Reduction of nutrients, microbes, and personal care products in domestic wastewater by a benchtop electrocoagulation unit

E. M. Symonds¹, M. M. Cook¹, S. M. McQuaig², R. M. Ulrich¹, R. O. Schenck¹, J. O. Lukasik³, E. S. Van Vleet¹ & M. Breitbart¹

¹University of South Florida, College of Marine Science, 140 7th Avenue South, St. Petersburg, Florida, USA, ²St. Petersburg College, 2465 Drew Street, Clearwater, Florida, USA, ³BCS Laboratories, Inc., 4609-A NW 6th Street, Gainesville, Florida, USA.

To preserve environmental and human health, improved treatment processes are needed to reduce nutrients, microbes, and emerging chemical contaminants from domestic wastewater prior to discharge into the environment. Electrocoagulation (EC) treatment is increasingly used to treat industrial wastewater; however, this technology has not yet been thoroughly assessed for its potential to reduce concentrations of nutrients, a variety of microbial surrogates, and personal care products found in domestic wastewater. This investigation’s objective was to determine the efficiency of a benchtop EC unit with aluminum sacrificial electrodes to reduce concentrations of the aforementioned biological and chemical pollutants from raw and tertiary-treated domestic wastewater. EC treatment resulted in significant reductions (p < 0.05, α = 0.05) in phosphate, all microbial surrogates, and several personal care products from raw and tertiary-treated domestic wastewater. When wastewater was augmented with microbial surrogates representing bacterial, viral, and protozoan pathogens to measure the extent of reduction, EC treatment resulted in up to 7-log₁₀ reduction of microbial surrogates. Future pilot and full-scale investigations are needed to optimize EC treatment for the following: reducing nitrogen species, personal care products, and energy consumption; elucidating the mechanisms behind microbial reductions; and performing life cycle analyses to determine the appropriateness of implementation.

In order to protect public and environmental health, innovative technologies are needed to reduce the concentrations of emerging microbes¹ and chemicals² from domestic wastewater prior to discharge into the environment and/or water reuse. Fecal-borne pathogens, encompassing known and emerging bacteria, helminths, protozoa, and viruses, substantially contribute to human disease and mortality worldwide¹,³. Furthermore, it has been postulated that the input of personal care products (PCPs; a chemically diverse group of over-the-counter medications, insect repellents, antibiotics, and disinfectants) into aquatic environments or the drinking water supply could negatively affect wildlife and humans, respectively⁴,⁵. Finally, it is well-understood that the removal of nutrients, principally nitrogen and phosphorus, from domestic wastewater is necessary to prevent the eutrophication of surface waters exposed to treated wastewater discharge. While many different wastewater treatment options exist, adequate reduction of all chemicals and microbes is extremely complex due to their great physical and structural diversity⁶,⁷. It is therefore important to evaluate treatment technologies for their ability to remove a diverse range of contaminants, since a combination of approaches will likely be required to ensure safe discharge of treated effluent and/or water reuse.

Electrocoagulation (EC) has become increasingly popular over the last 25 years to treat a wide-variety of wastewaters as technological advances have made this technique more cost- and energy-efficient⁷–¹⁰. The EC process applies electricity to sacrificial electrodes (typically aluminum or iron), which generates coagulants (e.g. aluminum hydroxide for an aluminum anode), destabilizes contaminants, enhances the suspension of particulates, and disrupts emulsions. Contaminants are either directly broken down or aggregated to form flocs that become buoyant as they associate with the gases generated by the concurrent electrolysis of water. Following EC, the floc is separated from the treated water via sedimentation and/or filtration. EC may be an advantageous treatment option as it does not require a constant supply of chemicals⁷–¹⁰ and consequently, may be more easily
implemented in a developing-country context where such chemicals are not readily available. It has also been suggested that EC technology could be an effective decentralized drinking water treatment technology and easily deployed as portable equipment for use in remote locations or in the event of emergencies.

The efficacy of EC to reduce various biological and chemical constituents found in water and wastewater under normal and emergency conditions has been investigated in several prior studies. When evaluating EC technologies for their use in treating potable water, Vik et al. determined significant removal of humic substances with EC treatment of surface waters and Zhu et al. ascertained effective removal of MS2 bacteriophages from synthetic freshwater. In another study, even though the use of iron electrodes in EC treatment reduced MS2 bacteriophage by up to 6.5-log10 in synthetic freshwater, natural levels of organic matter present in surface waters limited virus reductions to as little as 1.0-log10. Consequently, the use of aluminum electrodes was suggested to prevent the complexation of organic matter and iron ions that inhibit adequate flocculation and subsequent virus removal. Furthermore, EC treatment of surface waters is both technically and economically effective for the removal of algae and greatly reduces concentrations of fecal indicator bacteria. In a recent laboratory study, EC decreased concentrations of the antibiotic tetracycline by nearly 99% in laboratory-made aqueous solutions. With respect to the treatment of industrial wastewater, EC has also been extensively used, primarily with aluminum, iron, and steel electrodes, to reduce chemical oxygen demand (COD) as well as COD and turbidity. The incorporation of EC as a tertiary or polishing treatment has also been suggested as it can greatly reduce concentrations of fecal indicator bacteria. EC has also been extensively used, primarily with aluminum, iron, and steel electrodes, to reduce chemical oxygen demand (COD) as well as COD and turbidity. The incorporation of EC as a tertiary or polishing treatment has also been suggested as it can greatly reduce concentrations of fecal indicator bacteria. EC has also been extensively used, primarily with aluminum, iron, and steel electrodes, to reduce chemical oxygen demand (COD) as well as COD and turbidity. The incorporation of EC as a tertiary or polishing treatment has also been suggested as it can greatly reduce concentrations of fecal indicator bacteria.

Results and discussion

Nutrients. Significant (>95%; p < 0.0003, α = 0.05) reductions in phosphate were observed upon EC treatment of both raw wastewater and tertiary treated wastewater (Table 1). These results corroborate previous findings demonstrating up to 100% removal using aluminum sacrificial electrodes and further suggest that EC may be an especially useful treatment technology to achieve enhanced phosphorus reductions from domestic wastewater. Despite the consistent reduction of phosphate by EC, the extent of reduction for the other nutrients differed for raw wastewater compared to tertiary-treated wastewater (Table 1).

Significant decreases in nitrate + nitrite were observed (48.35%; p = 0.0007, α = 0.05) during the treatment of tertiary-treated wastewater; however, no significant reduction in nitrate + nitrite was achieved during the treatment of raw wastewater. Additionally, even though significant increases in nitrite and ammonia were observed during EC treatment of tertiary-treated wastewater, significant reductions (>14%; p < 0.0007; α = 0.05) were observed after EC treatment of raw wastewater. Previous studies on nitrate reduction from ground and surface water for potable water treatment have shown that EC with iron and aluminum blades is more efficient than chemical coagulation; however, the extent of nitrate reduction depended upon the EC conditions (e.g. current density applied, electrode connections) and the characteristics of the water under treatment (e.g. pH, initial nitrate concentration, total dissolved solids). Since up to 89.7% nitrate removal from aqueous solutions has been observed by Malakootian et al., future research is necessary to identify the optimum EC conditions for reduction of various nitrogen species from domestic wastewater and treated effluent.

Microbes. Six commonly used microbial surrogates were analysed using a combination of molecular- and culture-based techniques. The double-stranded DNA human polyomavirus (HPyV) and single-stranded RNA pepper mild mottle virus (PMMoV) were measured as surrogates for DNA and RNA viruses in wastewater, respectively.
using molecular techniques. Fecal-indicator bacteria (FIB; fecal coliforms and Enterococcus spp.) were measured as surrogates for wastewater-related bacteria using culture-based techniques as well as molecular techniques for Enterococcus spp. To quantify the extent of microbial reduction, the EC unit was used to treat domestic wastewater augmented with the aforementioned bacteria and viruses as well as two other commonly used microbial surrogates that are not typically found in wastewater at high concentrations: male-specific (F+) bacteriophages (MS2) and Bacillus subtilis spores (surrogate for wastewater-related, protozoan parasites; i.e Cryptosporidium). Both MS2 bacteriophages and B. subtilis spores were analysed using culture-based techniques.

EC treatment resulted in significant reductions ($p < 0.0286$, $\alpha = 0.05$), ranging from 81.567% to 99.999998%, of all microbial surrogates tested in all domestic wastewater samples (Table 2). These results suggest that EC with aluminum electrodes is an effective treatment for the wide-range of pathogen types present in domestic wastewater. Furthermore, EC treatment resulted in a greater than 4-log$_{10}$ reduction for all microbial surrogates in augmented domestic wastewater. Although this study does not attempt to discern the mechanisms behind “the observed reductions after” EC treatment, previous studies on synthetic freshwater and wastewater have suggested that the primary microbial removal mechanism during EC is due to the emmeshment of microbes to flocs and subsequent separation of flocs from treated water by filtration. It is also possible that the oxidants produced during EC (e.g. HO) oxidize O$_3$, H$_2$O$_2$) provide additional microbial reductions via disinfection as a result of cell capsid membrane damage. The effective reduction of FIB observed (as great as 7-log$_{10}$) in this study supports the results of previous investigations on EC treatment of domestic wastewater, which cite reductions as high as 4-log$_{10}$. Finally, this is the first study to our knowledge to demonstrate that EC can significantly reduce concentrations of viral and parasitic protozoan surrogates in domestic wastewater.

**Personal care products.** EC treatment of raw domestic wastewater significantly ($p < 0.05$, $\alpha = 0.05$) reduced concentrations of the following PCPs: acetaminophen, DEET, gemfibrozil, ibuprofen, iodopropynol, salicylic acid, triclocarban, and triclosan (Table 3). While the initial concentrations of many PCPs in tertiary-treated wastewater were below the process limit of detection (pLOD), EC treatment of tertiary-treated wastewater significantly ($p < 0.05$, $\alpha = 0.05$) decreased concentrations of iodopropynol, sulfamethoxazole, and thibendazole (Table 3). Although this study does not attempt to discern the EC removal mechanisms associated with the different PCPs, it is likely that PCP adsorption to flocs was a major removal mechanism, particularly for compounds with higher octanol-water partition coefficient ($K_{ow}$) values (e.g. gemfibrozil, ibuprofen, triclocarban, and triclosan). It is also possible that compounds with lower $K_{ow}$ values (e.g., acetaminophen, DEET, iodopropynol, salicylic acid, sulfamethoxazole, and thibendazole) were removed by the destabilizing effects of EC, which result in charge neutralization, decreased solubility, and ultimately, enhanced aggregation to flocs.

The differences in PCP removal by EC treatment observed for raw wastewater and tertiary-treated wastewater are likely the result of chemical differences between the two water types (e.g. total suspended solids, which differed on average by two orders of magnitude that influence chemical adsorption to flocs). (195 mg/L and 1 mg/L in raw wastewater and tertiary-treated wastewater, respectively; courtesy of South Cross Water Reclamation Facility). Since it has been previously reported that current intensity greatly influences the extent of tetracycline (a common antibiotic) removal from aqueous solutions during EC with aluminum electrodes, it is possible that the current intensity was suboptimal for maximizing PCP removal during this study. Future research is needed to optimize the EC treatment process for removal of a wide-range of PCPs from domestic wastewater after various primary and secondary treatments in order to understand the full potential of EC to reduce PCP concentrations.

**Conclusions**

This study demonstrates that a benchtop EC unit outfitted with aluminum electrodes can concomitantly reduce concentrations of phosphate, microbial surrogates representing several major pathogen types (DNA/RNA viral, bacterial, protozoan parasite), as well as several PCPs in domestic wastewater. By providing the first information about the ability of EC to reduce concentrations of viral and parasitic protozoan surrogates, as well as PCPs, this study enhances previous assertions that EC is a promising sustainable wastewater treatment technology for domestic wastewater. While these collective results highlight the potential of EC for domestic wastewater treatment, further research is needed to address a number of outstanding issues. First, future work should attempt to discern the mechanisms behind the observed reductions as well as to optimize EC configurations and conditions to enhance the removal of PCPs and nitrogen species from domestic wastewater. Second, it will be necessary to optimize the EC treatment conditions to minimize energy consumption and the incorporation of renewable energy sources. Future pilot-scale and full-scale studies assessing the effectiveness of EC treatment of domestic wastewater are needed to fully understand the feasibility of this treatment option with respect to removing nutrients, microbes, and PCPs both from raw wastewater as a stand-alone treatment or as a polishing technology for refining tertiary-treated wastewater from standard wastewater treatment plants. Additionally, full life-cycle assessments are needed in order to understand the appropriateness of EC technologies as an option for decentralized and/or centralized domestic wastewater treatment prior to their implementation.

**Methods**

**Benchtop electrocoagulation unit.** The demonstration, benchtop EC unit (United States patent number 7211185 B2 by Powell Water Systems, Inc.; Centennial, CO, USA) evaluated in this study was comprised of a non-conductive, acrylic-resin chamber (35.6 × 5.4 × 2.5 cm) with nine aluminum plates (each 3.68 × 2.5 × 0.3 cm) vertically arranged and spaced 0.3 cm apart such that they occupied approximately 45% of the chamber volume (Figure 1). A 110-volt AC to DC power converter, set to 98 volts, was used to supply electricity to the unit via three electrical connections to the first, fifth, and ninth blade, resulting in two anodes and one cathode. During EC treatment, the actual current delivered ranged from 8.5–15.0 amps for raw domestic wastewater and 12.0–15.5 amps for tertiary-treated domestic wastewater. A peristaltic pump (Cole-Parmer® Masterflex Peristaltic Pump System 77910; Vernon Hills, IL, USA) was used to pump wastewater up through the unit chamber, which recirculated wastewater throughout the benchtop unit at a rate of 0.94 L/min. Wastewater was recirculated for 1 min per every liter of wastewater being treated. The resulting flocculant was removed from the EC unit effluent via filtration with paper filters that retain 11 µm particles (Whatman Qualitative Grade Plain Circles Grade 1; GE Healthcare Bio-Sciences, Pittsburgh, PA, USA). Since the aluminum blades become oxidized over time, they were cleaned with a sandblaster after every 12 L of wastewater treated by the EC unit to physically remove the oxidized portion of the aluminum blade.

**Experimental design.** Raw influent (post-grit removal) and tertiary-treated (dechlorinated) effluent were collected in sterile, plastic HDPE carboys from South Cross Bayou Water Reclamation facility (activated sludge plant with tertiary treatment) in St. Petersburg, Florida, USA. The tertiary-treated domestic wastewater received the following treatment prior to collection: grit removal, primary clarification, secondary treatment with an activated sludge system, and finally tertiary treatment with sand filtration, chlorination, and de-chlorination. Carboys were stored at 4°C in the dark and all experiments were conducted within 12 h of collection. Given the large number of analytes and logistical limitations, twice the minimum anticipated number of trials (n = 4) were collected before and after EC treatment in order to test the reduction efficiency of the EC unit. Four trials were executed with both raw wastewater and treated effluent, with each trial requiring an 18-L sample. From each sample, 6.1 L were isolated before treatment and the remaining volume was treated with the EC unit and filtered as described above. The EC unit was cleaned with 1 L of analytical grade methanol and rinsed with 5 L DI water after each trial. Process controls, consisting of DI water that was recirculated through the EC unit, were collected after the second and fourth trial to ensure no cross-contamination between trials. All pre- and post-
Table 2 | Mean ± standard deviation (n = 4 unless indicated otherwise; n = 2 and n = 3) of bacteria and virus concentrations, two-tailed student’s t-test (t) or Wilcoxon Rank Sum test (S) results, and mean percent reduction from domestic wastewater before and after EC treatment. Analyte concentrations are described as less than the process limit of detection (<pLOD) when undetected: 0.01 fecal indicator bacteria cfu/ml, *2.08 × 10⁶* HPyV targets/ml, *1.09 × 10⁷* PMMoV targets/ml, *0.1* MS2 bacteriophage pfu/ml, and *1.00 × 10³* Enterococcus spp. targets/ml with IC-NASBA. For molecular analyses, analyte concentrations are considered positive but below the process limit of quantification (+BLOQ) when at least one replicate is +BLOQ. *5.00 × 10³* Enterococcus spp. targets/ml (qPCR), *2.00 × 10⁴* Enterococcus spp. targets/ml (IC-NASBA), and *2.19 × 10⁸* PMMoV targets/ml

| Wastewater Sample | Analyte                          | Pre-EC treatment | Post-EC treatment | Test statistic and p-value | % Mean reduction |
|-------------------|----------------------------------|------------------|-------------------|---------------------------|-----------------|
| Raw               | Enterococcus spp. (cfu)          | 4.701 ± 1.50 × 10² | <pLOD⁺| S = 26.00, p = 0.0286 | >99.998 |
|                   | Fecal coliform (cfu)             | 1.95 ± 2.29 × 10² | <pLOD⁺| S = 26.00, p = 0.0286 | >99.995 |
|                   | HPyVs (qPCR target)              | 2.69 ± 8.16 × 10² | <pLOD⁺| S = 26.00, p = 0.0286 | >92.252 |
|                   | PMMoV (qPCR target)              | 5.47 ± 1.64 × 10⁴ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.800 |
| Tertiary-treated  | Enterococcus spp. (cfu)          | 5.43 ± 2.54 × 10² | <pLOD⁺| S = 26.00, p = 0.0286 | >81.567 |
|                   | Fecal coliform (cfu)             | 7.40 ± 4.05 × 10¹ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.986 |
|                   | HPyVs (qPCR target)              | <pLOD⁺| | | |
|                   | PMMoV (qPCR target)              | <pLOD⁺| | | |
| Spiked raw        | Bacillus subtilis (cfu)          | 3.80 ± 3.74 × 10⁵ | 3.00 ± 1.41 × 10⁰ | S = 26.00, p = 0.0286 | >99.99996 |
|                   | Enterococcus spp. (cfu)          | 1.25 ± 1.56 × 10⁵ | 2.15 ± 4.12 × 10⁰ | S = 26.00, p = 0.0286 | >99.999998 |
|                   | Enterococcus spp. (qPCR target)  | 1.37 ± 1.40 × 10⁵ | +BPLOQ ¹ | S = 26.00, p = 0.0286 | >99.507 |
|                   | Enterococcus spp. (NASBA target) | 1.15 ± 1.72 × 10⁵ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.913 |
|                   | Fecal coliform (cfu)             | 1.22 ± 2.61 × 10⁵ | 8.13 ± 9.10 × 10⁻¹ | t = 9.33, p = 0.0026 | >99.9992 |
|                   | HPyVs (qPCR target)              | 6.29 ± 2.00 × 10⁵ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.998 |
|                   | PMMoV (qPCR target)              | 6.38 ± 2.35 × 10⁵ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.998 |
|                   | MS2 bacteriophage (pfu)          | 3.72 ± 3.82 × 10⁵ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.9997 |
| Spiked tertiary-treated | Bacillus subtilis (cfu)          | 3.60 ± 3.69 × 10⁵ | 7.45 ± 6.43 × 10² | t = 20.23, p = 0.0003 | >99.898 |
|                   | Enterococcus spp. (cfu)          | 1.44 ± 1.03 × 10⁵ | 5.67 ± 7.03 × 10¹ | t = 27.88, p = 0.0001 | >99.996 |
|                   | Enterococcus spp. (qPCR target)  | 1.68 ± 1.40 × 10⁵ | +BPLOQ ¹ | S = 26.00, p = 0.0286 | >99.702 |
|                   | Enterococcus spp. (NASBA target) | 1.56 ± 1.04 × 10⁵ | +BPLOQ ¹ | S = 26.00, p = 0.0286 | >99.872 |
|                   | Fecal coliform (cfu)             | 1.47 ± 6.24 × 10⁴ | 3.38 ± 4.59 × 10¹ | t = 47.03, p < 0.0001 | >99.998 |
|                   | HPyVs (qPCR target)              | 7.69 ± 2.81 × 10⁵ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.997 |
|                   | PMMoV (qPCR target)              | 1.24 ± 6.26 × 10⁴ | +BPLOQ ¹ | S = 26.00, p = 0.0286 | >99.998 |
|                   | MS2 bacteriophage (pfu)          | 2.98 ± 5.34 × 10³ | <pLOD⁺| S = 26.00, p = 0.0286 | >99.9996 |
Table 3 | Mean +/- standard deviation of personal care product concentrations before and after EC treatment of raw and tertiary-treated wastewater with the benchtop unit (n = 4, unless otherwise noted). Any undetected analytes are listed as less than the reported process limit of detection (<=pLOD). When a two-tailed student’s t-test (t) or Wilcoxon Rank Sum test (S) revealed a positive, significant difference (\(\alpha = 0.05\)) between pre- and post-EC treatment concentrations, the mean percent reduction was calculated. Results that exceeded the calibration range but did not saturate the instrument detector are indicated with an E. Results that were likely underestimations (laboratory control sample spike below the control limit) are indicated with *.

| Analyte          | Mean concentration +/- standard deviation (ng/L) | Test statistic and p-value | % Mean reduction | Raw wastewater | Tertiary-treated wastewater |
|------------------|-----------------------------------------------|----------------------------|------------------|----------------|----------------------------|
|                  | Pre-EC treatment | Post-EC treatment |                      | Pre-EC treatment | Post-EC treatment |                      | Pre-EC treatment | Post-EC treatment |                      |
| Acetaminophen    | 55750 +/- 28000f | 28000 +/- 4243f     | S = 26.00, p = 0.0286 | 49.78          |                | <pLOD                    | N/A               | N/A               |
| Caffeine         | 31250 +/- 3202f  | 26250 +/- 4787f     | S = 23.00, p = 0.1714 | N/A            |                | <pLOD                    | N/A               | N/A               |
| Carbamazepine    | 130 +/- 16       | 125 +/- 6           | S = 20.00, p = 0.6564 | N/A            |                | 165 +/- 6                | 178 +/- 10        | N/A               |
| DEET             | 4075 +/- 263     | 3400 +/- 4244f      | t = 2.70, p = 0.0354  | 16.56          |                | 29 +/- 1                 | 68 +/- 12          | N/A               |
| Gemfibrozil      | 2700 +/- 13      | 2350 +/- 82f        | t = 4.58, p = 0.0038  | 12.96          |                | <pLOD                    | N/A               | N/A               |
| Ibuprofen        | 12000 +/- 817f   | 9950 +/- 900f       | t = 3.37, p = 0.0150  | 17.08          |                | <pLOD                    | N/A               | N/A               |
| Iopromide        | 4600 +/- 653*    | 3325 +/- 619*f      | S = 26.00, p = 0.0286 | 24.77          |                | 505 +/- 31               | 350 +/- 26        | 31.00             |
| Metholopramine   | 655 +/- 48       | 660 +/- 82f         | t = 0.11, p = 0.9193  | N/A            |                | <pLOD                    | N/A               | N/A               |
| Naproxen         | 11250 +/- 500f   | 11250 +/- 500f      | S = 18.00, p = 1.0000 | N/A            |                | 588 +/- 24*              | 645 +/- 44*       | N/A               |
| Phenytoin        | <pLOD            | 130 +/- 0 (n=2)     |                             | N/A            |                | <pLOD                    | N/A               | N/A               |
| Primidone        | <pLOD            | <pLOD                |                             | N/A            |                | <pLOD*                   | N/A               | N/A               |
| Salicylic Acid   | 32500 +/- 5568   | 9425 +/- 961f       | S = 26.00, p = 0.0286  | 71.00          |                | 61 (n=1)                 | 78 +/- 15         | N/A               |
| Sulfamethoxazole | 1925 +/- 310     | 1700 +/- 548f       | t = 0.72, p = 0.5013   | N/A            |                | 24 +/- 5                 | <RL               | S = 26.00, p = 0.0286 | 58.76             |
| Thiabendazole    | <pLOD            | 15 +/- 1            |                             | N/A            |                | 20 +/- 1                 | 18 +/- 1          | t = 3.00, p = 0.0240 | 10.00             |
| Triclocarban     | 728 +/- 200      | 113 +/- 75          | t = 5.77, p = 0.0012   | 84.54          |                | <pLOD                    | N/A               | N/A               |
| Triclosan        | 1350 +/- 238     | 248 +/- 89          | t = 8.68, p = 0.0001   | 81.67          |                | <pLOD                    | N/A               | N/A               |
| Trimethoprim     | 488 +/- 54       | 508 +/- 48f         | t = 0.56, p = 0.5986   | N/A            |                | <pLOD                    | N/A               | N/A               |
| Warfarin         | <pLOD            | <pLOD                |                             | N/A            |                | <pLOD                    | N/A               | N/A               |
Electrical connections to power source

Figure 1 | The benchtop electrocoagulation unit with nine aluminum blades arranged vertically in the unit chamber. Electrical connections on the first, fifth, and ninth blades were connected to a 110-1 AC to DC power converter. A peristaltic pump re-circulated wastewater up through the unit chamber, into the post-treatment reservoir, and into the collection reservoir.

treatment samples, as well as process controls, were analyzed for nutrient, microbes, and PCPs. In order to quantify the reduction efficiency of microbes, 1-L wastewater influent and effluent samples were augmented separately with concentrated surrogates for bacteria (Enterococcus faecalis ATCC-29212™ and Escherichia coli strain C600), viruses (JC HPyV ATCC-VR-1583™, PMMoV (obtained from Scott Adkins; USDA), and MS2 bacteriophages), and parasitic protozoa (B. subtilis spores) (see Supplementary Information). Four trials were executed for both the raw wastewater and tertiary-treated effluent. Twenty-milliliter and 120-mL aliquots of the spiked wastewater were collected prior to treatment with the EC unit for samples augmented with bacteria and viruses, respectively. The remaining volume was treated with the benchtop EC unit as described above. The EC unit was cleaned between each trial and one process control was collected upon completion of the fourth trial.

Nutrient analyses. Four sets of pre- and post-EC treatment samples of raw wastewater and tertiary-treated wastewater samples, along with two process controls, were analyzed in duplicate by the Oceanic Nutrient Laboratory at the University of South Florida, College of Marine Science for nitrate + nitrite, nitrite, ammonium, and phosphate. Due to the high nutrient concentrations in raw wastewater, pre-EC treatment raw wastewater samples were diluted to 2.4% final concentration with deionized water prior to analysis. The analytical methods used for nitrate + nitrite, nitrite, ammonium, and phosphate followed the recommendations of Ref. 44 and were analyzed using a five-channel Technicon Autoanalyzer II (SEAL Analytical, Mequon, WI, USA) upgraded with new heating baths, proportional pumps, colorimeters, improved optics, and an analog to digital conversion system (New Analyzer Program v. 2.40; Waters Corporation, Milford, MA, USA). To extend the dynamic range to 30 μM, the ammonium technique was modified by decreasing the flow rates for the nitroprusside, hypochlorite, phenolate, citrate, sample, air bubble, and wash down to 50 μL, 50 μL, 50 μL, 320 μL, 600 μL, 160 μL, and 1200 μL per minute, respectively.

Nutrient standards were run in triplicate before and after analysis, as well as a check standard in the middle of the run to correct for any drift in sensitivity. The detection limits for nitrate + nitrite, nitrite, ammonium, and phosphate were 0.02 μM, 0.02 μM, 0.38 μM, and 0.09 μM, respectively. All method blanks were negative. Process controls for both experiments had low levels of nitrate + nitrite, nitrite, ammonium, and phosphate; however, the concentrations were less than the standard deviations for replicate samples.

Bacillus subtilis spores. All augmented pre- and post-EC treatment samples and process controls were inoculated with B. subtilis spores at 20 min to kill chlorine and other disinfecting bacteria and then maintained in the dark at 4 °C. Within 48 h of the experiment, aliquots of each sample were spread-plated in triplicate (all pre-EC treatment samples were diluted 1:10,000) onto tryptic soy agar and incubated at 36.5 ± 1 °C for 24 h. The resulting viable B. subtilis colonies (i.e. opaque in color and rough appearance) were enumerated and concentrations were back-calculated to account for dilutions. Since the maximum sample volume plated was 500 μL, the pLOD was 2 cfu/mL. No colonies grew on method blanks. While no colonies were present in the process control for the experiments with raw wastewater, the average B. subtilis concentration in the process control for the experiment with the tertiary treated wastewater was 39 cfu/mL.

MS2 bacteriophage. Since the wastewater samples were augmented with an MS2 bacteriophage culture prior to EC treatment, MS2 bacteriophage concentrations were quantified using the single-agar layer (SAL) protocol using E.coli Famp ATCC-70899™ for pre-EC treatment samples and the double-agar layer (DAL) protocol for pre-EC treatment samples that had been diluted four-fold45. Per US EPA method 1602, each pre-EC treatment sample was analyzed using the DAL protocol in triplicate and each post-EC treatment sample was analyzed in replicates of ten using the SAL protocol. All method blanks were negative. The pLOD was 1 plaque forming unit (pfu)/10 μL for the SAL protocol and 2,000 pfu/mL for the DAL protocol. The average concentrations of MS2 bacteriophage in the process controls were less than the pLOD.
triclocarban (10 ng/L), triclosan (50 ng/L), warfarin (20 ng/L), ibuprofen (25 ng/L), isopropamide (50 ng/L), meprobamate (10 ng/L), naproxen (50 ng/L), phenytoin (100 ng/L), sulfamethoxazole (10 ng/L), and trimethoprim (10 ng/L). For the analysis of raw wastewater prior to EC treatment, the LOD was an order of magnitude greater for all analytes.

No PCPs were detected in the two process controls collected during the experiment with tertiary-treated wastewater. However, low concentrations of acenaphthene (22 ng/L), acetylene (83 ng/L), DEF (180 ng/L), and salicylic acid (76 ng/L) were detected in the process controls collected during the experiment with raw wastewater. Since the detected concentrations of these analytes in the process controls are less than the standard deviations observed for raw wastewater samples before and after EC treatment, it is unlikely that the observed contamination influenced the results of this study.

**Statistical analyses.** Statistical analyses were executed in SAS v.9.3 (SAS Institute Inc., Cary, NC, USA) to identify significant ($\alpha = 0.05$) differences in the concentrations of all nutrients, microbes, and PCPs before and after EC treatment. If the data had normal distributions, a two-tailed t-test was performed with either the pooled method (for equal variances) or the Satterthwaite approximation (for unequal variances). If the data were not normally distributed, then the non-parametric Wilcoxon Rank Sum test was performed. For a given analyte, if a significant difference in pre- and post-EC treatment concentrations was determined with 95% confidence, then the average percent reduction was calculated. If concentrations were $>$ BLOQ or $<$ LOD, then the pLOD or pQLOD, respectively, were used to correctly test for statistical differences and to calculate the average percent reduction.

1. Nwachukwu, N. & Gerba, C. P. Emerging waterborne pathogens: Can we kill them all? Curr Opin Biotechnol 15, 175–180 (2004).
2. Petrović, M., Gonzalez, S. & Barceló, D. Analysis and removal of emerging pollutants in wastewater: A review. Trends Anal Chem 22, 685–696 (2003).
3. Ashbolt, N., Grawbow, W. O. K. & Snozzi, M. in Water quality guidelines, 2nd edn, Vol. 2, EC, WHO, 1999.
4. Brausch, J. M. & Rand, G. M. A review of personal care products in the aquatic environment: environmental concentrations and toxicity. Chemosphere 82, 1513–1521 (2010).
5. Touraud, E., Roig, B., Sumpter, J. P. & Coeister, C. Drug residues and endocrine disruptors in drinking water: risk for humans? Int J Hyg Environ Health 214, 437–441 (2011).
6. Jones, O. A. H., Vouillouf, N. & Lester, J. N. Human pharmaceuticals in wastewater treatment processes. Crit Rev Environ Sci Technol 35, 401–427 (2005).
7. Mollah, M. Y. A. et al. Fundamentals, present and future perspectives of electrocoagulation. J Hazard Mater 114, 199–210 (2004).
8. Emamjomeh, M. M. & Sivakumar, M. Review of pollutants removed by electrocoagulation and electrocoagulation/floation processes. J Environ Manag 90, 1663–1679 (2009).
9. Guzmán, C. Electrochemical technologies in wastewater treatment. Sep Purif Technol 38, 11–41 (2004).
10. Mollah, M. Y. A., Schennach, R., Parga, J. R. & Cocke, D. L. Electrocoagulation (EC) — science and applications. J Hazard Mater 84, 29–41 (2001).
11. Holt, P. K., Barton, G. W. & Mitchell, C. A. The future for electrocoagulation as a treatment for wastewater disinfection. J Environ Manag 59, 35–367 (2005).
12. Ghernaout, D. & Ghernaout, B. From chemical disinfection to electrodisinfection: The obligatory itinerary? Desalin Water Treat 16, 156–175 (2010).
13. Visk, E. A., Carlson, D. A., Eikum, A. S. & Gjesing, E. T. Electrocoagulation of potable water. Water Res 18, 1355–1360 (1984).
14. Zhu, B., Clifford, D. A. & Chellam, S. Comparison of electrocoagulation and chemical coagulation pretreatment for enhanced virus removal using microfiltration membranes. Water Res 39, 3098–3108 (2005).
15. Tannner, C. T. & Chellam, S. Mechanisms of virus control during iron electrocoagulation – Microfiltration of surface water. Water Res 46, 2111–2120 (2012).
16. Gao, S. et al. Electro-coagulation–floation process for algae removal. J Hazard Mater 177, 336–343 (2010).
17. Ghernaout, D., Badis, A., Kellil, A. & Ghernaout, B. Application of electrocoagulation in Escherichia coli culture and two surface waters. Desalination 219, 118–125 (2008).
18. Ouaisa, Y. A., Chabani, M., Aimra, A. & Bensmaila, A. Removal of tetracycline by electrocoagulation: kinetic and isotherm modeling through adsorption. J Environ Chem Eng 2, 177–184 (2014).
19. Koby, M., Can, O. T. & Bayramoglu, M. Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes. J Hazard Mater 100, 163–178 (2003).
20. Chen, X., Chen, G. & Yue, P. L. Separation of pollutants from restaurant wastewater by electrocoagulation. Sep Purif Technol 19, 65–76 (2000).
21. Gugli, D. Optimization of electrocoagulation of pistachio processing wastewaters using the response surface methodology. Desalin Water Treat 1–10, doi:10.1080/19443994.2014.907752 (2014).
Acknowledgments
The supplies for this research were funded by Powell Water Systems, Inc. (Centennial, CO, USA). E.M.S. was funded by the STAR Fellowship Assistance Agreement No. FP-91737601-3 awarded by the U.S. Environmental Protection Agency (EPA). This work has not been formally reviewed by the U.S. EPA; therefore, the views expressed in this paper are solely those of the authors and any mention of products does not constitute recommendation for use. Special thanks to Albert McAfee and the South Cross Bayou Water Reclamation facility for their cooperation in providing domestic wastewater. These experiments could not have been executed without the assistance of Bert Gerber (Gerber Pumps International, Inc.), Elizabeth Fahsbender (USF), Rachel Harbeitner (USF), Stephanie Lawler (USF), Bethany Levenson (USF), and Karyna Rosario (USF).

Author contributions
This study was designed by E.M.S., M.M.C., E.S.V.V. and M.B., with assistance from J.O.L. E.M.S., M.M.C., S.M.M., R.M.U. and R.O.S. executed the experiments with input from J.O.L. Microbial analyses were executed by R.O.S. (MS2 bacteriophage), S.M.M. (fecal indicator bacteria, B. subtilis spores, and HPyV), R.M.U. (fecal indicator bacteria), and E.M.S. (B. subtilis spores and PMMoV). The B. subtilis spores were provided by J.O.L. Statistical analyses were executed by E.M.S., with assistance from M.M.C. The manuscript text as well as tables and figures were written and prepared by E.M.S., with subject relevant contributions from all authors. All authors reviewed the manuscript.

Additional information
Supplementary information accompanies this paper at http://www.nature.com/scientificreports

Competing financial interests: All authors wish to disclose the following facts, which may be considered as potential conflicts of interest given the financial contribution to this work. While this study was funded by Powell Water Systems, Inc., the funders had no role in the study design, sample collection and analyses, decision to publish, or the preparation of the manuscript. All authors confirm that they have no known conflicts of interest associated with the publication of this manuscript and that the financial support received did not influence the outcome of this study. The authors declare no competing financial interests.

How to cite this article: Symonds, E.M. et al. Reduction of nutrients, microbes, and personal care products in domestic wastewater by a benchtop electrocoagulation unit. Sci. Rep. 5, 9380; DOI:10.1038/srep09380 (2015).

This work is licensed under a Creative Commons Attribution 4.0 International License. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in the credit line; if the material is not included under the Creative Commons license, users will need to obtain permission from the license holder in order to reproduce the material. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/