Elimination of COD, microorganisms and pharmaceuticals from sewage by trickling through sandy soil below leaking sewers

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Abstract

To simulate the filtration and/or degradation of trickling sewage from leaky sewers through the non-water-saturated underground, sewage was trickled through sand of 0.4–2 mm from the Rhine valley in glass columns of 125 cm length. For the same sewage the chemical oxygen demand (COD) removal was almost independent of low or high trickling rates. The COD removal efficiency varied, however, from 67% to 79%, for sewage from rain and dry weather periods, respectively. The water content of the moist sand increased from initially 80 ml kg\(^{-1}\) with increasing sewage trickling rates to 108 ml kg\(^{-1}\) sand. It remained at 108 ml kg\(^{-1}\) at higher trickling rates higher than 600 ml d\(^{-1}\).

Analyses of effluent of five consecutive 25-cm soil columns revealed that about 50% of the initial COD were filtrated off on top of the sand or degraded in the uppermost 25 cm at varying trickling rates. Another 6–12% of the COD were removed in the following 25–50 cm of sand, whereas almost no further COD removal was seen in the subsequent two or three 25-cm columns. The COD elimination during trickling of sewage through the segmented column (interrupted random flow) was slightly better than that in the non-segmented column.

Total and faecal coliform bacteria decreased faster with increasing trickling depth than that of total aerobic or anaerobic bacteria. After a filter/degradation stretch of 125 cm elimination of all bacteria reached 96.2–99.9%. The sewage contained low concentrations of at least 10 different pharmaceuticals or X-ray media. During trickling of sewage through sand, elimination of these compounds by adsorption onto sand and/or biodegradation varied from a complete removal, e.g. Ibuprofen or Naproxen, to almost no removal for several X-ray contrast media. Some of the medicals were removed as effectively as during conventional sewage treatment.

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1. Introduction

In urban settlements underground sewers transport sewage to wastewater treatment plants. About 17% of the public sewers in Germany are leaky and must be repaired, whereas another 14% are damaged [1]. In Great Britain 23% of the sewers are “in critical condition” of which 10% are in an “unsatisfactory condition” [2]. For the Greater London region a total loss of 5% of sewage by leaks was estimated [3]. If leaky sewers would be used for in-sewer storage of sewage for wastewater management in future [4], this might cause an even higher pollution of the underground.

Leaks of sewers are caused by sloppy connections of pipes, chemical and/or biochemical corrosion, abrasion or cracks. Raw sewage then passing through sand or gravel support trickles through the underground into the groundwater [5]. During trickling of sewage through sandy soil most of its organic material is retained by the
soil or is degraded by the indigenous flora [5,6]. About 6–8% of the chemical oxygen demand (COD) of sewage are not biodegradable. Thus, humic acid-like substances, halogen–organic compounds, pharmaceuticals from private or hospital medication [7], non-degradable material from estates of the chemical industry [2] or lysis products of the water and soil microflora [8] reach the groundwater, together with a small portion of the sewage or the soil population itself [9].

The aim of this investigation was to analyse the removal efficiency of organic matter of sewage during trickling through 1.25 m sandy soil at highly varying rates and to investigate the elimination of pharmaceuticals and of pathogenic bacteria.

2. Materials and methods

2.1. Soil column experiments

The fate of sewage that trickles into the underground below leaky sewers was analysed by two different experimental approaches:

(1) A glass column (150 cm length, 10 cm inner diameter) was filled to a height of 125 cm (average distance of sewers to the groundwater table) with sand from the Rhine valley (Fig. 1a). The sand (34% 0.4–1 mm grains, 66% 1–2 mm grains, coarse material sieved off, bulk weight 1.5 kg l⁻¹) was excavated from 2–6 m depths and represented the underground of Karlsruhe. Sewage was pumped onto the column to maintain 10 cm of hydrostatic pressure during undisturbed trickling through 125 cm. Sewage at the bottom outlet was collected with a funnel of 8.5 cm in diameter from the central zone, dewatering a volume of 7190 ml and from the peripheral ring zone (1.3 cm width from 8.5 to 9.8 cm), dewatering a volume of 2240 ml [6]. The wastewater that might have trickled along the glass walls was collected together with the peripheral drain water. Trickling conditions were anoxic and were considered stable when no changes of the COD of the effluent for 1 week were observed. To determine the water content of the sand, the column was placed on a balance.

(2) A set of five glass columns (25 cm length each, 10 cm in diameter) was filled with sand and connected bottom to top by rubber tubings with sampling ports for the trickling sewage (Fig. 1b).
Each of the lower columns obtained the trickling sewage from the column above it to avoid non-interrupted random flow over 125 cm length. The tubings were arranged like siphons to facilitate sewage withdrawal at different depths for analyses. This column was initially operated with 10 cm sewage on top of column S5 and later on was operated with a constant trickling rate, forced by the pump speed, as shown in Fig. 1b. Both columns were placed in a thermostated room at 18°C.

2.2. Sewage

Sewage was taken every 2 weeks from the sewage treatment plant of Karlsruhe (Germany) after mechanical treatment. The sewage was filtered through a sieve (pore size 1.5 mm) and was stored in a cold room at 4°C. It contained $10^5$ cfu ml$^{-1}$ bacteria, as compared to about $10^2$ cfu ml$^{-1}$ that were present in the sand. Analyses of the COD during 14 d of interim storage in the cold room were performed for sewage from dry weather periods and from periods of heavy rain falls. Maximally 5% of the COD were degraded. Nitrification was prevented by anaerobic storage. The variation of sewage from dry and rain weather is shown in Table 1. Every day the required portion was filled into the bottles for raw sewage at the columns and was continuously flushed with N$_2$ to prevent respiration and nitrification. The sewage in the bottles was stirred while being pumped onto the columns. Acidification of sewage in the storage bottles was negligible (< 0.1 mmol l$^{-1}$ of acetic, propionic or n-butyrac acid after 1 d).

2.2.1. Analyses

The COD, TKN, ammonia, nitrate, nitrite, the spectral absorbance coefficient (SAK) and phosphate were determined according to DEV [10]. The biochemical oxygen demand (BOD$_5$) was determined with a Sapromat (Selutech, Mössingen-Öschingen, Germany) according to DEV [10], the dissolved organic carbon (DOC) was analysed after filtration of samples through nitrocellulose filters (pore size 0.45 um) and acidification with HCl to remove bicarbonate and carbonate, by infrared spectroscopy with a TOCOR 2 DOC analyser (Maihak, Hamburg).

Fatty acids and gases (CO$_2$, H$_2$, CH$_4$) were analysed gas-chromatographically according to Gallert and Winter [11].

Pharmaceuticals and X-ray contrast chemicals in sewage and column effluent were analysed by Technologiezentrum Wasser, Karlsruhe, using solid-phase extraction, followed by GC–MS or HPLC-electrospray ionization MS–MS [12]. The sewage and column effluents were diluted with tap water to prevent matrix effects.

2.2.2. Bacterial counts

To determine colony-forming units (cfu) sewage was diluted up to $10^2$-fold with sterile, anoxic 0.9% NaCl, and 100 µl plated on Agar medium that contained (in g l$^{-1}$): yeast extract, 0.5; peptone, 0.5; caseine, 0.5; glucose, 0.5; starch, 0.5; pyruvate, 0.3; KH$_2$PO$_4$, 0.3; MgSO$_4$, 0.05; and agar–agar, 18. Plates of aerobic bacteria were inoculated in triplicate in a microflow safety cabinet (Nunc, Wiesbaden, Germany) and were incubated at 37°C for 2–3 d for colony growth. Plates for anaerobic bacteria were prepared in an anaerobic chamber (Coy Laboratories, Ann Arbor, USA) and incubated in N$_2$ atmosphere in stainless-steel jars for 2 weeks. For quantification of total and faecal coliform bacteria diluted sewage was plated onto Endo agar (containing (in g l$^{-1}$): peptone, 8.0; KH$_2$PO$_4$, 2.0; NaCl, 3.0; lactose, 10.0; Na$_2$SO$_3$, 2.5; Fuchsin, 0.3; and agar–agar, 18.0). Plates were incubated at 37°C for 1–2 d. The total coliforms (colourless colonies) and the faecal coliforms (black-greenish colonies) were counted.

2.2.3. Chemicals

All chemicals were obtained from Merck, Darmstadt. The source of the pharmaceuticals and X-ray media, used as standards, was as mentioned by Sacher et al. [12].

3. Results

3.1. Trickling behaviour of rain and dry weather sewage, moisture content and COD elimination in a 125-cm sand column

The total amount of sewage and the portions of sewage that left the sand in the periphery or the centre of the non-segmented 125-cm column varied with time.
Very high flow rates of around 121 d⁻¹ after start-up decreased within 2 weeks continuously to less than 21 d⁻¹, due to the formation of a filtering sludge layer on top of the sand. A complete clogging did, however, not occur within 2 years, during which about 2.5 cm of sludge accumulated on top of the sand. Methane was formed in the sludge and escaping biogas bubbles disturbed its compact structure periodically. This caused 10–20-fold increased trickling rates for 1–2 d every 2–3 weeks (not shown).

Sewage from rain weather periods contained 140–390 mg of total COD per litre (Table 1). After trickling through the sand column its COD was eliminated to finally slightly more than 100 mg l⁻¹ (Fig. 2a, weeks 1–5), irrespective of varying trickling rates from 500 ml d⁻¹ to more than 31 d⁻¹ (Fig. 2b). An almost identical COD concentration in the column effluent of the centre or the periphery was found, when 200–2200 ml d⁻¹ were trickling through the centre (weeks 1–5 in Figs. 2a and b), dewatering a volume of 7190 ml, and only around 400–1000 ml d⁻¹ were trickling through the periphery, dewatering a volume of 2240 ml. The same effluent COD concentrations were even found for lower trickling rates of sewage in the centre zone than in the periphery of the column (weeks 4–5 in Figs. 2a and b), representing highly different residence times.

The COD in the exfiltrating wastewater increased, however, from 100 to about 180 mg l⁻¹, when sewage from dry weather periods with a COD up to 1000 mg l⁻¹ was trickling through the sand. The COD of sewage from dry weather was not degraded below final concentrations of 150 mg l⁻¹, even at very low flow rates or long residence times (Figs. 2a and b, weeks 5–9).

The initial water content of 14.5 kg moist sand in the non-segmented column was 11. It increased with increasing elution rates of up to 600 ml d⁻¹ by 300 ml to 1350 ml and then remained constant (Fig. 3). To reach fully submerged conditions of the sand, a total volume of 3.3 l of water could be taken up by 13.5 kg dry sand. Thus, for trickling rates of 0–600 ml sewage per day the water saturation of the sand increased from 30% initially (moist sand, no trickling water) to 41% at a trickling rate of more than 600 ml d⁻¹.

3.2. Trickling of sewage through a segmented 125-cm sand column: elimination of carbon, nitrogen, pharmaceuticals and bacteria

In the segmented column the flow rate of sewage through the five consecutive 25-cm sand columns was also very high in the first 2 months with a maximum of more than 3500 ml d⁻¹. It then decreased to below 500 ml d⁻¹ (Fig. 4), presumably due to clogging by sludge on top of the sand of the uppermost column S5 and due to biofilm growth on the sand grains. Overall, the elution rate of sewage from the final column S1 was more even than that from the non-segmented column. The COD elimination during trickling of both more concentrated dry weather sewage and more diluted rain weather sewage was slightly better in the segmented column.

COD analyses in effluent of each of the five 25-cm columns revealed that column S5 contributed the major part to COD elimination from sewage of dry or rain weather (Fig. 5). The COD elimination continued during trickling of dry weather sewage through the columns S4,
S3, S2 and S1 to reach less than 180 mg l⁻¹ in the effluent (Fig. 5). If the initial sewage concentration was very high (supplementation of 0.5 g l⁻¹ glucose to the sewage during weeks 34–36, Fig. 5), the COD removal capacity of the uppermost column S5 was exceeded and a greater part of the COD was released into the subsequent columns S4–S1.

COD, DOC, total nitrogen and SAK of sewage from rain weather (Fig. 6a) or dry weather periods (Fig. 6b) decreased exponentially during trickling through the five 25-cm columns. Most of the particulate and biodegradable portion of the soluble COD of sewage from rain weather periods was removed after a trickling stretch of 50 cm (Fig. 6a, S4), whereas COD removal of sewage from dry weather periods required a trickling stretch of 100–125 cm (Fig. 6b, S1). The final COD concentration of column effluent after trickling of sewage from rain weather and after trickling of sewage from dry weather was 100 and 140 mg l⁻¹, respectively (Figs. 6a and b). The elimination with depth was faster for COD and DOC than for SAK and total nitrogen, indicating a slower biodegradability of aromatic and organic nitrogenous compounds.

In the segmented column the average COD elimination during more than 7 months of trickling dry weather sewage, with a COD_total of >600 mg l⁻¹ COD, was 79% for the total COD and 67% for the soluble COD (Table 2). The elimination efficiency was slightly better than in the non-segmented column, where 77% of the total COD were removed during dry weather conditions and 70% during rain weather conditions (Table 2). Ninety-four per cent of the ammonia and 66% of the organic nitrogen were removed during passage of the sewage through the five 25-cm columns of the segmented column (Table 2).

The sewage contained 2 × 10⁷ cfu of aerobic bacteria. The elimination of all bacteria was between 99.2% and 99.9% in the segmented column and between 96.2% and 99.9% in the single, 125-cm column (Table 3). Most of the coliform bacteria were retained in the first 25 cm of the sand and did not regrow, whereas aerobic bacteria were either washed into deeper sand layers or eliminated using alternative electron sources with 99% efficiency in the effluent of column S1. Only around 80% of anaerobic bacteria from the sewage were retained in the first 25 cm of sand. Within the next 50 cm of trickling the population density did not decrease, presumably due to an equilibrium of growth and adsorption or decay of bacteria. Only when there is no degradable COD left in columns S2 and S1 the number of anaerobic bacteria in the trickling sewage decreased to reach an elimination of 96.2% or 99.8% in the effluent of the single non-segmented column or in effluent of column S1 of the segmented column, respectively.

A wide spectrum of frequently medicated pharmaceuticals and iodinated X-ray contrast media was found
in sewage, of which only three of the pharmaceuticals were partially eliminated during sewage treatment (Table 4). Elimination of these three and the other pharmaceuticals by degradation, transformation to other substances or adsorption to the sand or the biofilm on the sand in the segmented column or the 125-cm column was either complete or at least proceeded to a higher extent than during sewage treatment (Table 4). Most of the X-ray contrast media, which were not at all eliminated during sewage treatment, were apparently little better eliminated in the segmented column than in the single 125-cm column.
4. Discussion

Leaking sewers or non-professionally constructed drain fields cause pollution of soil and groundwater by sewage. Visible leaks of sewers can be detected by in situ television inspection, whereas sewage in groundwater must be traced by analyses of non-degradable residues such as hormones and xenoestrogens [13], pharmaceuticals and X-ray media [12] or inorganic marker compounds, such as boric acid from detergents [2]. An estimation of pollution by sewage from groundwater analyses is, however, very inaccurate due to inaccurate quantification of the groundwater body and due to varying sewage compositions [14]. Both suspensa and the soluble COD in sewage reduce hydraulic conductivity of soil [15]. The solids cause a direct clogging of soil [16], whereas biofilm formation on sand particles along the trickling stretch changes the water content and the water conductivity.

After trickling of sewage through sand columns for more than 2 years the major clogging effect was caused by the sludge on top of the sand. When the sludge layer was removed to simulate back-flushing by infiltration of groundwater into sewers, the water conductivity of the sand increased for several weeks during application of diluted sewage from a rain period and for about 1 week during application of sewage from a dry weather period (data not shown). The initial water content of the moist
Sand increased with time from 30% (w/w) to 41% at increasing trickling rates up to 600 ml/d and then remained at 41% for higher trickling rates. Apparently the fine pores between sand and biofilm were saturated and a fast drainage through the large pores was prevalent.

The COD in column effluent depended mainly on the raw sewage and randomly on low or high trickling rates, as similarly observed in shallow sand filters [15]. Although the COD of raw sewage varied, the proportion of COD_{out}/COD_{in} during trickling of rain and dry weather sewage indicated a similar degree of stabilization (1:3.13 versus 1:3.33 mg l\(^{-1}\), respectively; Fig. 2), leaving behind the non-readily biodegradable COD. Aerobic sewage treatment or anaerobic sewage filtration through sand, followed by an aerobic post-treatment of the filtrate, was only able to remove maximally 94% of the total COD [6]. Thus, even if the distance of leaky sewers from the groundwater table would be much further than 1.25 m and anaerobic as well as aerobic conditions would exist, about 6% of the components of the sewage and/or soluble microbial lysis products [8] will reach the groundwater.

COD, DOC and SAK values of column effluent after 25–125 cm sand filtration (Fig. 6a) indicated that both the easily biodegradable COD or DOC and a major portion of components with a high SAK were removed during trickling of sewage through 25 cm of sand. Little more COD and SAK was degraded in the following column S4 and further elimination occurred in columns S3–S1.

The COD removal from sewage correlated with the dehydrogenase activity over the length of the sand column [6]. The highest dehydrogenase activity was found in the sludge layer on top and in the first few centimetres of the sand column. After trickling through 125 cm of sand only 1.3% of the initial activity were left. A similar elimination efficiency for \(\beta\)-glucosidase or phosphatase was reported after trickling of lake water through a cascade of gravel and sand filters in a water recharge plant [17]. In contrast to other reports on sand or soil filtration efficiencies (e.g. [18]) COD degradation was not complete after trickling through 25 or 50 cm of sand (Fig. 6b). More time and a longer trickling stretch was required.

Nitrogen removal depends on the availability of oxygen. If no oxygen was available, protein was desaminated and ammonia was released with the column effluent. If oxygen was not excluded, most of the ammonia was nitrified and then nitratized, leaving little ammonia and traces of nitrate in the effluent of the sand columns (data not shown). Whether anaerobic ammonia oxidation [19] was involved in nitrogen removal in the sand column, where only little COD for denitrification was available in the depth, remains to be tested. Nevertheless 66% of the total nitrogen of sewage was removed in the sand columns under anaerobic conditions, which was less than what was reported for a deeper sand filter, but much more than in a soil filter system [20].

More than 99% of the coliforms in sewage were effectively eliminated by the sludge layer on top of the sand and during trickling through the first 25 cm of sand, whereas the elimination of aerobic and anaerobic bacteria was reaching >99% only after 125 cm of sand filtration in the segmented column. In the non-segmented column, the column effluent contained some more bacteria. This could be due to non-interrupted random

<table>
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<tr>
<th>Substances</th>
<th>Sewage(^a) on columns (ng l(^{-1}))</th>
<th>Effluent of segm. column (ng l(^{-1}))</th>
<th>Effluent of 125 cm column (ng l(^{-1}))</th>
<th>Sewage after treatment(^b) (ng l(^{-1}))</th>
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<td>Benzafibrate</td>
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\(<\text{d.l.}>\) below detection limit.
\(^a\)Sewage of the sewage treatment plant of Karlsruhe after sand sedimentation and fat flotation.
\(^b\)Sewage effluent of the sewage treatment plant of Karlsruhe.
etchal. [23] found concentrations of 0.5–220 ng/l comparable with our data (Table 4). Hallin-Sorensen (1980) noted the effluent of a domestic sewage treatment plant, which was reduced to a level of 90%, Benzafibrate by 83% and Diclofenac by 69% in the sand columns. Ternes [7] reported elimination of Ibuprofen to a high degree in the sand columns, whereas others were almost not affected (Table 4). Barrett et al. [22] reported a reduction of faecal coliforms in a wetland system, constructed of a vertical sand/gravel filter followed by a coarse sand/gravel filter, at 10 d hydraulic retention time by 4.2 log10 decades. The removal of coliforms as an indicator of pathogenic organisms is important, although one cannot expect a complete removal of all bacteria, since even non-contaminated groundwater is not sterile.

Since most hospitals are releasing their wastewater into the public sewer system and private medication is leading to a significant contamination of sewage with all kinds of pharmaceuticals in the μg/l range, such as lipid lowering agents (e.g. benzafibrate), analgetics (e.g. Diclofenac, Ibuprofen, Naproxen) or X-ray media (e.g. Iopromide, Iomeprazole, Amidotrizoic acid, Iohexol or Iotalamic acid), these substances were also found in the sewage of Karlsruhe. Some of these pharmaceuticals were eliminated to a high degree in the sand columns, whereas others were almost not effected (Table 4). Ibuprofen, Benzafibrate and Diclofenac, for instance, were eliminated to a similar extent or even better than during a conventional sewage treatment process. However, residues are directly contaminating the groundwater. Together with the residual bacteria in sand-filtrated sewage this is not so much a problem of carbon release into groundwater than a problem of transferring antibiotic resistances and (xeno-)hormonal activities into the groundwater, which often serves as a source of drinking water.

Faecal coliforms were effectively retained after a few centimetres trickling through sand. Retention of other bacteria was little less effective, but reached efficiencies of 96–99.9% elimination after 125 cm trickling through sand.

5. Conclusions

Clogging by sludge reduced the sewage trickling rates through sand columns, but did not lead to a permanent self-healing of leaks.

The sand below leaking sewers increased its water content, presumably due to gel-like biofilm formation on the sand grains, reducing the void volume and causing a change in pore size distribution.

The percentage of COD elimination (COD_in:COD_out) after passage through 125 cm of sand was almost independent of the sewage. The residual soluble COD concentration was, however, significantly higher after trickling of dry weather sewage than after trickling of more dilute rain weather sewage. Most but not all of the COD removal takes place in the first 25 cm of sand at low trickling rates or in the first 50 cm of sand at higher trickling rates.

After trickling through 125 cm of sand pharmaceuticals were eliminated to a similar extent or even better than during a conventional sewage treatment process. However, residues are directly contaminating the groundwater. Together with the residual bacteria in sand-filtrated sewage this is not so much a problem of carbon release into groundwater than a problem of transferring antibiotic resistances and (xeno-)hormonal activities into the groundwater, which often serves as a source of drinking water.

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References


