



Comprehensive assessment of organic contaminant removal from on-site sewage treatment facility effluent by char-fortified filter beds

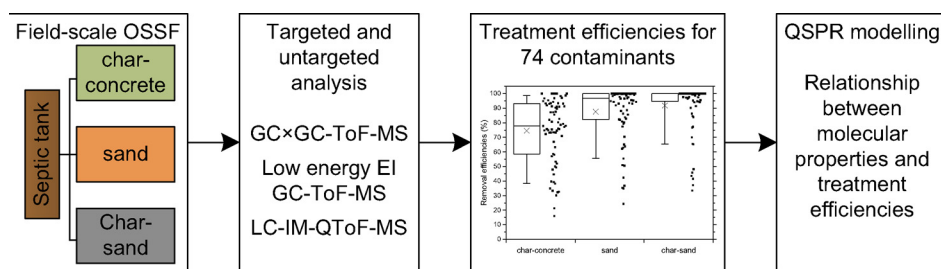
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GRAPHICAL ABSTRACT



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ABSTRACT

To remove organic contaminants from wastewater using cost-efficient and currently existing methods, our study investigated char-fortified filter beds for on-site sewage treatment facilities (OSSFs) in a long-term field setting. OSSFs are commonly used in rural and semi-urban areas worldwide to treat wastewater when municipal wastewater treatment is not economically feasible. First, we screened for organic contaminants with gas chromatography and liquid chromatography mass spectrometry-based targeted and untargeted analysis and then we developed quantitative structure-property relationship models to search for key molecular features responsible for the removal of organic contaminants. We identified 74 compounds (24 confirmed by reference standards) including plasticizers, UV stabilizers, fragrances, pesticides, surfactant and polymer impurities, pharmaceuticals and their metabolites, and many biogenic compounds. Sand filters that are used as a secondary step after the septic tank in OSSFs could remove hydrophobic contaminants. The addition of biochar significantly increased the removal of these and a few hydrophilic compounds (Wilcoxon signed-rank test, $\alpha = 0.05$). Besides hydrophobicity-driven sorption, biodegradation was suggested to be the most important removal pathway in this long-term field application. However, further improvements are necessary to remove very hydrophilic contaminants as they were not removed with sand and biochar-fortified sand.

1. Introduction

Wastewater from housing units that are not connected to any municipal sewage treatment plant (STP) is usually treated in decentralized

on-site sewage treatment facilities (OSSFs). In Sweden, OSSFs are in use for 28% of all housing units [1] and are mainly situated in semi-urban and rural areas. Typical treatment technologies are greywater separation, septic tanks followed by a soil infiltration system or a soil bed or,

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less frequently, an aerobic treatment system [2]. There is an increasing awareness of the consequences of sewage discharge into the environment that could lead to eutrophication and water quality related diseases. However, a few studies have evaluated the removal of organic contaminants in OSSFs [3] and have found that many contaminants were poorly removed. OSSF effluent concentrations from nanograms to several micrograms per liter have been reported [3–9]. These studies imply a need for improved OSSF technologies for the removal of organic contaminants.

One cost-effective option is to extend existing OSSFs with a filtration step utilizing low-cost and easily obtainable sorbents, for instance biochar. Biochar is a carbon-rich material that is produced by thermochemical treatment of biomass [10]. It has a large surface area and a high aromatic carbon content [11,12], which gives the material good sorption properties. Biochar produced at 400–700 °C showed improved removal of hydrophobic organic contaminants due to a higher microporosity, surface area and hydrophobicity [13]. Biochars are attractive for cost-sensitive applications, including privately-owned OSSFs and decentralized water treatment systems in developing countries. They have been evaluated for removal of specific organic water contaminants, such as pesticides [14,15], pharmaceuticals [16–18], and dyes [19,20]. To the best of our knowledge, removal of contaminants with biochar has not previously been systematically and comprehensively investigated under field conditions using untargeted analysis in combination with quantitative structure-property relationship modeling.

The current study is a follow-up to a field study by Kholoma et al. [21] that demonstrated a significantly better removal of phosphorus species in a field-scale OSSF sand filter (soil bed) when fortified with biochar or biochar-fortified gas concrete. It aims to investigate the removal of organic contaminants at the same study site after two years of operation using two-dimensional gas chromatography time-of-flight mass spectrometry (GC × GC-ToF-MS), gas chromatography time-of-flight mass spectrometry with low energy electron ionization (GC-ToF-MS), and liquid chromatography-ion mobility quadrupole time-of-flight mass spectrometry (LC-IM-QToF-MS). Targeted analysis for 26 environmentally-relevant wastewater contaminants was complemented with untargeted screening and the results were thoroughly evaluated using multivariate statistics with calculated chemical descriptors to understand better the relationship between molecular properties of the contaminants and their removal by biochar.

2. Experimental

2.1. Study site and sampling

The study site and experimental set-up is described in detail in Kholoma et al. [21]. Briefly, the wastewater originated from an OSSF operating for four households at Garns Ösby, Sweden. The three filter beds received septic tank wastewater after it was pretreated in a 200 L BioStep pond filter to reduce solids (Figure S1). The beds were installed in three parallel trenches and were housed in wooden containers (1.5 × 0.8 m, 0.75 m² upper surface area) that were lined with a polyethylene film. A 0.2 m soil layer and a polyethylene sheet covered the beds from the top and protected against precipitation and airborne contamination. Each container was first filled with 0.15 m of gravel (1–3 cm crushed bedrock), then with a plastic garden mesh (1.1 mm × 1.1 mm) followed by the specific filter media. The first container was filled with 0.5 m sand (0.3 to 2.5 mm, Hakungekrossen AB, Sweden) as reference, the second container was filled with a 0.3 m layer of the same sand, then with 0.2 m crushed and washed hardwood-derived biochar (0.5 to 20 mm, Skogens kol AB, Sweden) that was pyrolyzed at 500 °C by the manufacturer. The third container was filled with 0.3 m gas concrete (10–80 mm, Sorbulite®, Biotech AB, Sweden), followed by the same biochar as in container 2.

At regular time intervals, wastewater was pumped to a distribution

tank that fed the three filter beds by gravity flow. For the first three months, 5 L were added every 3 h; after that, the volume was adjusted to 2.5 L every 2 h to smooth the flow over each day. The set-up enabled even distribution, aeration, and removal of accumulated solids from the ingoing pipes and effluent sampling (using vertical pipes) from the outgoing pipes [21]. Field sampling was carried out in October 2016. One influent sample and one sample after each treatment (sand, char-sand, char-concrete) were collected using 24-hour time-integrated sampling in pre-burned and methanol-washed glass bottles.

2.2. Sample preparation

Extraction of the samples for GC analysis is summarized in the Supplementary Material and is described in detail in Blum et al. [8]. For LC analysis, 50 mL portions of each sample were centrifuged for 5 min at 3700 rpm and the supernatant was acidified with 0.1% formic acid. A 450 µL aliquot was spiked with 50 µL internal standard mixture (Table S2) and analyzed without further purification.

2.3. Two-dimensional gas chromatography high resolution time-of-flight mass spectrometry

The instrument set-up included a Pegasus 4D HRT mass spectrometer (Leco Corp., St. Joseph, MI, USA), equipped with a 7890 gas chromatograph (Agilent, Palo Alto, CA, USA). An operational summary can be found in the Supplementary Material and details of the instrument and operational parameters can be found in Blum et al. [22].

The data were initially processed using ChromaTOF-HRT software (V.1.90, Leco Corp., St. Joseph, MI, USA) to search for the target analytes with a given retention time and with characteristic ions within a 0.005 Da mass tolerance (Blum et al. 2017) [8]. Target analytes and quantification ions are listed in Table S4.

Untargeted data processing was carried out using the same software, including mass calibration, peak picking, retention index calculation and NIST-MS library search (similarity > 65%) for peaks with a signal-to-noise ratio (S/N) > 10. The accurate mass spectra of the resulting features were converted to nominal mass spectra and aligned (> 65% similarity) with the Java application GUINEU [23] resulting in 4 597 aligned features. Features appearing in blanks (n = 470) or with low NIST similarity score (n = 2 761) were excluded, leaving 1 499 provisionally identified compounds (Fig. 1). These compounds were further filtered by detection frequency (> 33%, n = 4 samples × 5 replicates = 20), resulting in 354 compounds that were manually investigated for the expected molecular ion and abundant fragments (from NIST-MS interpreter).

Seventy-nine tentatively identified compounds passed manual scrutiny and were included in a targeted data processing method using ChromaToF with a minimum of two fragments, 10 ppm mass accuracy and a < 30 s retention time window.

2.4. Gas chromatography time-of-flight mass spectrometry

The influent sample was re-run with a 7250 GC quadrupole TOF from Agilent Technologies using low energy electron ionization. Details can be found in the Supplementary Material.

Data were processed using Unknown Analysis (B.08.00) from Agilent Technologies, USA. The “Sure Mass” algorithm was used for deconvolution, and peaks in the 70 eV data were the subjects of a NIST library search (pure weight factor of 0.70, minimum match score 50). Tentatively identified compounds that lacked abundant molecular ions in the GC × GC-ToF-MS analyses (Section 2.3) were located in the 70 eV GC-ToF file. The corresponding 12 eV spectra were extracted at the same retention times and examined to find molecular and large fragment ion information. This information was used to accept or reject the tentative structure.

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