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Assessing the ecological long-term impact of wastewater irrigation on soil and water based on bioassays and chemical analyses

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ABSTRACT

The reuse of treated wastewater for irrigation and groundwater recharge can counteract water scarcity and reduce pollution of surface waters, but assessing its environmental risk should likewise consider effects associated to the soil. The present study therefore aimed at determining the impact of wastewater irrigation on the habitat quality of water after soil passage and of soil after percolation by applying bioassays and chemical analysis. Lab-scale columns of four different soils encompassing standard European soil and three field soils of varying characteristics and pre-contamination were continuously percolated with treated wastewater to simulate long-term irrigation. Wastewater and its percolates were tested for immobilization of *Daphnia magna* and growth inhibition of green algae (*Pseudokirchneriella subcapitata*) and water lentils (*Lemna minor*). The observed phytotoxicity of the treated wastewater was mostly reduced by soil passage, but in some percolates also increased for green algae. Chemical analysis covering an extensive set of wastewater-born organic pollutants demonstrated that many of them were considerably reduced by soil passage, particularly through peaty soils. Taken together, these results indicated that wastewater-born phytotoxic substances may be removed by soil passage, while existing soil pollutants (e.g. metals) may leach and impair percolate quality. Soils with and without wastewater irrigation were tested for growth of plants (*Avena sativa*, *Brassica napus*) and soil bacteria (*Arthrobacter globiformis*) and reproduction of collembolans (*Folsomia candida*) and oligochaetes (*Enchytraeus crypticus*, *Eisenia fetida*). The habitat quality of the standard and two field soils appeared to be deteriorated by wastewater percolation for at least one organism (enchytraeids, plants or bacteria), while for two pre-contaminated field soils it also was improved (for plants and/or enchytraeids). Wastewater percolation did not seem to raise soil concentrations of classical organic pollutants and priority substances, while a significant retention was found for zinc and several organic micropollutants, particularly in the peaty soils, thus matching these soils' observed higher removal efficiency. Overall, our results demonstrate that benefits of wastewater irrigation can come with the cost of deteriorating soil habitat quality and depend on the respective soil and considered test organism. The approach employed here represents a feasible tool to assess these integrated effects at lab-scale while being predictive for scenarios at field-scale.

1. Introduction

Urban areas of metropolises facing growing water scarcity together with increasing water demand could benefit from the reuse of treated wastewater on land (Lischeid and Nathkin, 2011). Moreover, this reuse could help to further improve the quality of treated wastewater and of receiving surface waters as demanded by the European Water Framework Directive (WFD, 2000). At a global scale, wastewater reuse by irrigation on land is widely practiced, particularly in arid areas (Hamilton et al., 2007). Concerns about wastewater irrigation relate to groundwater pollution and

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deterioration of the irrigated soil resulting from the accumulation of wastewater-born metals and organic pollutants (Munoz et al., 2009). However, soil contamination is often assessed with respect to risks for human health from crop consumption, while ignoring the potential impacts on soil habitat quality for terrestrial organisms (Prosser and Sibley, 2015). Environmental impacts of wastewater irrigation have been studied by monitoring individual wastewater-born pollutants in soil or water using laboratory or (semi-) field experiments (Ternes et al., 2007; Munoz et al., 2009; Grossberger et al., 2014) or by measuring the removal efficiency of wastewater-born pollutants and pathogens via constructed wetlands, bank filtration or soil aquifer treatment (Verlicchi and Zambello, 2014). The focus of such studies has thus been on either water or soil quality but rarely addressed the trade-off between these two compartments (Munoz et al., 2009; Orias and Perrodin 2013). Soil passage of treated wastewater can enhance the biodegradation of pollutants compared to the discharge into streams, however, adsorptive pollutants can also be retained in the soil and accumulate (Munoz et al., 2009). Selecting already contaminated instead of pristine land may relativize the pollution of soil by wastewater irrigation, but increase the risk for groundwater and surface water quality due to remobilization of old contaminants (Hamilton et al., 2007; Lottermoser, 2012). Usually, these risks are assessed by measuring already manifested effects during or *ex-post* studies at field-scale, but an experimental lab-scale method to estimate potential trade-offs *ex-ante* is missing.

Therefore, the present study aimed to assess in an integrative and predictive approach the impact of long-term irrigation with treated wastewater on the quality of soil as habitat for terrestrial organisms and the influence of soil passage on the water quality for aquatic organisms, thus considering both affected environmental compartments together. Saturated, packed soil columns were continuously percolated with treated wastewater in a condensed timescale to represent a worst-case scenario with regard to low degradation and high soil retention compared to field conditions. Moreover, the influence of soil characteristics in terms of physico-chemical properties and existing precontamination was investigated. To this end, an aquatic and a terrestrial bioassay battery encompassing standard ecotoxicity tests were combined with comprehensive chemical monitoring of priority and emerging pollutants.

The ecotoxicological hazard of a complex environmental sample depends on the chemical composition and usually differs among exposed species due to their individual sensitivity toward the single components and their mixture. Therefore, a battery of aquatic and terrestrial bioassays was applied representing different trophic levels and toxicity endpoints, to assess the quality of treated wastewater, its percolates after soil passage and the respective soils. In the context of wastewater irrigation, percolate quality can be seen as indicative for the impact on groundwater, adjacent surface water as well as soil pore water. Applying aquatic biotest batteries to assess the hazard of treated wastewater (whole effluent toxicity testing, WET) is widely established (Power and Boumphrey, 2004; Orias and Perrodin, 2013) and follows standardised test guidelines. Green algae as unicellular primary producers and crustacean as primary consumers are regularly required in WET, whereas water lentils as macrophytic primary producers are less frequently used in this context although being particularly suitable for the testing of turbid eluates (Moser and Römbke, 2009). These aquatic biotests proved also appropriate to assess the ecotoxicity of waste materials' leachates (Krüger et al., 2013), but have rarely been applied for the analysis of soil percolates (Abrantes et al., 2008). Until now, terrestrial bioassays are not considered in European legislation on soil protection, despite their successful usage in characterizing the ecotoxicity of contaminated sites (Hawthorne et al., 2005). The benefit and reliability of a terrestrial biotest battery such as that used in the present study was already demonstrated for the assessment of waste in a comprehensive international ringtest (Moser and Römbke, 2009). Assessing wastewater irrigated soil by applying biotests appears therefore promising, but has not been reported so far.

2. Materials and methods

2.1. Soil column apparatus

An experimental apparatus was constructed taking into account the recommendations on soil column design of Lewis and Sjöstrom (2010) for saturated packed soil columns. The columns consisted of stainless steel tubes of 20 cm height and 22 cm diameter (6532 cm^3 volume for soil) and lids of polyoxymethylene. The soil was held between two fixed steel meshes (mesh size 1.0 mm) covered at each side by a sieve fabric disk (polyester, mesh size 0.25 mm, SEFAR AG, Switzerland). The columns were continuously percolated from bottom to top to ensure water saturation of the soil (further details are described in Text S.1).

2.2. Validation experiment with climbazole-spiked treated wastewater

A four weeks percolation experiment was performed to demonstrate the suitability and reliability of the apparatus by percolating European standard soil (LUFA 2.3, LUFA, Speyer, Germany; identical with s4, Table 1) at 9.0 ± 0.1 kg soil d.w. per column ($n = 3$ replicate columns) with treated wastewater spiked with the fungicide climbazole. The percolation regime (6.9 ± 0.1 mL/min, equivalent to 10.0 ± 0.2 L/d, for 28 d) and spiking concentration (1 mg/L climbazole) were selected to yield a climbazole concentration of approximately 25 mg/kg soil d.w. (assuming 90% adsorption), expected to result in 50% effect in plant tests (Richter et al., 2013). After percolation, the soil of each column was thoroughly homogenized. One soil sample per soil column was stored frozen (<5 months) and analyzed in triplicate for climbazole (method see Table S.1 in supplements). The remaining soil was air-dried and used in growth inhibition tests with *Avena sativa* and *Brassica napus* (ISO, 2012a). Each of the technical replicates (i.e. soil columns) was tested with four biological replicates (i.e. plant pots), in parallel with four replicates of untreated (i.e., non-percolated) LUFA 2.3 soil.

2.3. Main experiment with treated wastewater and four soils

In order to experimentally simulate long-term irrigation (approximately 30 years assuming a realistic rate of 730 L/m^{2}*year, Ternes et al., 2007), four packed soil columns were continuously percolated with treated municipal wastewater (WW feed) for 77 days (6.9 ± 0.1 mL/min, equivalent to 10.0 ± 0.2 L/d or 276 L/m^{2}*d, approximately 54 cm/d filter velocity). Each of the four columns was packed with a different soil, encompassing one reference soil and three field soils representing various soil properties but also different levels of precontamination. The field soils were sampled at the study site Hobrechtsfelde, located close to Berlin, Germany, where raw wastewater had been irrigated onto almost 100 km^2 between the 1870s and the 1980s and where impacts on soil and groundwater quality have been extensively studied (Lottermoser 2012). For the last ten years, treated wastewater has been led into constructed wetlands and channels of this area in the course of pilot projects aiming to prevent falling water levels that are threatening forest and wetland biotopes and are promoting the remobilization of heavy metals from degraded organic matter