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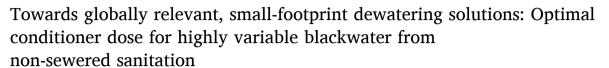
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Research article





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ABSTRACT

Globally, the sanitation needs of three billion people are met by non-sewered sanitation. Small-footprint treatment technologies are needed that are appropriate for dense urban areas. Blackwater (BW) (or fecal sludge), contains more than 95% liquid, and dewatering it without conditioning requires large footprints. Chemicallyenhanced dewatering with conditioners is a promising option to increase dewatering performance and reduce required footprints. However, before implementation of this solution there is a need for increased knowledge on selection and dosing of conditioners. This study evaluated bio-based and synthetic conditioners (chitosan, tannin-, and starch-based, synthetic with and without poly-acrylamide) with 14 types of BW from five countries. The supernatant after settling with jar-tests was analyzed to quantify optimal dose and dewatering performance. The reduction of total chemical oxygen demand (COD) was >55%, achieved by removal of particulate constituents with mainly soluble COD remaining in the supernatant. A reduction in particulate COD could lead to increased efficiency of soluble COD in supernatant treatment. Bio-based conditioners are as effective as synthetic conditioners, and when performance was variable, it was due to differing properties of TSS, TS, EC and pH. Optimal conditioner dose for synthetic conditioners and chitosan could be predicted using concentrations of total solids (TS) $(R^2 > 0.7)$, whereas optimal dose for starch- and tannin-based conditioners could be predicted with electrical conductivity (EC) ($R^2 > 0.8$), and colloid titration ($R^2 > 0.8$). In addition, real-time optical TSS and EC sensors could accurately predict chitosan dose for fresh BW treated at source ($R^2 = 0.97$, $R^2 = 0.95$). This study validates that use of conditioners for dewatering with highly variable BW can be implemented with real-time measurements for optimal dose, in globally relevant implementations.

1. Introduction

The sanitation needs of three billion people are met by non-sewered sanitation (Strande et al., 2014), (U. Habitat and WHO, 2021). Blackwater contains varying amounts urine, feces, flush water, and cleansing materials. Methods for safe management of the resulting blackwater (BW) are greatly needed, as in low-income countries discharge in urban environments places a huge burden on public and environmental health (Velkushanova et al., 2021), (Peal et al., 2020). In high-income

countries, it could provide more sustainable alternatives as sewer-based infrastructures reach their lifespan or capacity (Shaw et al., 2021). BW is used here as the wastewater that comes from toilets that are not connected to sewers (i.e. non-sewered sanitation). This BW can be treated directly at the source (e.g. off-grid or in-building), or stored onsite in containments and transported to treatment (e.g. pit latrines, septic tank), which is a type of BW typically referred to as "septage" or in low- and middle-income country contexts "fecal sludge" (Velkushanova et al., 2021). It consists of excreta (i.e. feces and urine), flush water,

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cleansing materials, and anything else entering the BW depending on level of separation (e.g. kitchen waste, greywater) (Rose et al., 2015). Knowledge cannot be directly transferred from municipal wastewater to non-sewered options, as the characteristics are 1–2 orders of magnitude more variable in total solids (TS), organic matter and nutrients, with varying levels of stabilization, due to the batch-wise collection, differences in toilet-level usage, and quality of construction (Velkushanova et al., 2021). Stabilization also varies if stored onsite, versus off-grid treatment at source (Ward et al., 2019), (Strande et al., 2018).

As the importance of non-sewered sanitation as a long-term sustainable solution has only recently been acknowledged, research lags substantially behind sewer-based solutions (Hayward, 2021). Existing fecal sludge treatment plants are scarce, and rely mostly on passive, land-intensive solutions (i.e., drying beds, settling tanks, stabilization ponds) (Tayler, 2018). BW is typically <5% TS, making dewatering a critical step in treatment (Velkushanova et al., 2021). As an improved understanding of fundamental mechanisms is being developed (Sam et al., 2022), (Ward et al., 2022), this knowledge can be applied for use of conditioners (Gold et al., 2016). Conditioners refer to coagulants and flocculants commonly used for chemically-enhanced dewatering in wastewater treatment (Bratby, 2016), (Metcalf and Eddy Inc., 2013).

Although floc formation and dewatering performance with conditioners has been widely studied for wastewater treatment, little is known about its application in BW treatment (Velkushanova et al., 2021), (Gold et al., 2018), (Kocbek et al., 2021). For example, Gold et al. (2018), reported use of synthetic polymers and bio-based conditioners for dewatering of fecal sludge only in Dakar Senegal, and did not report on optimization of process control. Kocbek et al. (2021) also reported use of conditioners but did not provide any specific operating conditions. Optimal dosing of conditioners is important, as under- and overdosing greatly impedes dewatering performance (Gold et al., 2016). The selection of conditioners commonly relies on empirical testing, as conditioner-solution interactions are complex (Bratby, 2016). Depending on the BW characteristics and conditioner type, flocs are formed by charge neutralization, polymer bridging or electrostatic patching, which are impacted by electrical charge, charge density, molecular weight and structure of conditioners (Lee et al., 2014). Organic particles in BW are negatively charged (Bratby, 2016), making a positively charged conditioner appropriate. However, many other characteristics influence floc formation and conditioner dose, such as high salt concentrations reducing the cationic charge of conditioners and reducing polymer bridging ('t Lam et al., 2016). High conductivity reduces dewatering performance, as monovalent ions weaken floc structure (Christensen et al., 2015). pH > 7 increases required dosage for cationic polymers (O'Brien and Novak, 1977), due to decreased solubility and charge density (e.g. tannin, chitosan) (Lau et al., 2017), (Ibrahim et al., 2021). Extra polymeric substances (EPS) decrease dewaterability, but BW is reported to have lower concentrations of EPS, potentially making conditioners more effective (Sam et al., 2022), (Christensen et al., 2015).

Optimal dosing of highly variable waste streams such as BW requires real-time monitoring. With wastewater, sensors for TS and total suspended solids (TSS) are commonly used to estimate conditioner dose (Metcalf and Eddy Inc., 2013). However, polymer dosage is not linearly proportional to the solids content, as the reduced particle counts lead to less particle interactions and floc formation (O'Brien and Novak, 1977). Real-time monitoring is lacking in non-sewered sanitation, with many operators relying on qualitative experience, for example color in relation to dewaterability (Ward et al., 2021). Rapid analytical field measurements could be used for process control to fill this gap, as proxies for metrics that normally require complicated or time-consuming laboratory analysis (Ward et al., 2019), (Ward et al., 2021). Promising reported field-measurements include image analysis of color and texture in photographs correlated to TS concentrations (Ward et al., 2019), (Ward

et al., 2021), and electrical conductivity (EC) and pH with dewatering performance (Ward et al., 2019), (Gold et al., 2018), (Ward et al., 2021).

The objectives of this study were to: 1) evaluate whether different types of BW (i.e., diverse regions and containment technologies) perform differently with bio-based and synthetic conditioners; 2) identify key parameters influencing dewatering performance; 3) evaluate whether rapid analytical field measurements can predict optimal dosage of conditioners; and 4) determine and present relevant information for the selection of conditioners.

2. Materials and methods

2.1. Sample collection

BW samples were collected from nine onsite containment technologies and one BW recipe mimicking an in-building source, in five different countries, in order to capture a range of stabilization (i.e., time since last emptied), BW consistency (i.e., liquid, slurry, and semi-solid), containment types, and usage patterns (i.e., household, commercial) (Table 1). Samples were collected, stored at 4 °C, and stored for no longer than 3 months, according to methods as described in Velkushanova et al. (2021). Additionally, all samples were characterized immediately prior to jar testing to provide an accurate baseline for further analysis.

2.2. Conditioners

Eighteen conditioners were identified and five selected based on supernatant turbidity as a metric how well suspended solids were removed from the liquid fraction, and observations of floc formation (reported in Supplementary Information). Chitosan Heppix A (Chitosan), Tanfloc SH (Tanfloc), and KCG750 (Emfloc) conditioners were selected as bio-based conditioners, and CP314 as a synthetic conditioner with polyacrylamide (PAM) and a cross-linked structure, and SFC100 without PAM and a linear structure. The combined use of coagulants and flocculants was ruled out, as all conditioners formed flocs without the use of coagulants, and coagulants alone form low strength flocs that are not appropriate for mechanical dewatering (Lee et al., 2014).

Chitosan was obtained from Biolog Heppe as a dry powder, which was dissolved in 1% acetic acid and distilled with water to a 0.5% (wt./vol.) stock solution. The Tanfloc was obtained from CDM Tannin as a dry

Table 1 Blackwater sample source overview.

Sample ID	Location	Containment Technology/Type of Establishment ^a	Volume Collected
BW01	Ghana	Cess Pit/Public Toilet	15 L
BW02		Septic Tank/Household	15 L
BW03		Septic Tank/Public Toilet	15 L
BW04		Septic Tank/Factory	15 L
BW05		Holding Tank/Public Toilet	15 L
BW06	Guatemala	Septic Tank/Restaurant	12 L
BW07	Lebanon	Septic tank/Household	10 L
BW08			10 L
BW09			10 L
BW10			10 L
BW11		Holding Tank/Informal Settlement	10 L
BW12	Canada	Septic Tank/Household	10 L
BW13	Switzerland	Septic Tank/Household	100 L
BW14		Mixture of feces, urine, toilet paper and tap water	100 L

Note

^a Self-reported containment type/type of establishment based on user surveys.

powder, which was diluted with tap water to a 5% (wt./vol.) stock solution. The polymers obtained from Emsland Chemie (Emfloc) and Flonex (CP314 and SFC100) were diluted with tap water to 1%, 0.5% and 0.5% (wt./vol.) stock solutions, respectively.

2.3. Analytical methods and data analysis

Jar tests were conducted to determine dewatering performance (Velkushanova et al., 2021), (Sawalha and Scholz, 2007). Incremental dosages of conditioner were added to five subsequent jars (400 mL), while maintaining one jar as a negative control. The system was stirred at a uniform speed (60 rpm for 2 min), settled for 5 min, and then supernatant was separated with a syringe for further analysis (Gold et al., 2016), (Dorea, 2006). A composite sample of settled portion including flocs was collected. Pre-conditioned (untreated) samples were homogenized prior to characterization. A complete table of characterization parameters are available in the Supplementary Information (SI).

Turbidity, pH, EC, TS, VS, TSS and VSS were analyzed according to standard methods (Velkushanova et al., 2021), (APHA, 2005). sCOD, COD (Method 8000), TN (Method 10,072) and NH₄-N (Method 10,031) were analyzed with Hach vials according to the manufacturer's directions and standard methods (Velkushanova et al., 2021), (APHA, 2005). Color and texture were obtained by analyzing photographs of 10 mL aliquots of BW, as described in Ward et al. (2021). Sand content was analyzed gravimetrically, after washing the residue of combustion with 0.1 mol/L hydrogen chloride following heating at 550 °C for 2 h (Velkushanova et al., 2021). Particle size distribution was determined gravimetrically by serial filtration (Metcalf and Eddy Inc., 2013). Zeta potential was measured using a Zetasizer (Zen3600), with 1 mL samples of undiluted BW and reported in mV. Colloid titration method was performed with small modifications as described in Gold et al. (2018) from the method by Ueno and Kina (1985). Results were measured in meq/gTS. Following centrifugation at 3300 rpm for 20 min, supernatant was decanted and turbidity measured using a HACH 2100 N turbidity meter and reported in Nephelometric Turbidity Units (NTU), adapted from the method described in Ward et al. (2021). CST was quantified as a metric of filtration time. CST was measured using a Triton 319 Multi-CST apparatus with 18 mm funnel, according to Method 2710 G (APHA,

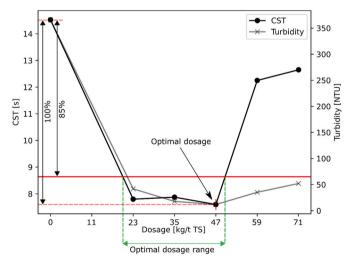


Fig. 1. Results from a single jar test to illustrate optimal dosage range calculation from measured CST and turbidity results.

2005), as adapted in Velkushanova et al. (2021). CST values are reported in seconds and were standardized by subtracting the CST of tap water. Settled BW sludge height was measured for each jar during jar testing as a way to qualitatively differentiate between sludge floc formation, volume, and strength. The full dataset is available in the SI.

In this study, both CST and supernatant turbidity were used as metrics of dewaterability to determine the appropriate optimal dosage range for each conditioner and BW sample, to account for differences due to treatment mechanisms, and variable BW and conditioner characteristics. As illustrated in Fig. 1, an optimal dosage range for each jar test was based on the difference between CST of unconditioned BW (the control), and the lowest CST among the other five samples in the jar test. The range was then defined as the intersection points of the 85% reduction line and the curve obtained by plotting dosage against CST.

Predictions of the optimal dosage of chitosan were evaluated for fresh BW from flush-toilets based on pH, EC and TSS measurements. Flush water, toilet paper, feces, and urine were combined in different ratios to obtain eight representative sub-samples of BW14 (BW recipes one to eight, see the SI). TSS was measured with an optical TSS probe that used light scattering principles at 90° and 135° (Endress + Hauser Turbimax CUS51D) (Velkushanova et al., 2021), (APHA, 2005).

2.4. Quality assurance & quality control

A partial factorial design was utilized to evaluate all relevant combinations of the parameters that were hypothesized to affect dewatering (Velkushanova et al., 2021). For each conditioner and each BW at least one set of jar testing was carried out in triplicate, verifying the reproducibility of the analytical methods, and identifying the extent of variability due to BW characteristics. Analysis for at least 10% of the samples were performed in triplicate (APHA, 2005), and the error that was obtained from the triplicates was extrapolated to the entire sample set for representative absolute error values. Every CST measurement was replicated four times and relative standard error on the replicates of jar tests and parameters was calculated.

3. Results and discussion

3.1. Results of analysis

Results of characterization parameters for BW are presented in Table 2, together with values from literature for comparison. In this study, parameters were defined as rapid analytical field characteristics if they could be done in the field and did not require access to a laboratory (Ward et al., 2021), (Junglen et al., 2020). This included pH, EC, turbidity, (Table 2). Additional parameters such as color, texture, colloid titration and zeta potential are available in the SI.

3.2. Parameters affecting conditioner performance

Presented in Fig. 2 are the percent removals of COD, TSS and TN in supernatant following conditioning and settling for all BW samples and all five conditioners. Detailed information regarding percent removals are available in the SI. Points of the same color and shape illustrate a triplicate of a jar test, as described in section 2. Presented in Table 3 are the average turbidity, TSS, COD, sCOD and TN measurements in the supernatant following conditioning and settling. The dewatering performance varied among samples, but in general, the reduction of COD was high (>55%) for all conditioners and samples. Clustering of points in Fig. 2 indicates that variation in performance for the five conditioners among the different BW was relatively low except for BW4, BW7 and BW14, indicating that the overall variability of dewatering performance can mostly be attributed to differences between individual BW

Table 2 Conventional laboratory and rapid analytical field characteristics of blackwater samples used in this study. Reported mean values and absolute error are calculated from the laboratory triplicates. Values from the literature are provided for comparison.

Sample	pH^h	ECh [mS/cm]	Turbidity ^h [NTU]	TS [g/L]		VS [g/L]		Sand Content [%TS]		TSS [g/L]	
				Mean	Stdev	Mean	Stdev	Mean	Stdev	Mean	
BW01	6.3	5.4	>10,000	39.8	± 1.6	33.1	± 2.0	6	± 1	30.3	
BW02	7.2	2.4	>10,000	18.2	\pm 0.7	13.7	\pm 0.8	11	± 2	12.0	
BW03	7.3	8.0	>4000	24.9	\pm 1.0	19.0	\pm 1.1	9	± 2	8.3	
BW04	7.6	2.1	323	1.2	\pm 0.1	0.5	\pm 0.0	72	\pm 12	0.4	
BW05	7.0	11.7	>10,000	17.1	\pm 0.7	12.1	\pm 0.7	4	± 1	10.2	
BW06	5.6	2.3	>10,000	36.2	$\pm~1.5$	30.2	\pm 1.8	7	± 1	25.5	
BW07	7.2	3.3	1058	9.0	\pm 0.4	5.1	\pm 0.3	18	\pm 3	7.5	
BW08	7.3	3.3	1704	5.3	\pm 0.2	2.8	\pm 0.2	8	± 1	3.7	
BW09	7.4	3.7	1436	7.1	\pm 0.3	5.2	\pm 0.3	3	± 1	5.8	
BW10	7.2	3.4	8583	10.6	\pm 0.4	5.9	\pm 0.4	22	± 4	10.8	
BW11	7.4	10.0	976	6.5	\pm 0.3	2.5	\pm 0.2	9	± 4	2.7	
BW12	6.5	1.3	1716	3.1	\pm 0.1	2.3	\pm 0.1	2	± 0	2.0	
BW13	6.7	1.4	1977	2.5	\pm 0.1	2.9	\pm 0.2	Nil	-	2.0	
BW14	8.2	1.2	140	1.2	\pm 0.1	1.9	\pm 0.1	Nil.	-	0.5	
Septic Tank Values from Literature	6.9–7.9 ^{a,b,c,d,e,}	2.3-15.4 ^{a,b,c,d,e}	_	$1-52^{a,e}$	-	1-30 ^e	-	$10-30^{a}$	-	$2-30^{c}$	
Lined Pit Latrine Values from Literature	$7.1 - 8.2^{a,d,e}$	12.1-14.6 ^{a,e}					-	$5-30^{a}$	-		
Sewered municipal wastewater	$7.1-7.9^{f}$	_	$2-15^{f_j}$	0.50^{g}	-	0.35^{8}	-	_	-	$0.12-0.30^{f,g}$	

Note: Nil = negligible.

NR = Not Reported. Italicized values indicate acceptable measurement error (i.e., VS/VSS > TS/TSS) when values were close to negligible.

a(Gold et al., 2018).

b(Gold et al., 2016).

c(Ward et al., 2019), (Strande et al., 2018), (Ward et al., 2022).

d(Velkushanova et al., 2021), (Englund et al., 2020).

e(Strande et al., 2018), (Ward et al., 2021), (Bassan et al., 2019).

f(Metcalf and Eddy Inc., 2013).

g(Henze et al., 2008).

hOnly single measurement, no stdev available.

iMean values where n (No. of samples) > 180.

iAfter indicated treatment.

characteristics, rather than certain conditioners performing significantly better than others. Factors contributing to performance variability are evaluated in the following paragraphs.

All conditioners were effective at removing pCOD. The observed variability in total COD reduction could be attributed to BW04, BW05. BW11 and BW14 having a higher fraction of sCOD (sCOD/COD of 0.30-0.37) compared to the other samples (sCOD/COD <0.15). In BW05, TSS removal was high (>90%), but COD reduction was low as most of the COD was soluble. For BW04, low total COD reduction due to its remarkably higher sand content (78% of total TS concentration) compared to other samples (0-25%) (Metcalf and Eddy Inc., 2013). BW01, BW03, BW05 and BW06 had high supernatant turbidity following centrifugation without conditioning (>500 NTU, Table 2), but remarkably less turbidity after passive settling with conditioning (<150 NTU), illustrating the importance of conditioners to achieve solid-liquid separation, even with mechanical dewatering.

High pH (7.4-8.6) can account for reduced removal of TSS, as cationic polymer dosage increases exponentially above pH 7 (O'Brien and Novak, 1977)- (Ibrahim et al., 2021), as shown with TSS reduction in BW04, BW11 and BW14. As observed with the variability of TSS removal in BW14, high charge density conditioners are less affected by pH (i.e. SFC100) (Tarleton and Wakeman, 2006). High salinity can reduce the efficiency of bridging mechanisms, due to coiling and reduced mobility of polymers ('t Lam et al., 2016), (Tarleton and Wakeman, 2006). In this study, high EC samples (i.e. BW05, BW11) visibly showed poor floc formation, which has been quantified in other studies (Velkushanova et al., 2021), (Ward et al., 2019). Particulate nitrogen reduction was also achieved, as demonstrated by samples with

high TS, TN and low NH₄-N having high removal of TN (i.e. BW06), whereas samples with high TS and high NH₄-N had much lower removal (i.e. BW05, BW11). Soluble nitrogen comes from urine. The variability of usage patterns at the toilet level has a double impact on treatment, as high pH and EC reduce conditioner performance (Udert et al., 2003). and elevated NH4⁺ following dewatering requires additional treatment. Fine particles are responsible for poor dewatering of BW, and also reduce conditioner efficiency (Motta et al., 2018) (Ward et al., 2022), as demonstrated with low TSS removal in BW11 and BW14 (>30% particles <10 μm). Particle size distribution, VS/TS, COD/N and pCOD/COD are metrics of stabilization that have been linked to dewaterability of fecal sludge (Ward et al., 2022). In this study, clear trends with stabilization and dewatering performance were not seen, and differences in BW04, BW11, and BW14 appear to be dominated by pH and conductivity.

3.3. Parameters affecting optimal dosage

Presented in Fig. 3 are optimal dosage (point) and optimal dosage range (defined in Fig. 1) for the five conditioners with a selection of samples. By dosing based on the lower end of the range rather than the optimal point, operating costs could be greatly reduced, without significant impacts on treatment performance. Not all manufacturers provided proprietary information on compositions or concentrations, so absolute comparison on a molar basis is not possible. Chitosan requires the lowest dosage with respect to the weight of the raw delivered product per influent concentration of TS (Fig. 3), which is in agreement with literature (Gold et al., 2016). As expected, the samples with lower

TSS [g/L]	VSS [g/L]		$\begin{array}{c} \text{Particle Size}^h \\ \text{[\% < 10 } \mu\text{m]} \end{array}$	Turbidity of supernatant after centrifugation ^h [NTU]	COD [mg/L]		sCOD [mg/L]		TN [mg/L]		NH ₄ –N [mg/L]	
Stdev	Mean	Stdev	Mean	Mean	Mean	Stdev	Mean	Stdev	Mean	Stdev	Mean	Stdev
± 3.6	29.3	± 2.6	14.1	962	72,233	± 2167	5796	± 728	2800	\pm 168	339	± 7
\pm 1.4	11.3	$\pm~1.0$	10.7	398	25,400	\pm 762	860	$\pm~108$	820	\pm 49	149	\pm 3
\pm 1.0	8.5	\pm 0.8	9.6	870	18,700	\pm 561	600	± 75	1785	$\pm~107$	907	\pm 18
\pm 0.1	0.8	\pm 0.1	32.3	25.8	490	$\pm~15$	178	\pm 22	192	\pm 12	141	\pm 3
\pm 1.2	11.7	\pm 1.1	11.4	954	32,850	\pm 986	12,150	\pm 1526	2190	$\pm~131$	1260	\pm 25
\pm 3.1	28.0	\pm 2.5	4.4	2950	71,800	\pm 2154	4460	\pm 560	1670	$\pm~100$	121	\pm 2
\pm 0.9	5.2	\pm 0.5	10.3	117	8300	\pm 249	615	± 77	312	± 19	149	± 3
± 0.4	2.9	\pm 0.3	9.0	58.1	5150	\pm 155	272	± 34	312	\pm 19	169	\pm 3
± 0.7	5.2	\pm 0.5	11.5	39.4	2295	\pm 68.9	196	\pm 25	399	\pm 24	291	\pm 6
\pm 1.3	6.7	\pm 0.6	22.3	108	9250	\pm 278	303	\pm 38	502	\pm 30	266	± 5
\pm 0.3	1.9	\pm 0.2	15.1	156	4085	\pm 123	1205	$\pm~151$	1010	\pm 61	895	\pm 18
± 0.2	2.4	\pm 0.2	6.1	119	3220	± 97	467	\pm 59	170	$\pm~10$	55	± 1
\pm 0.2	2.4	\pm 0.2	20.3	182	3650	± 110	127	\pm 16	189	± 11	72	± 1
\pm 0.1	1.0	\pm 0.1	30.2	8.2	1170	\pm 35	422	\pm 53	242	\pm 15	29	± 1
-	$1-20^{c}$	-	4–27°	180 ^{i,e}	7600–72,100 ^{a,b,d,e}	-	_	-	129-918	-	$18-60^{a,b,d,e}$	-
-		-		850 ^{i,e}	10,900-129,000 ^{a,d,e}	-	_	-	_	-	140-320 ^{a,d,e}	-
-	0.17-0.25 ^g	-	-	-	250-550 ^{f,g}	-	163–360 ^g	-	41–80 ^g	-	12.4 ^f	-

TS concentrations prior to conditioning had higher optimal dosage ranges due to fewer particle interactions (Bratby, 2016), (Metcalf and Eddy Inc., 2013). Based on observation, Tanfloc and Emfloc had noticeably smaller and weaker flocs, and floating flocs were occasionally observed with chitosan.

Presented in Table 4 are the Pearson correlation coefficients of TS, TSS, colloid titration (surface charge), pH, EC, color and texture, in terms of their relation to optimal dose. TS, TSS, EC and colloid titration were the most promising overall. TS and TSS have a stronger correlation for synthetic conditioners, while colloid titration had a stronger correlation for the bio-based conditioners. Colloid titration could be a better predictor of bio-based conditioners due to charge neutralization mechanisms (lower molecular weight), whereas TS and TSS for physical entrapment mechanisms (higher molecular weight) (Bratby, 2016), (Lee et al., 2014), (Tarleton and Wakeman, 2006). The weak correlation of CP314 to EC and colloid titration could be due to its cross-linked structure, which is less sensitive to higher conductivity (Palomino

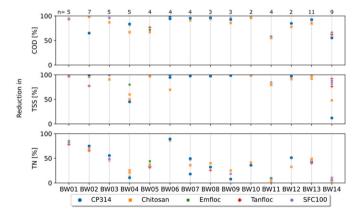


Fig. 2. Percent reduction of COD, TSS and TN in the supernatant after conditioning compared to original sample.

et al., 2012).

Zeta potential of untreated samples (11 of 14 BWs) did not show a clear pattern to conditioner performance (see Section 3.2). This was unexpected, as zeta potential and colloid titration are both metrics of surface charge, a key parameter in determining how well conditioners adsorb to particles, and particles to agglomerate (Ward et al., 2019), (Bratby, 2016), (Metcalf and Eddy Inc., 2013). However, colloid titration has been shown to adequately describe the zero surface charge point in wastewater, and is easier and less expensive to implement (Bratby, 2016), (Gold et al., 2018).

3.4. Conditioner selection: planning

As presented in Table 5, the selection of conditioners for different applications depends on factors such as supply chain, costs, ecological aspects, resource recovery, and available resources (e.g. financial resources and land availability). Supply chain issues have been identified as a common reasons for treatment plant failures in low-income countries (Bassan et al., 2015). Conditioners that can be produced from locally sourced bio-based materials could alleviate this (Gold et al., 2016), (Bassan et al., 2019), (Ssekatawa et al., 2021), such as chitin, tannin, cellulose, starch, sodium alginate, gums, and mucilage (Lee et al., 2014). Although it required the lowest dosage, chitosan in this study had the highest normalized cost, providing an example of how costs could be reduced through local production with waste from shrimp and crab shells, fungus, weevils, or fish scales (Gold et al., 2016), (Ssekatawa et al., 2021). Chitosan produced in India and Senegal had an estimated cost of 90% less than the one purchased in this study (Gold et al., 2016).

For resource recovery from the dewatered solids, it is important to consider biodegradability, energy potential, and toxicity of conditioners as they have a direct impact on calorific value, protein production (e.g. animal feed), and sustainability of soil conditioners (Diener et al., 2014). The environmental impact of PAM, which is a component of many synthetic conditioners, is not entirely clear. It is reported to undergo degradation by a variety of mechanisms, significantly increasing its mobility and potentially leading to the release of acrylamide monomer,

Table 3

Turbidity, TSS, COD, soluble COD and TN in the supernatant after conditioning. The mean and the standard deviation (stdev) were calculated over all supernatant measurements grouped by BW, independent from type of conditioner. Replicates included in average.

	Turbidity [NTU]		TSS [g/L]		COD [mg/L	COD [mg/L]		L]	TN [mg/L]	
Sample	Mean	Stdev	Mean	Stdev	Mean	Stdev	Mean	Stdev	Mean	Stdev
BW01	7.98	± 44.3	0.44	± 0.40	4566	± 471	4600	± 316	524	± 71
BW02	296.9	\pm 313.3	0.55	\pm 0.97	1578	\pm 3229	1574	\pm 3209	250	\pm 25
BW03	102.5	\pm 97.0	0.19	\pm 0.33	1154	\pm 728	1233	\pm 432	905	\pm 69
BW04	40.5	\pm 18.5	0.16	$\pm~0.05$	113	± 43	128	\pm 71	157	\pm 12
BW05	125.0	\pm 112.8	0.23	$\pm~0.07$	17,925	\pm 17,399	16,775	\pm 18,468	1390	\pm 124
BW06	85.0	\pm 32.6	2.33	\pm 3.66	2918	\pm 1223	2549	\pm 703	198	\pm 28
BW07	148.7	\pm 38.5	0.19	$\pm~0.05$	471	$\pm~170$	392	\pm 121	195	\pm 39
BW08	14.3	\pm 12.1	0.09	$\pm~0.01$	253	\pm 65	234	$\pm~32$	211	\pm 22
BW09	19.3	\pm 13.5	0.09	$\pm~0.01$	198	$\pm~119$	210	± 47	331	\pm 35
BW10	78.3	\pm 12.0	0.12	$\pm~0.03$	327	± 70	279	\pm 39	308	$\pm~19$
BW11	394.0	\pm 124.0	0.51	$\pm~0.06$	1825	\pm 51	1254	\pm 128	954	\pm 27
BW12	42.3	\pm 23.3	0.12	$\pm~0.08$	604	$\pm~162$	539	\pm 53	99	\pm 22
BW13	61.9	\pm 21.0	0.08	\pm 0.03	475	$\pm~132$	459	$\pm~109$	104	± 5
BW14	23.0	\pm 16.6	0.14	\pm 0.13	441	\pm 42	390	\pm 142	243	± 42

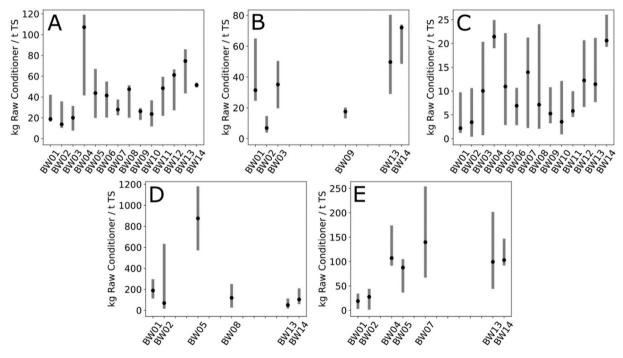


Fig. 3. Optimal dosage and optimal dosage range of blackwater samples in this study (A) CP314 (B) SFC100 (C) Chitosan (D) Tanfloc and (E) Emfloc; bars represent the optimal dosage range indicated as 85% of the maximum achievable CST reduction.

Table 4
Optimal conditioner dosage (kg raw/tonne TS) correlation coefficients in relation to untreated sample characteristic parameters; strongest related parameters shown in dark grey and supporting parameters shown in light grey.

Variables	CP314	SFC100	Chitosan	Emfloc	Tanfloc
TS	0.8	0.9	0.7	0.4	0.5
TSS	0.8	0.8	0.5	0.4	0.5
pН	-0.7	-0.6	-0.4	-0.4	-0.4
EC	0.2	0.8	0.5	0.8	1
Colloid Titration	-0.5	-0.6	-0.9	-0.8	-0.9
Color	0.4	0.6	0.3	0.0	0.5
Texture	-0.4	-0.5	-0.5	-0.7	-0.4

a known toxin and potential carcinogen (Bratby, 2016). However, there is also evidence that supports minimal concerns over the degradation products and its use in agriculture (Entry et al., 2008), (Holliman et al., 2005). PAM could also limit the methanogenesis pathway during anaerobic digestion (Wang et al., 2018).

3.5. Dewatering process control and implementation

Rapid field measurements are required to predict optimal conditioner due to the highly variable nature of BW, and the effect of TS or TSS, pH and EC or salinity on flocculation. As shown in Fig. 4, TS, EC, and pH sensors were evaluated for use as predictors of optimal dosage of chitosan for BW from flush-toilets. TSS and EC readings were good predictors ($R^2 = 0.97$ and $R^2 = 0.95$, respectively), whereas pH was a poor predictor ($R^2 = 0.24$). One sample with urine and no feces had a high EC that did not correlate with optimal dosage, indicating that combinations of sensors could be required for in-building, or treatment of individual flushes, increasing operational complexity. This would not be the case for fecal sludge BW, as it is mixed in containment and during collection and delivery to treatment. The predictive capacity of this model based on one conditioner and one type of BW, provides validation that instantaneous feedback for process control is possible. With further experience and more sophisticated models, use of sensors could include prediction of doses based on the lower optional dosage range.

Prior to selecting conditioners as an option for dewatering technologies, a study needs to be conducted to evaluate local quantities and qualities (Q&Q) of BW to be treated (Velkushanova et al., 2021). As illustrated in Fig. 2, if BW11 and BW14 were most characteristic, other alternatives could perform better, or as also demonstrated by BW14, the selection of conditioner based on actual BW characteristics can directly impact performance. Considerations should include source (e.g. restaurant, factory, public toilet, household), and characteristics that are specifically related to flocculation performance in addition to "conventional" parameters (e.g. pH, colloid titration, EC, sand, TSS, particle size distribution). In this study, conditioner performance was evaluated in laboratory-studies based on supernatant after settling. However, as summarized in Table 5, other dewatering technologies that can further reduce required footprints are of interest, such as gravity belt filter, filter press, screw press and centrifuge. Although conditioners are required for mechanical dewatering, higher shear stress applied to flocs also needs to be considered, which with weaker flocs will result in more particles released into the liquid stream (supernatant), affecting the overall TSS and COD reduction. Therefore, pilot testing is needed to select optimal combinations of conditioners and dewatering technologies, to achieve appropriate floc formation for subsequent dewatering technologies.

The reduction in pCOD with conditioners could open up the possibility for more compact treatment of the resulting sCOD in the liquid supernatant, assuming it is more readily biodegradable. For example,

Gold et al. (2016) reported that the use of chitosan, lime or synthetic polymers could reduce dewatering times on drying beds by 57-97% to achieve 50% TS or 9-15% and 15-26% to achieve 90% TS at loading rates of 100 and 150 kg TS/m2, respectively. Similar results would be expected with the conditioners used in this study because the reduction of TSS in the supernatant of samples conditioned were in the same range. However, due to the high variability of BW, the variability of supernatant streams going to further treatment will also be highly variable, and most likely intermittent, which needs to be accounted for in design and operation. For example attached growth processes (e.g. moving bed bioreactor or rotating biological contactor), which are more resistant to variable loadings could still lead to additional space savings (Metcalf and Eddy Inc., 2013). The dewatered solids could then be readily transported and managed with more well-established options for resource recovery (Andriessen et al., 2019). Therefore, savings from the reduction in required land with treatment plants due to use of conditioners could include increased loadings to existing settling tanks or drying beds, mechanical dewatering technologies, supernatant treatment, and/or more efficient solids handling.

4. Conclusions

To the best of our knowledge, this is the first study of its kind looking extensively at the use of conditioners for dewatering of BW from non-sewered sanitation. The main conclusions of the study are:

- Synthetic and bio-based (i.e., starch, tannin, and chitosan) conditioners removed total COD in BW by more than 55%. Removal of pCOD results in a significant reduced loading for the removal of sCOD in dewatering supernatant.
- Differences in COD and TSS in dewatering supernatant are affected by BW characteristics (TSS, TS, EC and pH) and selection of dewatering technology.
- The implementation of conditioners will require real-time monitoring for accurate dosing. The feasibility of inline sensors for TSS and EC were demonstrated.
- To alleviate supply chain issues, the local resource recovery-based production of bio-based conditioners from available waste streams should be explored.
- The results of this study are based on a range of BW (i.e., directly from flush toilet or stored in containment, low-, middle-, and highincome countries, with- and without greywater), indicating they are globally transferable for more compact and efficient treatment of BW.

Author contributions

Kelsey Shaw & Michael Vogel: Methodology, Investigation, laboratory analysis, Data curation, Formal analysis, co-lead in writing. Nienke Andriessen: interpretation of data, Writing – review & editing. Tommy Hardeman: laboratory analysis and Data curation. Caetano Dorea: interpretation of data, Writing – review & editing. Linda Strande: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Funding acquisition.

Declaration of competing interest

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Table 5Conditioner application overview for use by practitioners for evaluation of suitability for further investigation and implementation.

	General						ance l Dose [kg/t TS] es [g/L]:			Flocculation Formation	Minimum Costs based on Optimal Dosage Range (\$USD/t TS) ^c TS ranges [g/L]:	
Conditioner	PAM and/or Hydro Carbons	Charge Density ^a	Molecular Weight	Availability	Raw Material	<2.5	2.5–15	15–25	>25	Visual overdose observed (i.e., Floating)/ Floc Size	<2.5	2.5–15
CP314	+	Medium (40% mol)	High	Worldwide	Proprietary	47–85	25–51	14–43	20–47	Floating; large	205	109
SFC100	-	High (50–100% mol)	High	Worldwide	Proprietary	49–73	22–49	13–32	25–64	Not floating; large	218	97
Chitosan	-	High (50–100% mol)	Medium	Can be produced from locally sourced materials	Chitin	19–25	4–17	2–17	2–10	Floating; medium/large	379- 3775	82–814
Emfloc	-	Medium (25% mol)	Low		Plant Starch	95–158	58–225	22–72	6–31	Not Floating; small and stringy	164	101
Tanfloc	-	Medium	Low		Acacia mearnsii bark	72–196	34–169	306–894	126–283	Not Floating; small	233	110

Notes:

aConditioners can be regarded as having low medium or high charge densities around 10%, 25% and 50–100%mol (Bolto and Gregory, 2007). bDepends on sludge characteristics.

cBased on ordering/delivering in Europe; locally sourced options would be expected to have different costs. The price ranges of chitosan covers two prices: Supplier from Germany (max) and supplier from India (min).

dPyrolosis and incineration is the preferred method as there is currently no clear evidence that disposal of PAM is harmful if disposed of in this way. However other end-uses (i.e., composting) may be appropriate depending on the situation.

eUnless indicated otherwise information sourced from personal communications with conditioner supplier.

Minimum based or Optimal Range (S TS) ^c TS range [g/L]:	Dosage SUSD/t	Ecological Aspects			Further Treatm	ient
15–25	>25	Biodegradability	Production	End-Use ^d	Resistance to shear stress	Recommended use ^b
61	86	Low	Must be produced commercially based on inclusion of crude oil products and/or industrially synthesized polymers, low production price.	Pyrolysis/Incineration	High	Settling, Geotubes, Gravity belt filter, Filter press, Screw press, Centrifuge
56	112	Low		Pyrolysis/Incineration	Medium	Settling, Geotubes, Gravity belt
31–313	45–446	Acceptable: Readily Biodegradable	Locally available materials; Requires chemicals and laboratory knowledge/access.	Good biodegradability; therefore end uses not	Medium	filter (Filter press, Screw press, Centrifuge)
38	10	Acceptable: Meets German Fertilizer Ordinance Regulations		limited.	Low to Medium	Settling, Geotubes, Gravity belt filter
988	407	Acceptable: Readily Biodegradable			Low	Coagulation, Settling

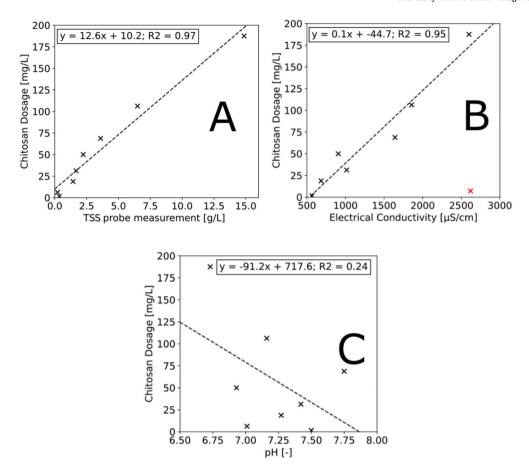


Fig. 4. Results of linear regression model predicting optimal dosage with chitosan and eight synthesized BW recipes (A) is a function of TSS concentrations only, (B) is a function of EC concentrations only, and (C) is a function of pH only. One outlier marked as a red "X" was removed from the model.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2022.115961.

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