Occurrence and Distribution of Pharmaceutical Organic Compounds in the Groundwater Downgradient of a Landfill (Grindsted, Denmark)

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Introduction

Usually landfill leachates contain specific organic compounds as BTEXs (benzene, toluene, ethylbenzene, and xylenes), chlorinated aliphatic hydrocarbons and chlorobenzenes originating from household chemicals and waste from small businesses (*1*). However, where industrial waste has been landfilled, the leachate may contain many other organic compounds (*2*).

Another paper of ours (*3*) described the distribution of commonly found organic compounds in the leachate plume downgradient of the Grindsted Landfill and discussed the fate of the organic compounds in view of the redox environments determined in the plume (*4*). In this paper, we describe the occurrence and distribution of organic compounds originating from waste from the pharmaceutical industry in the groundwater downgradient of the same landfill. According to our knowledge, this is the first report on pharmaceutical compounds in a leachate plume.

Pharmaceutical Compounds

A short presentation of the pharmaceutical compounds studied at the Grindsted Landfill is given. In the 1930s, sulfanilic amide was discovered to influence the course of bacterial infections. On the basis of systematical research for effective pharmaceuticals for infection control, more than 5000 different *sulfonamides* have been synthesized, all derivatives of sulfanilamide. However, less than 100 compounds have been produced commercially. In the 1970s, the use of sulfonamides declined, but was still the preferred medicine for control of urinary infections. The development of resistant bacteria, the discovery of numerous side effects, and the availability of other antibiotics have restricted the use of sulfonamides, which is an inexpensive pharmaceutical, to control nocardiosis (acute or chronic infection in the lungs) and toxoplasmosis (protozoan disease in central nervous system) and for veterinary purposes. Sulfonamides are today produced in China, Poland, and Hungary. The first synthetic step in the production of sulfonamides is a reaction between acetanilide and chlorosulfonic acid forming *N*-acetylsulfanilyl chloride. Sulfonamides are formed by reaction between the relevant amines and *N*-acetylsulfanilyl chloride and finally removal of the acetyl group by hydrolysis. Sulfonamides with heterocyclic groups can also be synthesised by a reaction between *N*-acetylsulfaguanidine and the appropriate β-diketo compound. As commerical chlorosulfonic acid contains small amounts of chlorosulfonic acid anhydride, low levels of *o*- and *p*-chloroacetanilide are formed by chlorination of acetanilide. These byproducts will be converted into *o-chloroaniline*, *p-chloroaniline*, and aniline, respectively, by the hydrolysis step in the sulfonamide production.

Early in this century, the sleep-inducing properties of barbiturates were discovered. Until the 1970s, barbiturates were the most common active substance in sleeping pills. However, in many western countries barbiturates are no longer important in the production of sleeping pills but are still used for certain medical treatments. Barbiturates are today produced in the U.K., France, Switzerland, United States, China, Hungary, Russia, and Denmark. 5,5-*Diallylbarbituric acid* (DBA) is produced by condensation of 5,5-diallyl malonic ethyl ester with urea. 2-*Methyl*-2-*n*-*propyl*-1,3-*propanediol* (MPP) is an intermediate of the sedidative pharmaceutical meprobamate, 2-methyl-2-*n*-propyl-1,3-propanediol dicarbamate. MPP is produced by a Cannizzaro reaction between 2-methylpentanal and formaldehyde. MPP is then reacted with ethyl carbamate to form meprobamate.

Propyphenazone (1,2-dihydro-1,5-dimethyl-4-(1-methylethyl)-2-phenyl-3*H*-pyrazol-3-one) is synthesized by condensation of acetyl acetic acid ethyl ester with phenylhydrazine and finally reacted with acetone. Propyphenazone is a mild analgesic (pain-relieving) pharmaceutical that is normally used in combination with other analgesics. Today, it is produced in Japan and Eastern Europe. *Tris*(2-*methylpropyl*) phosphate is a common antifoaming agent used in pharmaceutical production.

The above-mentioned chemicals have in the period from the early 1940s to the mid 1970s been included in pharmaceutical production in many countries all over the world. As a common practice in that period, waste from these pharmaceutical industries has been disposed of at landfills with no leachate collection systems. The chemicals as a part of the leachates may have entered into the surrounding aquifers.

The compounds studied, originating from a pharmaceutical production, are shown in Figure 1, and their chemical properties summarized in Table 1.

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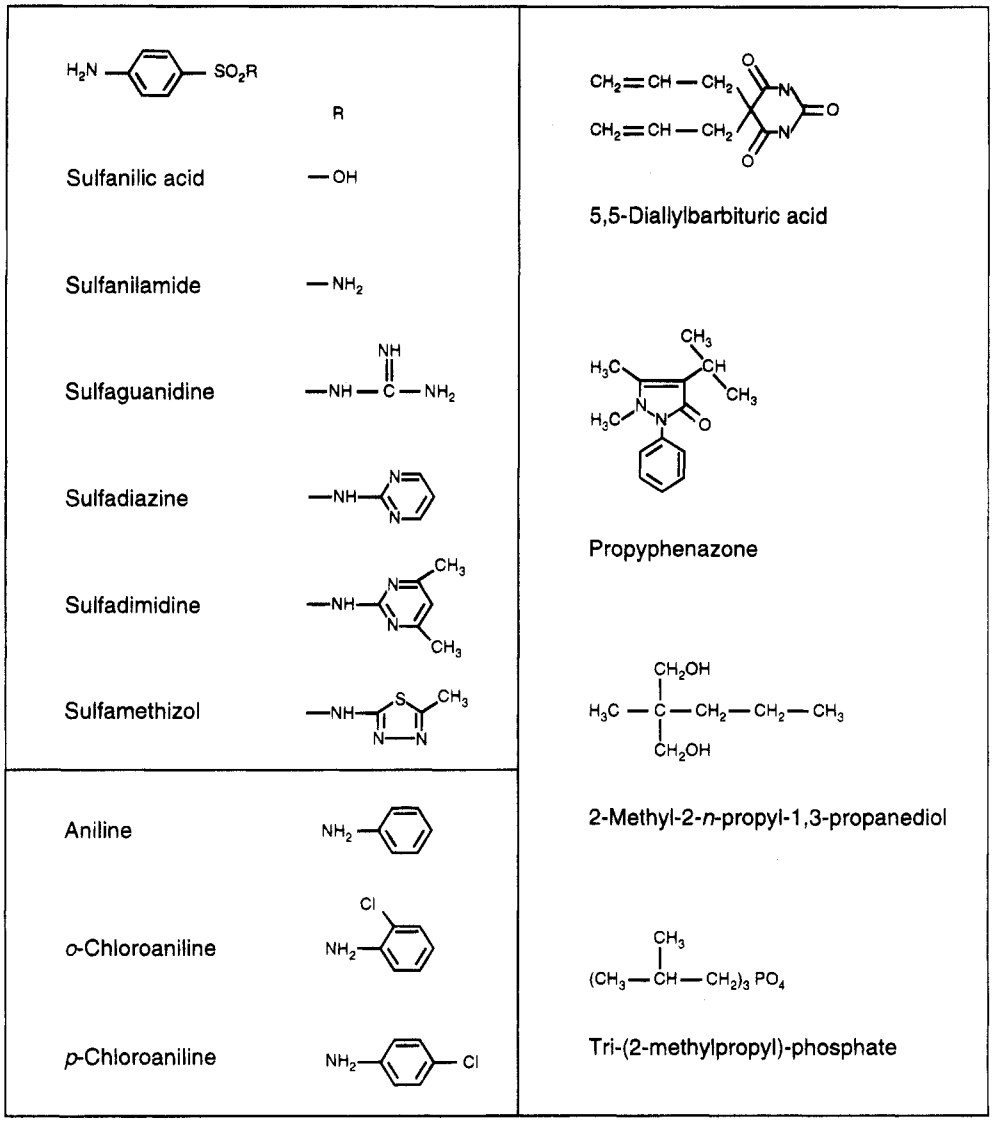

FIGURE 1. Chemical structure of the compounds studied.

Materials and Methods

Grindsted Landfill Site. The landfill is located directly on the surface in a flat landscape and has no leachate collection system (*5*). Landfilling of household waste took place from 1930 until 1977 when the landfill was closed. In the period 1962–1975, the landfill was also used for the disposal of waste from pharmaceutical production, both solid and liquid waste. The estimated amount of chemical waste from the pharmaceutical production is 85 000 t. The main part of the pharmaceutical waste was activated carbon and filter aid used in the purification of sulfonamides, barbiturates, and some water-soluble vitamins, but also calcium sulfate and sodium chloride including some pharmaceutical compounds and distillation residues were disposed of at the landfill.

The hydrogeology of the sandy aquifer underlying the landfill site and the redox conditions in the groundwater downgradient of the landfill have been investigated in detail as described by Bjerg et al. (*4*). Close to the landfill, methanogenic/sulfate-reducing conditions seem to exist, followed by an iron-reducing zone, a manganese-reducing zone, a nitrate reducing-zone, and finally an aerobic zone. The redox zonation indicates several transition zones where more than one redox process probably takes place at the same time.

Wells and Sampling. Groundwater samples for this study originate from 23 sampling points at nine distances from the landfill (Figure 2). The groundwater sampling procedure has been described by Rügge et al. (*3*). The transect was established along a flowline up to 300 m downgradient of the landfill (*4*). The wells consisted of iron pipes supplied with 10-cm screens as described by Lyngkilde and Christensen (*6*). At each sampling point, 1 L of sample was collected.

Analytical Methods. The analytical program was chosen with due respect of the waste landfilled. The analytical procedures used for determination of pharmaceutical compounds have been used for about 10 years to monitor industrial wastewater from the pharmaceutical production at Grindsted Products. Quantification was made by comparison with pure standard components. For all analyses, external standard method was used as the quantification principle. HPLC analyses were made by direct injection of the sample after filtration. For compounds determined by HPLC, the identity was verified by comparing UV spectra of the chromatographic peaks with references. Recovery for HPLC components were better than 95%, and no correction for recovery was made. Recovery for one component, 2-methyl-2-*n*-propyl-1,3-propanediol analyzed by GC was determined to 23 ± 3%,

TABLE 1

Chemical Properties of Compounds Studied[a]

	solubility (mg L^{-1})	log K_{ow}	pK_a
sulfanilic acid	10000	$<-2.0^c$	3.21[f]
sulfanilamide	7500	−0.83	10.43
sulfaguanidine	1000	−1.22	pK_b = 11.25[g]
sulfadiazine	80	−0.13	6.52
sulfadimidine	1000	0.27	7.00
sulfamethizole	250	0.54	5.45
5,5-diallylbarbituric acid	3300	1.19	–
propyphenazone	2400	2.32[d]	–
aniline	35000	0.90	pK_b = 9.30
o-chloroaniline	–	1.90	pK_b = 11.35
p-chloroaniline	3000[b]	1.83	pK_b = 9.85
tris(2-methylpropyl) phosphate	6000	–	*h*
2-methyl-2-*n*-propyl-1,3-propandiol	–	0.70[e]	*h*

[a] Log K_{ow} data are based upon compilation by Leo et al. (*13*), solubility and pK_a data are from ref 14, and pK_b data are from ref 15. A minus sign (−) indicates that data were not available. [b] Solubility from ref 8. [c] Log K_{ow} for sulfanilic acid was determined at pH = 7 (sodium salt) by the flask-shaking method following OECD Guideline (*9*) and UV spectroscopy. [d] Log K_{ow} for propyphenazone was determined by HPLC on reversed phase following the procedure described by Eadsworth and Moser (*10*). [e] Log K_{ow} value for 2-methyl-2-*n*-propyl-1,3-propanediol was determined by HPLC by flask-shaking method and GC (see Materials and Methods) following OECD Guideline (*9*). [f] pK_a value from ref 11. [g] pK_b value from ref 12. [h] No acid or base properties.

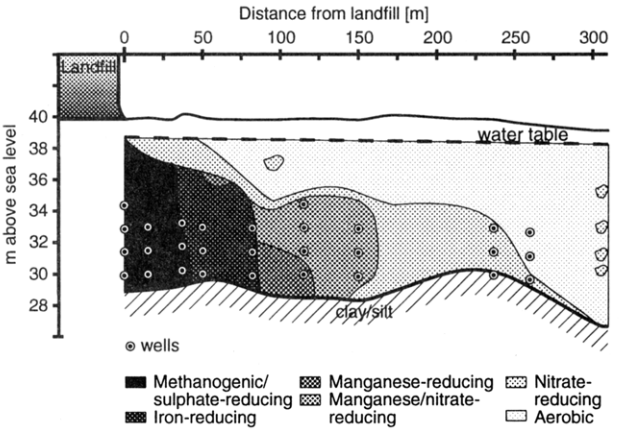

FIGURE 2. Location of sampling points and distribution of the proposed redox zones in the groundwater downgradient of the Grindsted Landfill.

and correction was made. Other GC components have recovery better than 92%, and no correction was made. Compounds analyzed by GC were identified by mass spectroscopy on a Hewlett Packard Model 5970.

Sulfonamides. HPLC determination was made using the Hewlett Packard 1090 HPLC system with binary gradient and diode-array UV detector. The 250 × 4.6 mm column packed with Lichrosorb RP-18 10-μm particles was from Merck and was operating at room temperature. Mobile phase A consisted of 0.5% of 20% tetrabutylammonium hydroxide in water, 5.0% methanol, and 94.5% of 0.5% acetic acid in water. Mobile phase B consisted of 0.5% tetrabutylammonium hydroxide in water, 30.0% methanol, and 69.5% of 0.5% acetic acid in water. The gradient program started with 100% A, linear gradient for 20 min to 100% B, which was kept until 50 min after injection, and then linear gradient for 1 min to 100% A, which was kept for 9 min before a new injection was made. Flow was 1 mL min^{-1}. Injection volume was 25 μL. The sample passed through a 0.45-μm filter before injection. Quantification was at 254 nm. Detection limit was 20 μg L^{-1}. Precision is 5% relative for results above 30 μg L^{-1}.

5,5-Diallylbarbituric Acid. HPLC determination was by the same equipment and column type as for sulfonamides, except that the particle size was 5 μm. Mobile phase A consisted of 10% acetonitrile and 90% of 10 mM ammonium carbonate in water. Mobile phase B consisted of 50% acetonitrile and 50% of 10 mM ammonium carbonate in water. The gradient program started with 100% A, linear gradient for 30 min to 100% B, and then linear gradient to 100% A over 1 min, which was kept for 14 min before a new injection was made. Flow was 1 mL min^{-1}. Injection volume was 25 μL. The sample passed through a 0.45-μm filter before injection. Quantification was at 240 nm. Detection limit was 10 μg L^{-1}. Precision is 10% relative for results in the range 10−300 μg L^{-1}.

GC-Identified components were determined using a modified method from the U.S. Environmental Protection Agency, EPA 625 (*7*). In a sample preparation, 1 L of sample was extracted with 75 mL of dichloromethane followed by two extractions with 60 mL. The combined phases were concentrated to 1−2 mL using a Kuderna−Danish evaporator. A total of 1 μL of the residue was injected into a United Packard GC Model 433 equipped with a Gerstel programmable temperature vaporizer Model KAS 2. The column was a 25-m FFAP column, 0.32 mm i.d. and 0.25 μm film thickness. Helium was used as the carrier gas. Injection was by a PTV-injector using 30 °C as the starting temperature, programming with 2 °C/s to 50 °C, which is kept for 1 min, and then programming with 12 °C/s to 300 °C. The detector was a FID at 250 °C. Detection limit was 10 μg L^{-1} for all compounds except tris(2-methylpropyl) phosphate, which is 1 μg L^{-1} because of the high hydrogen to oxygen ratio in the molecule that gives a better response at a FID detector. Precision is 30% relative for results below 100 μg L^{-1} and 15% relative for results in the range 100−5000 μg L^{-1}.

Results and Discussion

Organic Compounds Identified. The quantitative results for the 13 identified compounds are shown in Table 2 for sulfonamides and in Table 3 for the GC-identified compounds. The analytical program included 10 different sulfonamides, 10 barbiturates and intermediates, and byproducts from the production of vitamin B1 and B6, but only compounds identified above the detection limit are listed.

Meprobamate was not included in the analytical program, although it is expected to be present in the leachate plume. Because meprobamate is a dicarbamide, it cannot be determined by GC with the technique used. When meprobamate is present in a sample, its primary hydrolysis products, 2-methyl-2-*n*-propyl-1,3-propanediol monocarbamate (MPPMC), is observed in the GC chromatogram. A peak at that position was present in the samples closest to the landfill. This is believed to be an indicator for the presence of meprobamate. MPPMC has not been determined due to the nonquantifiable conditions for such high-boiling components. Whether MPPMC was present itself or only appeared as a GC-degradation product of meprobamate cannot be elucidated.

Distribution of Pharmaceutical Compounds in Leachate Plume. Many of the identified organic com-

TABLE 2

Distribution of Sulfonamides, NVOC, and Chloride in Groundwater as a Function of Distance from Grindsted Landfill[a]

		distance from landfill (m)								
compound	depth (m)	0	15	37	50	82	115	150	237	260
sulfanilic acid	5.5	1380	*b*	*b*	*b*	*b*	<20	*b*	*b*	*b*
	7	6470	6280	1000	90	50	*c*	*c*	<20	<20
	8.5	930	10440	5530	70	*c*	<20	<20	<20	<20
	10	5300	*c*	3160	1610	*c*	45	<20	*c*	<20
sulfanilamide	5.5	60	*b*	*b*	*b*	*b*	<20	*b*	*b*	*b*
	7	170	210	30	<20	<20	*c*	*c*	<20	<20
	8.5	40	300	140	<20	*c*	<20	<20	<20	<20
	10	120	*c*	170	40	*c*	<20	<20	*c*	<20
sulfaguanidine	5.5	110	*b*	*b*	*b*	*b*	<20	*b*	*b*	*b*
	7	1600	900	110	<20	<20	*c*	*c*	<20	<20
	8.5	190	280	480	<20	*c*	<20	<20	<20	<20
	10	1070	*c*	360	540	*c*	<20	<20	*c*	<20
sulfadiazine	5.5	100	*b*	*b*	*b*	*b*	<20	*b*	*b*	*b*
	7	480	720	170	<20	<20	*c*	*c*	<20	<20
	8.5	100	1160	440	<20	*c*	<20	<20	<20	<20
	10	480	*c*	410	80	*c*	<20	<20	*c*	<20
sulfadimidine	5.5	100	*b*	*b*	*b*	*b*	<20	*b*	*b*	*b*
	7	900	540	50	<20	<20	*c*	*c*	<20	<20
	8.5	230	900	310	20	*c*	<20	<20	<20	<20
	10	640	*c*	180	140	*c*	<20	<20	*c*	<20
sulfametizole	5.5	70	*b*	*b*	*b*	*b*	<20	*b*	*b*	*b*
	7	310	190	<20	<20	<20	*c*	*c*	<20	<20
	8.5	60	330	190	<20	*c*	<20	<20	<20	<20
	10	210	*c*	70	70	*c*	<20	<20	*c*	<20
NVOC	5.5	32.9	*b*	*b*	*b*	*b*	4.3	*b*	*b*	*b*
	7	79.1	74.0	45.4	39.2	47.8	*c*	*c*	1.3	1.2
	8.5	69.9	91.4	55.6	34.8	*c*	14.8	2.2	1.5	1.3
	10	82.9	*c*	65.3	44.3	*c*	15.3	3.7	*c*	1.3
chloride	5.5	163	*b*	*b*	*b*	*b*	119	*b*	*b*	*b*
	7	96	129	185	124	98	*c*	*c*	71	60
	8.5	89	135	256	94	*c*	21	43	42	56
	10	114	*c*	189	149	*c*	40	29	*c*	117

[a] Sulfonamides are in $\mu g\ L^{-1}$, NVOC is in mg of C L^{-1}, and chloride is in mg of Cl L^{-1}. [b] No sampling point. [c] Not enough water for analysis could be sampled at this depth.

pounds appeared in high concentrations (up to 18 000 $\mu g\ L^{-1}$) in the sampling points close to the landfill. Especially sulfanilic acid, propyphenazone, and 2-methyl-2-*n*-propyl-1,3-propanediol were present in extremely high concentrations compared to what has been observed for the specific organic compounds originating from household waste and waste from small businesses at the Grindsted Landfill (*3*).

The pharmaceutical compounds identified account for approximately 5% (with 1% and 24% as the extremes) of the NVOC (nonvolatile organic carbon) measured in the groundwater within the first 50 m downgradient of the Grindsted Landfill (Table 3). The sulfur content of the sulfonamides can explain about 5–25% of the organic sulfur determined at the landfill border.

The vertical distribution of the compounds showed a significant variation. At two sampling points at the landfill border with a vertical distance of 1.5 m, the concentration of 2-methyl-2-*n*-propyl-1,3-propanediol varied from 60 to 18 000 $\mu g\ L^{-1}$. At a distance of 50 m, it seemed like the concentration in the bottom part of the aquifer was larger than in the central part of the aquifer. A rapid decrease in concentration of the identified compounds (Tables 2 and 3) with distance can be observed, although there is a considerable scatter in the results from adjacent sampling points. At a distance of 150 m downgradient of the landfill, none of the pharmaceutical compounds could be detected in the groundwater.

Attenuation Processes. The disappearance of the pharmaceutical compounds may be caused by dilution, sorption, or degradation as discussed in Rügge et al. (*3*) for the other organic compounds found in the plume. The reduction in concentrations within a distance of 115 m is for some of the pharmaceutical compounds more than 50 times, which cannot be explained by dilution, when compared to chloride acting as a conservative tracer in this part of the aquifer (Table 2). All the compounds shown in Tables 2 and 3 are highly water-soluble with low octanol/water distribution coefficients, indicating that they will be highly mobile and probably migrate with the same velocity as the groundwater in the aquifer. Among the identified compounds, propyphenazone has the highest octanol/water distribution coefficient ($\log K_{ow} = 2.32$), which means that propyphenazone is expected to show the strongest retardation in the aquifer. However, propyphenazone is present at 70 $\mu g\ L^{-1}$ in a distance of 115 m from the landfill, where the concentrations of all other compounds studied except sulfanilic acid are below the detection limit. This observation combined with the expected high mobility of the studied compounds indicates that sorption probably is not significant in this case. The observed attenuation within a short distance from the landfill border may therefore be caused by degradation; however, no direct proof can be presented as no compounds that exclusively can be regarded as degradation products were identified. Sulfaguanidine, sulfanilamide, and sulfanilic acid can be

TABLE 3

Distribution of Different Compounds Identified in Groundwater as a Function of Distance from the Grindsted Landfill[a]

compound	depth (m)	distance from landfill (m)						
		0	15	50	115	150	237	260
5,5-diallylbarbituric acid	5.5	–	*b*	*b*	*b*	*b*	*b*	*b*
	7	135	–	10	*c*	*c*	–	–
	8.5	–	205	–	<10	–	–	<10
	10	–	*c*	25	–	–	*c*	–
propyphenazone	5.5	300	*b*	*b*	*b*	*b*	*b*	*b*
	7	4000	–	–	*c*	*c*	–	–
	8.5	1000	–	30	–	<10	<10	–
	10	2900	*c*	300	70	–	*c*	<10
aniline	5.5	<10	*b*	*b*	*b*	*b*	*b*	*b*
	7	720	–	–	*c*	*c*	–	–
	8.5	<10	–	70	–	<10	<10	–
	10	1100	*c*	100	<10	–	*c*	<10
o-chloroaniline	5.5	<10	*b*	*b*	*b*	*b*	*b*	*b*
	7	110	–	–	*c*	*c*	–	–
	8.5	30	–	<10	–	<10	<10	–
	10	15	*c*	10	<10	–	*c*	<10
p-chloroaniline	5.5	<10	*b*	*b*	*b*	*b*	*b*	*b*
	7	20	–	–	*c*	*c*	–	–
	8.5	50	–	<10	–	<10	<10	–
	10	35	*c*	<10	<10	–	*c*	<10
tris(2-methylpropyl) phosphate	5.5	<1	*b*	*b*	*b*	*b*	*b*	*b*
	7	80	–	–	*c*	*c*	–	–
	8.5	<1	–	<1	–	<1	<1	–
	10	70	*c*	8	<1	–	*c*	<1
2-methyl-2-*n*-propyl-1,3-propanediol	5.5	60	*b*	*b*	*b*	*b*	*b*	*b*
	7	18000	–	–	*c*	*c*	–	–
	8.5	440	–	230	–	<10	<10	–
	10	720	–	80	<10	–	–	<10

[a] All concentrations in $\mu g\ L^{-1}$. (–), not analyzed. [b] No sampling point. [c] Not enough water for analysis could be sampled at this depth.

degradation products of the heterocyclic sulfonamides, but the concentrations in the leachate plume do not indicate any increase in concentrations of these compounds with distance.

The prevailing redox conditions in the part of the plume where the pharmaceutical compounds seem to be degraded are strongly anaerobic (Figure 2). Most of the compounds are attenuated in the zone characterized as methanogenic/sulfate-reducing and iron-reducing according to ref 4. Sulfanilic acid and propyphenazone have been found in the mixed transition zone between the manganese-reducing zone and the nitrate-reducing zone. However, also for these compounds the largest reduction seems to take place under more reduced conditions.

The degradation of the sulfonamides will release sulfur, which may be oxidized and observed as sulfate in the plume. However, calculations of the theoretical concentrations indicate that this is not a major source of sulfate, and the increasing sulfate concentration observed 100–120 downgradient of the landfill probably has another origin as discussed by Bjerg et al. (*4*).

Comparison with Other Studies. The large attenuation observed within the first 50 m from the landfill apparently takes place under methanogenic/sulfate-reducing and iron-reducing conditions. This is in accordance with the observed attenuation pattern for the aromatic and the bicyclic compounds at the Grindsted Landfill (*3*). However, this is to our knowledge the first study where the attenuation of pharmaceutical compounds has been demonstrated in a landfill leachate plume. Based on a literature survey, the published reports on anaerobic degradation of the identified pharmaceutical compounds are extremely few. Kuhn and Suflita (*2*) have studied in a laboratory study with aquifer slurries amended with leachate the degradation of sulfanilic acid and aniline under methanogenic and sulfate-reducing conditions. Sulfanilic acid was neither degradable under methanogenic nor sulfate-reducing conditions. Aniline was recalcitrant under methanogenic conditions while biodegraded after a lag phase of several months under sulfate-reducing conditions. The reported recalcitrance of sulfanilic acid under methanogenic and sulfate-reducing conditions support our findings in the Grindsted leachate plume.

The results presented here indicate strong attenuation of pharmaceutical compounds under strongly anaerobic conditions. However, further investigation will be necessary in order to provide direct evidence of the degradability of the pharmaceutical compounds under specified redox conditions.

Acknowledgments

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