et al., 2008; Goldstone et al., 1990a, 1990b, 1990c; Östman et al., 2017). As, Cr and Ni concentrations appeared to be slightly lower relative to studies conducted in Central Europe, while Cu concentrations are comparable or slightly higher (except studies from Goldstone and co-workers) (Buzier et al., 2006; Cantinho et al., 2016; Carletti et al., 2008; Goldstone et al., 1990a, 1990b, 1990c; Östman et al., 2017). Concentrations of Ni, Co, Zn, Cd and Pb are comparable to those recently reported in a study analyzing pollutants in 11 Swedish WWTPs, with determined Cu concentrations being slightly higher in our study (Östman et al., 2017). Similarly, the average concentrations of total P, Al and Fe correspond well with average concentrations in UK wastewater (Gardner et al., 2013).

Gd anomalies in wastewater and receiving water bodies deriving from medical imaging have been previously reported and are gaining increasing attention (Kummerer and Helmers, 2000). In the current study, Gd reached maximum influent concentrations of 0.53 μg L^{-1} in LARA and 0.71 μg L^{-1} in HØRA (Table 1). This is consistent with the main application of Gd being in MRI imaging and reflects the location of the main hospital in Trondheim being in the HØRA catchment area. This is also in good agreement with a previous study analyzing Gd in the raw influent wastewater from the city Münster, Germany (Telgmann et al., 2012). Gd was below the <LOD in raw influent wastewater in three out of 12 samples, with two cases occurring at the weekend (Sun–Mon). A similar trend has been previously reported and was suggested to correspond to reduced MRI imaging activity in weekends (Telgmann et al., 2012).

The effluent concentrations of Cr were <LOD (0.63 μg L^{-1}) in all samples, while As, Co and Gd were <LOD in several effluent samples. Gd was only <LOD in effluent samples corresponding to influent concentrations <LOD. Effluent concentrations in this study are not directly comparable to those reported in most other studies, where element concentrations are reported for WWTPs applying secondary or tertiary treatment. However, it is possible to compare the removal efficiencies of elements to those of other primary treatment steps as described below.

Element concentrations in sewage sludge (ww and calculated dw) are given in Table 1. Despite higher concentrations determined in our study in the raw influent wastewater, Cu concentrations in the sludge

were lower (approximately 147 mg kg^{-1}) compared to those reported in sludge samples from a Swedish WWTP (323 mg kg^{-1}), indicating lower Cu removal in Trondheim (Östman et al., 2017). However, Cu concentrations in our study are comparable to those reported in sewage sludge in one of the most industrialized regions in Poland, which reached from 104 to 194 μg L^{-1} (Tytła, 2019). Ni concentrations were similar to those reported in the Swedish study, with Co, Zn, Cd and Pb being lower in Trondheim (Östman et al., 2017). These elements were also within sludge quality criteria 0-I for use in agriculture in Norway, while concentrations of Cu were close to quality class II (>150 mg kg^{-1}) (Landbruks og matdepartementet, 2003).

3.2. Element removal efficiency

The removal efficiencies of all elements in LARA and HØRA, expressed as a percentage, are presented in Fig. 1. In HØRA (Fig. 1A–B) removal efficiencies were >80% for Al, V, Cr, Fe and Co. However, removal was <50% for P, Cu, S, Mg, Mn, Ni, Gd, Cd and As. In LARA, removal efficiencies >80% were only observed for Al and Cr, being 50–60% for P, V, Cu, Zn, Ba and Pb (Fig. 1B). Interestingly, the concentrations of Na, Mg, S, K, Ca, Mn, Fe, Co and Ni were found to be increased in the effluent in LARA relative to influent concentrations (Fig. 1A). It has previously been shown that the use of the inorganic flocculant $ClFeO_4S$ can result in increased effluent levels of Ti (Polesel et al., 2018). It is therefore likely that other inflow streams, such as flocculant dosing, are the source of increased levels of some elements in effluent, including the priority pollutant elements Ni and Co (Fig. 1A). This is in agreement with Buzier et al. (2006), who quantified Ni and Co at mg L^{-1} levels in commercial dosing chemicals. In contrast, P removal was significantly higher in LARA compared to HØRA due to the use of the $ClFeO_4S$ flocculant. This reflects the application of iron, alum, or lime as the main processes for removing phosphorus through chemical precipitation (de-Bashan and Bashan, 2004). The removal efficiencies of elements determined in this study are comparable to those reported for primary treatment stages in full-scale WWTPs (Buzier et al., 2006; Cantinho et al., 2016; Gardner et al., 2013), with removal efficiencies of most elements being related to the association to the particulate fraction, as described in the next section.

3.3. Element distributions associated with size fractions

The relative distribution of most elements in the three size fractions (particulate, colloidal and dissolved) is presented in Fig. 2A. These were calculated using the concentrations determined in the corresponding non-fractionated 8 h samples, which were assumed to be 100%. This approach was used as the concentrations of the non-fractionated samples and the sum of the fractions were generally in good agreement for most elements, indicating the reliability of the methodology. However, the sum of the fractions was <80% of the total concentrations of Mn and Fe, which are therefore also presented as the relative sum of the fractions in Fig. 2A. The elements Zn and Ba are not presented due to high background levels in fractionated samples, potentially deriving from contamination by the filter material. Since Cd and Gd exhibited not detectable concentrations in one or more of the fractions, a reliable mass balance could not be established.

The removal efficiency of most priority elements can be related to their association with settling particles or aggregates, as most of these elements were strongly associated with the particulate fraction (>0.7 μm) in the raw influent wastewater of both HØRA and LARA (Fig. 2A). In HØRA, removal efficiencies for most elements were strongly correlated with the association of the elements with the particulate size fraction (Fig. 2B; not determined for LARA). Exceptions were Cu and Pb, which were predominantly found in the particulate fraction (>0.7 μm) in the raw influent wastewater but exhibited comparably low removal efficiencies. Furthermore, Ni occurred equally in the particulate (42%) and colloidal phase (42%), with approximately 13% being present in the dissolved phase. This is in agreement with a previous study, reporting a similar size fraction distribution of Ni in raw influent wastewater (Hargreaves et al., 2017). Choubert and co-authors even reported that almost 60% of Ni occurred in the dissolved phase in samples taken in several WWTP in France (Choubert et al., 2011). However, the authors used a different cut-off size (0.45 μm) to distinguish between the particulate and dissolved phase, and did not include a colloidal phase separation, which may explain the differences in findings (Choubert et al., 2011).

Several elements were also detected as micron- or nano-sized particles in raw influent wastewater samples using STEM analyses (Fig. 3). In

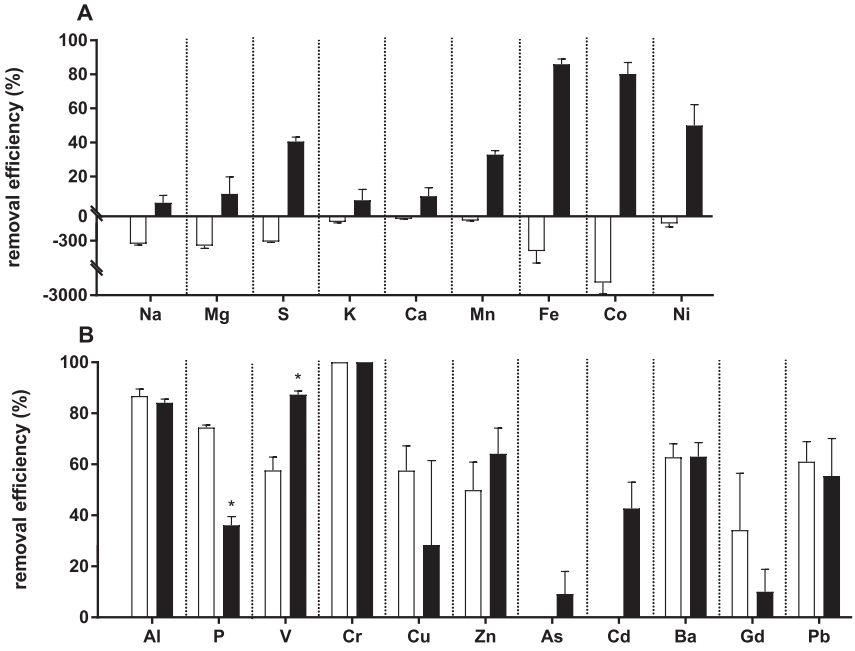


Fig. 1. Relative percentage removal of elements in wastewater streams at LARA (white bars) and HØRA (black bars). Elements with negative removal rates in LARA are shown in A, while elements with positive removal in both LARA and HØRA are presented in B. Data are shown as mean + SE. Asterisks indicate a significant difference ($p < 0.05$; unpaired *t*-test) in concentrations between LARA and HØRA.

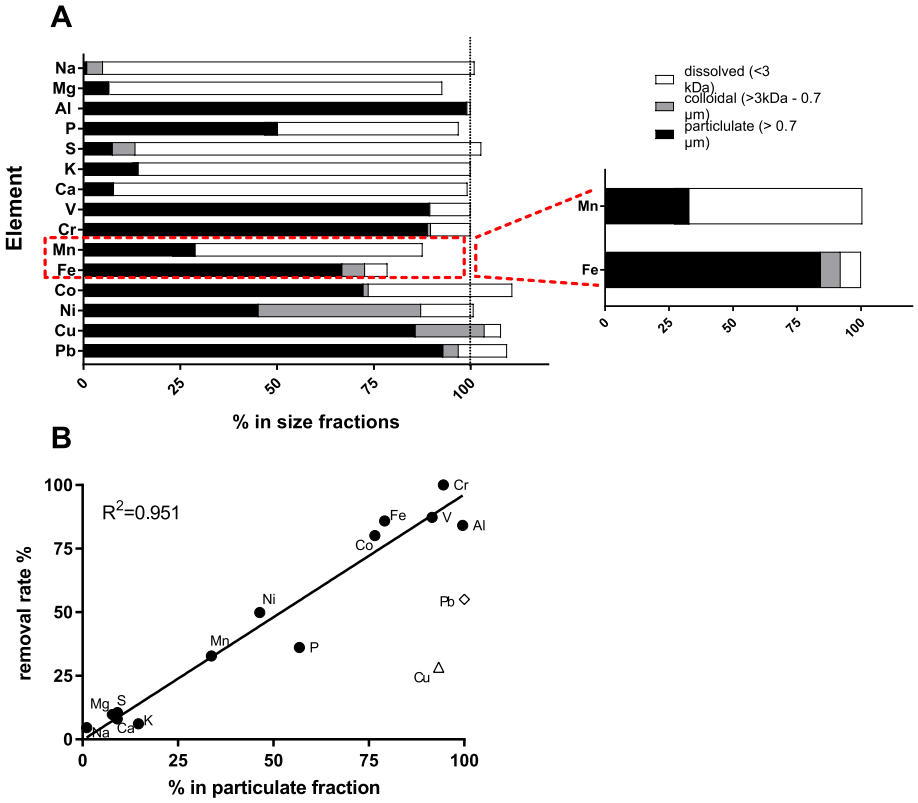


Fig. 2. A) Relative concentrations (% of total) of elements in particulate (black: <0.7 µm), colloidal (grey: >0.7 µm–3 kDa) and dissolved (white: <3 kDa) fractions in raw influent wastewater from LARA and HØRA. Elements with high backgrounds resulting from filter digestion (Zn, Ba) or low concentrations in some fractions (As, Cd and Gd) are not shown. The fraction of the elements Mn and Fe are additionally presented as relative amounts related to the sum of fractions. B) The correlation of relative removal rate (%) and relative amount (%) of the elements in the particulate fraction in HØRA.

particular, Cu and Cu/Zn/S containing particles in the nanometer size range were frequently found (Fig. 3A–C). It is therefore possible that Cu and Cu/Zn/S nanoparticles are small enough to undergo limited removal during the relatively short residence time (contact and settling) in LARA and HØRA (approximately 2 h). Micron- and nanosized particles containing Al, Ba, Fe and/or P were also frequently detected in the present study. Most of these particles were associated with other particles and/or aggregates of organic origin, which is in agreement with the size fractionation data. However, it should also be considered that such associations and agglomerations can be drying artefacts in sample preparation and that particle size and association may be overestimated to some degree. While the size fractionation data indicates that Ca and Mg were predominantly present in the dissolved fraction, Ca- and Mg-containing particles were also frequently detected in the STEM images (data not shown). This is most likely explained by their high concentrations in the raw influent wastewater relative to most other elements.

3.4. Diurnal profiles: experimental results and model simulations

Elemental concentrations measured in the 8 h raw influent wastewater samples from LARA and HØRA are summarized in Fig. 4A–B. A number of elements exhibited a clear diurnal profile in both treatment facilities, with peak concentrations in the morning (M) and/or the evening (E) samples, and minima at night (N). These elements included Al, Cd, Co, Cr, Cu, Fe, Pb, V, which showed more than a 50% reduction in night-time concentrations relative to other day periods in some cases. This type of profile, matching the raw influent wastewater loading pattern, suggests the predominant release of these elements occurs via anthropogenic sources (e.g. households and industrial activities). This hypothesis was subsequently verified by determining the enrichment factors (EF) for each of the elements based on their accumulation in the sewage sludge. Conversely, no distinct pattern was observed for the other elements (Ca, Mg, K, Na, S), with slightly higher concentrations measured during night periods. For these elements, sewer infiltration (e.g., of groundwater, shown to be considerable for Trondheim; Beheshti and Sægrov, 2018) may represent a major contribution in their release to WWTPs. Interestingly, the M, E and N distributions for all elements were similar across both LARA and HØRA, suggesting the main sources of the elements are common in both catchment areas. It should be noted that a significant contribution to element loading from the water supply could be excluded for several elements (Ca, Cu, Mg, Na, Fe, S, V), where concentrations in Trondheim's main water supply source (Jonsvatnet) were typically one order of magnitude lower than in WWTP influents (Seither et al., 2012).

Fig. 4C–D presents a comparison between the measured and the simulated diurnal profiles for two selected elements (Al and Cu) that exhibited significant correlation with raw influent wastewater TSS concentrations. For most elements, significant correlations were shown with conventional pollutant indicators, and TSS concentrations were used as reference indicator for modelling purposes as significant correlation was shown in highest number of cases for LARA and HØRA. Conversion factors (mass element per mass TSS) were shown to be generally consistent for LARA and HØRA influents, indicating the potential for extending this approach for predicting element occurrence to other catchments. Conversely, no or negative correlation with conventional pollutants was observed for Na, Mg, Ca and S in LARA and HØRA, indicating a negligible anthropogenic contribution to WWTP influent loading (in addition to limited affinity with TSS and organics). Overall, the modelling approach used in the study was generally capable of reproducing empirical measurements well, indicating that correlation-based influent generation can be employed as a simple and consistent tool for predicting the total input of inorganic elements

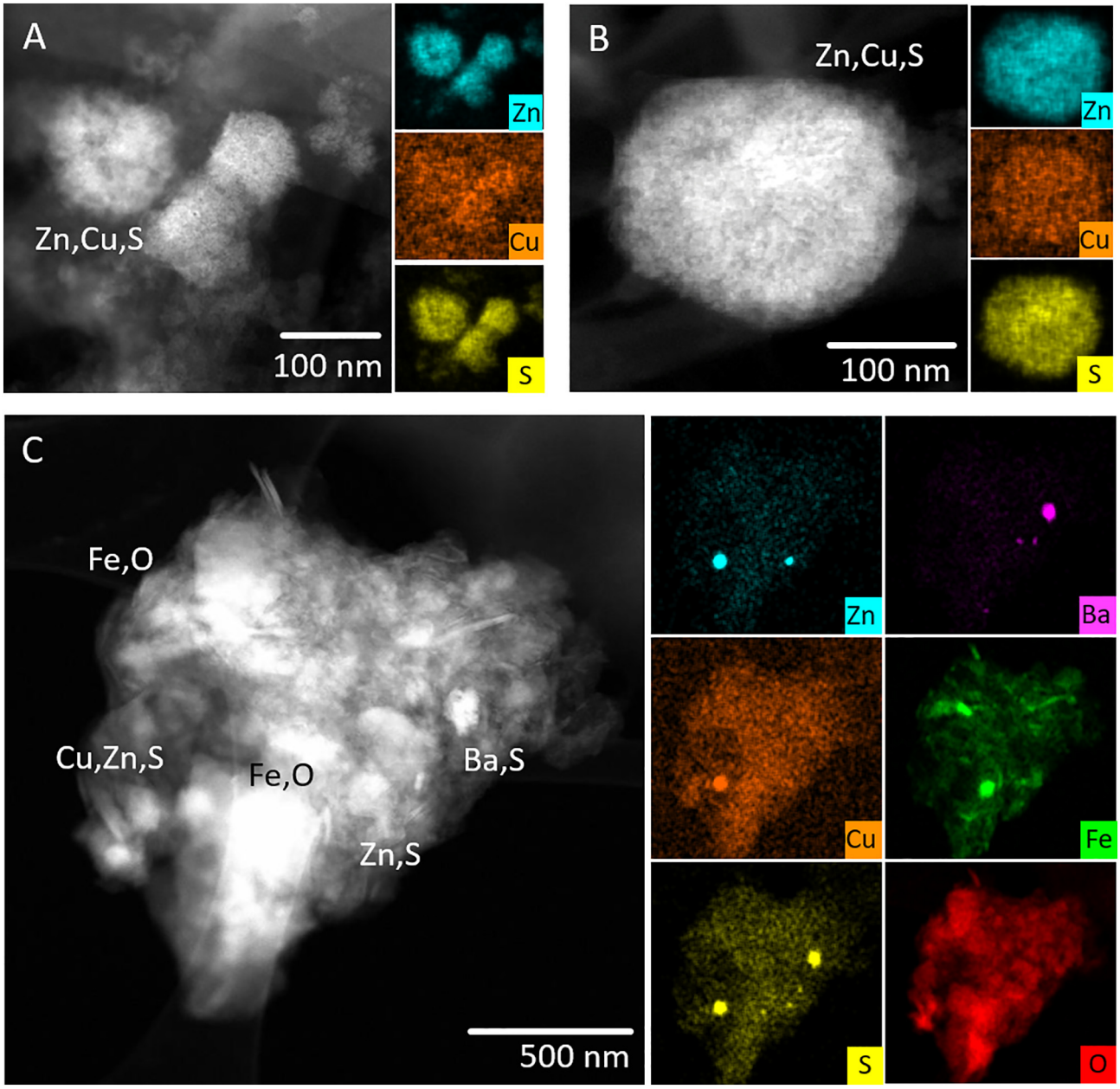


Fig. 3. Elements found as particles in raw influent wastewater. A and B) Cu/Zn/S particles in the nano size range. C) Organic aggregate with associated metal particles (Fe/O, iron oxide; Cu/Zn/S; Ba/S, barium sulphate).

in WWTPs. Modelling results for other elements in both LARA and HØRA, conversion factors from TSS mass and correlation matrices for element and conventional pollutant concentrations can be found in SI.

3.5. Multivariate analyses

The PCA for the diurnal influent patterns of 8 h composite flow-proportional (FP) samples of raw influent wastewater for selected elements, conventional pollutant indicators (TSS, COD, N, P, NH_4-N) and flow resulted in three significant components (PCs) explaining 83.7% of the total variation. Two first PCs accounted for 71.8% of the total variation (56.8 and 15% for PC1 and PC2 respectively). Score values show distinct clusters of samples corresponding to variations in the diurnal pattern in elemental and pollutant composition. This corresponds to high PC1 score values for evening and morning periods, and low values for night-time periods (Fig. 5A). Evening and morning raw influent wastewater samples are characterized by high loading values which correspond to high concentrations of many elements (e.g. Al, V, Cr, Co, Cu, Zn, and Pb; especially those present in the particulate fraction), high levels of particulates, pollution indicators and high influent flow rates (Fig. 5C). In contrast to evening and morning, the elements Na, Mg and S were more closely associated with the night flow. These results are in good agreement with findings from the diurnal patterns established in the current study, indicating anthropogenic sources for elements related to M and E maxima.

In contrast, PC2 distinguishes between the two WWTP (Fig. 5B). HØRA is characterized by higher flow and Ca and Mn concentrations, and to a lesser extent Cd, Fe, Gd and Ba. Notably, Gd and Ba are both used in medical applications and correspond with HØRA being the WWTP receiving hospital effluents. In contrast, LARA is characterized by high COD_{sol} (Fig. 5C), which can be associated with the discharges from food processing industries and a brewery in the catchment that contribute to the influent load (Trondheim Kommune, 2015). While PCA has previously found multiple

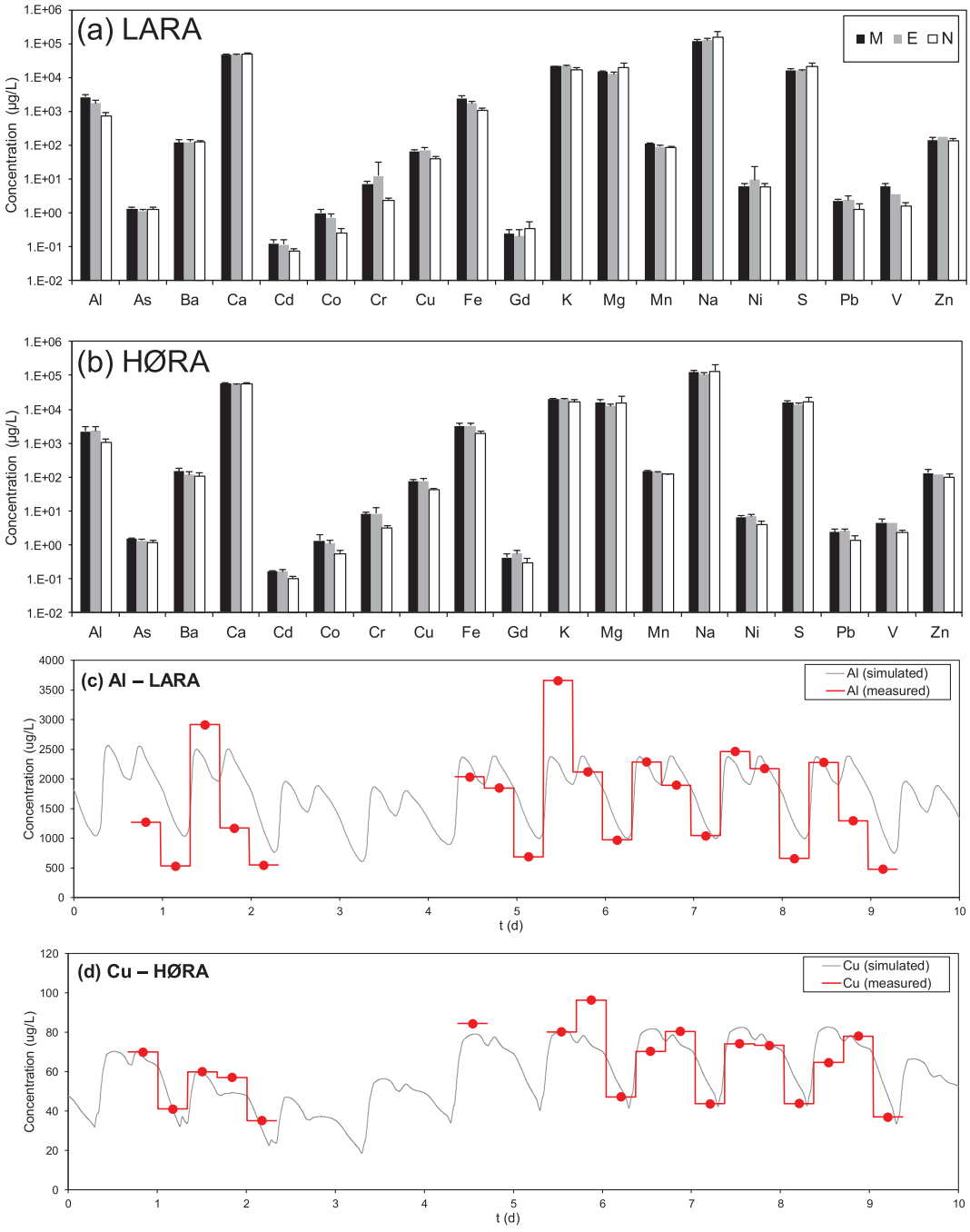


Fig. 4. Overview of diurnal variations in raw influent wastewater concentrations (8 h samples) for all elements in LARA and HØRA, and comparison between measured and simulated profiles for Al in LARA and Cu in HØRA.

applications in understanding sources of heavy metal discharges in the environment (e.g., Singh et al., 2005; Bhardwaj et al., 2017), this study represents one of the first applications for elemental source elucidation in WWTP influents.

3.6. Enrichment factors in biosolids

Enrichment factors (EF) were calculated for each element in the biosolids to determine the potential for them to deriving from anthropogenic sources. Elements with EF values close to 10 or above are presented in Fig. 6 and indicate that anthropogenic sources dominate rather than natural sources (e.g. weathering of earth crust) (Westerhoff et al., 2015). Enrichment was most obvious for P (EF > 100) and was more pronounced in LARA compared to HØRA, which may reflect a more efficient P removal when employing $ClFeO_4S$ during treatment. Cu, Zn and Cd also showed high EFs, with little difference between the two WWTPs. High EFs have previously been reported for sludge from different WWTPs in the USA, especially for P, Cu, Zn, Cd and Pb (Westerhoff et al., 2015). The EF values for Fe are over 10 in LARA and are probably related to the addition of an inorganic flocculant, which will at least partly settle and be enriched in sludge. It should be highlighted that using Al as a reference element is not optimal, as both PCA and diurnal loading dynamics have indicated a likely anthropogenic influence of Al. This can potentially lead to an underestimation of EF values for other elements. In this specific study, however, the application of inorganic flocculant in LARA made it unfeasible to use other elements (e.g., Fe) as a reference for EF calculations.

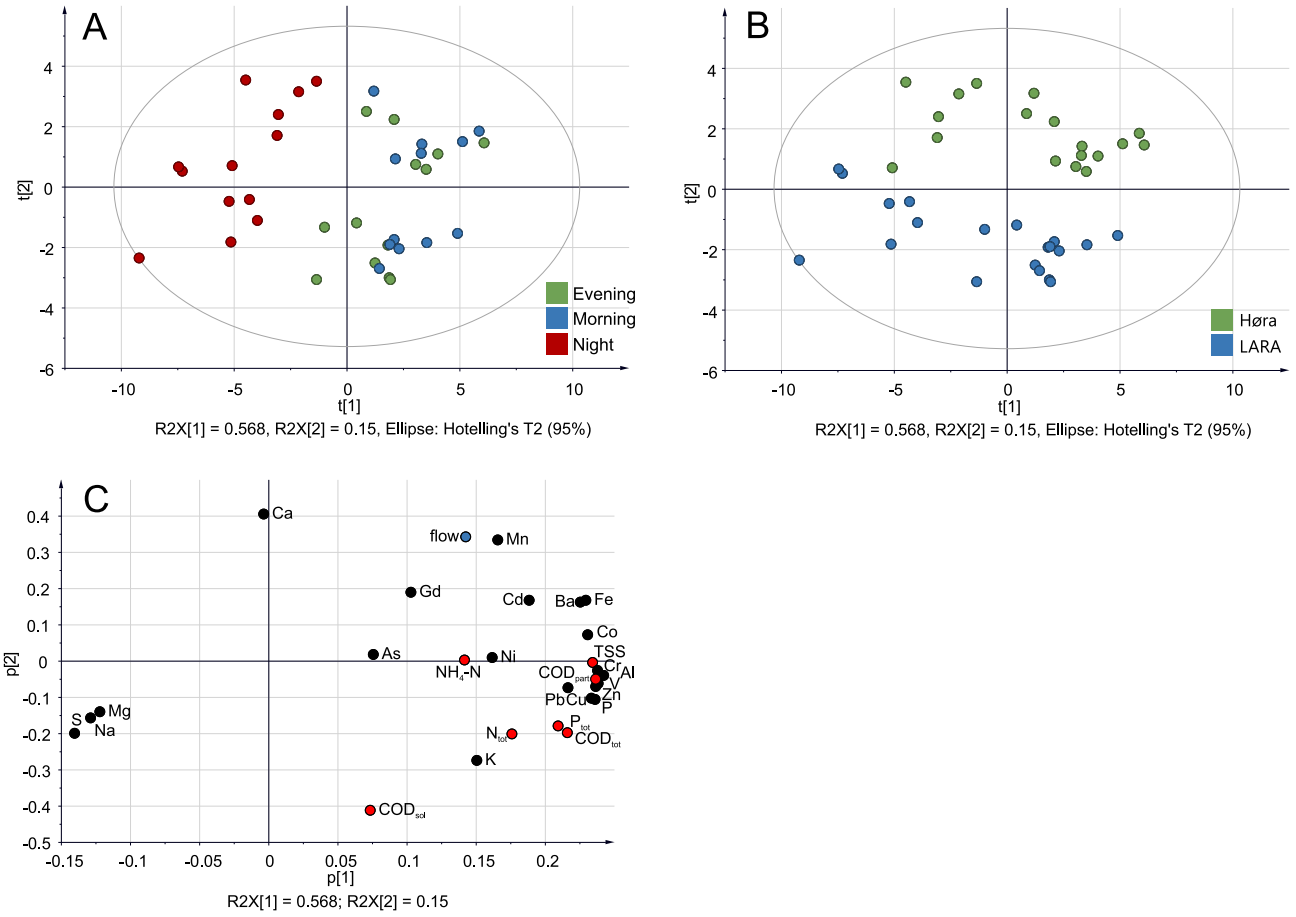


Fig. 5. Principal component analysis (PCA; PC1 vs. PC2) scores (A, B) and loading (C) plots.

4. Conclusions

This study investigated the occurrence and fate of 20 elements in two distinct WWTPs applying primary treatment. A comprehensive approach was adopted, combining full-scale sampling and element characterization (with a unique methodology for a heterogeneous set of elements) with advanced statistical analyses and influent generation modelling. The approach proved particularly suitable for elucidating temporal release patterns, sources of release and anthropogenic influences on element loading in wastewater influents, and it can be applicable to catchments in other geographical regions. It is envisaged that the outcome of this study can facilitate (i) improved design of targeted sampling campaigns and refinements in empirical data collection, (ii) improved prediction of the occurrence and fate dynamics of elements in WWTPs, as well as their impact on treatment processes (e.g., resource recovery), and (iii) improved interpretation for priority substances in view of risk assessment associated with the release into and from WWTPs and, where necessary, identification of possible source minimization strategies.

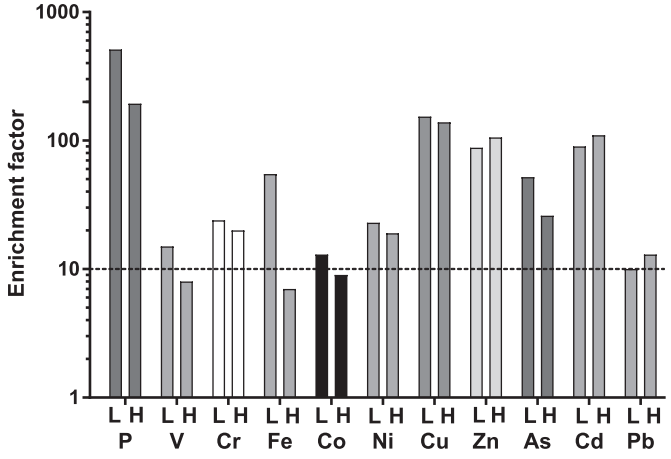


Fig. 6. Enrichment factors (EF) of elements in sludge (recalculated for dw) from LARA (L) and HØRA (H) with values above 10 in at least one of the WWTPs. EFs are calculated relative to Al, and EF values of 10 and above can indicate anthropogenic influence. Elements with EF values below 10 are not shown.

CRediT authorship contribution statement

Julia Farkas: Conceptualization, Formal analysis, Investigation, Writing - original draft, Writing - review & editing, Visualization. **Fabio Polesel:** Conceptualization, Formal analysis, Investigation, Writing - original draft, Writing - review & editing, Visualization, Software. **Marianne Kjos:** Investigation. **Patricia Almeida Carvalho:** Investigation. **Tomasz Ciesielski:** Formal analysis, Visualization. **Xavier Flores-Alsina:** Software, Funding acquisition, Methodology, Recources, Writing - review & editing. **Steffen Foss Hansen:** Conceptualization, Funding acquisition. **Andy M. Booth:** Conceptualization, Funding acquisition, Writing - original draft, Writing - review & editing, Project administration, Supervision.

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.

Acknowledgements

This study was supported by the Norwegian Research Council project NanoWASTE (GA: 238972). The authors are particularly grateful to WWTP operators at LARA and HØRA, especially Grete Klippenvåg Støen. Dr Flores-Alsina gratefully acknowledges the financial support of the Danida Fellowship Centre (DANIDA) project ERASE (18-M09-DTU) and the funds provided by the Danish Council for Independent Research under the project GREENLOGIC (project number: 7017-00175B).

References

de-Bashan, L.E., Bashan, Y., 2004. Recent advances in removing phosphorus from wastewater and its future use as fertilizer (1997–2003). Water Res. 38 (19), 4222–4246.
Batstone, D., 2009. Towards a generalised physicochemical modelling framework. Rev. Environ. Sci. Bio/Technol. 8, 113–114.
Bau, M., Dulski, P., 1996. Anthropogenic origin of positive gadolinium anomalies in river waters. Earth Planet. Sci. Lett. 143 (1–4), 245–255.
Becouze-Lareure, C., Thiebaud, L., Bazin, C., Namour, P., Breil, P., Perrodin, Y., 2016. Dynamics of toxicity within different compartments of a peri-urban river subject to combined sewer overflow discharges. Sci. Total Environ. 539, 503–514.
Beheshti, M., Sægrov, S., 2018. Sustainability assessment in strategic management of wastewater transport system: a case study in Trondheim, Norway. Urban Water J. 15 (1), 1–8.
Benn, T.M., Westerhoff, P., 2008. Nanoparticle silver released into water from commercially available sock fabrics. Environ. Sci. Technol. 42 (11), 4133–4139.
Berge, G.M., Sæther, M.S., 2018. Kommunale avløp 2017, statistics Norway. https://www.ssb.no/natur-og-miljo/artiklerogpublikasjoner/_attachment/370465?_ts=1675fa996e8.
Bhardwaj, R., Gupta, A., Garg, J.K., 2017. Evaluation of heavy metal contamination using environmetrics and indexing approach for River Yamuna, Delhi stretch, India. Water Sci. 31 (1), 52–66.
Buzier, R., Tusseau-Vuillemin, M.H., Meriadec, C.M.D., Rousselot, O., Mouchel, J.M., 2006. Trace metal speciation and fluxes within a major French wastewater treatment plant: impact of the successive treatments stages. Chemosphere 65 (11), 2419–2426.
Cantinho, P., Matos, M., Trancoso, M.A., dos Santos, M.M.C., 2016. Behaviour and fate of metals in urban wastewater treatment plants: a review. Int. J. Environ. Sci. Technol. 13 (1), 359–386.
Carletti, G., Fatone, F., Bolzonella, D., Cecchi, F., 2008. Occurrence and fate of heavy metals in large wastewater treatment plants treating municipal and industrial wastewaters. Water Sci. Technol. 57 (9), 1329–1336.
Choubert, J.-M., Pomiès, M., Martin Ruel, S., Coquery, M., 2011. Influent concentrations and removal performances of metals through municipal wastewater treatment processes. Water Sci. Technol. 63, 1967–1973.
Council of the European Union, 1986. Pollutants in Urban Waste Water and Sewage Sludge. Council Directive of 12 June 1986 on the Protection of the Environment, and in Particular of the Soil, When Sewage Sludge Is Used in Agriculture (86/278/EEC). Council of the European Union.
De Keyser, W., Gevaert, V., Verdonck, F., De Baets, B., Benedetti, L., 2010. An emission time series generator for pollutant release modelling in urban areas. Environ. Model. Softw. 25, 554–561.
Deycard, V.N., Schäfer, J., Blanc, G., Coynel, A., Petit, J.C.J., Lanceleur, L., Dutruch, L., Bossy, C., Ventura, A., 2014. Contributions and potential impacts of seven priority substances (As, Cd, Cu, Cr, Ni, Pb, and Zn) to a major European Estuary (Gironde Estuary, France) from urban wastewater. Mar. Chem. 167, 123–134.
Drozdova, J., Raclavska, H., Raclavsky, K., Skrobankova, H., 2019. Heavy metals in domestic wastewater with respect to urban population in Ostrava, Czech Republic. Water Environ. J. 33, 77–85.
European Environmental Agency, 2017. Urban waste water treatment - indicator assessment. https://www.eea.europa.eu/data-and-maps/indicators/urban-waste-water-treatment/urban-waste-water-treatment-assessment-4.
Farkas, J., Peter, H., Christian, P., Gallego Urrea, J.A., Hassellöv, M., Tuoriniemi, J., Gustafsson, S., Olsson, E., Hylland, K., Thomas, K.V., 2011. Characterization of the effluent from a nanosilver producing washing machine. Environ. Int. 37 (6), 1057–1062.
Flores-Alsina, X., Kazadi Mbamba, C., Solon, K., Vrecko, D., Tait, S., Batstone, D.J., Jeppsson, U., Gernaey, K.V., 2015. A plant-wide aqueous phase chemistry module describing pH variations and ion speciation/pairing in wastewater treatment process models. Water Res. 85, 255–265.
Gardner, M., Jones, V., Comber, S., Scrimshaw, M.D., Coello-Garcia, T., Cartmell, E., Lester, J., Ellor, B., 2013. Performance of UK wastewater treatment works with respect to trace contaminants. Sci. Total Environ. 456–457, 359–369.
Gernaey, K.V., Flores-Alsina, X., Rosen, C., Benedetti, L., Jeppsson, U., 2011. Dynamic influent pollutant disturbance scenario generation using a phenomenological modelling approach. Environ. Model. Softw. 26 (11), 1255–1267.
Goldstone, M.E., Atkinson, C., Kirk, P.W.W., Lester, J.N., 1990a. The behavior of heavy-metals during waste-water treatment. 3. Mercury and arsenic. Sci. Total Environ. 95, 271–294.
Goldstone, M.E., Kirk, P.W.W., Lester, J.N., 1990b. The behavior of heavy-metals during waste-water treatment. 1. Cadmium, chromium and copper. Sci. Total Environ. 95, 233–252.
Goldstone, M.E., Kirk, P.W.W., Lester, J.N., 1990c. The behavior of heavy-metals during waste-water treatment. 2. Lead, nickel and zinc. Sci. Total Environ. 95, 253–270.
Government of Canada, 2017. Municipal wastewater treatment. https://www.canada.ca/en/environment-climate-change/services/environmental-indicators/municipal-wastewater-treatment.html.
Hargreaves, A.J., Vale, P., Whelan, J., Constantino, C., Dotro, G., Campo, P., Cartmell, E., 2017. Distribution of trace metals (Cu, Pb, Ni, Zn) between particulate, colloidal and truly dissolved fractions in wastewater treatment. Chemosphere 175, 239–246.
Kaegi, R., Ulrich, A., Sinnet, B., Vonbank, R., Wichser, A., Zuleeg, S., Simmler, H., Brunner, S., Vonmont, H., Burkhardt, M., Boller, M., 2008. Synthetic TiO2 nanoparticle emission from exterior facades into the aquatic environment. Environ. Pollut. 156 (2), 233–239.
Kaegi, R., Sinnet, B., Zuleeg, S., Hagendorfer, H., Mueller, E., Vonbank, R., Boller, M., Burkhardt, M., 2010. Release of silver nanoparticles from outdoor facades. Environ. Pollut. 158 (9), 2900–2905.
Kiser, M.A., Westerhoff, P., Benn, T., Wang, Y., Perez-Rivera, J., Hristovski, K., 2009. Titanium nanomaterial removal and release from wastewater treatment plants. Environ. Sci. Technol. 43 (17), 6757–6763.
Kummerer, K., Helmers, E., 2000. Hospital effluents as a source of gadolinium in the aquatic environment. Environ. Sci. Technol. 34 (4), 573–577.
Landbruks- og matdepartementet, Klima- og miljødepartementet, Helse- og omsorgsdepartementet, 2003. Forskrift om organisk gjødsel. https://lovdata.no/dokument/SF/forskrift/2003-07-04-951#KAPITTEL_5 (In Norwegian).
Li, L., Hartmann, G., Döblinger, M., Schuster, M., 2013. Quantification of nanoscale silver particles removal and release from municipal wastewater treatment plants in Germany. Environ. Sci. Technol. 47 (13), 7317–7323.
Martin, C., Vanrolleghem, P.A., 2014. Analysing, completing, and generating influent data for WWTP modelling: a critical review. Environ. Modell. Softw. 60, 188–201. https://doi.org/10.1016/j.envsoft.2014.05.008.
Östman, M., Lindberg, R.H., Fick, J., Bjorn, E., Tysklind, M., 2017. Screening of biocides, metals and antibiotics in Swedish sewage sludge and wastewater. Water Res. 115, 318–328.
Park, S., Choi, M., Jang, D., Joe, D., Park, K., 2020. Distribution and sources of dissolved and particulate heavy metals (Mn, Co, Ni, Cu, Zn, Cd, Pb) in Masan Bay, Korea. Ocean Sci. J. https://doi.org/10.1007/s12601-020-0001-2.
Polesel, F., Farkas, J., Kjos, M., Carvalho, P.A., Flores-Alsina, X., Gernaey, K.V., Hansen, S.F., Plosz, B.G., Booth, A.M., 2018. Occurrence, characterisation and fate of (nano)particulate Ti and Ag in two Norwegian wastewater treatment plants. Water Res. 141, 19–31.
Rabiet, M., Brissaud, F., Seidel, J.L., Pistre, S., Elbaz-Poulichet, F., 2009. Positive gadolinium anomalies in wastewater treatment plant effluents and aquatic environment in the Herault watershed (South France). Chemosphere 75 (8), 1057–1064.
Rogowska, J., Olkowska, E., Ratajczyk, W., Wolska, L., 2018. Gadolinium as a new emerging contaminant of aquatic environments. Environ. Toxicol. Chem. 37 (6), 1523–1534.
Sabin, L.D., Lim, J.H., Stolzenbach, K.D., Schiff, K.C., 2005. Contribution of trace metals from atmospheric deposition to stormwater runoff in a small impervious urban catchment. Water Res. 39 (16), 3929–3937.
Seither, A., Eide, P.E., Berg, T., Frengstad, B., 2012. The Inorganic Drinking Water Quality of some Groundwaterworks and Regulated Wells in Norway. Geological Survey of Norway (report 2012.073).
Shafer, M.M., Overdier, J.T., Armstong, D.E., 1998. Removal, partitioning, and fate of silver and other metals in wastewater treatment plants and effluent-receiving streams. Environ. Toxicol. Chem. 17 (4), 630–641.
Singh, K.P., Malik, A., Sinha, S., Singh, V.K., Murthy, R.C., 2005. Estimation of source of heavy metal contamination in sediments of Gomti river (India) using principal component analysis. Water Air Soil Pollut. 166, 321–341.
Snip, L.J.P., Flores-Alsina, X., Plosz, B.G., Jeppsson, U., Gernaey, K.V., 2014. Modelling the occurrence, transport and fate of pharmaceuticals in wastewater systems. Environ. Model. Softw. 62, 112–127.
Sorme, L., Lagerkvist, R., 2002. Sources of heavy metals in urban wastewater in Stockholm. Sci. Total Environ. 298 (1–3), 131–145.
Sörme, L., Lagerkvist, R., 2002. Sources of heavy metals in urban wastewater in Stockholm. Sci. Total Environ. 298 (1), 131–145.
Taylor, S.R., McLennan, S.M., 1995. The geochemical evolution of the continental crust. Rev. Geophys. 33 (2), 241–265.
Telgmann, L., Wehe, C.A., Birka, M., Künnemeyer, J., Nowak, S., Sperling, M., Karst, U., 2012. Speciation and isotope dilution analysis of gadolinium-based contrast agents in wastewater. Environ. Sci. Technol. 46 (21), 11929–11936.
Trondheim Kommune, 2015. Årsrapport 2011–2014 Kildekontroll av miljøgifter i Ladehammeren rensedisrtikt (In Norwegian).
Tytła, M., 2019. Assessment of heavy metal pollution and potential ecological risk in sewage sludge from municipal wastewater treatment plant located in the most industrialized region in Poland—case study. Int. J. Environ. Res. Public Health 16, 2430.
Vogelsang, C., Grung, M., Jantsch, T.G., Tollefsen, K.E., Liltved, H., 2006. Occurrence and removal of selected organic micropollutants at mechanical, chemical and advanced wastewater treatment plants in Norway. Water Res. 40 (19), 3559–3570.
Westerhoff, P., Song, G., Hristovski, K., Kiser, M.A., 2011. Occurrence and removal of titanium at full scale wastewater treatment plants: implications for TiO2 nanomaterials. J. Environ. Monit. 13 (5), 1195–1203.
Westerhoff, P., Lee, S., Yang, Y., Gordon, G.W., Hristovski, K., Halden, R.U., Herckes, P., 2015. Characterization, recovery opportunities, and valuation of metals in municipal sludges from U.S. wastewater treatment plants nationwide. Environ. Sci. Technol. 49 (16), 9479–9488.
Yoshida, H., Christensen, T.H., Guildal, T., Scheutz, C., 2015. A comprehensive substance flow analysis of a municipal wastewater and sludge treatment plant. Chemosphere 138, 874–882.
Ziolko, D., Martin, O.V., Scrimshaw, M.D., Lester, J.N., 2011. An evaluation of metal removal during wastewater treatment: the potential to achieve more stringent final effluent standards. Crit. Rev. Environ. Sci. Technol. 41, 733–769.