

HUMIC SUBSTANCE FORMATION DURING WASTEWATER INFILTRATION

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Soil infiltration of wastewater effluents is a widely practiced method of treatment and disposal/reuse throughout the world. Systems employing this method range from small, individual household septic tank drainfields to large, municipal infiltration basins. Renovation of the wastewater results from a wide variety of complex physicochemical and biological processes. One set of processes is speculated to involve the accumulation of organic matter by filtration and sorption followed by formation of humic substances (Siegrist 1987). This humic substance formation can affect the performance of soil treatment systems by contributing to soil pore clogging and a reduction in hydraulic capacity, and by yielding reactive substances and an enhancement of purification processes.

While there has been a wealth of research into the nature and genesis of humic substances in terrestrial environments (e.g. Schnitzer and Khan 1978, Stevenson 1982, Aiken et al. 1985), there has been limited research of humic substance formation during soil infiltration of wastewater. The purpose of the research reported herein was to determine if humic substances can form under conditions typical of those present during wastewater infiltration into natural soil systems. This work was conducted during 1989 to 1990 as a collaborative effort between the Centre for Soil and Environmental Research, located in Aas, Norway and the Institute for Water, Soil and Air Hygiene located in Langen, West Germany. A synopsis of the research is provided herein while additional details may be found elsewhere (Siegrist et al. 1991).

OVERVIEW OF HUMIC SUBSTANCES IN SOILS

Natural Soil Humic Substance Properties and Genesis

For mineral soils, organic matter contents are often less than 5 to 6% (by wt.) near the surface decreasing to less than 1% with depth (Stevenson 1982). This organic matter is comprised of a wide variety of compounds that generally can be grouped into non-humic and humic substances. The non-humic fraction includes well-defined chemical compounds such as carbohydrates, proteins, amino acids, fats, waxes and low-molecular weight organic acids (Schnitzer and Khan 1978). These compounds do not persist for extended periods since they are readily attacked by soil microbes and utilized for energy or synthesis purposes. Soil humic substances include a heterogeneous mix of organic materials which are biogenic, refractory, hydrophilic, yellow-black in color and of high molecular weight (Schnitzer and Khan 1978, Aiken et al. 1985).

Humic substances (or humus) are traditionally divided into three fractions (i.e. humins, fulvic acids and humic acids) based upon solubility in water as a function of pH. Humins are comprised of the alkali insoluble fraction of humus. Humic acids represent dark colored organic materials which can be extracted from soil with various reagents, but which are insoluble in dilute acid.

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Fulvic acids are the colored organic materials which remain in solution after removal of the humic acids by acidification (Stevenson 1982). The fulvic and humic acids are not distinctly different groups of compounds, but rather two ends of a continuum.

Humic substances are macromolecules built upon basic structural units of aromatic rings of di- or trihydroxybenzene. Humic acids include nitrogen incorporated into cyclic positions as well as peripheral side chains. Carbohydrate residues are also present. Covalent bonding is involved in the stability of the macromolecule. Molecular weights may exceed several hundred thousand. Fulvic acids are built upon the same basic aromatic units but include less nitrogen within the structure and hydrogen bonding is primarily responsible for binding individual ring units together. Humic substances can exist naturally in fibers and shredded sheet structures (Schnitzer and Khan 1978, Tan 1985). Individual fibers have been observed with diameters as large as 0.5 to 4 um while net-like associations have been observed to extend over substantial areas (e.g. 100 um x 100 um). At the pH's normally encountered in soils, humic substances form sponge-like structures capable of trapping inorganic and organic molecules by virtue of the voids present or the large surface areas and adsorption capacities.

Humic substances exert profound effects on the physical and chemical properties expressed by the bulk soil in which they are present. In mineral soils they commonly form clay-metal-humus complexes which coat the surfaces of mineral particles (Schnitzer and Khan 1978, Aiken et al. 1982). Humic substances are hydrophilic and able to retain up to 20 times their own weight in water. They react with a variety of non-humic organic materials and make them less available for biodegradation. The reaction of organic molecules with mineral colloids has also been reported to alter water flow by forming physical barriers in the interlayer spaces as well as by changing mineral surfaces from hydrophilic to organophilic. In contrast, the reaction of organic molecules with mineral colloids can also lead to aggregation and improved permeability.

While many theories have been put forth to explain the genesis of humic substances in natural soils (Schnitzer and Khan 1978, Stevenson 1982, Aiken et al. 1985), there is consensus that humus synthesis is a multistage process that primarily includes the following: 1) decomposition of all organic residues, including lignin into simpler monomers, 2) metabolism of monomers with an accompanying increase in soil biomass, 3) repeated recycling of the biomass carbon (and nitrogen) with synthesis of new cells, and 4) concurrent polymerization of reactive monomers into high molecular weight polymers (humic substances). Amino compounds appear important in the genesis of humic substances. For example, in the polyphenol pathway, phenolic compounds are released during the degradation of lignin as well as during microbial decomposition of low molecular weight, non-aromatic compounds. The phenolic compounds are oxidized to quinones by phenoloxidase enzymes or by autooxidative processes. The quinones can self-condense under the conditions found in soils, but the reaction is greatly enhanced in the presence of amino compounds (e.g. amino acids). An alternative pathway, although believed to be of lesser importance, involves the condensation of amino compounds with reducing sugars.

Residual organic material and humic substances form from a wide variety of precursors, even readily degradable organic compounds such as sugars. Cool, humid, restricted aeration conditions with an influx of organic and nitrogenous materials can stimulate the accumulation of soil organic materials and the development of soil humic substances (Schnitzer and Khan 1978, Stevenson 1982). With time, higher molecular weight compounds are synthesized through condensation reactions, such as through the reaction of amino acids with reducing sugars and phenols. Ultimately, the true humic substances can be altered to coal-like products.

Formation of Humic Substances During Wastewater Infiltration

Researchers have speculated on the formation of humic substances during soil infiltration of wastewater, based largely on observations that organic materials accumulate in these systems under conditions which also favor humic substance formation (e.g. high influx of organic matter and nutrients including amino compounds, low temperatures, alternating redox potentials) and that the organic materials are slowly biodegradable (McGauhey and Krone 1967, Kristiansen 1982, Siegrist 1987). While the properties and genesis of humic substances in soil environments support this speculation, direct evidence has been lacking. Only recently, has relevant direct evidence been derived from other research where humic substances were shown to form during decomposition of municipal wastes deposited in a landfill (Newman et al. 1987, Filip and Smed-Hildmann 1988).

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MATERIALS AND METHODS

To evaluate whether humic substances could form during soil infiltration of wastewater, a laboratory experiment was carried out using column lysimeters. The experimental conditions were chosen to represent those typical of infiltration systems and to include a series of controls (Table 1). Some details regarding the materials and methods employed in this research are highlighted below while other details are given in Siegrist et al. 1991.

Experimental Apparatus

The two soils used for this study were naturally occurring, surface soils of sand and silt loam texture. A bulk volume of soil (upper 50 cm) was collected from each of two locations in southern Norway. In the laboratory, the field moist soil was sieved (8-mm mesh) and then two composite samples were collected from each volume and duplicate subsamples were analyzed for various soil properties by standard methods (Table 2). The field moist soil was then packed into column lysimeters constructed as shown in Fig. 1.

A total of 12 columns were packed for this experiment: four columns with each of the two test soils, two control columns for tapwater (TW) loading, and two columns as blanks (i.e. no liquid loading) (Table 1). After packing, each soil column was analyzed with a computer-assisted x-ray tomograph (Siemens Somatom 2). In the sand columns there were no apparent cracks or channels that might have been conduits for preferential flow through the column. However, in the silt loam columns, the interpedal pore channels associated with the crumb structure (4 to 8 mm diameter crumbs) were apparent. In all of the columns, at the soil infiltrative surface there was a thin zone (4 to 8 mm) of gravel/soil intermingling. After x-ray analysis, each soil column was mounted in a darkened, temperature controlled laboratory.

Septic tank effluent (STE) was collected weekly from a lift station serving 11 homes in a residential development near Aas, Norway. The collected STE was sampled for characterization purposes and then stored at 4°C pending application to the soil columns. Tapwater (TW) was obtained from a cold water faucet in the laboratory, the supply to which was untreated, surface water. Each column was initially dosed with 1 cm of TW on 16 December 1988. Six days later each column received 10 cm of the respective liquid (i.e. TW or STE). On the next day, once a day dosing of 5 cm/d was initiated and then continued for 60 days.

Monitoring

Samples of each volume of STE collected were taken at the time of collection and analyzed for various characteristics. Column infiltration rates were measured weekly by measuring the time from the initial application of a 5 cm dose to the completion of infiltration. A tracer study was initiated on the 9th day of operation when 1 mL of tritiated water was added to each column during normal dosing (final ³H concentration = 3,200 becquerels/mL). Tritium in the column percolate was monitored over time and breakthrough curves were developed.

Table 1. Experimental Conditions Evaluated.

| Treatment | Column No. | Soil Texture (USDA) | Soil Temperature (°C) | Wastewater Type ^a | Wastewater Loading (cm/d) |
|---------------------|------------|------------------------|--------------------------|------------------------------|------------------------------|
| <u>Test Columns</u> | | | | | |
| 1 | C2, C4 | Sand | 7 | STE | 5 |
| 2 | C8, C9 | Silt Loam | 7 | STE | 5 |
| 3 | C1, C3 | Sand | 20 | STE | 5 |
| 4 | C7, C10 | Silt Loam | 20 | STE | 5 |
| <u>Controls</u> | | | | | |
| 5 (=3) | C5 | Sand | 20 | TW | 5 |
| 6 (=2) | C6 | Silt Loam | 7 | TW | 5 |
| Blank | B1 | Sand | 7 | None | None |
| Blank | B2 | Silt Loam | 20 | None | None |

^a STE = domestic wastewater after septic tank pretreatment. TW = unsoftened tapwater derived from untreated surface water.

Table 2. General Properties of the Two Soils Used in this Experiment.

| Property ^a | Units | Sand | Silt Loam |
|-----------------------------|-------------------|--------|-----------|
| USDA Texture | - | Sand | Silt Loam |
| Moist Bulk Density (Packed) | g/cm ³ | 1.65 | 1.33 |
| Water Content | wt. % | 7.85 | 16.65 |
| pH | - | 6.15 | 5.05 |
| Total Organic Carbon | wt. % | 0.2 | 1.75 |
| Cation Exchange Capacity | meq/100g | 2.65 | 13.38 |
| Bacteria - Direct Count | Cell/g | 3.43E8 | 94.7E8 |
| Bacteria - YP Media | CFU/g | 5.22E6 | 60.4E6 |
| Bacteria - CSEA Media | CFU/g | 3.54E6 | 72.9E6 |

^a All results are based on duplicate analyses of field moist soil expressed on a soil dry weight basis.

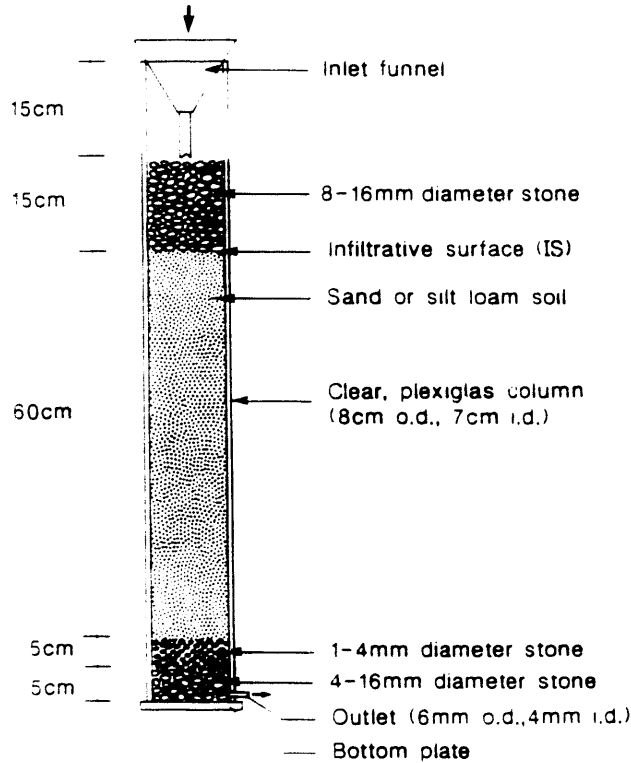


Figure 1. Schematic of the Soil Column Lysimeters Used in this Experiment.

After 30 days of operation, the percolate generated from each column following a dose was collected and characterized for pH, total organic carbon (TOC), total nitrogen and ammonium nitrogen.

After 60 days of operation, soil samples were collected from 0 to 2 and 10 to 12 cm depth in each of the columns. In addition, a soil sample from 0 to 2 cm depth was collected from a soil column containing a similar sand which had been dosed with the same STE for about 1 year as part of another experiment (column X). Soil TOC was determined by dry combustion. Total bacteria were measured by fluorescence microscopy while total heterotrophic bacteria were determined by plate counts made on two media: yeast extract (YP) and cold-extracted soil extract agar (CSEA). The viable bacteria count (i.e. colony forming units (CFU)) was estimated as the plate count.

Humic substance assays were made on liquid and soil samples as follows. Liquid samples (i.e. STE and TW) were filtered through a 0.45 µm membrane filter and acidified with HCL to pH 1.5.

Humic substances were separated by adsorption on microporous polystyrene beads (XAD 2, Serva 40820, FRG) and eluted with 0.2M NaOH. The eluate was acidified again (pH 1.5) and humic acids were allowed to precipitate overnight. They were then separated by centrifugation (10,000 g for 20 min at 4°C), dialyzed against deionized water for 3 days and freeze dried. The supernatant (fulvic acid fraction) was purified on a cation-exchange resin (AG MP-50, Bio-Rad 143-0821, USA) and freeze dried.

Soil samples were dried at 50°C until a constant weight, and then finely crushed in a mortar. Their organic carbon contents (C_{ox}) were estimated oxidimetrically by a wet combustion technique. Portions of 50 g of the dry soil were extracted with 200 mL of 0.1M H₂SO₄ to obtain a free fulvic acid fraction. An extraction mixture (400 mL) composed of 0.1M NaOH plus 0.1M Na₄P₂O₇ (1:1 v/v) was then used for a 24 hr soil extraction under N₂. The extracted soil was separated by centrifugation (5,500 g for 15 min at 4°C). The supernatant was acidified with HCL to pH 1.5 and humic acids were allowed to precipitate overnight. The precipitate was then separated by centrifugation, dialyzed and freeze dried as described above. Some preparations were dialyzed in the presence of 0.3M HF. The humic substance fractions were analyzed for optical properties as well as carbon, hydrogen and nitrogen contents. Electron microscopic analyses were also made.

RESULTS AND DISCUSSION

Soil Column Dynamics

During 60 days of operation, each column received a total of 3 m (11.5 L) of liquid characterized as outlined in Table 3. Long-term BOD measurements revealed that slowly biodegradable organic matter and nitrogenous substances were present, typical of this type of effluent (Siegrist 1987). The high TOC in the STE can be attributed to a fulvic acid fraction. The UV spectra of this fraction resembled fulvic acid with an absorption maximum of free phenols at 270 nm (Siegrist et al. 1991). Neither STE nor TW contained humic acids.

The dose infiltration rate (DIR) measurements are shown in Fig. 2 and 3. The DIR are expressed as a % of the DIR on the 7th day of loading to normalize the data for comparison as well as to eliminate any initial DIR effects due to entrapped air. In the sand columns there was a gradual decline in the DIR until the eighth week when a more dramatic loss occurred (Fig. 2). The DIR loss was more pronounced in the 7°C columns as compared to the 20°C columns and after 60 days of operation, these two columns were continuously ponded. The DIR gradually increased in the TW sand column. In the silt loam columns, the DIR loss was more gradual (Fig. 3). In contrast to the sand columns, the rate of DIR loss in the silt loam columns was greater at 20°C than at 7°C. The DIR of the TW control column declined somewhat, presumably due to gradual settlement/slaking of the silt loam crumb structure. Two soil clogging mechanisms are believed to be principally responsible for the DIR loss observed in this experiment. In the sand columns, biological clogging by organic matter is believed to be the most important while in the silt loam columns, mechanical clogging by settlement/slaking is believed most important. Observation of the soil columns following the experiment revealed that the silt loam columns had compressed in length by as much as 7%.

Based on the tracer study, percolation in the sand columns resulted in an average hydraulic retention time (HRT) of about 3 days (range of 2 to 6 days), while in the silt loam columns, the average HRT was about 4 days (range of <1 to 15 days). These data suggest that percolation through the silt loam columns may have occurred within the pedol micropores.

After 30 days of operation, the percolate samples collected from the sand columns were clear and free of particulates, while the samples from the silt loam columns were turbid and contained minor amounts of particulate matter. TOC removals were 85% or higher in all eight of the test columns (Table 4). In the sand columns, the TOC removals were higher at 20°C than at 7°C while just the opposite was observed in the silt loam columns. Complete nitrification was observed in all columns except the sand N columns at 7°C. In the sand columns the total N removals were higher at 20°C than at 7°C while the opposite was observed in the silt loam columns. The highest total N removals were achieved in the sand columns with the lowest nitrification. These results suggested that poor aeration within the sand columns at 7°C may have inhibited aerobic biological activity reducing the oxidation of amino compounds. The high N removals in these same columns may have been due to ammonium sorption and organo-incorporation, perhaps into humic substances.

Table 3. Composition of the STE Used in this Experiment.

| Characteristic | Units | Samples | Average | Std. Dev. | Range |
|-------------------------|---------|---------|---------|-----------|-------------|
| Temperature | °C | 8 | 7.2 | 0.6 | 6.5 - 8.5 |
| pH | - | 8 | - | - | 6.9 - 7.1 |
| Conductance | uS/cm | 7 | 734 | 24.9 | 731 - 763 |
| BOD ₅ | mg/L | 9 | 185 | 35.7 | 135 - 240 |
| TOC | mg/L | 4 | 91 | 46.5 | 98 - 130 |
| Total Solids | mg/L | 5 | 413 | 72.8 | 323 - 494 |
| Suspended Solids | mg/L | 3 | 37 | 12.2 | 23 - 45 |
| Total Nitrogen | mg-N/L | 5 | 75.3 | 8.3 | 64 - 85 |
| Ammonium | mg-N/L | 5 | 20.1 | 12.9 | 9.1 - 36.2 |
| Total Phosphorus | mg-P/L | 5 | 7.87 | 1.20 | 6.5 - 9.4 |
| Total Sulfur | mg-S/L | 5 | 24.2 | 3.8 | 19.5 - 30.1 |
| Bacteria - Direct Count | Cell/mL | 1 | 2.88E8 | - | - |
| Bacteria - YP Media | CFU/mL | 1 | 0.20E6 | - | - |
| Bacteria - CSEA Media | CFU/mL | 1 | 0.15E6 | - | - |

Infiltration Effects on Soil Properties and Humic Substance Formation

As a result of 60 days of operation, the sand columns increased in weight by ca. 3 to 5% compared to nearly 8% for the silt loam columns. The sand columns at 7°C exhibited higher weight increases. This difference may be attributed to the increased soil clogging suspected at the lower temperatures and the moisture retention associated with accumulated organic materials and decreased pore sizes.

The two sand columns operated at 7°C exhibited the greatest DIR loss (Fig. 2). At the time of soil sampling (i.e. after 60 days of loading), these two columns were gray-black in color at the infiltrative surface. This effect was only observed around the gravel locations, not underneath the gravel at the gravel/soil contact. The infiltrative surface of column X was black in color to a depth of 30 cm or more. This was presumably due to reducing conditions since these columns were continuously ponded for a prolonged time immediately prior to sampling.

For the sand columns, the water contents increased, with the increase most notable in the columns operated at 7°C (Table 5). For all STE sand columns, the water content at 0-2 cm depth was always higher than at 10-12 cm. Moreover, the TOC contents appeared to be elevated at the soil infiltrative surface as compared to 10-12 cm depth (ca. 20 to 40% higher). The exception was the TW column (C5) which had a depressed water content and TOC at the infiltrative surface. There were no similar apparent trends in water content and TOC for the silt loam column data.

Total bacteria levels were elevated in all sand columns as compared to fresh soil, and appeared to be directly correlated with the TOC levels (Table 5). In contrast, the bacteria levels in the silt loam columns did not change as a result of wastewater infiltration. The light and transmission electron microscopy of microorganisms from blanks, tapwater or wastewater treated soils showed the microflora being composed of both gram-positive and gram-negative bacteria, mainly short rods irrespective of the soil or treatment.

Clean sandy soil held at 7°C and the soil samples from sand columns percolated only with TW lost about 15% Cox during the 60-day operation period (Table 6). STE percolation at both temperatures resulted in an increased Cox content of 23% at both depths. This increase was not due to a higher content of fulvic or humic acids but rather humins, i.e. the humic material which was not extracted by acid or alkali. In all of the STE sand columns, the content of humins increased when compared with the fresh soil and the blank column (Table 6). An increase of 60% was estimated in the top 2-cm soil layer of the 7°C sand columns. This firmly bound humic material apparently contributed to the increase in soil water content and TOC, and perhaps to the clogging effects (Fig. 2).

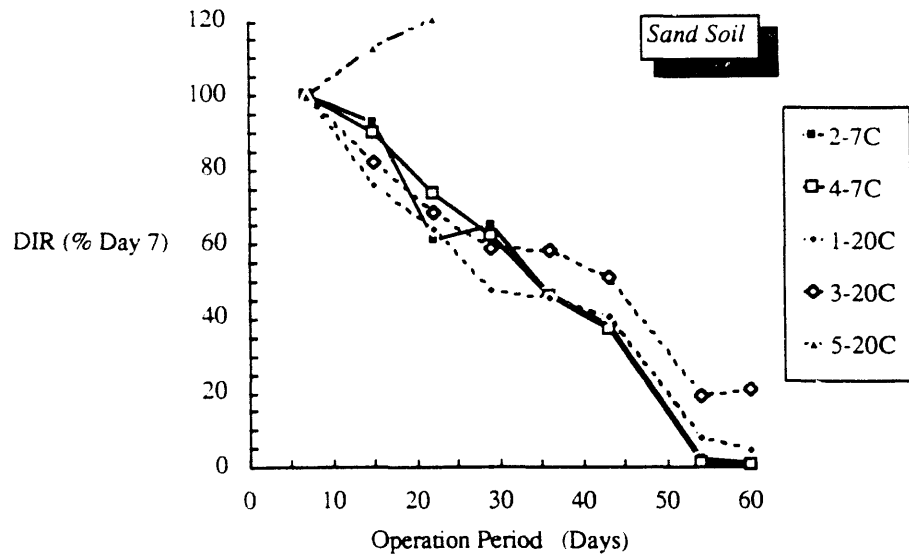


Figure 2. Sand Column Dose Infiltration Rate Versus Days of Loading.
 (Legend is column no. - temperature; all columns received STE, except no. 5)

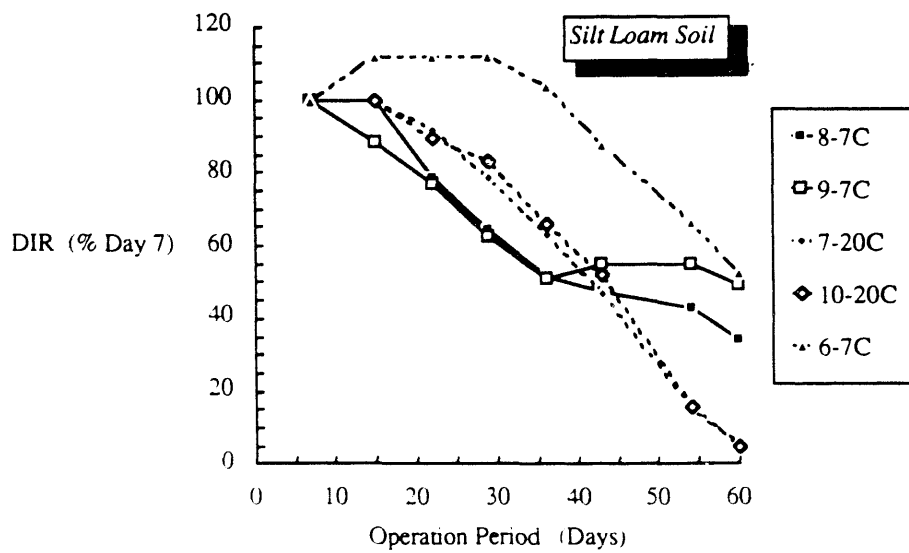


Figure 3. Silt Loam Column Dose Infiltration Rate Versus Days of Loading.
 (Legend is column no. - temperature; all columns received STE, except no. 6)

Table 4. Soil Column Percolate Composition After 30 days of Loading.

| Column | Soil | Temp. (°C) | pH | Conductance (µS/cm) | TOC (mg/L) | Total N (mg-N/L) | Ammonium (mg-N/L) |
|------------------------|-----------|---------------|------|------------------------|---------------|---------------------|----------------------|
| <u>Test Columns</u> | | | | | | | |
| - | STE | - | 7.0 | 763 | 115.0 | 75.8 | 36.2 |
| C2 | Sand | 7 | 6.95 | 654 | 13.8 | 43.0 | 26.8 |
| C4 | Sand | 7 | 7.01 | 637 | 13.8 | 44.0 | 23.0 |
| C1 | Sand | 20 | 6.46 | 560 | 7.85 | 51.5 | < 0.1 |
| C3 | Sand | 20 | 6.89 | 563 | 8.24 | 52.6 | < 0.1 |
| C8 | Silt Loam | 7 | 6.30 | 734 | 16.2 | 72.0 | < 0.1 |
| C9 | Silt Loam | 7 | 6.45 | 731 | 15.5 | 72.0 | < 0.1 |
| C7 | Silt Loam | 20 | 6.53 | 678 | 10.9 | 57.5 | < 0.1 |
| C10 | Silt Loam | 20 | 6.24 | 675 | 11.9 | 59.7 | < 0.1 |
| <u>Control Columns</u> | | | | | | | |
| - | TW | - | 7.3 | 180 | - | - | - |
| C5 | Sand | 20 | 6.66 | 183 | 9.17 | 3.7 | < 0.1 |
| C6 | Silt Loam | 7 | 6.95 | 172 | 5.52 | 5.0 | < 0.1 |

In silt loam soil percolated with water, the Cox content decreased by 9%, and remained almost unchanged in columns treated with STE irrespective of soil depth and temperature. The humin content, and also the overwhelming percentage of humins in the total Cox did not change substantially in the individual soil samples. The acid soluble fulvic fraction ("free" fulvic acids) in all soil column samples showed the same low concentration as in the blanks. However, fulvic acids which were extracted by alkali jointly with humic acids, were about 25% less than when compared with the original silt loam soil. The optical characteristics of all of the humic substance preparations are described in detail elsewhere (Siegrist et al. 1991).

When wastewater effluents are continually applied to soils, considerable changes in several soil properties may result, including the content and distribution of soil humic substances. Filip and Muller (1984) observed changes in the quantity and properties of soil humic substances when a brown soil was enriched over several weeks by liquid waste rich in easily utilizable carbon, such as fermentation residue. The quantity of humic substances increased with the amount of the waste applied, but their quality (optical density) showed an opposite correlation (Filip and Muller, 1984). In the experiment reported here, humins insoluble in alkali or acid showed increasing content in sandy soil treated for 60 days with STE. A slight decrease in the optical density was observed, but no difference was found in the content of fulvic and humic acids among individual samples. A special sample of a clogged soil (Column X), which was analyzed in parallel and which showed almost the same fulvic and humic concentrations as the test column soil samples, had a distinctly higher content of humins (1.72 % by wt.). This suggests that long-term infiltration of wastewater can yield substantial accumulations of humins in sandy soils.

SUMMARY AND CONCLUSIONS

A laboratory experiment was conducted to determine if humic substances can form and accumulate under environmental conditions typical of those found in soils during wastewater infiltration. Columns of two different soils (sand, silt loam) were intermittently infiltrated with 5 cm/d of septic tank effluent for a period of 60 days. Monitoring included observations of infiltration capacity and purification efficiency during operation, and the changes in soil properties as a result of 60 days of wastewater infiltration. Based on the results of this experiment, it can be concluded that humic substances can form in sandy soils during wastewater infiltration and that humins are the dominant form which accumulate. In silt loam soils, a similar effect could not be observed, due in part to the high initial concentration of these substances in the natural soil and the short 60-day observation period. Further investigation is necessary and appropriate to define the rate of humic substance accumulation and its role in soil pore clogging and wastewater purification.

Table 5. Basic Soil Properties After 60 Days of Wastewater Infiltration.

| Column | Soil | Temp. (°C) | Sample Depth (cm) | Water Content (wt.%) | TOC (wt.%) | Total Bacteria (Cellx10 ⁸ /g) |
|------------------------|-----------|---------------|-------------------------|----------------------------|----------------|--|
| <u>Test Columns</u> | | | | | | |
| C2 | Sand | 7 | 0 - 2 10 - 12 | 14.5 9.9 | 0.284 0.244 | 68.8 - |
| C4 | Sand | 7 | 0 - 2 10 - 12 | 12.7 9.6 | 0.305 0.243 | 63.9 - |
| C1 | Sand | 20 | 0 - 2 10 - 12 | 8.1 7.6 | 0.354 0.246 | 89.9 - |
| C3 | Sand | 20 | 0 - 2 10 - 12 | 8.4 6.9 | 0.312 0.244 | 52.5 - |
| C8 | Silt Loam | 7 | 0 - 2 10 - 12 | 23.5 24.8 | 1.70 1.76 | 125.1 - |
| C9 | Silt Loam | 7 | 0 - 2 10 - 12 | 24.4 24.3 | 1.73 1.69 | 111.9 - |
| C7 | Silt Loam | 20 | 0 - 2 10 - 12 | 23.8 24.6 | 1.68 1.63 | 141.1 - |
| C10 | Silt Loam | 20 | 0 - 2 10 - 12 | 23.1 24.7 | 1.56 1.66 | 88.8 - |
| <u>Control Columns</u> | | | | | | |
| C5 (TW) | Sand | 20 | 0 - 2 10 - 12 | 5.9 6.9 | 0.16 0.21 | 17.8 - |
| C6 (TW) | Silt Loam | 7 | 0 - 2 10 - 12 | 23.4 23.5 | 1.63 1.62 | 138.2 - |
| B1 | Sand | 7 | 0 - 2 | 8.2 | 0.223 | 53.2 |
| B2 | Silt Loam | 20 | 0 - 2 | 20.5 | 1.61 | 116.4 |
| <u>Fresh Soil</u> | | | | | | |
| - | Sand | | | 7.85 | 0.2 | 3.43 |
| - | Silt Loam | | | 16.65 | 1.75 | 94.7 |
| <u>Column X</u> | Sand | 7 | 0 - 2 | 16.6 | 0.214 | 38.8 |

Table 6. Contents of Soil Humic Substances After 60 Days of Wastewater Infiltration.

| Treatment | Temp (°C) | Depth (cm) | Total Cox (mg/100g) | Free Fulvic Acids (mg/100g) | Bound Fulvic Acids (BFA) (mg/100g) | Humic Acids (HA) (mg/100g) | Humins (mg/100g) |
|------------------|--------------|---------------|---------------------------|--------------------------------------|---|-------------------------------------|---------------------|
| <u>Sand</u> | | | | | | | |
| Clean Soil | - | - | 140 | 10 | 10 | 1 | 120 |
| Blank | 7 | - | 120 | 10 | 3 | 1 | 100 |
| TW | 20 | 0-2 | 120 | 10 | 10 | 1 | 110 |
| TW | 20 | 10-12 | 130 | 10 | 10 | 1 | 110 |
| STE | 7 | 0-2 | 180 | 10 | 10 | 1 | 160 |
| STE | 7 | 10-12 | 150 | 10 | 4 | 1 | 140 |
| STE | 20 | 0-2 | 180 | 10 | 10 | 2 | 170 |
| STE | 20 | 10-12 | 180 | 10 | 10 | 1 | 160 |
| <u>Silt Loam</u> | | | | | | | |
| Clean Soil | - | - | 1330 | 10 | 50 | 30 | 1250 |
| Blank | 20 | - | 1180 | 10 | 40 | 30 | 1110 |
| TW | 7 | 0-2 | 1200 | 10 | 40 | 30 | 1110 |
| TW | 7 | 10-12 | 1240 | 10 | 30 | 30 | 1160 |
| STE | 7 | 0-2 | 1360 | 10 | 40 | 30 | 1270 |
| STE | 7 | 10-12 | 1270 | 10 | 40 | 30 | 1190 |
| STE | 20 | 0-2 | 1290 | 10 | 40 | 30 | 1200 |
| STE | 20 | 10-12 | 1350 | 10 | 30 | 30 | 1280 |

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