Supporting Information S5 Data for:

Integration of silicate minerals for ammonium and phosphate removal with an on-site wastewater treatment prototype.

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Authors’ Note: Some language in the following report has been edited lightly to maintain consistent nomenclature with the main manuscript text. These edits do not affect the results or substantive content of the report.
Field testing of the Nutrient Removal Module (NRM) integrated with the Duke Center for WaSH-AID on-site wastewater treatment system in Coimbatore

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Introduction
The Duke Center for WaSH-AID on-site wastewater treatment system (OWTS) prototype was installed and tested at a privately-owned textile mill in Coimbatore, India, beginning in March 2019. Results indicate good removal of TSS and COD and disinfection, however the system had modest nutrient removal, specifically only 10% removal for total N. On February 27th 2020, a Nutrient Removal Module (NRM) was integrated in the system, that operated by flowing blackwater through zeolites (clinoptilolite), which capture ammonia via ion exchange. A 12 L tank was filled with zeolite (clinoptilolite) shipped from Duke University. The module was operated for 11 cycles in 9 days in March 2020 prior to COVID-19 lockdown. This report describes total N, ammonia, turbidity and other data collected during this initial study.

Materials and methods

Test site and system description
The OWTS prototype is installed in a shared women’s toilet facility in Coimbatore in early March 2019 with approximately 20 to 50 users per day. The toilet effluent is processed through custom-designed components for solid-liquid separation and settling of the liquid waste. The solid waste after separation is collected in an accumulation tank. The effluent of the settling tanks feeds into the OWTS prototype.

The OWTS prototype was tested until the end of February 2020, without the nutrient removal module (NRM), as shown in the left panel of Figure 1. It comprises three sub-systems operating in series: ultrafiltration (UF) membrane, a granular activated carbon column (GAC), and electrochemical oxidation (EC) to achieve disinfection.

The NRM was added to the system and tested in March 2020 for only 9 days. The feed tank liquid is pumped through the UF column, the effluent from the UF flows through a GAC packed column in an up-flow direction. The effluent from the GAC column is gravity fed to the NRM in an up-flow configuration (Figure 1, right).

The NRM is a 12L tank filled with ~8.4Kg of clinoptilolite (zeolite) for ammonia removal. The zeolites were washed thoroughly with tap water before being installed in the nutrient removal tank. Zeolites remove ammonium ions by means of ion-exchange and, at higher concentration, adsorption. The ammonium ions present in the wastewater are exchanged for sodium ions.

The effluent from the NRM is gravity fed to the electrochemical disinfection chamber where electrochemical oxidation of blackwater takes place in batches of 12L.

A single treatment cycle of the system comprises two phases: a UF, GAC and nutrient removal combined phase, and an electrochemical oxidation phase. The system process was controlled by a programmable logic controller (PLC) connected to capacitive level sensors attached to the outside of the tanks to detect
the water level. The process logic begins a treatment cycle whenever the level sensors detect water in the blackwater feed tank.

![Schematic of the OWTS prototype before (left) and after (right) installation of the nutrient removal module.](image)

**Figure 1.** Schematic of the OWTS prototype before (left) and after (right) installation of the nutrient removal module.

**Analytical measurements**

Influent samples were collected from a 200 L pre-feed tank at the OWTS prototype inlet and effluent samples from the effluent liquid line so that an individual batch treatment would be analyzed rather than a composite of multiple effluent batches. A sterile disposable serological pipette was used to collect samples which were then stored in sterile polypropylene centrifuge tubes.
Conductivity, pH and ORP were measured using a Myron L 6PFCE Ultrameter II. Turbidity was measured using the HACH 2100Q IS Portable Turbidimeter. Color was measured with a HACH CO-1 test kit.

Water chemistry analyses were performed using the appropriate HACH reagent kits and methods (total N, HACH method 10072; and NH3, HACH method 10031), and results were measured with a HACH DR 900 colorimeter. A HACH DRB200 reactor was used for methods requiring digestion (total N). Ultrapure water was used for samples requiring dilution prior to analysis.

One measurement was conducted on total suspended solids (TSS) according to EPA method 160.2.

**Results and discussion**

**Operational**

The NRM was added, and then the system was operated for 11 cycles, for 33 hours, treating 132 L of wastewater. The field testing of the zeolite had to be halted due to COVID-19. Prior to NRM installation, the system was operated for nearly 350 cycles for collectively 581 hours of operation treating 4708 L of blackwater.

The system was set up for 12 L batch and the operational run time was around 3 hours and 10 minutes (160 minutes for UF and 30 minutes for EC).

<table>
<thead>
<tr>
<th>Table 1. Operational Results</th>
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<tbody>
<tr>
<td>OWTS prototype without NRM</td>
</tr>
<tr>
<td>Days of Operation</td>
</tr>
<tr>
<td>Start date</td>
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<tr>
<td>End date</td>
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**Figure 4.** Graph showing UF runtime versus number of treatment cycles.
**Water quality**

We examine the effect of the NRM module on the OWTS prototype effluent on selected individual effluent parameters with plots as function of the trial run and provide a summary table of average values at the end of this section.

**Ammonia and total N removal**
The influent prior to NRM installation featured an average total N of $167 \pm 24$ mg/L, comprising over 90% of ammonia $156 \pm 24$ mg/L. The system provides in average 10% total N removal (Table 2) in agreement with the fact that the UF removes the organic fraction of N, but NH$_3$ remains in the effluent. With the NRM installation, the ammonia and total N effluent was measured as zero for the first 7 cycles (after an initial value attributed to washover from previous run). The ammonia value exhibited increasing values of 1, 7, 13 mg/L starting at cycle 9.

![Figure 5: Influent and effluent ammonia before and after the NRM upgrade.](image)

![Figure 6: Influent and effluent total N (TN) before and after the NRM upgrade.](image)
**pH and turbidity**

We observed a consistent pH increase from 8 to 8.5 with zeolite addition (Figure 7).

![Figure 7: Influent and effluent pH before and after the NRM upgrade](image)

Prior to NRM installation, the average turbidity removal was approximately 48%. In the first 4 cycles after installation of NRM, relatively high turbidity was observed (Figure 8), despite the zeolite being washed thoroughly with tap water before installation. This effect possibility due release of dust from zeolite appears to be going away in the last cycles that were measured with turbidity at or below 10 NTU. TSS was measured once at cycle 7 after zeolite installation and was found to be a modest 15 mg/L.

![Figure 8: Influent and effluent turbidity before and after the NRM upgrade](image)
Chlorine

Chlorine production was null for many cycles before the NRM installation. We had exhausted our attempts at troubleshooting the issue with chlorination and we just ran the system to evaluate the UF performance (Figure 9 and Figure 10). It is remarkable that immediately after installation of zeolites, and with no changes to the electrochemical cell, the system began to produce chlorine (total chlorine of 10 ± 8 mg/L, free chlorine of 6 ± 7 mg/L.)

Figure 9: Effluent free chlorine before and after the NRM upgrade

Figure 10: Effluent total chlorine before and after the NRM upgrade
**Electrical conductivity**

The electrical conductivity of the influent varies between 3600 and 4600 mS/cm and historically contained adequate chloride (average 490 mg/L) for electrochemical oxidation with no need for chloride additives. The conductivity of the system effluent is reduced (Figure 11). It is hard to establish whether NRM installation had an impact on EC. Similarly, ORP and color were measured but no specific effects due to NRM installation were observed.

![Figure 11: Influent and Effluent electrical conductivity before and after the NRM upgrade](image)
Table 2 reports influent and effluent quality parameters averaged over the entire testing period, for both with and without the nutrient removal module, and compares the results to ISO 30500 standards.

Table 2. Summary of influent and effluent water quality for before and after the addition of the NRM. ‘n’ is the number of samples measured for the parameter. Data are mean ± standard deviation (range). Removals are calculated as the percent reduction of the average influent and effluent values.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Before nutrient removal module</th>
<th>After nutrient removal module</th>
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<tbody>
<tr>
<td></td>
<td>Influent</td>
<td>Effluent</td>
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<tr>
<td>pH</td>
<td>7.8 ± 0.2 (6.9 - 8.3)</td>
<td>8.0 ± 0.2 (7.1 - 8.5)</td>
</tr>
<tr>
<td>TSS (mg/L)</td>
<td>22.7 ± 12.2 (10.4 – 60)</td>
<td>10.7 ± 9.9 (0 - 40)</td>
</tr>
<tr>
<td>COD (mg/L)</td>
<td>269 ± 130 (127- 615)</td>
<td>105 ± 38 (61 - 195)</td>
</tr>
<tr>
<td>total N (mg/L)</td>
<td>144 ± 28 (99 – 191)</td>
<td>129 ± 27 (96, 176)</td>
</tr>
<tr>
<td>total P (mg/L of PO4³⁻)</td>
<td>45 ± 4.9 (40-51)</td>
<td>21 ± 2.2 (18-24)</td>
</tr>
<tr>
<td>NH3 (mg/L)</td>
<td>139 ± 46 (60-270)</td>
<td>129 ± 58 (42-380)</td>
</tr>
<tr>
<td>E. coli (CFU/L)</td>
<td>2.13 e6 ± 2.88 e6</td>
<td>&lt; 1000³⁵</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>48.7 ± 44.3 (1.4 – 283)</td>
<td>23.3 ± 24.2 (1.2 – 136)</td>
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³ Limit of detection for this assay was 1/mL or 1000/L; all measurements were below the detection limit.

Conclusion

Despite the limited testing period, the NRM demonstrated from the very first run of the system effective ammonia and total N removal. The NRM had a negative impact of turbidity, that may have been transient, and induced a slight increase in pH. Remarkably the NRM brought back chlorine production from a system where we had exhausted our attempt at troubleshooting the issues with chlorination. The ammonia removal effect appeared to begin to lose efficiency around trial 10, but the testing period was interrupted by COVID-19 lockdown.