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1                   **Design and preliminary implementation of onsite**  
2                   **electrochemical wastewater treatment and recycling**  
3                   **toilets for the developing world**

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6                   Clément A. Cid<sup>1</sup>, Yan Qu<sup>1,2</sup>, and Michael R. Hoffmann\*<sup>1</sup>

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8  
9                   <sup>1</sup>Linde-Robinson Laboratories, California Institute of Technology,  
10                   1200 E California Blvd, Pasadena, CA 91125

11                   <sup>2</sup>Trussell Technologies Incorporation,  
12                   232 N Lake Ave., Suite 300, Pasadena, California, 91101, USA

13  
14                   *Prepared for submission to Environmental Science: Water Research and Technology*

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16  
17  
18  
19  
20                   \*corresponding author: Contact information: [mrh@caltech.edu](mailto:mrh@caltech.edu)

## 21      **Abstract**

22      Self-contained toilet wastewater treatment system prototypes based on  
23      electrochemical oxidation of feces and urine using bi-layered semiconductor anodes  
24      ( $[\text{Bi}_2\text{O}_3]_z[\text{TiO}_2]_{1-z}/\text{Ir}_x\text{Ta}_y\text{O}_2/\text{Ti}$ ) have been designed, constructed, and implemented in  
25      regions where access to proper and sufficient sanitation is limited. Prototypes were  
26      designed to fit in shipping containers in order to provide toilets and onsite  
27      wastewater treatment with clean water recycling. Units were designed to handle the  
28      waste of 25 users per day (or 130 L of toilet wastewater). The first prototype was  
29      tested on the Caltech campus (Pasadena, California) followed by improved second-  
30      generation prototypes that were subsequently installed in India (Ahmedabad, Gujarat  
31      and Kottayam, Kerala) and China (Yixing, Jiangsu) for open use in various public  
32      settings. The prototypes were able to provide for the disinfection of pathogens  
33      ( $<10$  MPN *Total coliforms* and  $<1$  MPN *Fecal coliform* indicator organisms per  
34      100 mL), reduction of chemical oxygen demand ( $<100$  mg  $\text{O}_2$   $\text{L}^{-1}$ ), ammonia  
35      ( $<10$  mg  $\text{N}$   $\text{L}^{-1}$ ), and color at an average energy consumption of less than 180 Wh per  
36      user per day. The treated wastewater was recycled for use as toilet flushing water.

## 37      **Keywords**

38      onsite sanitation; electrochemical wastewater treatment; chlorine disinfection

## 39 **Water impact statement**

40 More than 2.6 billion people do not have access to sanitation. With an average  
41 energy usage of less than 180 Wh per user per day, electrochemical toilet  
42 wastewater disinfection, chemical oxygen demand removal, and ammonia removal  
43 can provide a viable and cost-effective alternative to traditional sewers. Onsite  
44 electrochemical wastewater treatment systems were tested and performed  
45 without critical breakdown during the field testing.

46

## 47 **1. Introduction**

48 In February 2011, The Bill & Melinda Gates Foundation (BMGF) announced a  
49 major challenge to university researchers to “Reinvent the Toilet.” The primary goal of  
50 the BMGF was to engage universities in the development of new and innovative  
51 methods to treat human bodily wastes at the site of origin without discharge to the  
52 ambient environment or discharge to conventional sewer systems, septic tanks,  
53 cesspools, or open drainage systems. The overarching goal of the BMGF Global  
54 Development Program within the context of their Water, Hygiene and Sanitation  
55 initiative was to develop practical low-cost solutions that could be implemented in  
56 regions of the world that lack access to safe and affordable sanitation. The primary  
57 challenge was to develop a comprehensive approach to design, development, testing,  
58 and prototyping of systems that could collect and process human waste on-site at the  
59 source of origin and at the same time produce useful byproducts including fertilizer,  
60 mineral salts, energy, purified, and disinfected water with no solid or liquid discharge  
61 to the environment. The overarching objective is to provide suitable sanitary systems  
62 for the 2.6 billion people who currently lack access to safe and affordable sanitation  
63 (Figure 1). A cost constraint was set at a maximum of \$0.05 per person per day  
64 include capital costs and operating expenses.

65 The development of integrated networks and facilities for the transport and  
66 subsequent treatment of domestic wastewater has been a key factor in the growth and  
67 development of modern urban environments. Sanitation has accompanied human

68 development from early civilizations with rudimentary systems (1) to mid-19th  
69 century first large-scale sewer networks in American and European cities (2-4).  
70 Although a well-constructed modern urban sewer network can be hygienic and  
71 efficient due to economies of scale (5), they also have major drawbacks, which include  
72 nuisance odors from improper operation and maintenance (6), local groundwater  
73 contamination due to leakage from improper connections and corrosion of pipes and  
74 concrete sewers (7, 8), or prohibitively expensive capital investments (9).

75 For these reasons, developing countries have often turned to non-sewered  
76 sanitation (NSS) systems for the disposal of human bodily waste. Furthermore, in  
77 areas with limited access to water, technologies have traditionally been restricted to  
78 dry or manual pour-flush types of toilets such as composting toilets or pit latrines  
79 (10). Although these waterless technologies appear attractive because of their limited  
80 need for water, they do not provide the olfactory or sanitary comfort of flush toilets  
81 (11). In addition, they are not always reliable for disinfection, pathogen removal (12),  
82 or for preventing subsequent pollution by latrine waste soils and groundwater (13).  
83 In areas that have access to water, decentralized toilets using flush technologies are  
84 most often connected to septic tanks. Septic tank treatment systems require large land  
85 surface areas to build effective leaching fields that are necessary for the safe  
86 elimination of pathogens (Title V septic system in the United States) and can often  
87 lead to fecal contamination of local water sources if improperly installed and  
88 maintained (14-16).

89 Therefore, a technology capable of treating and recycling toilet wastewater at low  
90 cost would have significant advantages over traditional NSS solutions (*vide supra*). In  
91 this regard, Radjenovic and Sedlak have identified electrolysis processes such as  
92 potentiostatic electrochemical oxidation as “potential next-generation technologies  
93 for the treatment of contaminated water” (17). Electrochemical oxidation of  
94 wastewater has been investigated for more than 30 years with a focus on organic  
95 pollutant degradation (18, 19), most systems rely on the anodic formation of free  
96 hydroxyl radical OH<sup>·</sup> from water oxidation or the direct oxidation of the compounds of  
97 interest. Both processes consume a lot of energy to achieve appropriate contaminant  
98 removal (20).

99 Weres and collaborators (21-23) investigated the use of multilayer semiconductor  
100 anodes to generate surface-bound hydroxyl radicals OH<sup>·</sup> for organics degradation (24-  
101 26). These multilayer semiconductor anodes have a low overpotential for the  
102 oxidation of chloride to chlorine (27). This capability makes the multi-layer  
103 semiconductor anodes particularly suitable for the direct formation of Reactive  
104 Chlorine Species (RCS) from the oxidation of the chloride naturally present in the  
105 human wastewater (28-31). Although the previously published laboratory results  
106 have shown the feasibility of anodic oxidation for toilet wastewater treatment (29-  
107 32), there is no literature available about the automated, autonomous on-site  
108 electrochemical treatment of toilet wastewater under actual field operating and  
109 testing conditions. Herein, we present the results of field studies employing

110 electrochemical wastewater treatment for the removal of chemical oxygen demand  
111 and for recycling of disinfected and clarified water for use as toilet flushing water.

## 112 **2. Guidelines, materials and methods**

### 113 *2.1. Health considerations for an onsite wastewater recycling systems*

114 The primary sources of biological and chemical contamination entering onsite  
115 wastewater treatment systems are from human excreta. The amount and the  
116 composition of human excreta varies greatly from one individual to another (33, 34)  
117 with an average of 1 L to 1.5 L of urine and 300 g to 450 g of feces per adult per day.  
118 Feces are often the major carrier of pathogens in human excreta (35) with an average  
119 number of  $10^{11}$  CFU (colony-forming units) of bacteria per gram of feces for a healthy  
120 adult individual. Given that pathogen die-off times in untreated human excreta are  
121 between one and three months for bacteria and viruses, and several months for  
122 helminth eggs (36), a reliable and rapid removal of pathogens down to acceptable  
123 levels is crucial for the success of an onsite human wastewater treatment technology.  
124 For example, the World Health Organization considers that a safe pathogen level  
125 appropriate for water reuse in agriculture is less than 1 CFU per 100 mL for typical  
126 indicator organisms *E. coli*. (37).

127 When onsite wastewater treatment systems are installed close to their users in  
128 order to minimize installation costs, such systems can become potential threats to the  
129 health of humans living nearby when the wastes are not properly contained (38) or  
130 sufficiently treated. Natural barriers such as the leaching fields for septic tanks or clay



131 or concrete walls for dry latrine pits are not always effective barriers (13, 39). Risks of  
132 contamination are further increased when the users come into contact with effluent  
133 streams (*e.g.*, treated water and/or biosolids) produced by onsite sanitation systems.  
134 Thus, the treated and recycled waters that are processed onsite must be free of  
135 pathogens and have an acceptable physicochemical composition that meets  
136 conventional water quality standards for reuse.

137 However, technical standards that have been adopted in many countries help to  
138 regulate the composition of recycled water for domestic reuse but they are often  
139 limited to large scale indirect and direct potable reuse of conventional wastewater  
140 treatment plant effluents (40) or the treatment and reuse of non-fecal contaminated  
141 water (greywater) primarily from sinks, washers, and showers (NSF/ANSI Standard  
142 350). Therefore, a toilet wastewater recycling system has to produce an effluent that  
143 does not damage the system itself, is safe for users, and contains enough residual  
144 disinfecting capacity to prevent subsequent chemical and microbial contamination  
145 due to exposure to the treated and recycled water.

146 In addition to the meeting the basic sanitary and water quality requirements, a self-  
147 contained toilet and wastewater treatment system for use in developing countries  
148 needs to be affordable, durable, and functional in an off-grid environment with limited  
149 access to electricity, fresh water, and sewers.

## 150 *2.2. Choice of prototype testing locations*

151 Four pre-alpha prototypes of a similar design (Figure 2 and section 3) were tested  
152 in the USA, India, and China (Figure 1). All the testing sites had a connection to sewers

153 in case the prototypes would fail during testing. The first pre-alpha prototype was  
154 tested on the campus of the California Institute of Technology (Caltech) in Pasadena,  
155 California (PAS prototype, Figure 2 a) for preliminary data gathering and early design  
156 adjustments; two additional pre-alpha prototypes (AMD and KYM) were designed and  
157 built on the Caltech campus and then shipped to India for 6-month field trials in two  
158 different locations. The quantity of treated wastewater was recorded for each  
159 prototype and the frequency of usage of the toilet was recorded for the prototype  
160 designated as AMD only. All prototypes were designed to be running full time. The  
161 prototype designated as AMD (Figure 2 c) was installed in a public park in the city of  
162 Ahmedabad in Gujarat State of northwest India. Ahmedabad has a semi-arid climate  
163 and is the sixth largest city of India with more than 6.3 million inhabitants. The  
164 prototype designated as KYM (Figure 2 b) was installed in the campus of Mahatma  
165 Gandhi University near the School of Environmental Sciences in Amalagiri district of  
166 Kottayam City in the State of Kerala, which is located in southwestern India. Kottayam  
167 has a tropical climate with a population of 200,000 inhabitants. A fourth pre-alpha  
168 prototype was constructed and tested in the Municipal Yixing Elementary School of  
169 Yixing, China (YXG prototype, Figure 2 d) in collaboration with Yixing Eco-Sanitary  
170 Manufacture Co.

### 171 2.3. Monitoring and evaluation methods

172 Chemical oxygen demand (30), total nitrogen (41), total suspended solids (TSS),  
173 and indicator organisms *E. coli*, *Total coliform*, and *Fecal coliform* bacteria were  
174 measured to assess the wastewater treatment efficacy.

175 The voltage at the electrodes and the current delivered by the power supply to the  
176 electrode arrays were continuously measured using a personalized data logger  
177 (Programmed Scientific Instruments, Arcadia CA) at regular intervals (*e.g.*, every 10  
178 seconds). The activation and deactivation of pumps as well as the status of water level  
179 sensors were monitored and recorded using the same data logger. At each recording  
180 time, the data logger stored a new line of values of the different components of the  
181 system in a daily Comma Separated Values (CSV) file. The data was tagged with the  
182 local time and date and stored in a solid-state device in the computer controlling the  
183 system. The CSV files were regularly retrieved by an operator.

#### 184 2.4. Analytical methods

185 Unfiltered COD was measured using a reflux digestion system with water  
186 condensers followed by titration according to Standard Method 5220 (42) or via  
187 colorimetric method similar to Hach Method 8000 (Hach Company, Loveland CO). TN  
188 was determined using persulfate digestion (Hach Method 10071). Total Kjeldahl  
189 Nitrogen (TKN) was determined by distillation (Indian Standard 5194-1969).  $\text{Cl}^-$ ,  $\text{NH}_4^+$   
190 +  $\text{NH}_3$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  concentrations were determined by ion chromatography  
191 (Dionex ICS 2000; AS19G anions, CS12A cations).

192 Disinfection was assessed by estimating the quantity of indicator organisms *E. coli*,  
193 *Total coliforms*, and *Fecal coliforms* with the following respective EPA methods:  
194 1103.1 (43), 9132 (44), and 1680 (45) with appropriate dilutions. Free chlorine (FC)  
195 was measured by reaction with N,N-diethyl-p-phenylenediamine (DPD) indicator in  
196 accordance with Standard Method 4500-Cl G (42) and Hach Method 8021. Total

197 chlorine (TC) was measured by the Amperometric Titration Method in accordance  
198 with Standard Method 408 C (42).

199 Cathodic and anodic potentials relative to Normal Hydrogen Electrode (vs. NHE)  
200 were measured using a 3.5 M Silver/Silver Chloride (Ag/AgCl,  $E_0 = 0.205$  V vs. NHE)  
201 reference electrode (RE-5B, Bioanalytical Systems Inc., USA) connected to a three-  
202 electrode potentiostat (Biologic, France) measuring the potential between the  
203 reference electrode and the anode used for chlorine production (see below).

### 204 ***3. Design of the self-contained toilet and treatment systems***

#### 205 *3.1. Sizing considerations*

206 A flow diagram of the overall toilet wastewater treatment process is presented  
207 Figure 3 and Figure S1 and a picture of a typical treatment system as installed in the  
208 field-tested prototypes (PAS, AMD, KYM) is reproduced Figure 4. When the flush toilet  
209 was used, the mix of urine, feces, and flush water (toilet wastewater) was macerated  
210 and pumped (Jabsco Macerator Pump 18590-2094, Xylem USA or Saniflo Sanigrind  
211 Grinder Pump for Bottom Outlet Toilets, SFA France) into a 2-m<sup>3</sup> polypropylene  
212 sedimentation tank for a residence time of  $\tau_{\text{bio}} \geq 15$  days. The maceration reduced the  
213 feces to particles of 3 mm or less according to the manufacturer's description. In the  
214 sedimentation tank, the toilet wastewater underwent decantation and some level of  
215 anaerobic digestion, similarly to a septic tank (46). The decanted solids remained at  
216 the bottom of the sedimentation tank without further active processing. The  
217 prototypes were designed for treating the toilet waste from approximately 25 daily

218 uses: this is the equivalent to having a single toilet for a family of five people, the  
219 average household size in India in 2011 (47). Each of the family members flushing five  
220 times per day on average (48) with a flush volume of 1.28 US gallons or 5 L (US EPA  
221 WaterSense) and considering that one person produces approximately 1.5 L of urine  
222 in one day (41), the total daily volume of toilet wastewater to treat was then  
223 estimated to be  $V_d = 132 \text{ L day}^{-1}$  so the sedimentation tank should be sized to hold at  
224 least  $\tau_{\text{bio}} * V_d \approx 2 \text{ m}^3$ .

225 After decanting in the sedimentation tank, the wastewater was macerated and  
226 pumped (Jabsco Macerator Pump 18590, Xylem USA) to an electrochemical reactor  
227 (ECR) system (see below for description) for batch processing at constant voltage  
228 with active recirculation ( $10 \text{ L min}^{-1}$ ) during a period  $\tau_{\text{elec}}$ , the electrochemical  
229 residence time. The working volume needed for the ECR  $V_{\text{ECR}}$  was determined by (eqn.  
230 1) with  $\alpha$  the fraction of the daily time during which the ECR is running.

$$231 \quad V_{\text{ECR}} = \frac{V_d}{24 \alpha \tau_{\text{elec}}} (= 22 \text{ L}) \quad (40)$$

232 After electrolysis in the ECR for a period  $\tau_{\text{elec}}$ , the water was filtered through a  
233  $200 \mu\text{m}$ -mesh microfiltration unit (Grainger USA). The treated effluent was then  
234 pumped with a drain pump (Jabsco PAR Max 3, Xylem USA) to a storage tank to use  
235 for flushing the toilet. The treated water tank (TWT) was capable of storing flushing  
236 water for one day of operation. An overflow mechanism was installed in the TWT in  
237 case excess treated water needed to be discharged from the system occasionally. The  
238 sizing of the polypropylene sedimentation tank as well as the water losses through

239 evaporation during the process significantly limited the necessity to discharge treated  
240 water. Four complete toilet and associated wastewater treatment and recycling  
241 systems were operated in the US, China, and India (Table 1).

### 242 3.2. Electrochemical reactor system

243 The ECR tank body is made of a poly(methyl methacrylate) (PMMA) welded  
244 together (Nanopac, Yongin-Gun, South Korea) in a rectangular cuboid shape with the  
245 following dimensions: 63.5 cm \* 35.6 cm \* 16.5 cm (height \* width \* depth; Figure S2).  
246 Two 6-mm thick PMMA plates of respective dimensions 25 cm \* 14 cm and  
247 56 cm \* 14 cm with 1.5 cm diameter holes spaced every 2.5 cm were used to hold the  
248 electrode array in place at a distance of 7 cm above the bottom of the ECR. 0.75-inch  
249 and 1-inch diameter National Pipe Tapered (NPT) thread holes were drilled on the  
250 side and the bottom of the ECR tank to connect sampling ports and plumbing.  
251 Circulation of fluid inside the ECR tank was assured by a brushless centrifugal pump  
252 (Fortric ZKWP04 24V, Fortric China). The ECR tank was connected to the other  
253 components of the system using braid-reinforced polyurethane hose or polyvinyl  
254 chloride (PVC) pipes of sufficient diameter.

255 At the core of the ECR, electrode plates were assembled as an array in alternate  
256 configurations of doubly-coated anodes sandwiched between two stainless cathodes  
257 (*e.g.*,  $CA_nCA_{n+1}...C$ , etc.) where each electrode plate was separated by a 3-mm spacing;  
258 nylon screws, nuts, and washers were used for structural integrity. The arrays  
259 composed of eight stainless-steel (316 Grade) cathodes “C” and seven doubly-layered  
260 semiconductor anodes “A” ( $[Bi_2O_3]_z[TiO_2]_{1-z}/Ir_xTa_yO_2/Ti$ ) (Nanopac, South Korea) that

261 were coated on both sides. The manufacturing process and the effect of the outer layer  
262 composition ( $[\text{Bi}_2\text{O}_3]_z[\text{TiO}_2]_{1-z}$ ) have previously been described in the literature (28,  
263 31). The total exposed surface area of the anodes ( $S_A$ ) was  $1.8 \text{ m}^2$ . The ECR electrode  
264 array was powered at an electrical potential between 3.3 V and 3.5 V using a  
265 potentiostatic power supply (Program Scientific Instruments, USA).

### 266 3.3. Automation for the wastewater treatment and recycling

267 The daily number of users and the frequency of usage of the toilets were not  
268 controlled in any of the systems. For this reason, it was necessary to ensure that a  
269 sufficient amount of treated water was available for flushing at all time to support the  
270 continuous operation of the treatment system without direct supervision. This was  
271 achieved with a computer-controlled automation algorithm (Figure 3) programmed  
272 on a dedicated software package (Program Scientific Instruments, Arcadia CA)  
273 running on a Panel PC PPC-L62T (Advantech, China) with Windows 7 operating  
274 system (Microsoft, USA). Capacitive level sensors CD50 DC (Carlo Gavazzi, Italy) were  
275 used as triggers for the automation mechanism. Pumps with a programmed maximum  
276 running time were used as actions (*e.g.*, macerator pump turns on) or as triggers  
277 when they changed state (*e.g.*, circulation pump stops running). ECR power status (or  
278 change of status) was used as a trigger, an action, and a feedback for the automation  
279 algorithm.

280 The algorithm is composed of two main parts (Figure 3): the start of a treatment  
281 cycle (lines 1-3) and the treatment cycle loop (lines 4.1 – 4.5). A treatment cycle starts  
282 when the level of water in the treated water tank falls below the sensor TWT\_S1 (line

283 1), placed approximately at 20% of the tank's height. The macerator pump MP  
284 switches on for a fixed duration to fill up the ECR tank, the ECR power supply starts,  
285 and the circulation pump CP starts. At the end of the treatment cycle ( $\tau_{elec}$ ), the ECR  
286 tank is emptied by the drain pump DP into the treated water tank TWT. After that, if  
287 the water level in TWT is still below TWT\_S1, a new treatment cycle begins. The  
288 treatment cycles will continue until the level of the treated water tank is above the  
289 TWT\_S2 sensor, positioned close to 90% of the tank's height.

#### 290 *3.4. Energy distribution across to the system*

291 The energy consumption of the entire system and ECR was monitored using non-  
292 invasive current sensors installed on the wires connecting the control system to its  
293 power source. The power source was a combination of grid electricity at 220 – 240 V  
294 AC when available, and 24 V DC power source from a 330 W solar panel (Xunlight,  
295 USA) stored in two 12 V Blue Top lead-acid backup batteries (Optima, USA) via a  
296 Conext MPPT 60 PV (Schneider Electric, Germany) charge controller (Figure S3). A  
297 backup battery recharge was also implemented using a TRUECharge 2 40 A (Xantrex,  
298 USA) battery charger connected to grid electricity.

#### 299 *3.5. Integration*

300 All components of the entire system were housed in customized steel shipping  
301 containers with an integrated public bathroom when necessary (Figure S4). AMD and  
302 PAS prototypes were modified 10-ft and 30-ft long containers cut from standard  
303 length 20-ft and 40-ft international shipping containers, respectively. KYM and YXG  
304 prototypes were repurposed 20-ft standard shipping containers. All containers were



305 insulated and retrofitted with in-wall electrical wiring and on-wall plumbing in  
306 copper, cross-linked polyethylene (PEX), or PVC pipes. Doors and windows were  
307 added to improve access to the treatment system, increase air circulation, and provide  
308 a physical work environment for sampling and on-site measurements (Figure 4).

#### 309 **4. Results and discussions**

##### 310 *4.1. Free chlorine production*

311 Chlorination is a cost-effective way to remove pathogens in water, if allowed  
312 sufficient contact time for a given free or total chlorine concentration (49, 50). In our  
313 prototype systems, chlorine is continuously generated via the electrochemical  
314 oxidation of chloride, (i.e., the Chlorine Evolution Reaction, CER), at a fixed potential  
315  $3.5 \text{ V} \pm 0.25 \text{ V}$  across the electrodes. The CER is an apparent first-order reaction with  
316 respect to the concentration of chloride in solution. In a large-electrode array (ECA) as  
317 shown in (Figure S5), the CER rate is shown to vary quasi-linearly from 11 to 17 ppm  
318  $\text{Cl}_2 \text{ min}^{-1}$  with respect to the ratio  $S_A/V_{\text{ECA}}$  in 20 mM NaCl in water. After an extended  
319 period of operation of the system with wastewater, the CER rate in tested in 20 mM  
320 NaCl in water stabilized at a near constant level (Figure S6).

321 Due to a consistent input of urine into the system, the  $[\text{Cl}^-]$  was found to variable at  
322 any point in time, in part, because the treated water that was recycled also allowed for  
323 a build-up of total chloride in the anaerobic holding tank. Variations in  $[\text{Cl}^-]$  were  
324 observed depending on the type and frequency of usage as well as the location of the  
325 unit (Table 2). In theory, when the system is running at full capacity after an initial set

326 up period, the steady-state  $[Cl^-]_{ss}$  should be approximately equal to the concentration  
327 of chloride in urine, which ranges between 53 mM to 240 mM (41). However, lower  
328 concentrations (typically 10 – 20 mM) were actually observed (Table 2). The lower  
329 concentrations were most likely due to the following factors: the treated water tank  
330 was periodically loaded with tap water at the beginning of the testing period until the  
331 wastewater in the sedimentation tank reached at least 40% of volume and also  
332 because of the use of excess non-recycled water for additional flushing or for personal  
333 hygiene in the public bathrooms.

334 For an electrical potential of 3.5 V between the anodes and cathodes, the measured  
335 anodic potential is  $1.4 \pm 0.2$  V vs. NHE and the cathodic potential is  $-2.1 \pm 0.2$  V vs.  
336 NHE. The cathodic potential is sufficient for the reduction of protons to hydrogen as it  
337 was previously described in the literature. This potential is sufficient for the  
338 production of surface-bound hydroxyl radicals on titanium dioxide at pH between 6  
339 and 9 (51, 52) but is not sufficient to generate free hydroxyl radicals ( $E_0(HO^\cdot) = 2.31$  V  
340 vs. NHE) in solution (53).

341 The detailed electrochemical surface reactions (Figure 5 (1) through (40)),  
342 previously identified and classified by Comninellis (18) consist of a two-step electron  
343 transfer for the oxidation of water on the surface of the metal-oxide electrode. In the  
344 case of  $TiO_2$ , a one-electron surface oxidation of water locally reduces titanium from  
345 Ti(IV) to Ti(III) with chemisorption of  $\cdot OH$  (40); then the surface metal hydroxy  
346 adduct undergoes deprotonation with the concomitant release of an electron (2) and  
347 the corresponding oxidation of Ti(III) back to Ti(IV) coupled with the formation of  $O_2$

348 or the direct surface oxidation of organic matter (3). Comninellis has also shown that  
349 the metal-hydroxyl bond ( $>\text{Ti(III)OH}\cdot$ ) can directly oxidize electron-donating organic  
350 matter, leading to subsequent mineralization or simply oxidize Ti(III) back to Ti(IV)  
351 with deprotonation and liberation of  $\text{O}_2$  (4). In the presence of chloride and a  
352 sufficient concentration, the surface-bound hydroxyl radical of  $>\text{Ti(III)OH}\cdot$  can directly  
353 oxidize  $\text{Cl}^-$  to form  $\text{Cl}_2$  and subsequently  $\text{HOCl}$  due to hydrolysis of molecular chlorine  
354 (40) (27). FC is defined as the sum of concentrations of hypochlorous acid,  $[\text{HOCl}]$ , and  
355 hypochlorite ion,  $[\text{ClO}^-]$ , which are in an acid-base equilibrium  
356 ( $\text{HOCl} \rightleftharpoons \text{H}^+ + \text{ClO}^-$ ,  $\text{pK}_a = 7.53$ ).

357 The measured CER rate in 20 mM NaCl solutions varied from  
358  $11 \pm 0.5 \text{ ppm Cl}_2 \text{ min}^{-1}$  before the electrodes had any contact with wastewater and  
359 decreased to  $7 \pm 0.7 \text{ ppm Cl}_2 \text{ min}^{-1}$  after approximately 50 hours of electrolysis of  
360 toilet wastewater (Figure S6). The observed decrease in the rate of the CER was most  
361 likely due to the formation of a layer of organic compounds on the surface of the  
362 anodes. The net effect was a reduction in the CER rate by almost 40%; however, after  
363 stabilization of the CER, the removal of organic matter was stable (*vide infra*).

#### 364 4.2. Removal of undesired organic and inorganic contaminants

365 The electrochemically produced FC (Figure 5 (6)) can oxidize ammonia to form  
366 chloramines (40) while also oxidizing organic matter (8) present in the wastewater  
367 (54). The ammonia present in the collected toilet wastewater was formed primarily  
368 from the hydrolysis of urea (9). Although bicarbonate formed from the hydrolysis of  
369 urea combined with that generated via the oxidation of organic matter could interfere

370 with the CER by adsorption on active anodic surfaces (40), thereby limiting the sites  
371 available for the oxidation of  $\text{Cl}^-$  to FC. The increase in TC concentration during an  
372 electrolysis batch was correlated with the removal of TKN because formation of  
373 chloramines (Figure 6). The TC concentration reached a steady state due to the  
374 reduction of chloramines to  $\text{N}_2$  in the solution as well as the reduction of  $\text{ClO}^-$  back to  
375  $\text{Cl}^-$  at the cathode surfaces (10). For instance, the ammonia monitored in the YXG  
376 prototype over the first 30 days of operation (Figure 7) showed 70% of  $\text{NH}_3$  removal  
377 efficiency once the system stabilized. The stabilization period was probably due to the  
378 fact that the system was started with significant amount of non-recycled water in the  
379 wastewater tank. This led to a high dilution of urine and feces for the first 15 days of  
380 operation.

381 The oxidation and mineralization of the organic matter (Figure 5 (4) and (8)) was  
382 observed through the decrease of COD during electrolysis (Figure 6). For instance, the  
383 COD monitored in the AMD prototype over 1000 hours of operation showed between  
384 70% and 80% removal efficiency when the chloride concentration was above 500  
385 ppm and the applied potential was 3.5 V (Figure S9 and Table 4). Furthermore, COD  
386 removal kinetics from prototype MGU (designated as KYM above) (Figure S7) were  
387 consistent with the first-order kinetic model expressed by Martinez-Huitle and Ferro  
388 (51) for transport-limited electrolytic oxidation with fitting coefficients reproduced  
389 Table S1.

### 390 4.3. Disinfection

391 A summary of disinfection analysis performed at the MGU (KYM) unit installed in  
392 Kottayam, India is reproduced Table 3: disinfection occurred after 2 to 3 hours of  
393 treatment (equivalent to 10 – 20 Wh L<sup>-1</sup> electrolysis energy), as indicated by the levels  
394 of the major indicator organisms being below the detection limit (*Fecal coliforms* and  
395 *E. coli*) or below drinking water safety standards (*Total coliforms*). These results are in  
396 accordance with the amount of chlorine and chloramines produced as well as the  
397 residence time in the ECR: 25 ppm TC assumed to be mostly chloramines because  
398 breakpoint chlorination was not reached; the equivalent contact time Ct value for 4  
399 hours operation was greater than 6,000 mg min L<sup>-1</sup>, which is more than 5 times higher  
400 than the recommended Ct value for 3-log inactivation of *Giardia* cysts at 20°C, and  
401 almost 10 times of recommended Ct value for 4-logs virus inactivation at 20°C (55).  
402 Similar results were observed in AMD and PAS units.

### 403 4.4. Energy consumption

404 The amount of electrical power drawn by the electrode arrays and by the overall  
405 system was measured on a regular basis (Figure S8). On average, 35 Wh were needed  
406 to treat 1 L of toilet wastewater, among which more than 95% of the electricity was  
407 used by the electrochemical treatment itself and the remaining 5% was used to  
408 compensate the power supply losses and to power the pumps. A large share of the  
409 energy used during electrolysis is for COD removal, especially when more than 200  
410 mg O<sub>2</sub> L<sup>-1</sup> removal is needed (Figure 8), and the electrolysis energy consumption is  
411 between 30 and 40 Wh L<sup>-1</sup>. Despite drastic changes in the input COD level and over the

412 course of close to 700 h of toilet wastewater electrolysis, the COD removal energy  
413 requirements remained relatively stable at approximately 10 Wh L<sup>-1</sup> for 100 mg O<sub>2</sub> L<sup>-1</sup>  
414 and up to 40 Wh L<sup>-1</sup> for 200 mg O<sub>2</sub> L<sup>-1</sup> initial COD.

#### 415 *4.5. Applicability of the technology in the context of a developing country*

416 The AMD prototype unit was connected to a public toilet produced by ERAM  
417 Scientific; the “eToilet” had remote monitoring capacity. All of the eToilet uses were  
418 recorded over the course of the testing period as well as the number of treatment  
419 cycles logged by the AMD unit (Figure S10). The treatment capacity of the unit was  
420 adequate for the number of users since there was no limitation in the number of  
421 eToilet uses from lack of treated water. Issues related to the engineering connections  
422 between the eToilet-AMD unit prevented use for more than 6 months. Mechanical and  
423 electrical issues detected by the maintenance engineer in residency on the AMD  
424 testing site were solved with remote or on-site assistance of the authors. The parts  
425 that were replaced during the testing period included pumps that failed for  
426 mechanical reasons and failures in the electrical energy storage subsystem  
427 (TRUECharge 2 40 A grid to 24 V converter and 12 V Blue Top lead-acid backup  
428 batteries, Figure S3). The mechanical failures of the pumps were due to fatigue and  
429 solids (sand) abrading the impeller and/or the diaphragms. The electrical failures of  
430 the energy storage subsystem were probably due to over-drainage events of the  
431 batteries when the system was used in the park but disconnected from the grid for  
432 very long periods (12 hours or more) and several grid electricity failures. These issues  
433 highlight the necessary trade-off between increasing the overall capital expenditure of

434 a system with components prone to less failure such as higher-grade pumps or  
435 sufficient solar panels to provide a backup source of power, and managing the  
436 operational expenditures due to frequent replacement of parts and grid electricity  
437 costs. These issues also highlight the necessity for frequent monitoring of the toilet  
438 wastewater treatment system in order to minimize the potential negative health  
439 impact on the users. A solution could be in the form of an automatic detection and  
440 maintenance system that could investigate the status of the treatment system via a  
441 suite of sensors and potentially self-repair or provide a step-by-step guide for repairs  
442 that necessitate the presence of a technician or a lesser qualified person.

#### 443 *4.6. Possible prototype improvements for commercialization*

444 The efficacy of the electrochemical treatment technology to clarify and disinfect  
445 toilet wastewater by generating chlorine without addition of water or chemicals  
446 makes this technology attractive as a non-sewered sanitation system, especially since  
447 it does not depend on the type of toilet used (*e.g.*, “western-style” flush toilet, squat  
448 pan) and does not require specific training or any change of behavior of the user.

449 Nevertheless, several improvements to the pre-alpha prototypes can be made to  
450 increase the robustness and energy efficiency of this electrochemical technology to  
451 meet the goals of the RTTC. A replacement of the sedimentation tank at the input of  
452 the process (Figure S1) by more advanced biological pre-treatment technologies such  
453 as small-size coupled aerobic/anaerobic systems (5) or microbial fuel cells (56) could  
454 effectively decrease the amount of undesired organic and inorganic contaminants  
455 entering the electrochemical reactor. This approach would drastically reduce the

456 operational expenses of the system by lowering the amount of electricity needed to  
457 complete the electrochemical treatment. Also, the biosolid residuals from the pre-  
458 treatment step as well as the filtered materials (Figure S1) should be properly  
459 decontaminated before being extracted from the system via a targeted  
460 decontamination process such as ohmic heating (57).

## 461 5. **Summary**

462 In response to the Bill and Melinda Gates Foundation challenge to “Reinvent the  
463 Toilet”, our research group at Caltech developed several self-contained, decentralized  
464 waste treatment systems that were designed to treat human domestic toilet waste at  
465 its source with discharge to the environment. After toilet flushing the discharged  
466 waste is stored in a wastewater tank. After some decantation, the effluent water from  
467 the wastewater tank is pumped into an electrochemical reactor array upon demand  
468 for the electrochemical oxidation of the residual organic and inorganic constituents.  
469 Disinfection is achieved via *in situ* chlorine generation resulting from anodic oxidation  
470 of chloride. Electrons released during anodic oxidation flow to the electronically  
471 coupled cathodes to produce molecular hydrogen via water reduction. The sequential  
472 biological and electrochemical treatment reduces the COD and microbial levels to  
473 below WHO agricultural reuse standards, while denitrification takes place due to  
474 breakpoint chlorination. In the field-level prototype systems, the treated black water  
475 is recycled into flush water reservoirs without significant discharge to the  
476 surrounding environment.



477 **6. Conflict of interest**

478 There are no conflicts of interest to declare.

479 **7. Acknowledgments**

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481 Ahmedabad, Gujarat), Mr. Arun Babu, and Mr. Shalu Achu (Mahatma Gandhi  
482 University, Kottayam, Kerala) for their help with the technical analyses in the  
483 respective field testing sites, as well as Ms. Heather Crammer (California Institute of  
484 Technology, Pasadena, California) for her help with the bacterial analyses and Mr.  
485 Asghar Aryanfar for the CAD rendering reproduced Figure S2 and Figure S4. This  
486 research was supported by the Bill and Melinda Gates Foundation under RTTC Grants  
487 OPP 1069500 and OPP 1111246.

488 **Table 1: Information about the different toilet wastewater treatment and**  
 489 **recycling units installed in the world.**

Configuration	Ref	Location	Testing period	Average daily usage during testing
<b>Self-contained bathroom + wastewater treatment and recycling unit in a shipping container</b>	PAS	Pasadena, USA	06/2013 to 06/2016	<5
	KYM	Kottayam, India	04/2014 to 01/2016	6
	YXG	Yixing, China	12/2014 to 05/2015	35
<b>Wastewater treatment and recycling unit connected to an “eToilet” public toilet (Eram Scientific, Trivandrum, Kerala, India)</b>	AMD	Ahmedabad, India	04/2014 to 01/2016	7

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492 **Table 2: Average toilet wastewater composition in the different prototypes.**

493 **Ranges are given when available.**

Parameter	Unit	Prototype reference			
		PAS <sup>1</sup>	AMD <sup>2</sup>	KYM <sup>3</sup>	YXG <sup>3</sup>
<b>COD</b>	mg O <sub>2</sub> L <sup>-1</sup>	150-250	100	335	550
<b>Cl<sup>-</sup></b>	mmol L <sup>-1</sup>	11-20	11	15	24
<b>NH<sub>3</sub> + NH<sub>4</sub><sup>+</sup></b>	mg NH <sub>3</sub> L <sup>-1</sup>	80	30-40	235	480
<b>PO<sub>4</sub><sup>3-</sup> + HPO<sub>4</sub><sup>2-</sup></b>	mmol L <sup>-1</sup>	0.64	-	-	-
<b>Alkalinity as CaCO<sub>3</sub></b>	mmol L <sup>-1</sup>	17	-	10.7	27
<b>pH</b>	-	8.3	7.4	7.5	8.5

<sup>1</sup> after 16 months of collection and 6 months of recycling water,

<sup>2</sup> no recycled water used

<sup>3</sup> after 2 months of running

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499 **Table 3: Indicator organisms *Total coliform*, *Fecal coliform*, and *E. coli* detection**  
 500 **test results during electrochemical treatment cycles. Analysis performed by the**  
 501 **Topical Institute of Ecological Sciences of Mahatmah Gandhi University**  
 502 **(Kottayam, Kerala, India) and Albio Technologies (Kochi, Kerala, India).**

Reaction time (energy consumed)	<i>Total coliforms</i> MPN/100ml	<i>Fecal coliforms</i> MPN/100ml	<i>E. coli</i> CFU/ml
<b>11/17/14</b>			
<b>0 h (0 Wh L<sup>-1</sup>)</b>	>1100	>1100	200
<b>2 h (11 Wh L<sup>-1</sup>)</b>	<1	<1	<1
<b>4 h (22 Wh L<sup>-1</sup>)</b>	<1	<1	<1
<b>3/28/15</b>			
<b>0 h (0 Wh L<sup>-1</sup>)</b>	>2400	75	Present
<b>1 h (4.1 Wh L<sup>-1</sup>)</b>	1100	0	Absent
<b>2 h (8.2 Wh L<sup>-1</sup>)</b>	23	0	Absent
<b>3 h (12 Wh L<sup>-1</sup>)</b>	15	0	Absent
<b>7/25/15</b>			
<b>0 h (0 Wh L<sup>-1</sup>)</b>	>2400	120	Present
<b>1 h (6.7 Wh L<sup>-1</sup>)</b>	1100	75	Present
<b>2 h (13 Wh L<sup>-1</sup>)</b>	93	4	Present
<b>3 h (20 Wh L<sup>-1</sup>)</b>	43	3	Present
<b>4 h (27 Wh L<sup>-1</sup>)</b>	9	0	Absent
<b>9/18/15</b>			
<b>0 h (0 Wh L<sup>-1</sup>)</b>	75	0	Absent
<b>2 h (12 Wh L<sup>-1</sup>)</b>	23	0	Absent
<b>4 h (24 Wh L<sup>-1</sup>)</b>	9	0	Absent

503

504 **Table 4: Typical wastewater quality parameters measured before and after a 3-**  
 505 **hour electrolysis cycle over the course of the field testing of the AMD prototype.**  
 506 **Values are average of three replicates.**

Date (dd/mm/yyyy)	Unfiltered COD (mg O <sub>2</sub> /L)		TKN or NH <sub>3</sub> (mg N/L)		TSS (mg/L)		Chloride (mg Cl <sup>-</sup> /L)		Total/Free Chlorine (mg Cl <sub>2</sub> /L)	
	Before	After	Before	After	Before	After	Before	After	Before	After
18/09/2014	32	-	n.d.	-	-	-	-	-	-	-
14/10/2014	100	-	31	-	100	-	-	-	-	-
03/11/2014	90	n.d.	-	-	100	50	182	35.1	-	-
29/11/2014 <sup>a</sup>	43	11	15	2	-	-	100	30	-	-
23/02/2015	100	48	-	-	-	-	-	-	-	-
27/08/2015	-	-	-	-	-	-	-	-	0 <sup>b</sup>	39 <sup>b</sup>
01/12/2015	240	40	43	5	-	-	-	-	-	-
16/01/2016	223	26	-	-	-	-	235	203	<1 <sup>c</sup>	-
02/02/2016	371	95	-	-	-	-	886	382	<1 <sup>c</sup>	2.73 <sup>c</sup>
31/03/2016	320	50	1.05 <sup>d</sup>	2.04 <sup>d</sup>	245	87	-	-	-	-
11/04/2016	234	56	25 <sup>d</sup>	40 <sup>d</sup>	180	35	425	390	<1 <sup>c</sup>	<1 <sup>c</sup>

507

508 <sup>a</sup> Analyses performed by third-party (Ahmedabad Municipal Corporation Central Lab). Total  
 509 Nitrogen was measured.

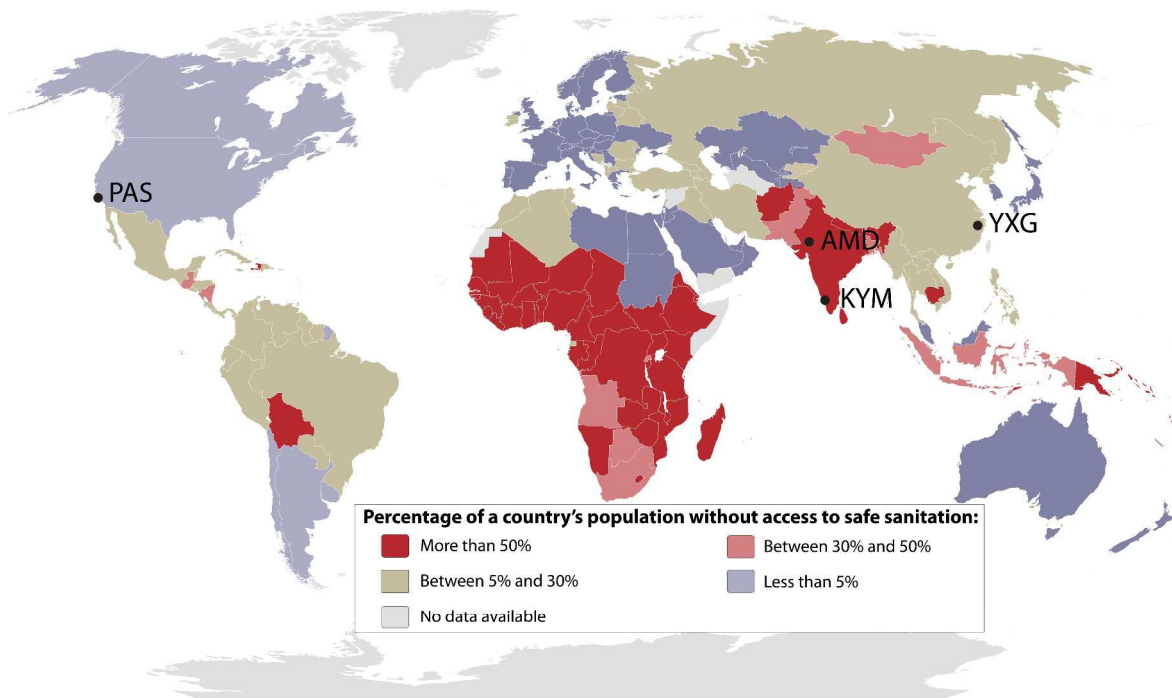
510 <sup>b</sup> Total Chlorine

511 <sup>c</sup> Free Chlorine

512 <sup>d</sup> Potential Interference for Ammonia measurement

513

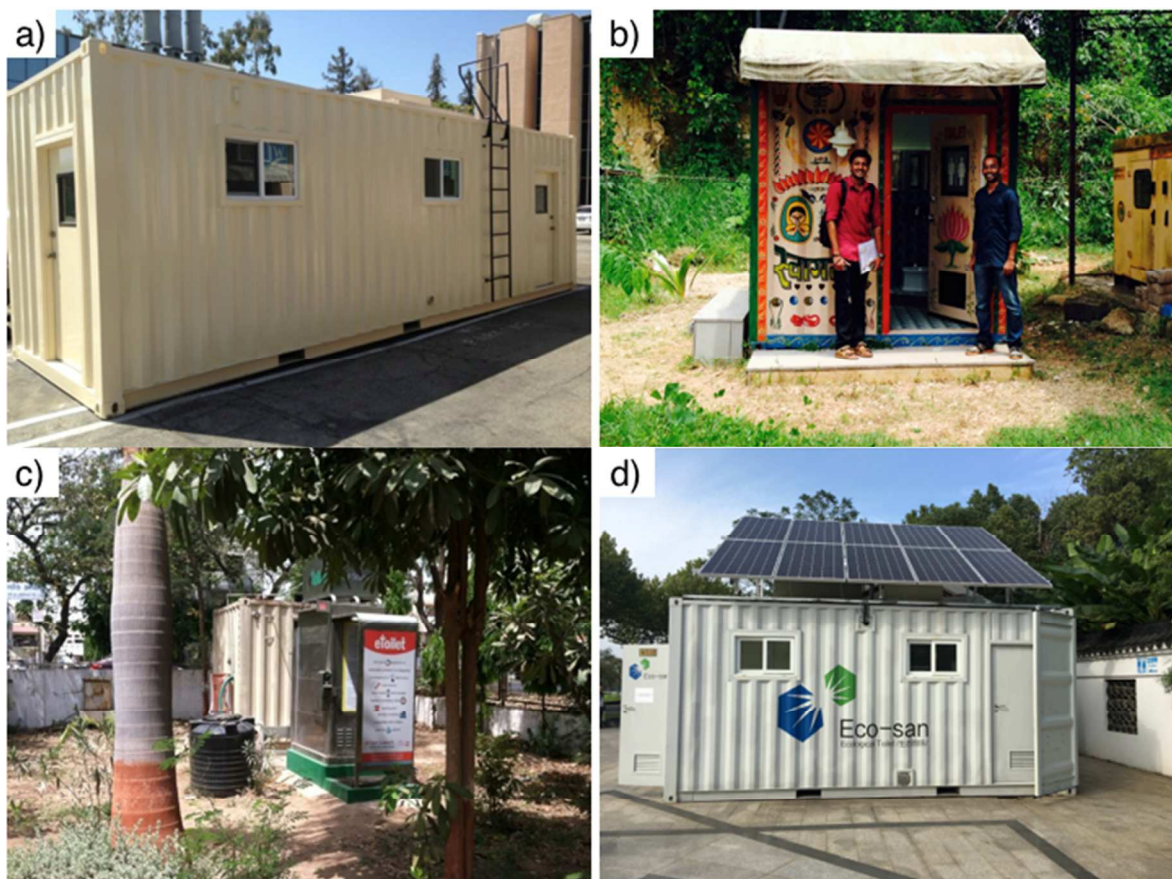
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516 **Figure 1: Percentage of a country's population without access to safe sanitation**  
517 **in 2015 according to the World Health Organization (58). Location of the four**  
518 **prototype testing sites across the world: PAS, Pasadena, California, USA; AMD,**  
519 **Ahmedabad, Gujarat, India; KYM, Kottayam, Kerala, India; YXG, Yixing, Jiangsu,**  
520 **China.**

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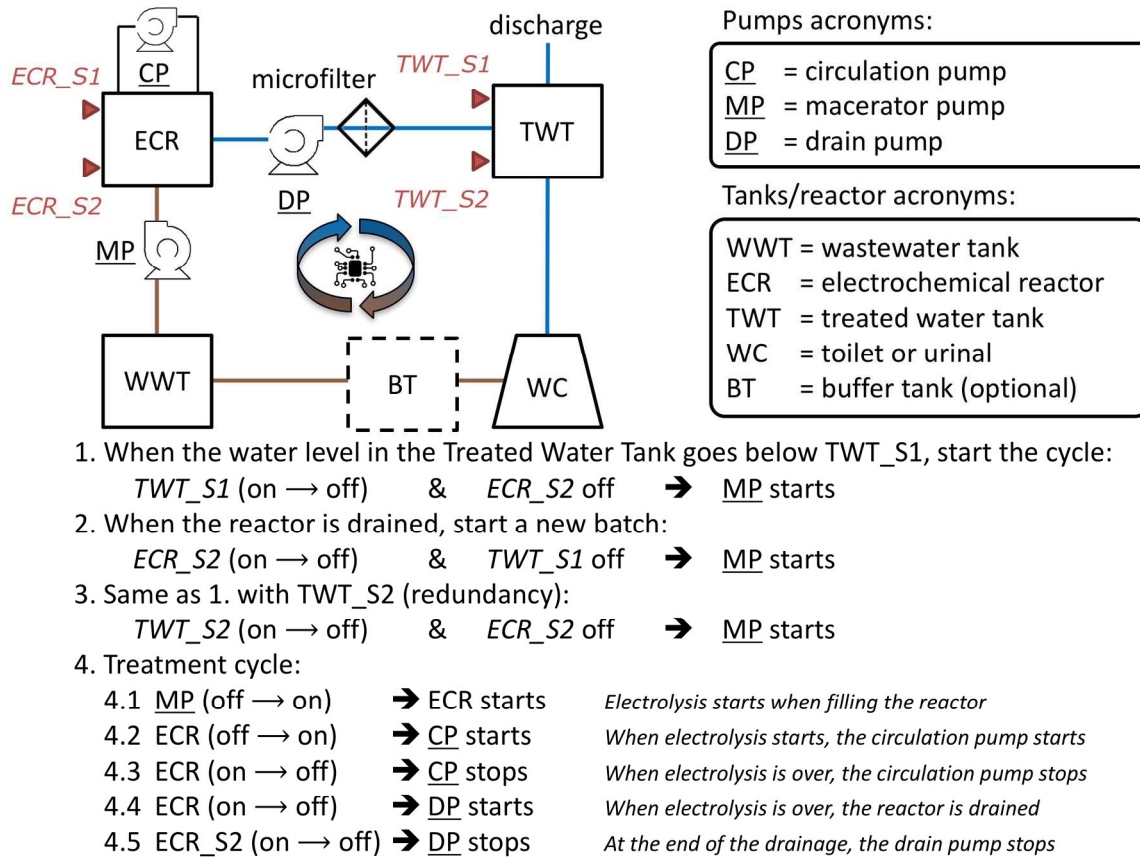


522

523 **Figure 2: Caltech Solar Toilet system prototypes: a) Prototype PAS (Pasadena,**  
524 **CA); b) Prototype KYM (Kottayam, Kerala, India); c) Prototype AMD**  
525 **(Ahmedabad, Gujarat, India); d) Prototype YXG (Yixing, Jiangsu, China).**

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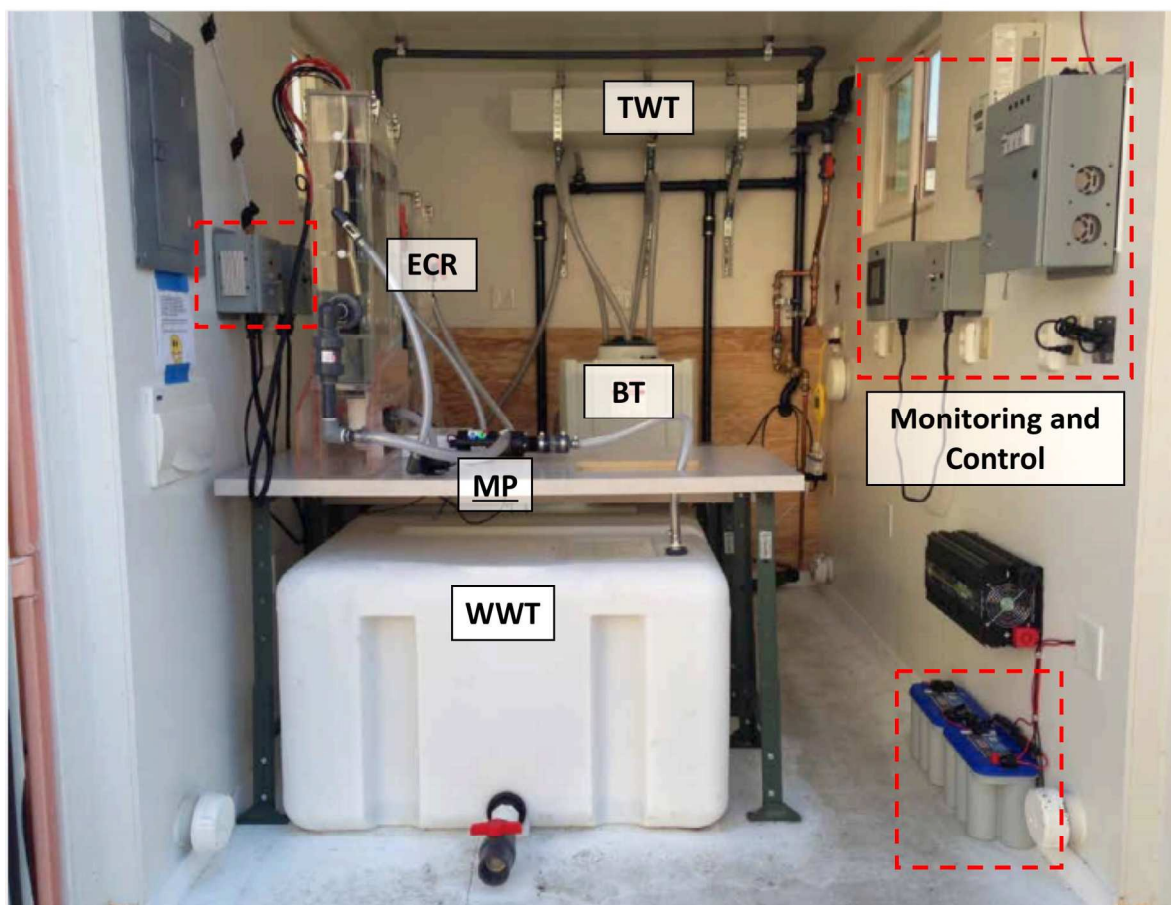


528

529 **Figure 3: System flow diagram (top left, see Figure S1 for volumes and residence**  
 530 **times) with automation algorithm description for the onsite toilet wastewater**  
 531 **treatment and recycling systems. Pumps are underlined. Capacitive level**  
 532 **sensors are represented by red triangles. Brown lines illustrate the flow of**  
 533 **untreated wastewater while blue lines illustrate the flow of treated and**  
 534 **recycled wastewater.**

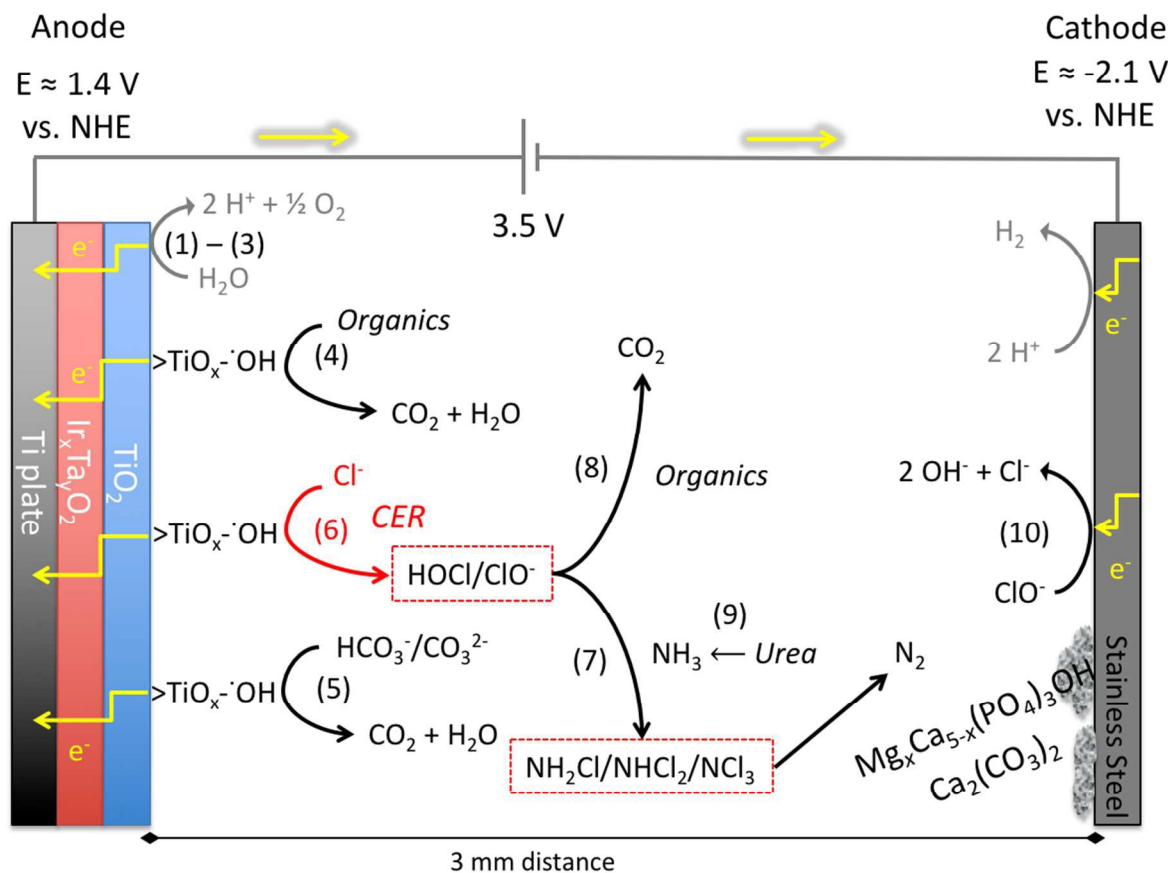
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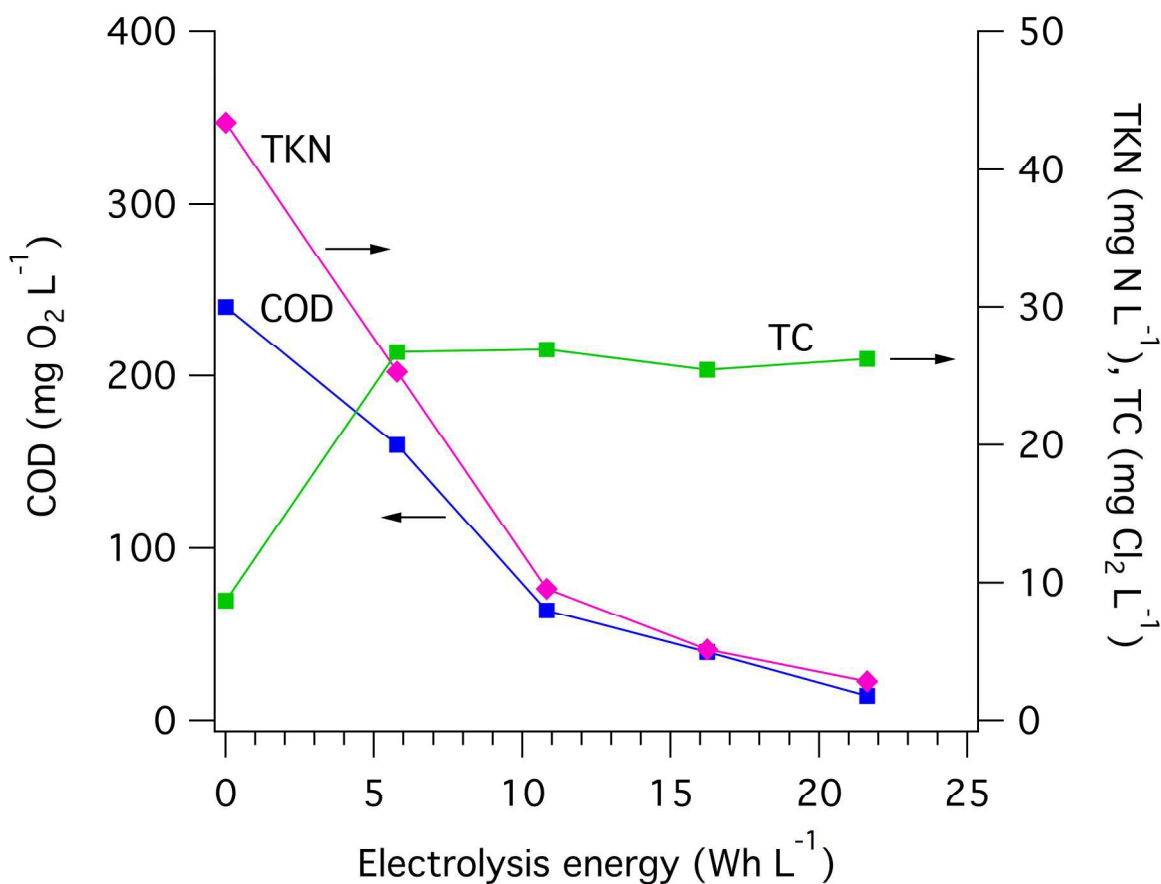
536

537 **Figure 4: Photograph of the layout of one of the self-contained electrochemical**  
538 **treatment prototypes installed in the field. The combined power, monitoring,**  
539 **and control system is highlighted in red dashes. Refer to Figure 3 for definition**  
540 **of the acronyms.**



541

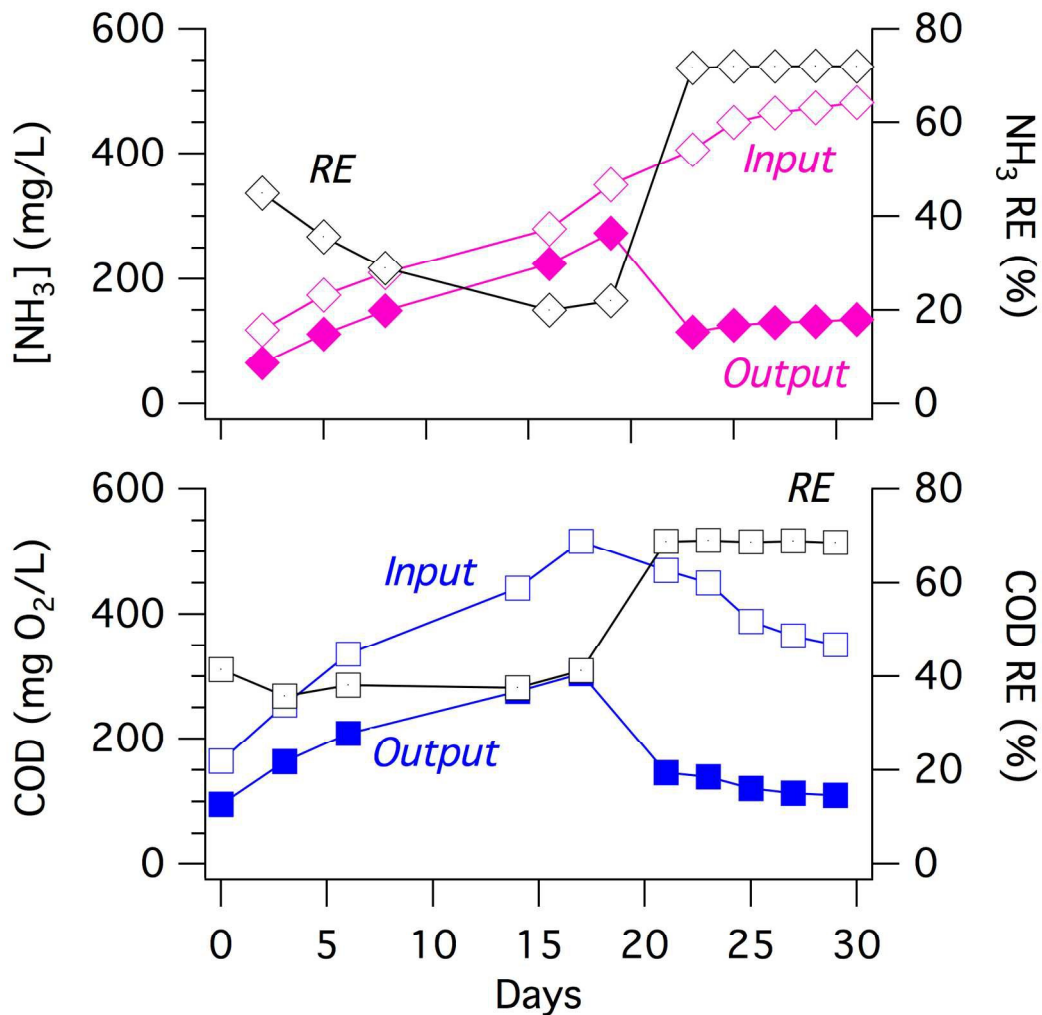
542 **Figure 5: Electrons flow and main chemical reactions in the electrochemical**  
 543 **reactor. (1) - (3) illustrate the oxidation of water on the TiO<sub>2</sub>-semiconductor**  
 544 **oxide surface. (4) represents the direct oxidation of bicarbonates to CO<sub>2</sub>. (4)**  
 545 **and (8) illustrate the removal of organics by direct (4) or indirect (8) oxidation.**  
 546 **(4), (6), (8), and (10) illustrate the production and the fate of free chlorine**  
 547 **(HOCl/ClO<sup>-</sup>) during electrochemical treatment. The yellow arrows represent the**  
 548 **flow of electrons in the electrodes and across the wires.**



549

550 **Figure 6: Evolution of the COD, TKN, and TC during the treatment of toilet**  
551 **wastewater in a single electrochemical treatment cycle in AMD prototype on**  
552 **01/12/2015. Each point represent the average of three triplicate**  
553 **measurements.**

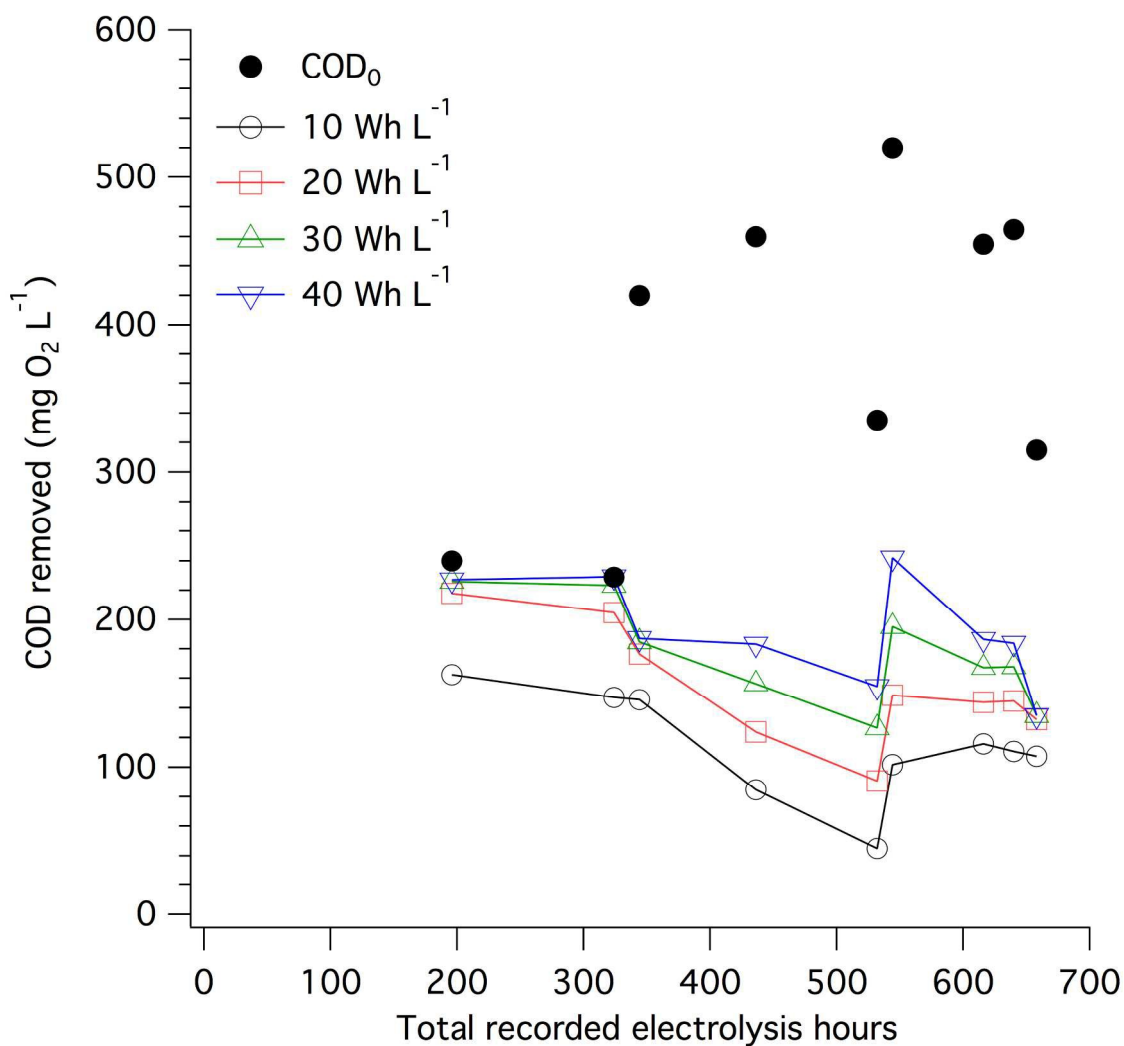
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556 **Figure 7:  $\text{NH}_3$  (top) and COD (bottom) averaged concentrations before (input)**  
 557 **and after (output) an electrochemical treatment cycle of 4 hours with respective**  
 558 **Removal Efficiencies (RE) for 30 continuous days of operation of YXG prototype.**  
 559 **Day 0 corresponds to the beginning of usage of the prototype.**

560



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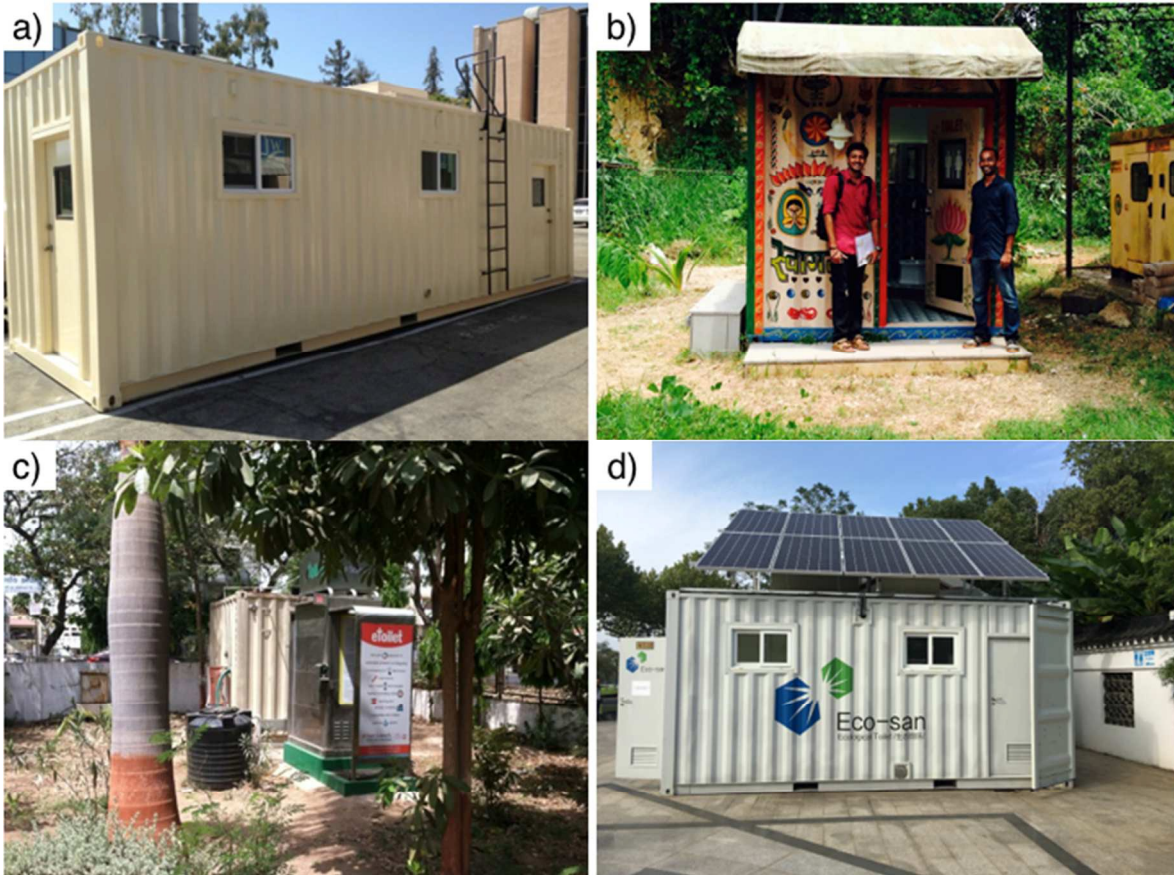
562 **Figure 8: Extrapolated COD removal from toilet wastewater at different levels of**  
563 **electrical energy consumption (10, 20, 30, and 40 Wh L<sup>-1</sup>) and over the course of**  
564 **the field testing of the AMD unit. The extrapolation calculation is based on a**  
565 **first-order kinetic model for electrochemical COD removal developed by**  
566 **Martinez-Huitle and Ferro (51), see Figure S7 and Table S1 for curve fittings and**  
567 **calculations. The black dots indicate the COD<sub>0</sub> value of the wastewater before**  
568 **electrochemical treatment.**

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570

571 **Graphical abstract**

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