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## 2 **Opportunities for rotating belt filters in novel wastewater treatment**

#### 3 plant configurations

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#### 12 Abstract

Novel wastewater treatment plants (WWTPs), which are based on a partial nitritation-13 14 anammox (PN-anammox) process, enable higher chemical oxygen demand (COD) recovery to produce biogas and lower treatment costs. In this study, rotating belt filters 15 (RBFs) were examined in different configurations to identify the opportunities for RBFs 16 to be included in novel WWTP configurations. RBFs enable recovery of 22-37% of the 17 influent COD and removal of 34-56% of hydrophobic organic micropollutants (OMPs). 18 However, the effluent was not suitable for treatment in a PN-anammox process due to 19 20 its high COD. Chemically enhanced settling (CES) enabled these limitations to be 21 overcome and caused an increase in OMP removal to 73-94%. However, a dose of 300 22 mg/L of ferric chloride was required to produce a suitable effluent for a PN-anammox reactor. The combination of RBF and CES not only derived effluents suitable for 23 treatment in PN-anammox units but also decreased the alkalinity consumption and the 24 required chemical dose 3-fold to achieve comparable COD recovery and OMP removal. 25 26 The methane yield of the combined sludges that were produced (184 L(N) CH<sub>4</sub>/kg COD<sub>influent</sub>) was 75% higher than that obtained in conventional wastewater treatment 27 28 (105 L(N) CH<sub>4</sub>/kg COD<sub>influent</sub>), and the electricity requirements decreased from 0.54 to 29 0.41 kWh/m<sup>3</sup> of treated wastewater. The energetic calculations showed that a WWTP 30 incorporating this combined treatment could attain energy autarky with 29% lower operational costs than that of conventional treatment (0.022 vs  $0.031 \text{ } \text{€/m}^3$ ) as long as a 31 32 minimum alkalinity-to-ammonium ratio of 1-1.25 g IC to g NH4<sup>+</sup>-N was ensured in the effluent of the combined treatment. 33

- 34 Keywords: cellulosic sludge, chemically enhanced settling, energy self-sufficiency,
- 35 organic matter recovery, organic micropollutants.

#### Introduction 36

The discovery of the autotrophic nitrogen removal process (anammox process)<sup>1</sup>, which 37 38 does not require an organic carbon source for denitrification, introduces new possibilities for conceiving more energetically efficient wastewater treatment plants 39 (WWTPs).<sup>2</sup> Although anammox-based processes are already applied at full scale to treat 40 the supernatants of anaerobic sludge mesophilic digesters, the implementation of this 41 technology in the mainstream of WWTPs is currently under investigation since they 42 operate at considerably lower temperatures (10-20°C).<sup>3</sup> 43 In the first stage of these novel WWTPs, pre-concentration technologies can recover 44 most of the chemical oxygen demand (COD) from sewage to produce energy (methane), 45 which enables WWTPs to gain energy self-sufficiency or even become energy-46 producing facilities.<sup>4</sup> Some studies have suggested that the energy contained in 47 wastewater is nearly 5-fold the electrical energy that is used to drive conventional 48 wastewater treatment.<sup>5</sup> Therefore, as long as 20% of the total energy in domestic 49 50 wastewater can be completely converted to electrical energy, WWTPs may be energetically self-sufficient. 51 The energy contained in wastewater can be recovered either directly in an anaerobic 52 reactor in moderate climates<sup>6</sup> or indirectly in an organic matter pre-concentration step in 53 the form of sludge, which is subsequently treated by anaerobic digestion. Several pre-54 concentration alternatives exist: physical (e.g., sieving),<sup>7</sup> chemical (e.g., precipitation),<sup>8</sup> 55 biological (e.g., A-stage)<sup>9</sup> or combinations of these alternatives. 56 Fine mesh rotating belt filters (RBFs) offer a very low footprint solution for recovering 57

58	COD. They have been successfully applied as a replacement for conventional primary
59	treatment (CPT) in traditional WWTPs <sup>10,11</sup> and achieve total suspended solids (TSS)
60	removals that are similar to those reported for CPT (~50%). <sup>12,13</sup> A maximum of 50%
61	dry matter content of RBF sludge (also known as cellulosic sludge) can be achieved
62	with a very high percentage of cellulose (maximum of 79% of TSS), <sup>7</sup> which facilitates
63	its use as a soil conditioner in agriculture, fuel in a biomass-based power plant, and feed
64	stock in the fermentation industry for the production of biofuels <sup>6</sup> or chemicals, such as
65	volatile fatty acids (VFA). <sup>14</sup> However, the most straightforward method for onsite
66	valorisation is energy recovery by mesophilic AD. <sup>15</sup>
67	RBFs for municipal wastewater are commonly employed with a mesh size of 350
68	$\mu$ m. <sup>7,16</sup> Their effluents show important COD content, which is derived not only from
69	soluble COD that remain unaffected in the RBFs but also from particulate COD that
70	corresponds to small TSS that pass the RBFs. Therefore, the implementation of
71	additional steps prior to autotrophic nitrogen removal treatment is required. <sup>17</sup>
72	Chemically enhanced primary treatment (CEPT) or chemically enhanced settling (CES)
73	is an alternative that overcomes the limitations of RBF. In CEPT processes, using some
74	chemical additives (coagulants and/or flocculants), TSS and COD removal can
75	eliminate a maximum of 90% and 70%, respectively, of TSS and COD, and an increase
76	in the sedimentation rate decreases the size of the settling tank. <sup>18</sup> Chemical coagulation
77	has been shown to completely eliminate viruses from wastewater. <sup>19</sup> However, these
78	processes require a considerable amount of chemicals and generate large volumes of
79	sludge, with subsequent excessive costs of reagents and sludge disposal. <sup>20</sup>

Therefore, the combination of RBF and CEPT may have a synergistic effect that 80 overcomes the limitations of separately applying both technologies. The first effect is 81 82 the incapacity of RBF to achieve high COD removal efficiencies and generate suitable effluents for autotrophic nitrogen removal process, and the second effect is the large 83 chemical doses that are required in CES processes to achieve this goal.<sup>21</sup> 84 The objective of this study is to assess the potential of RBFs to recover COD in novel 85 WWTP configurations via its combination with a CES. The system was technically, 86 energetically and economically evaluated. Considering the different characteristics of 87 88 RBF sludge that influence the interaction with OMPs, an additional goal is to assess their fate in RBFs and compare it with the removal in conventional and chemically 89 enhanced primary treatment. 90

91 Materials and methods

#### 92 **RBF systems**

The technical performance of two RBF systems with a mesh size of 350 µm and located 93 in the Blaricum WWTP (1,600 m<sup>3</sup>/h) and Aarle-Rixtel WWTP (2,600 m<sup>3</sup>/h) in The 94 Netherlands was evaluated. Influent, effluent and sludge samples were collected and 95 stored at 4 °C in aluminium bottles prior to analysis. Wastewater samples were 96 characterised in terms of total solids (TS, g TS/kg) and volatile solids (VS, g VS/kg), 97 total suspended solids (TSS, g TSS/kg) and volatile suspended solids (VSS, g VSS/kg), 98 total chemical oxygen demand (CODtot, g O2/L) and soluble chemical oxygen demand 99 (COD<sub>sol</sub>, g O<sub>2</sub>/L), total Kjeldahl (TKN, g N-TKN/L), total ammonium nitrogen (g N-100 TAN/L) and OMPs concentrations. Dewatered RBF sludge samples were collected in 101

102	both WWTPs, whereas WWTP raw RBF sludge (the sludge generated in the RBF
103	without the dewatering process) was sampled in the Aarle-Rixtel WWTP. The sludge
104	samples were characterised in terms of TS, VS, CODtot, TKN and OMPs concentrations.
105	Eighteen commonly employed OMPs with different physico-chemical properties were
106	considered in this study: three musk fragrances, galaxolide (HHCB), tonalide (AHTN)
107	and celestolide (ADBI); three anti-inflammatories, ibuprofen (IBP), naproxen (NPX)
108	and diclofenac (DCF); four anti-biotics, sulfamethoxazole (SMX), trimethoprim (TMP),
109	erythromycin (ERY) and roxithromycin (ROX); four neurodrugs, fluoxetine (FLX),
110	carbamazepine (CBZ), diazepam (DZP) and citalopram (CTL); one endocrine
111	disrupting compound, triclosan (TCS); and three hormones, estrone (E1), $17\beta$ -estradiol
112	(E2) and 17α-ethinylestradiol (EE2).
113	CES tests
114	Chemically enhanced settling (CES) assays were carried out with both RBF influent and
115	RBF effluent of the Aarle-Rixtel WWTP in a Jar-Test device with vessels that contain 1
116	L of liquid volume following the protocol described by Carballa et al. <sup>22</sup> , but without pH
117	correction. The influence of the dose of ferric chloride (0-300 mg/L) on the removal of
118	TSS, COD and OMPs was analysed at 25°C. The test included an initial 3 min period of
119	rapid stirring (150 rpm) after the addition of the coagulant, followed by 5 min of slow
120	mixing (50 rpm) for emulsion breaking and floc formation and a 1 hour period without
121	mixing for floc separation, after which 500 mL of supernatant were collected for the
122	characterisation.

## 123 Biomethane potential tests

124 The biomethane potential (BMP) of the RBF sludge and of the sludges generated after RBF (settling without chemicals and chemically enhanced settling using 100 mg/L of 125 ferric chloride) was carried out following a protocol that was described elsewhere.<sup>23</sup> The 126 inoculum was flocculant biomass (11.8 g VS/L) from the sludge anaerobic digester of a 127 128 WWTP. 129 The assays were conducted in 500 mL bottles (375 mL of working volume) by triplicate and were carried out with an inoculum-to-substrate ratio (ISR) in terms of VS of 2. 130 Methane production of the blank (inoculum without substrate) was also determined by 131 132 triplicate. The reactors were filled with macro- and micro-nutrient solutions, and the pH was adjusted to 7.2-7.5 with NaOH or HCl when necessary. After flushing the head 133 space with nitrogen, the bottles were incubated at 37°C. Accumulated methane 134 135 production was monitored over time to determine the COD fraction that was converted into methane. The assays continued until the methane production during three 136 consecutive days was less than 1% of the total production.<sup>23</sup> Methane production by 137 each sludge was calculated as the difference between the average production in the 138 bottles with substrate and the average production in the blank. BMP was calculated as 139 140 the experimental ultimate methane production, which was expressed in L(N)/kg VS fed, where N denotes the normal conditions (1 atm, 0°C). Anaerobic biodegradability was 141 expressed as the percentage of the initial COD of the substrate converted to methane. 142 At the end of the test, bottles were opened and the pH and VFAs concentrations were 143 measured to confirm that acidification did not occur. 144

#### 145 Solid-water distribution coefficient (K<sub>d</sub>) tests

146	A common approach to determining the fraction of OMPs sorbed onto sludge is the use			
147	of the solid-water distribution coefficient (K <sub>d</sub> , L/kg). A spike of the 18 selected			
148	compounds was performed on raw RBF sludge at different concentrations in the three			
149	tests. Sodium azide (10 mg/L) was added to avoid biological activity. After 12 hours of			
150	mixing at room temperature to achieve equilibrium conditions, the samples were			
151	centrifuged and liquid and solid phases were separately analysed, as explained in the			
152	section analytical methods of this document.			
153	Analytical methods			
154	COD, pH, PA, TA, TSS, VSS, TS, VS, N-TKN and N-TAN were determined according			
155	to standard methods. <sup>24</sup> Total inorganic carbon (IC) concentrations were measured with a			
156	Shimadzu analyser (TOC-5000). In BMP tests, biogas production was measured by a			
157	pressure transducer (Centrepoint Electronics) and its composition was determined by			
158	gas chromatography (HP 5890 Series II). VFAs were measured by gas chromatography			
159	with flame ionisation detection (FIC, HP 5890A).			
160	To determine the OMP concentrations in the wastewater samples, the latter were			
161	centrifuged, pre-filtered (AP4004705, Millipore) and filtered by 0.45 mm			
162	(HAWP04700, Millipore) before performing solid phase extraction (SPE) with 200 mg			
163	OASIS HLB cartridges (Waters, Milford, MA, USA), as described by Fernandez-			
164	Fontaina et al. <sup>25</sup> The quantification of musk fragrances (HHCB, AHTN, ADBI), anti-			
165	inflammatories (IBP, NPX, DCF) and endocrine disrupting compound TCS was			
166	accomplished using a gas chromatograph (Varian CP-3900) coupled with an ion trap			
167	spectrometer (Varian CG-2100). Antibiotics (ERY, ROX, SMX, TMP), neurodrugs			

(FLX, CBZ, DZP, CTL) and hormones (E1, E2, EE2) were quantified using an Agilent 168 G1312A liquid chromatograph with a binary pump and automatic injector HTC-PAI 169 170 (CTC Analytics) connected to a mass spectrometer API 4000 triple quadrupole (Applied Biosystems).<sup>26</sup> 171 172 For influents and effluents, the sample volume that was analysed was 1 L, and the final 173 volume of the extract was 3 mL, which generated an enrichment factor (concentration in the extract compared with the source) of 333 L<sub>supernatant</sub>/L<sub>extract</sub>. For the liquid phase of 174 RBF sludge, the analysed volume was 100 mL and the final volume of extract was 3 175 176 mL, which yielded an enrichment factor of 33 L<sub>supernatant</sub>/L<sub>extract</sub>. The limits of quantification for each case are shown in Table S1 in Supporting Information. 177 The frozen solid phases of the influent and effluent of RBF and raw and dewatered SS 178 179 were lyophilised to perform ultrasonic solvent extraction following a procedure based on the procedure described by Alvarino et al.<sup>27</sup> Three sequential extractions with 180 methanol and two sequential extractions with acetone were performed on the freeze-181 182 dried samples (0.5-1 g). In each extraction, samples were sonicated for 15 min and centrifuged at 1500 rpm for 5 min. The resulting supernatants were combined and 183 184 filtered through glass wool. The resulting volume was evaporated to 1 mL (TurboVap LV, Biotage) flowing nitrogen (200 kPa, 30 °C) and resuspended in 100 mL of Milli-Q 185 water prior to SPE. SPE and OMPs quantification were performed as previously 186 described for liquid samples. The enrichment factor was 166 gsludge/Lextract. 187 **Results and discussion** 188

189 Technical performance of RBF systems

190	The physico-chemical characterisation of the influent and effluent of the two RBF
191	sampled systems are shown in Table 1. Both influents showed similar average values of
192	TSS (320 and 275 mg/L), VSS (300 and 255 mg/L), VS (600 and 570 mg/L), COD <sub>tot</sub>
193	(680 and 600 mg O <sub>2</sub> /L), COD <sub>sol</sub> (230 and 260 mg O <sub>2</sub> /L) and TKN (87 and 75 mg TKN
194	N/L), which is consistent with previously reported values for the Blaricum WWTP $^7$ and
195	for other urban WWTPs in The Netherlands. <sup>28</sup> Conversely, the TS concentration in the
196	influent of the Blaricum WWTP (770 mg/L) was considerably lower than that measured
197	in the influent of the Aarle-Rixtel WWTP (1,260 mg/L), which indicates a lower salts
198	dissolved concentration.
199	The removal efficiency of TSS (~50%), VSS (~50%), $COD_{sol}$ (~0%) and TKN (~10%)
200	were similar in both RBF systems. Regarding $COD_{tot}$ , a higher removal efficiency
201	(37%) was determined in the Blaricum WWTP than in the Aarle-Rixtel WWTP (22%),
202	which is explained by its higher $COD_{sol}$ -to- $COD_{tot}$ ratio. The removal efficiencies
203	determined in this study are similar to those reported by other authors regardless of the
204	TKN, <sup>7,17</sup> for which the removal efficiencies achieved in both scenarios were
205	approximately 10%—a value that is slightly higher than the that reported elsewhere
206	(~1%). <sup>7</sup>
207	The physico-chemical properties of dewatered RBF sludge are shown in Table 2. TS
208	(21.5-27.5%), VS (20.0-25.8%), COD <sub>tot</sub> (273-356 g O <sub>2</sub> /kg) and TKN/VS ratio (12-16
209	mg N/g VS) are in accordance with the values reported for RBF sludge from the
210	Blaricum WWTP <sup>15</sup> . VS represents approximately 95% of the TS of the sludge, and the
211	COD <sub>tot</sub> -to-VS ratio was approximately 1.3, which is in accordance with the mean values

212	obtained by Paulsrud et al. <sup>29</sup> for 19 Norwegian WWTPs that apply RBF technology.
213	Ghasimi et al. <sup>15</sup> reported higher COD <sub>tot</sub> /VS ratios (1.6-1.8), which may be indicative of
214	a lower cellulose concentration.
215	Raw RBF sludge of the Aarle-Rixtel WWTP was ten times less concentrated than the
216	dewatered RBF sludge, which shows similar physico-chemical characteristics (data not
217	shown), as expected.

218 Technical performance of CES

The results of applying a CES to the influent of RBFs are shown in Figure 1. Ferric 219 220 chloride was selected as a coagulant rather than aluminium salts, considering its lower price <sup>18</sup> and lower required doses. <sup>22</sup> Effluent quality that is comparable to that obtained 221 222 in RBFs was obtained after conventional settling (CODtot removal and TSS removal of 223 36-38% and 47-49%, respectively), without removal of COD<sub>sol</sub>. By the addition of 100 mg/L of ferric chloride, the removal of COD<sub>tot</sub> and TSS increased to 66 and 86%, 224 respectively. This decrease was primarily attributed to a large removal of suspended 225 226 matter, considering that the maximum COD<sub>sol</sub> removal achieved did not exceed 25%. Heterotrophic denitrifiers may increase nitrogen removal efficiency in anammox 227 reactors with low influent COD/N ratios;<sup>30</sup> however, system failure was reported with 228 COD/N ratios higher than 2 due to the growth of fast-growing heterotrophic denitrifiers, 229 which compete with the slow-growing anammox bacteria for nitrite.<sup>31</sup> To fulfil this 230 condition, the required ferric chloride dose should be increased to 300 mg/L, which is 231 similar to that reported by Carballa et al.<sup>22</sup> 232

233 Technical performance of combined RBF and CES technologies

RBFs are suitable to partial removal of COD<sub>part</sub> from wastewater but they are unable to
remove COD<sub>sol</sub>. CES enables complete removal of COD<sub>part</sub> and partial removal of
COD<sub>sol</sub> with a requirement of high chemical doses. Thus, the combination of RBF and
CES was analysed (Figure 2) to attempt to overcome the limitation of both technologies

and compare with the CES system (Figure 1).

239 The combination of RBF followed by settling without chemicals addition produced a

removal of almost 100% of the COD<sub>part</sub> of wastewater, whereas 50% of removal was

241 achieved when the test was carried out for the RBF influent (Figure 1), which may

242 indicate that the presence of cellulose in wastewater limits the settlement of other

suspended solids due to its tendency to float. COD<sub>sol</sub> remained unaffected. When 100

244 mg/L of ferric chloride were added, COD<sub>part</sub> was totally removed, which generates an

effluent with approximately 10 mg TSS/L, and more than 50% of COD<sub>sol</sub> was removed

246 (Figure 2), which accounts for COD<sub>tot</sub> removal of 84%. To obtain comparable results

without an RBF system, a considerably higher dose of FeCl<sub>3</sub> (approximately 300 mg/L)

was needed (Figure 1).

Note that a side effect exists of the addition of the coagulant on the alkalinity, with a

consumption of 22 mg IC for each 100 mg of FeCl<sub>3</sub>.<sup>32</sup> Taking into account that the

251 further PN-anammox step requires a minimum alkalinity-to-ammonium ratio of 1-1.25

252 g IC per g NH<sub>4</sub><sup>+</sup>-N,<sup>33</sup> the need for an external bicarbonate addition to avoid acidification

in the PN-anammox unit, which considers an ammonium concentration of  $67 \text{ mg NH}_4^+$ -

254 N/L (Table 1), would be much higher if the RBF system is not present.

#### **Biomethane potential of the sludges**

Figure 3 shows the results of the BMP tests of RBF sludge, which comprise the sludges 256 generated after RBFs (settling without chemicals and settling with the addition of 100 257 258 mg/L of ferric chloride). Three of the sludges showed similar methane production rates and needed 26 days to complete the test, following the criteria proposed by Holliger et 259 al. <sup>23</sup> The neutral pH values (7.3-7.7) and the absence of VFA (<2.5 ppm acetic acid) at 260 261 the end of the tests (data not shown) indicated that the performance of the tests was adequate and acidification did not occur. 262 Dewatered RBF sludge showed the highest BMP value (386 L(N) CH<sub>4</sub>/kg VS), which is 263

264 consistent with other results in the literature.<sup>29</sup> However, the anaerobic biodegradability 265 (80%) was higher than that the obtained by Ghasimi et al. <sup>15</sup> (64%), which is explained 266 by its lower  $COD_{tot}/VS$  ratio.

267 The sludges generated during conventional settling and CES after RBF showed similar

268 BMP values (327 L(N) CH4/kg VS and 310 L(N) CH4/kg VS, which corresponds to

269 60% of anaerobic biodegradability and 58% of anaerobic biodegradability,

270 respectively), but values lower than those for dewatered RBF sludge. These values are

271 comparable to those reported for conventional primary sludge (259-325 L (N) CH<sub>4</sub>/kg

272 VS).<sup>29</sup> Thus, the installation of RBFs seems to not affect the BMP of sludge obtained by

conventional settling, which may be explained by the low proportion of cellulose

recovered in the latter system.<sup>7</sup> The similar results obtained for both sludges do not

agree with those obtained by Kooijman et al.,<sup>34</sup> who reported an important increase in

the BMP of the sludge generated during CES compared with conventional settling due

277 to the higher readily degradable biomass removed by flocculation. However, Romero-

278	Güiza et al. <sup>35</sup> reported that Fe <sup>3+</sup> reduction can limit the conversion of organics to
279	methane as $Fe^{3+}$ reduction is more thermodynamically favourable than methanogenesis.
280	The data from the BMP test and the results of the CES tests enabled calculation of the
281	methane production of each sludge in relation to the COD recovery in each treatment
282	step. RBF enables a very limited energy recovery (62 L(N) CH4/kg COD <sub>influent</sub> ), which
283	is explained by lower $\text{COD}_{tot}$ recovery compared with that of other tested technologies
284	(Figure 2).
285	The system RBF followed by settlement without chemicals enabled an increase in the
286	energy recovery to 85 L(N) CH <sub>4</sub> /kg COD <sub>influent</sub> ). However, the highest energy recovery
287	was achieved with the combination RBF and CES with 100 mg/L of ferric chloride,
288	which boosted it to 122 L(N) CH <sub>4</sub> /kg COD <sub>influent</sub> due to the higher COD capture
289	determined with the addition of chemicals (Figure 1).
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289 290 291 292 293	determined with the addition of chemicals (Figure 1). <b>Fate of organic micropollutants in the RBF and CES systems</b> The concentrations of the 18 selected OMPs in the influent of both RBF systems are reported in Table S2. The highest concentrations of OMPs in both WWTPs were observed for the anti-inflammatories IBP and NPX (3.47-4.89 μg/L), although DCF was
289 290 291 292 293 294	determined with the addition of chemicals (Figure 1). <b>Fate of organic micropollutants in the RBF and CES systems</b> The concentrations of the 18 selected OMPs in the influent of both RBF systems are reported in Table S2. The highest concentrations of OMPs in both WWTPs were observed for the anti-inflammatories IBP and NPX (3.47-4.89 μg/L), although DCF was not detected in either of the two WWTPs. These concentrations are consistent with other
<ol> <li>289</li> <li>290</li> <li>291</li> <li>292</li> <li>293</li> <li>294</li> <li>295</li> </ol>	determined with the addition of chemicals (Figure 1). <b>Fate of organic micropollutants in the RBF and CES systems</b> The concentrations of the 18 selected OMPs in the influent of both RBF systems are reported in Table S2. The highest concentrations of OMPs in both WWTPs were observed for the anti-inflammatories IBP and NPX (3.47-4.89 μg/L), although DCF was not detected in either of the two WWTPs. These concentrations are consistent with other concentrations in the literature. <sup>13,36,37</sup> Musk fragrances (HHCB, AHTN and ADBI) and
<ol> <li>289</li> <li>290</li> <li>291</li> <li>292</li> <li>293</li> <li>294</li> <li>295</li> <li>296</li> </ol>	determined with the addition of chemicals (Figure 1). <b>Fate of organic micropollutants in the RBF and CES systems</b> The concentrations of the 18 selected OMPs in the influent of both RBF systems are reported in Table S2. The highest concentrations of OMPs in both WWTPs were observed for the anti-inflammatories IBP and NPX (3.47-4.89 μg/L), although DCF was not detected in either of the two WWTPs. These concentrations are consistent with other concentrations in the literature. <sup>13,36,37</sup> Musk fragrances (HHCB, AHTN and ADBI) and the endocrine-disrupting compound TCS were detected in the range 0.95-2.16 μg/L, in
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289 290 291 292 293 294 295 296 297 298	determined with the addition of chemicals (Figure 1). <b>Fate of organic micropollutants in the RBF and CES systems</b> The concentrations of the 18 selected OMPs in the influent of both RBF systems are reported in Table S2. The highest concentrations of OMPs in both WWTPs were observed for the anti-inflammatories IBP and NPX (3.47-4.89 µg/L), although DCF was not detected in either of the two WWTPs. These concentrations are consistent with other concentrations in the literature. <sup>13,36,37</sup> Musk fragrances (HHCB, AHTN and ADBI) and the endocrine-disrupting compound TCS were detected in the range 0.95-2.16 µg/L, in accordance with other authors. <sup>36,38,39</sup> The four antibiotics (SMX, TMP, ERY, and ROX) and the four neurodrugs (FLX, CBZ, DZP and CTL) were detected in the influent of

300	WWTP. In general, the measured concentrations (LOD-235 ng/L) were in the lower
301	range of the literature. <sup>36,37,39,40</sup> This fact can be explained by the fact that the
302	Netherlands has the lowest human antibiotic consumption rate of Europe. <sup>41</sup> The
303	concentrations of hormones ranged between LOQ and 57 ng/L, which is the same range
304	of those reported in the literature). <sup>39</sup>
305	The OMP removal efficiencies achieved in the different evaluated systems (RBF, CPT,
306	and RBF+CES using 100 mg/L of ferric chloride and CES using 300 mg/L of ferric
307	chloride) are shown in Figure 4. Only fragrances and TCS were removed in a significant
308	percentage in the evaluated scenarios. In the RBFs of the Blaricum WWTP (WWTP 1),
309	the removal efficiencies of HHCB, AHTN and ADBI were 39%, 46% and 34%,
310	respectively, and 45% for TCS. In the RBFs of the Aarle-Rixtel WWTP, the determined
311	removal efficiencies of HHCB, AHTN and ADBI were slightly higher (53%, 54% and
312	44%, respectively). TCS was not detected in this WWTP. In conventional settling of the
313	Aarle-Rixtel WWTP (WWTP 2), the elimination of HHCB, AHTN and ADBI were
314	54%, 56% and 45%, respectively, compared with those obtained during CPT of hospital
315	wastewater. <sup>42</sup> Thus, no difference was found between RBF and CPT. These results can
316	be explained by the similar removal efficiencies of TSS that are achieved in both
317	systems and the similar affinity of these OMPs to the solid phase of conventional
318	primary and RBF sludges, which is confirmed by the results of the solid-liquid
319	distribution coefficients (Figure S1) and the comparable concentration of these OMPs in
320	both sludges (in Supporting Information S3, a detailed discussion is included). In the
321	Aarle-Rixtel WWTP (WWTP 2), the combination of RBF and CES enabled

322	improvement in the removal efficiencies of HHCB and AHTN to 96% and
323	improvement in the removal efficiency of ADBI to 75%, which is similar to those
324	achieved in the CES. For the remaining OMPs, the elimination of different RBF
325	systems ranged from -8 to 14% (Figure 3). The increase in concentration of some OMPs
326	was likely attributed to the analytical deviation caused by the distinctive characteristics
327	of the wastewaters. These results show agreement with the literature. <sup>13,42,43</sup>
328	It seems to be a consensus in the literature that most OMPs are poorly removed during
329	coagulation-flocculation processes, however, some exceptions such as musks, a few
330	pharmaceuticals (e.g. DCF) and nonylphenol were found. <sup>39</sup> Moreover, some authors
331	reported that the different composition of wastewater can play a major role on OMPs
332	removal during CES. For example, high fat content in wastewater was reported to
333	improve the removal of hydrophobic compounds. <sup>42</sup> Dissolved humic acid could also
334	enhance the elimination of some pharmaceutical compounds, such as DCF or IBP. <sup>44</sup> On
335	the contrary, the presence of $\text{COD}_{sol}$ , especially low-molecular-weight fractions, can
336	possibly inhibit the OMPs removal due to the preferential removal of $\text{COD}_{\text{sol}}$ through
337	coagulation. Negatively charged COD <sub>sol</sub> could react with positively charged coagulants,
338	leading to a less amount of coagulant available for elimination of OMPs. <sup>45</sup>
339	Although the system RBF+CES of the proposed alternative did not enable extra
340	removal of hydrophilic compounds compared with conventional settling, it enabled an
341	increase in the removal efficiency of hydrophobic compounds, which achieved results
342	that were as good as those obtained in the CES with higher doses of ferric chloride.
343	Energetic and economic assessment of the proposed WWTP configuration

344	Among the evaluated alternatives, the combination of RBF and CES drove the
345	maximum energy production with the lowest ferric chloride and alkalinity consumption
346	and to the highest OMPs removal. Figure 5 shows a comparison of this alternative with
347	the conventional WWTP scheme in terms of energy production/consumption.
348	In conventional WWTPs, aeration usually consumes the largest energy fraction
349	(maximum of 70%), <sup>4,46</sup> which causes a total energy consumption of wastewater in the
350	range 0.6-1.2 kWh/kg CODtot, with an average value of 0.9 kWh/kg CODtot.47
351	Therefore, the treatment of 0.54 kWh/m <sup>3</sup> wastewater is needed, considering 0.6 kg
352	$COD_{tot}/m^3$ for the Aarle-Rixtel WWTP (Table 1), in accordance with other reported
353	values in the literature. <sup>48</sup> The potential energy in typical domestic wastewater has been
354	estimated as 4.5 kWh/kg COD <sub>tot</sub> in the influent $^{5,49}$ , and the COD <sub>tot</sub> transformed into
355	methane usually does not exceed 30%, <sup>50,51</sup> of which a maximum of 35% is converted to
356	electricity via its combustion. <sup>47</sup> As approximately 0.47 kWh per kg of $COD_{influent}$ are
357	commonly recovered, a maximum of 50% of self-produced energy can be achieved
358	(Figure 5a). Considering an electricity cost of 0.12 €/kWh, <sup>52</sup> a treatment cost of 0.031
359	$\epsilon/m^3$ for treated wastewater is estimated.
360	In the proposed scenario (Figure 5b), the electricity demand of the PN-anammox unit
361	was considered to be 60% of the nitrification-denitrification electricity demand,
362	according to Schaubroeck et al., <sup>51</sup> which yielded 0.23 kWh/m <sup>3</sup> of treated wastewater.
363	The maximum electricity consumption of RBF was reported to be approximately 0.04
364	kWh/m <sup>3</sup> , <sup>54</sup> and the costs due to pumping and sludge treatment were assumed to be $0.17$
365	kWh/m <sup>3</sup> , which is equivalent to those in conventional configurations <sup>47</sup> . Thus, the

366	electricity demand was 0.44 kWh/m <sup>3</sup> , which is 19% lower than that in the conventional
367	configuration (0.54 kWh/m <sup>3</sup> ). The combination of RBF followed by CEPT enabled 84%
368	recovery of the COD <sub>tot</sub> of the influent as sludge (Figure 2). During AD, 52% of the
369	influent COD was transformed into methane and yielded electricity generation of 0.82
370	kWh per kg of $COD_{influent}$ (0.49 kWh/m <sup>3</sup> of wastewater treated), which almost doubles
371	the electricity generation in the conventional WWTP. This fact and the lower electrical
372	requirements of this configuration enable an electrical self-production of 100-110%.
373	To calculate the operational costs associated with chemical addition, the FeCl <sub>3</sub> price
374	was estimated at approximately 220 $\epsilon$ /ton <sup>18</sup> and the sodium bicarbonate price was
375	estimated at approximately 200 €/ton (1,400 €/ton IC). Two different scenarios were
376	evaluated: in the first scenario, the alkalinity of wastewater was assumed to be
377	sufficiently high bear the consumption produced in CES; thus, the influent to the PN-
378	anammox would have an appropriate alkalinity-to-ammonium ratio; in the second
379	scenario, an external alkalinity addition as sodium bicarbonate was considered to
380	compensate its consumption in the CES stage. The first scenario yielded a treatment
381	cost that was 29% lower than the treatment cost in the conventional WWTP ( $0.022$
382	versus 0.031 $\text{€/m^3}$ ). In the second scenario, the calculated treatment costs increased to
383	0.053 €/m <sup>3</sup> ; thus, it was distinctly uncompetitive compared with the conventional
384	WWTP.

385 Conclusion

386 The implementation of rotating belt filters before chemically enhanced settling was

387 proven effective for generating suitable effluents for a nutrient removal stage,

maximising the energy recovery via the anaerobic digestion of the generated sludges,

attaining 110% of the electrical autarky and achieving OMP removal that is as

390 substantial as that obtained in chemically enhanced settling, and decreasing the required

dose3-fold. However, a minimum alkalinity level in wastewaters was shown mandatory

to consider this WWTP configuration as more economically favourable with respect to

393 the conventional WWTP configuration.

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#### 483 Table legends

- 484 **Table 1.** Physicochemical characteristics of influent and effluent in rotating belt sieves
- 485 from Blaricum and Aarle-Rixtel wastewater treatment plants (n=3).
- 486 **Table 2.** Physicochemical characteristics of dewatered sieved sludge generated in
- 487 rotating belt sieves of Blaricum and Aarle-Rixtel wastewater treatment plants (n=3).
- 488 **Table 3.** Calculated electricity and chemical operational costs of the proposed WWTP
- 489 configuration.

490 Figures captions

491 Fig. 1. Influence of ferric chloride dose on the removal of soluble (□) and particulate
492 (□) COD and TSS (●) on raw wastewater of Aarle-Rixtel WWTP. 0 mg FeCl<sub>3</sub>/L refers
493 to conventional settling without chemicals.

- 494 Fig. 2. Influence of ferric chloride dose on the removal of soluble (□) and particulate
  495 (□) COD and TSS (●) on the RBF effluent of wastewater of Aarle-Rixtel WWTP. 0 mg
  496 FeCl<sub>3</sub>/L refers to conventional settling without chemicals.
- 497 **Fig. 3.** Average BMP results of dewatered sieved sludge (■), conventional settling
- 498 sludge after RBF ( $\blacktriangle$ ) and chemically enhanced settling sludge after RBF ( $\bigcirc$ ).
- **Fig. 4.** Removal efficiencies of the organic micropollutants in RBF system in WWTP 1 ( $\blacksquare$ ), in RBF in WWTP 2 ( $\blacksquare$ ), in CPT in WWTP 2 ( $\blacksquare$ ), RBF + CES in WWTP 2 ( $\blacksquare$ ), and CES in WWTP 2 ( $\square$ ).
- Fig. 5. COD balance (in relation to 1 kg in the influent) and energy flows for the
  traditional WWTP configuration (adapted from Wan et al. <sup>50</sup>) and the proposed WWTP
  configuration. \* Represents the energy required for sludge management and pumping,
  = ► Refers to energy inputs for the wastewater treatment.

#### 506 **Table 1**

	Blaricum WWTP		WWTP Aarle-Rixtel	
	Influent	Effluent	Influent	Effluent
TS (mg/L)	$770 \pm 30$	$610 \pm 20$	$1260 \pm 0$	$1140 \pm 10$
VS (mg/L)	$600\pm20$	$440 \pm 20$	$570\pm5$	$450 \pm 20$
TSS (mg/L)	$320\pm10$	$160 \pm 0$	$275\pm5$	$145 \pm 10$
VSS (mg/L)	$300\pm20$	$150\pm0$	$255\pm10$	$130 \pm 5$
COD <sub>tot</sub> (mg O <sub>2</sub> /L)	$680\pm10$	$440 \pm 10$	$600 \pm 30$	$470 \pm 10$
COD <sub>sol</sub> (mg O <sub>2</sub> /L)	$230\pm10$	$220 \pm 10$	$260\pm20$	$240\pm10$
TKN-N (mg/L)	$87 \pm 2$	$75 \pm 1$	$75 \pm 4$	$69 \pm 6$
TAN-N (mg/L)	nd	nd	$67 \pm 3$	$61 \pm 4$

507 TS: total solids; VS: volatile solids; TSS: total suspended solids; VSS: volatile

508 suspended solids; COD: chemical oxygen demand; TKN-N: total Kjeldahl nitrogen,

509 TAN-N: total ammonium nitrogen, n.d.: not determined.

510

### 511 **Table 2**

	Blaricum WWTP	Aarle-Rixtel WWTP
TS (g/kg)	$215 \pm 10$	$279 \pm 10$
VS (g/kg)	$200 \pm 10$	$261\pm10$
COD <sub>tot</sub> (g/kg)	$273 \pm 18$	$350 \pm 20$
TKN-N (g/kg)	$3.3 \pm 0.2$	$4.2 \pm 0.1$

512 TS: total solids; VS: volatile solids; COD<sub>tot</sub>: total chemical oxygen demand; TKN: total

513 Kjeldahl nitrogen.

#### **Table 3**

	Alkalinity supply	No alkalinity supply
Electricity requirements (kWh/m <sup>3</sup> )	0	0
Ferric chloride dosage (kg/m <sup>3</sup> )	0.1	0.1
Alkalinity supply (g IC/m <sup>3</sup> )	0.022	0
Ferric chloride cost (€/m <sup>3</sup> )	0.022	0.022
Sodium bicarbonate cost (€/m <sup>3</sup> )	0.031	0
Operational costs (€/m <sup>3</sup> )	0.053	0.022















#### Figure 5 519

