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The Fate of Organic Pollutants in Wastewater and Sludge Treatment: Significant Processes and Impact of Compound Properties

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Abstract. The fate of organic pollutants during wastewater and sludge treatment is determined by three main processes: gas exchange, sorption to suspended solids, and biodegradation. The influence of these processes differs strongly depending on the physicochemical properties of the individual compound and the particular treatment stage. For the assessment of the fate of trace pollutants in wastewater treatment, the impact of these processes must be evaluated. An acceptable removal in mechanical-biological wastewater treatment is achieved for hydrophilic compounds if they are rapidly degradable under aerobic conditions. Substances with lipophilic or amphiphilic properties should be degradable under aerobic and anaerobic conditions in order to prevent accumulation in digested sewage sludges. This article presents recent and current investigations at EAWAG which deal with the fate of selected organic substances in municipal wastewater and sludge treatment.

Introduction

Discharge to wastewater streams is one of the principal disposal routes of chemicals used in households and industry. Therefore, it is highly important to understand the mass flow and changing composition of chemicals as they pass through wastewater treatment plants (WWTP). Wastewater treatment plays an important role in the life cycle of chemicals by reducing the discharges to the environment, by altering the composition of chemical mixtures, and by acting as

point sources of chemicals to the aquatic environment. Field studies conducted at full-scale WWTPs are needed to validate fate predictions that are based upon laboratory studies. In addition, improved exposure and subsequent risk assessment of chemicals toward aquatic organisms, ecosystems, and water quality can be better evaluated.

Before an organic or inorganic chemical is manufactured and used in large quantities and eventually released to the environment, its physical, chemical, and biological properties must be assessed. This is especially true for substances that, because of their application, are released into wastewater. Among the latter are surfactants, complexing agents, pharmaceuticals, and aromatic sulfonates.

Field studies are based on two different approaches. In *monitoring studies*, pollutant concentrations are measured over long periods of time in order to determine changes as a function of time. Also spatial differences can be covered by monitoring. Monitoring data allow to assess the impact of regulatory and voluntary measures. However, for the understanding of the environ-

mental fate of pollutants *process-oriented field studies* are needed. Process-oriented field studies focus on mass flows and the dynamic behavior in defined systems such as a WWTP. These studies enlarge our knowledge about the dominant processes affecting the environmental fate of pollutants and they are significantly enhanced when mathematical modeling is applied to interpret these results.

This article describes activities at EAWAG concerning the behavior of organic pollutants in wastewater and sludge treatment. After the discussion of research concepts, several case studies are presented which focus on different compound classes.

Research Concepts for Process-Oriented Field Studies in Wastewater Treatment Plants

A municipal wastewater treatment plant usually consists of a mechanical treatment (screening, grit removal, and primary clarification), biological treatment (*e.g.*, removal of organic compounds, nitrification, and often simultaneous phosphate precipitation in an activated sludge system), and sludge treatment (mesophilic digestion and dewatering) (*Fig. 1*). The excess sludge (secondary sludge) formed during activated sludge treatment is led back into the inflow to the primary clarifier and is then thickened with primary sludge. Relevant to the environment are the treated wastewater effluents, the exhaust gases from aeration, and the sludge which is often used as fertilizer in agricultural soils.

The tiered research concept for field studies on the fate of organic pollutants in mechanical-biological wastewater treatment are outlined in *Table 1*. Prerequisite for successful investigations is the availability of suitable analytical methods for the specific and quantitative determination of chemicals and their most important transformation products in complex environmental matrices. The research approach can be described on the basis of four distinct levels of sophistication. TIER 1 should be only used in screening studies to obtain preliminary results. It should rather be attempted to reach at least TIER 2 yielding conclusive elimination values and reliable information on mass flows and mass balances. TIER 3 needs an extensive sampling program, but gives the most detailed assessment on the fate of chemicals in a WWTP. In TIER 4, the concentrations in WWTPs are calculated with mathematical models including different trans-

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port, transfer, and transformation processes. These processes determine the temporal and spatial distribution of a pollutant in a WWTP.

Significant Processes Governing the Environmental Fate – Properties of Compounds

To evaluate the environmental effects of a chemical compound, its environmental fate has first to be studied for assessing possible damaging effects. The behavior of chemicals during wastewater treatment is mainly determined by the following three processes [1]:

- gas exchange with the atmosphere,
- sorption to the suspended solids and to the formed biomass,
- aerobic and anaerobic biodegradation.

The influence of these three processes differs strongly depending on the physico-chemical properties of the particular compounds and on the type of treatment stage.

Gas Exchange with the Atmosphere: Air/Water Partitioning

Air/water partitioning of a volatile compound is important for the stripping of a compound during biological treatment and aerated grit removal.

At thermodynamic equilibrium, the distribution between gaseous and aqueous phases may be described by Henry's law: $H = C_g/C_w$

C_g, C_w are the concentrations of a compound in the gaseous and in the aqueous phase, respectively. The less volatile a compound, the smaller its Henry's law constant (H), and the faster the compound achieves equilibrium in a rising air bubble with the dissolved compound in the aqueous phase. Compounds with $H < 2$ reach almost saturation equilibrium. This is the case e.g., for chloroform ($H = 0.13$), trichloroethylene ($H = 0.41$), and tetrachloroethylene ($H = 0.77$) [1]. The behavior in wastewater treatment is currently not in the focus at EAWAG despite the general importance of volatile organic chemicals.

Sorption to Particles: Water/Solid Partitioning

The distribution of a compound between the solid phase (suspended matter: particles and formed biomass) and the aqueous phase significantly influences the behavior and biodegradation of a compound during wastewater treatment. Hydrophobic compounds adsorb to a large extent to the solid organic phase. Hydrophilic and ionized compounds remain primarily in the aqueous phase.

Table 1. Research Concept for Field Studies in Wastewater Treatment Plants

PREREQUISITE:	– analytical method for individual compound determination – parent compounds – transformation products
TIER I:	– grab samples of influents, effluents, and sludges – concentrations \Rightarrow 'elimination'
TIER II:	– composite (24 h) samples of influents, effluents, and grab samples of sludges – dissolved and particulate fractions – mass balance in wastewater treatment – concentrations \Rightarrow mass flows \Rightarrow elimination
TIER III:	– diurnal fluctuation (input/output) – dynamic behavior
TIER IV:	– mathematical modeling using transport, transfer, and transformation processes – steady state – dynamic behavior

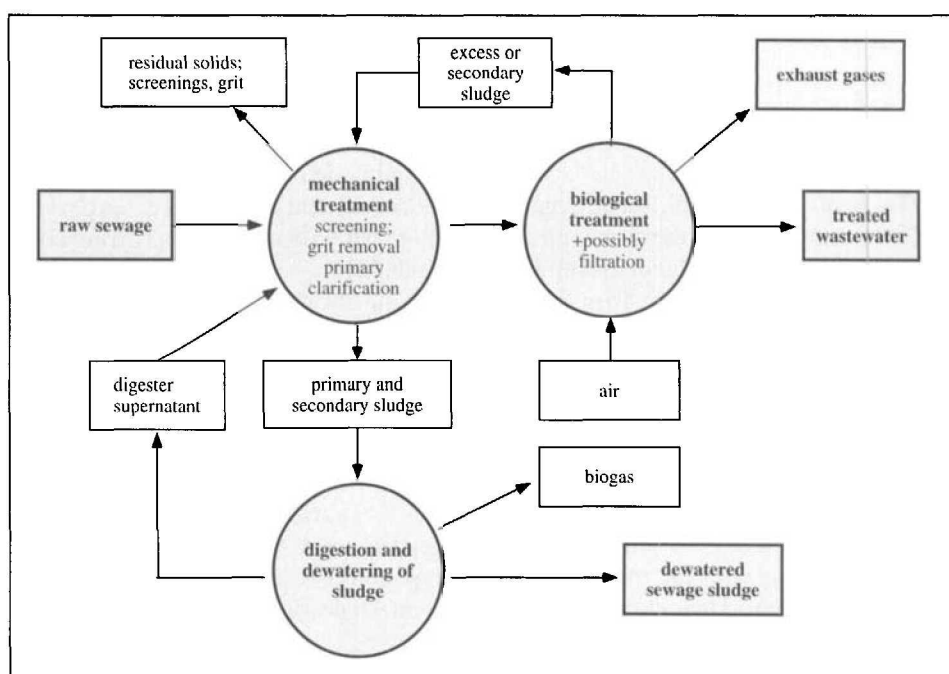


Fig. 1. Scheme of wastewater treatment with mechanical and biological stages including anaerobic digestion and dewatering of sludge [1]

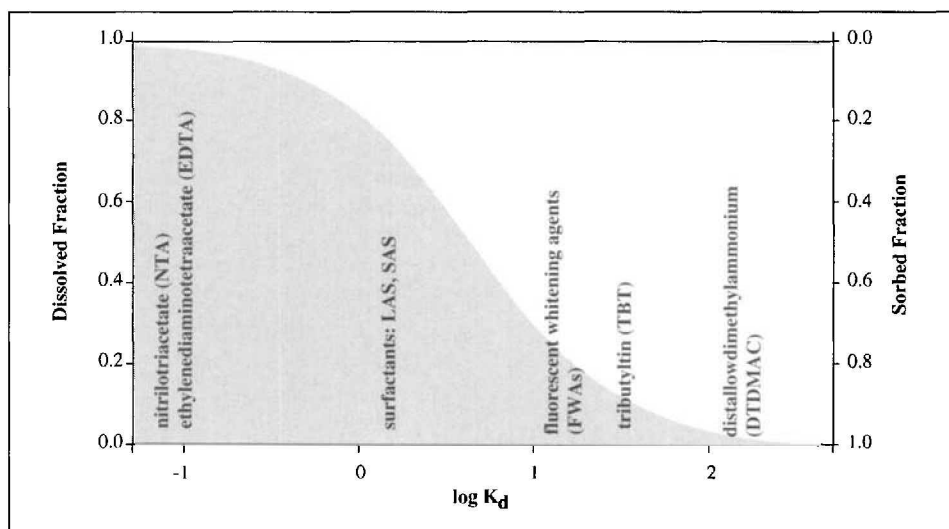


Fig. 2. Dissolved and sorbed fractions of organic trace pollutants as a function of the distribution coefficient of sorption (K_d ; $m^3 kg^{-1} d.m.$). The concentration of suspended matter of $200 g m^{-3}$ in the influent to a municipal wastewater treatment plant is assumed [1].

Amphiphilic, surface-active chemicals, are adsorbed to solids despite their polar or charged moieties (*e.g.*, linear alkylbenzenesulfonates (LAS), quaternary ammonium surfactants). This behavior is caused by the hydrophobic alkyl and/or aryl substituents.

The higher the sorbed fraction of a compound, the more efficiently it is removed in the mechanical stages of treatment. Compounds which mainly remain in the aqueous phase are transferred to the biological treatment stage.

The simplest approximation for the solid/water partitioning is given by:

$$K_d = C_s/C_w [m^3 kg^{-1} d.m.]$$

where K_d is the distribution coefficient given by the ratio of the concentration in the solid phase (C_s ; $g kg^{-1} d.m.$) and in the aqueous phase (C_w ; $g m^{-3}$) (*d.m.* = dry matter). In this equation an approximately linear sorption for trace compounds is assumed.

The distribution of different organic trace pollutants between the dissolved and sorbed fraction in the influent of a municipal WWTP is shown in *Fig. 2*.

Aerobic and Anaerobic Biodegradation

Depending on the concentrations of important electron acceptors (O_2 , NO_3^- , SO_4^{2-} , CO_2) in a specific environment, those organisms will establish that are able of utilizing the most favorable and most abundant electron acceptor, *i.e.*, the electron acceptor with the most positive reduction potential. This sequence results

in environments of characteristic microbial metabolism including aerobic respiration, denitrification, sulfate reduction, and methanogenesis. If sufficient molecular oxygen is present, aerobic respiration dominates.

Microbial degradation of trace compounds in WWTPs occurs in the presence of naturally occurring carbon compounds originating from the decomposition of plant and animal material. For heterotrophic microorganisms, which are responsible for the bulk of carbon elimination in WWTP, the bioavailability of carbon compounds is a main limiting growth factor. Because the hydrolysis rate of particulate organic matter is slow, a long residence time of bacteria in a system allows a more complete degradation of the polymeric carbon compounds by the bacterial population. Trace pollutants are estimated to make up only a minor part of the total carbon used for a microbial growth (usually less than 1%). Consequently, degradable synthetic organic chemicals found in the environment are in most cases degraded together with naturally occurring compounds [2].

In wastewater treatment the biodegradability of a compound depends on the redox conditions of the water and the average washout rate (dilution rate = D_x) of bacteria in the system. The smaller the washout rate, *i.e.*, the longer the residence time (solid retention time $SRT = 1/D_x$), the more diversified the biomass becomes; degradation of normally nondegradable compounds becomes possible. In the biological stage of treatment, aerobic and

denitrifying conditions prevail with biomass residence time of $SRT = 4-15$ d. In Switzerland, during digestion of sludge, anaerobic conditions predominate with hydraulic residence times of 20-40 d [1].

Furthermore, the hydrophilicity of a compound determines its fate in the WWTP. Dissolved pollutants reach the activated sludge reactor where aerobic degradation will occur. Compounds with hydrophobic and amphiphilic moieties are transferred to the anaerobic sludge treatment directly by the way of primary clarification because of their association to wastewater particles. Consequently, pollutants that are not biodegradable under methanogenic conditions will accumulate in the digested sewage sludge.

Fate of Trace Compounds – Case Studies

Table 2 summarizes the results of several recent and current case studies carried out at EAWAG dealing with the fate of organic compounds occurring in municipal wastewater.

Chelating Agents: Different Fate of NTA and EDTA

The chelating agents NTA (nitrilotriacetate) and EDTA (ethylenediaminetetraacetate) were comprehensively investigated [14-16]. In nitrifying wastewater treatment plants, NTA is easily biodegradable; EDTA in contrast is not [3][4]. For both chemicals, sorption to particles is less than 1%. Average concentrations in

Table 2. Behavior of Trace Pollutants in Wastewater Treatment Plants at Nitrifying Conditions and at Temperatures above 15°

Substance	Sorption to sludge [%]	Aerobic biodegradation [%]	Residual levels in secondary effluent [%] of influent	Refs.
Nitrilotriacetate (NTA)	–	95	< 5	[3] [4]
Ethylenediaminetetraacetate (EDTA)	–	–	100	
3-Nitrobenzenesulfonate	–	98	2	
Naphthaleneosulfonates	–	98	2	[5]
Naphthalene-1,5-disulfonate	–	5	95	
Naphthalene-1,6-disulfonate	–	96	4	
Linear alkylbenzenesulfonates (LAS)	26	73	1	[6]–[9]
Secondary alkylsulfonates (SAS)	16	84	< 1	[9]
Ditallowdimethylammonium chloride (DTDMAC)	23–51	36–43	6–41	[10] [11]
Fluorescence whitening agents (FWA)	53–98	–	2–47	[12]
Tributyltin (TBT)	90	–	10	[13]

influent are ca. 100–1000 $\mu\text{g NTA/l}$ and 10–500 $\mu\text{g EDTA/l}$. Mass balances comprising sampling periods of several days showed that no significant elimination of EDTA occurred by biological or chemical processes during wastewater treatment, whereas NTA was efficiently degraded (> 95%). The NTA degradation was successfully simulated with a simple activated sludge model that includes growth of NTA degraders on NTA [17].

Because EDTA is persistent, its load in the secondary effluent is five times higher than that of NTA (Fig. 3). Besides from naturally occurring complexing agents among the degradation products, the dissolved fraction of heavy metals is dependent on EDTA. Field studies showed that EDTA speciation changes from input to output because FeEDTA complex is formed from other metal EDTA complexes (mainly calcium and magnesium) after addition of iron(II)-containing solutions for the phosphate removal in the aeration tanks. As long as the speciation of EDTA in wastewaters is dominated by FeEDTA, and aerobic conditions are maintained, the remobilization of common heavy metals out of sewage sludge is not significant [4].

In the case of NTA, biological degradation plays the key role in its removal from wastewater and surface water. Other processes, e.g., photochemical transformation or sorption are not significant. At EAWAG, a number of different NTA-degrading microbial strains have been isolated and characterized [18][19]. Using antibodies that bind specifically to the cell surface of these strains in combination with immunolabelling techniques, strong evidence was obtained that NTA-degrading microorganisms are ubiquitously distributed in the environment. In activated sludge, the fraction of NTA-degraders was as high as 0.1–1% of the total microbial population, whereas this range was an order of magnitude lower in natural waters. The fact that the number of NTA-degrading bacteria was not significantly different in treatment plants that varied considerably with respect to their NTA elimination efficiency, indicated that NTA degradation might be regulated at the level of enzyme synthesis and not of enrichment of NTA-degrading organisms. This indication was further substantiated by recent results obtained for the presence of the key enzyme of NTA metabolism in aerobic NTA-degrading bacteria, i.e., the NTA monooxygenase. The data suggest that NTA-degrading micro-organisms are present in treatment plants at fairly high concentrations, growing mainly with alternative substrates (together with a little

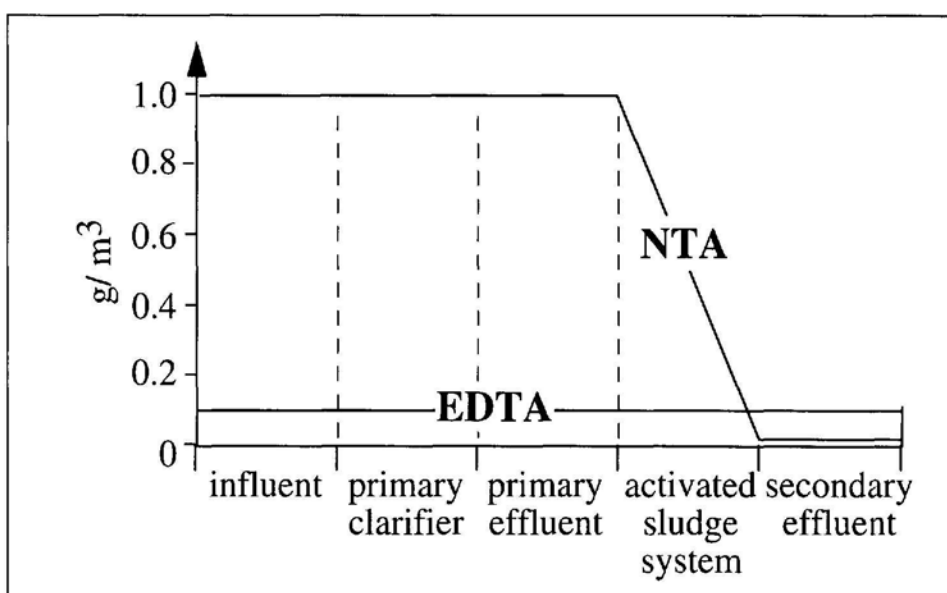


Fig. 3. Concentration profiles of NTA and EDTA in a municipal wastewater treatment plant [1]

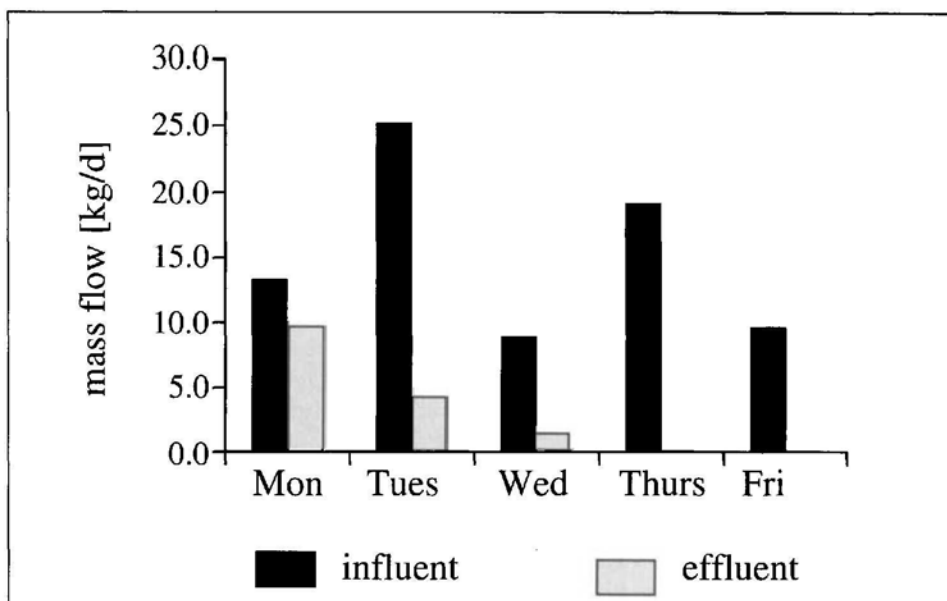


Fig. 4. Adaption in the degradation of 3-nitrobenzenesulfonate in the two-stage nitrifying wastewater treatment plant in Herisau [5]

bit of NTA) and that they fully induce the NTA-degrading enzyme system only if NTA contributes a significant amount of the total carbon that can be utilized [2].

Hydrophilic Pollutants in Industrial Wastewaters: Substituted Benzene- and Naphthalenesulfonates

Substituted benzene- and naphthalenesulfonates are highly soluble anionic compounds that are widely used in chemical, pharmaceutical, and textile industries. In a field study, six benzene- and nine naphthalenesulfonates were found in raw wastewaters from the municipal WWTP Herisau (Switzerland) which receives wastewaters from three textile-manufacturing plants [5]. 3-Nitrobenzenesulfonate was found as a major pollutant in

concentrations up to 5 mg/l. Elimination of nitrobenzene- and naphthalenemonosulfonates was > 98%. Naphthalenedisulfonates had elimination rates between 5% (naphthalene-1,5-disulfonate) and 96% (naphthalene-1,6-disulfonate). However, high wastewater flows (due to rainy weather) and longer periods with no sulfonate reduced the elimination efficiency. The fact that no adsorption of aromatic sulfonates to suspended solids was observed, indicates that the elimination occurred primarily by biodegradation.

The kinetics of biodegradation are dependent to a large extent on the adaptation of the bacteria to the compound they are to remove. If a compound occurs irregularly or the input load is interrupted for weeks, its degradation can be reduced drastically,

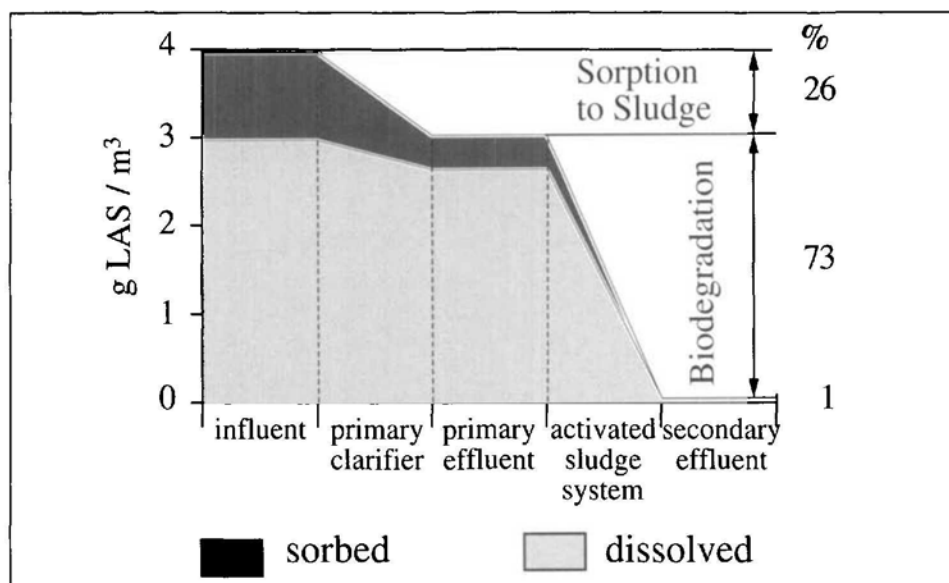


Fig. 5. Concentration profiles of LAS in a municipal wastewater treatment plant [1]

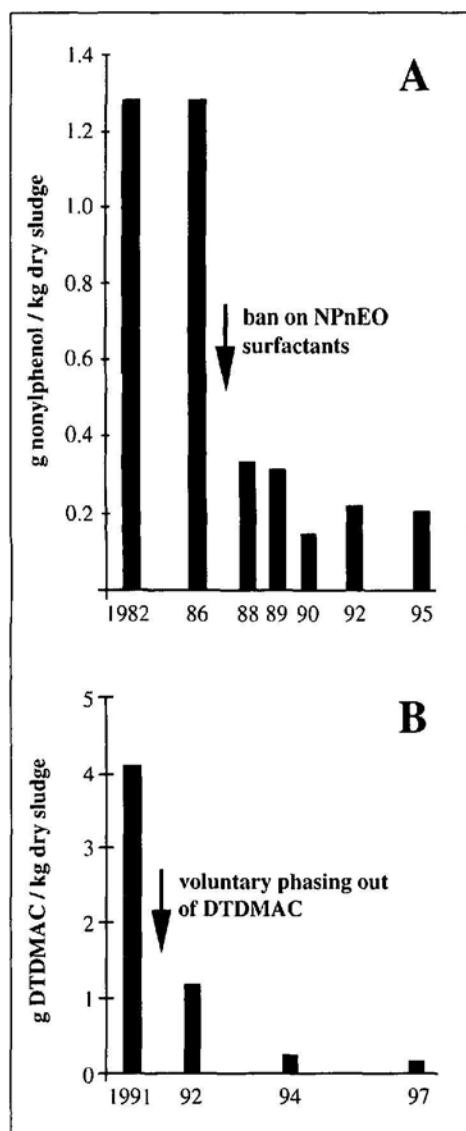


Fig. 6. Accumulation of nonylphenol (A) and DTDMAC (B) in digested sewage sludges. The decrease of nonylphenol concentrations after 1986 is caused by the Swiss ban on the use of NPnEO surfactants in laundry detergents [15]. The drop in DTDMAC concentrations is due to the producers' voluntary phasing out in the second half of 1991.

even though it is completely degraded when present in continuous supply. An example of adaptation of activated sludge to 3-nitrobenzenesulfonate is shown in Fig. 4. This compound is usually degraded efficiently (> 95%). After a four week interruption of the operation of the textile plant, the degradative capacity of the wastewater treatment recovered within a few days.

Anionic Surfactants: Accumulation in Digested Sludges

Linear alkylbenzenesulfonates (LAS) are the anionic surfactant most widely used in laundry detergents and surface cleaners although newer surfactants such as secondary alkanesulfonates (SAS) are an alternative, because of their faster biodegradability and the absence of a phenyl moiety. Technical LAS products typically are composed of components having alkyl chains of C_{10} to C_{14} . SAS mixtures normally contain homologs ranging from 13 to 17 carbon atoms in the alkyl chain. Each homolog is made up of several isomers each defined by the carbon atom to which the benzenesulfonate or the sulfonate group is attached.

The extend of LAS removal from the waste stream during aerobic sewage treatment (98–99%) (Fig. 5) [7–9] [20] is equal to that of SAS (99.7%) [9]. In samples of secondary effluent [9], the concentrations of LAS ranged from 60 to 100 $\mu\text{g/l}$ and of SAS from below detection limit (< 1 $\mu\text{g/l}$) to 14 $\mu\text{g/l}$.

The surface-active compounds LAS and SAS sorb to a substantial degree to solids despite their negatively charged sulfonate groups. Both compounds are degradable under aerobic conditions in the activated sludge system but are stable un-

der anaerobic conditions. Approximately 26% of the LAS and 16% of the SAS mass flows entering the treatment are transferred to the anaerobic sludge digester. In 1994, average LAS concentration in digested sludges of 18 WWTPs in the Canton of Zürich was 4.1 g/kg dry sludge. The concentration of SAS of 0.7 g/kg dry sludge was found in a field study [9]. Assuming those average concentrations in anaerobically digested sludges and an annual disposal of 110000 t per year sludge dry matter to Swiss farmland, ca. 450 t of LAS and 77 t of SAS are annually applied to Swiss agricultural soils. Given the current practice in Switzerland of applying sludge to agricultural soils and assuming a maximum loading capacity of 5 t per year sludge dry matter to farmland every three years, the allowed maximum annual loading rates of LAS and SAS to agricultural soils are 670 mg/m^2 and 120 mg/m^2 , respectively.

Phenolic Nonionic Surfactant: Formation of Toxic Transformation Products in Wastewater Treatment

Nonylphenol polyethoxylate surfactants (NPnEO, $n = 3-20$) are efficiently eliminated during biological treatment [6] [21]. However, the overall rate of biotransformation was limited due to the formation of biorefractory products, including nonylphenol (NP), nonylphenol mono- and diethoxylate (NP1EO and NP2EO), and nonylphenoxy carboxylic acids (NPEC). Nonylphenol is a highly toxic compound which accumulates in anaerobically digested sludge because of its hydrophobicity and persistence under anoxic conditions.

As a consequence of the results of the EAWAG studies, the use of NPnEO in laundry detergents in Switzerland was banned in 1986. In order to determine the effect of this regulatory measure, a program was initiated in 1982 to monitor nonylphenol concentrations in digested sludges in the Canton of Zürich (Fig. 6, A). The results indicate significantly lower NP levels after the ban imposed by the Swiss ordinance for environmentally dangerous compounds (Stoff-VO) in 1986. It can also be inferred that products containing nonylphenol-based compounds are still in use and are released into municipal wastewaters.

Based on studies in several WWTPs, it was estimated that ca. 19% of all nonylphenolic compounds introduced to WWTPs are released to the environment as NPEC, 11% as lipophilic nonylphenol ethoxylates (NP1EO + NP2EO), 25% as NP, and 8% as untransformed NPnEO.

Almost all of the released NPnEO and NPEC, as well as the majority of NP1EO and NP2EO, are discharged into natural receiving waters *via* secondary effluents, which are responsible for 60% of the total input of nonylphenolic compounds into the environment. In contrast, most NP (> 90%) is transferred to the anaerobic sludge digester, representing *ca.* 40% of the total input.

Softer than Soft: Cationic Surfactant as a Fabric Softener

Quaternary ammonium surfactants show a high affinity for negatively charged surfaces, making them suitable for industrial applications and as components of consumer products. These chemicals are mainly used as fabric softeners and anti-static agents in laundry detergents. The most widely applied active ingredient in fabric softeners has been the aliphatic quaternary ammonium compound DTDMAC (ditallowdimethylammonium chloride).

DTDMAC adsorbs strongly to suspended solids because of hydrophobic and electrostatic interactions with negatively charged surfaces. Owing to its physico-chemical properties and its biorefractory character in anoxic environments, DTDMAC is substantially enriched in digested sludges. Therefore, DTDMAC has been replaced in Europe by new quaternary ammonium compounds that contain ester functions in the long hydrophobic chains and are expected to have an improved degradability in the environment. To monitor the impact of this measure, DTDMAC amounts were determined in digested sludges of 14 WWTPs in the Canton of Zürich. Mean concentrations of DTDMAC decreased from 4.15 g/kg (in 1991) to 1.16, 0.24, and 0.18 g/kg dry sludge (Fig. 6, B; [11][22] unpublished results).

The monitoring study allowed us to follow changes in the DTDMAC concentrations in the environment caused by changing usage at the source by comparing the percentage consumption data with the measured values in digested sludges. The consumption estimated by the Swiss detergent industry decreased from *ca.* 1200 t (1990, 100%) to 350 t (1991, 29%), to 160 t (1992, 13%), and to 100 t (1994 and 1997, 8%) because of the phasing out of DTDMAC. As DTDMAC was replaced in the second half of 1991, the measured mean value of 4.15 g/kg was arbitrarily defined as 100% for this study. The mean concentrations in these 14 WWTPs followed the consumption. The concentrations dropped from 100% (1991), to 28% (1992), to 6% (1994), and to 4% (1997).

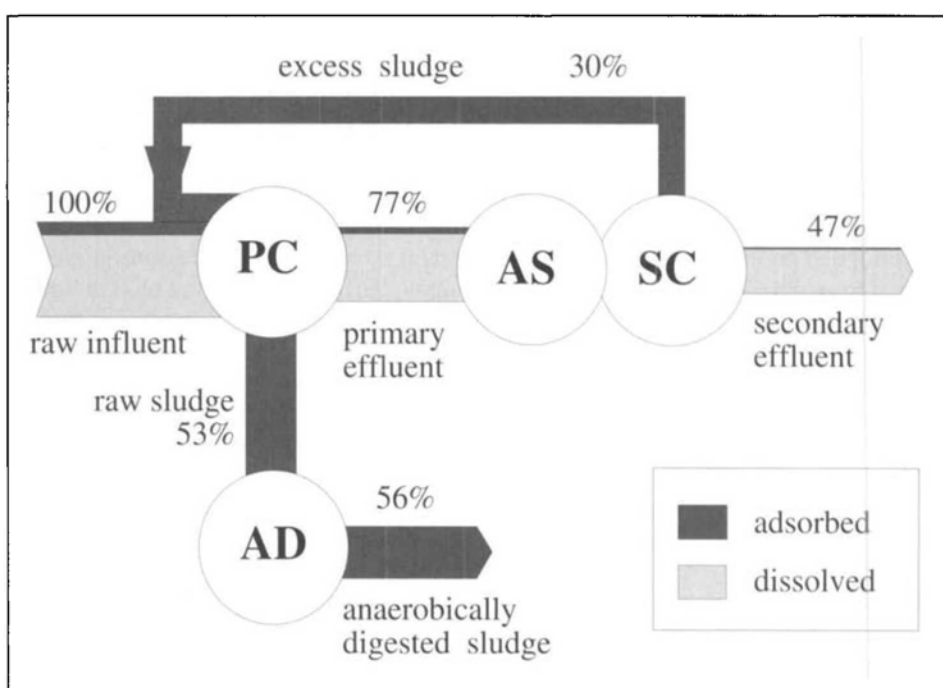


Fig. 7. Mass fluxes of the fluorescent whitening agent DSBP in the wastewater treatment plant Zürich-Glatt [12]. The WWTP serves a population of 120000. 100% influent corresponds to 461 g d⁻¹. (PC: primary clarifier, AS: activated sludge treatment, SC: secondary clarifier, AD: anaerobic digester.)

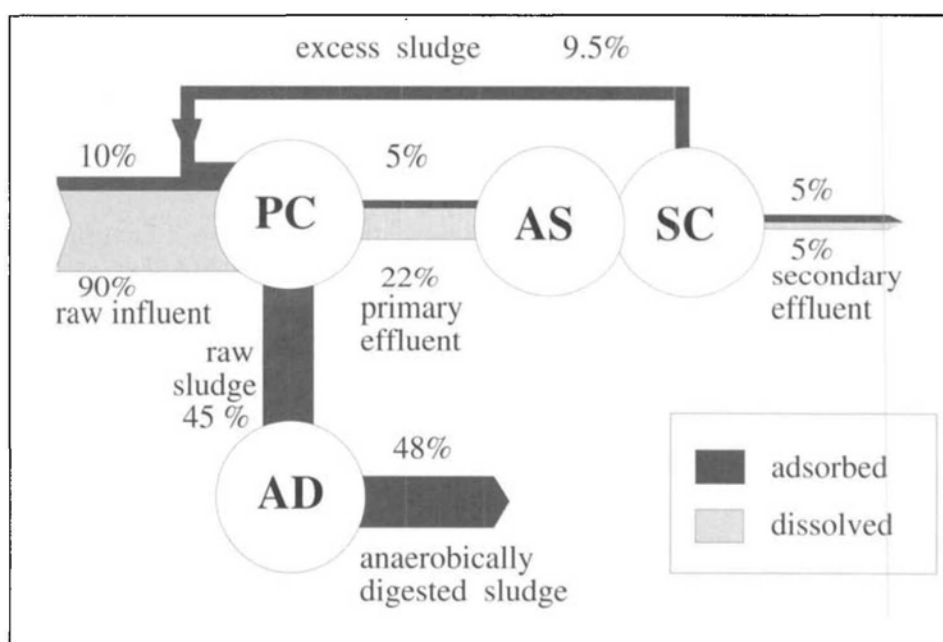


Fig. 8. Mass fluxes of organotin compounds in the wastewater treatment plant Zürich-Werdhölzli [13]. The WWTP serves a population of 500000. Data are given on a daily (24 h) base, with percentages relative to 100% influent corresponding to 122 g d⁻¹. (PC: primary clarifier, AS: activated sludge treatment, SC: secondary clarifier, AD: anaerobic digester.)

This analytically confirmed drop of 95% in DTDMAC concentrations in digested sludges is due to the replacement of this compound and a clear illustration of the effects of the producers' voluntary phasing out.

Whiter than White: Fluorescence Whitening Agents

Fluorescence whitening agents (FWAs) that are used in laundry deter-

gents are moderately water-soluble organic compounds with a high affinity for cellulosic material. When adsorbed to textiles, the strong blue fluorescence of FWAs eliminates the typical yellowish cast of white fabrics and makes them look whiter. Residual FWAs not incorporated into the washed goods are discharged with the washing liquor.

Of the FWAs entering the WWTP, 53–98% are eliminated during wastewater

treatment by adsorption to primary and activated sludge [12]. The extent of removal of the investigated FWAs corresponds well with their differing affinity to suspended solids. During anaerobic sludge treatment, FWAs are not biodegraded and are thus discharged to farmland with sewage sludge. The average FWA levels in sludges from nine WWTPs around Zürich were 118 mg/kg dry weight (85–170 mg/kg).

Residual FWAs in primary effluent were also not biodegraded aerobically in the activated sludge facility, so that all FWAs not bound to sludge were discharged with the treated wastewater. Residual FWA concentrations in secondary effluent ranged from 2.6 to 8.9 µg/l.

Sorption as the only process affecting the fate of FWAs in WWTPs per se does not reduce the amounts discharged to the environment. Instead, it leads to a redistribution between the two points of discharge, sewage effluent and sludge, as shown in Fig. 7 for DSBP (= 4,4'-bis(2-sulfostyryl)-biphenyl), the second most important detergent FWA. As 50% of the sludge in Switzerland is used as fertilizer in agriculture, ca. 11 t FWA per year are discharged to Swiss farmland, corresponding to a maximum annual FWA load of 18 mg/m². Another 20 t FWA per year are discharged to surface waters. Predicted average surface-water concentrations of ca. 0.5 µg FWA/l correspond well with concentrations found in Swiss rivers ranging from 0.001 to 1 µg/l and are well below predicted no-effect concentrations of 100 µg/l.

Toxic Organometallic Chemicals: Organotins

Trisubstituted organotin compounds represent environmental contaminants with high ecotoxicological risks for aquatic ecosystems. Organotin compounds find various applications in industry and agriculture. Tributyltin (TBT) compounds have been included in antifouling paints on ships because of their high toxicity toward aquatic organisms. After regulation in many countries, tributyltin compounds find application nowadays mainly as fungicides in timber and other materials. Monobutyltins (MBT) and dibutyltins (DBT) are used as stabilizers and as catalysts for polyurethane foams and silicones, and in industrial processes.

In raw sewage, MBT, DBT, and TBT compounds were detected in the range of 136–564, 127–1026, and 64–217 ng/l, respectively [13]. Because they were primarily associated with suspended matter (61–93% of MBT, 87–97% of DBT, and 83–92% of TBT), they were transferred from waste-

water to sludge in the primary clarifier of the WWTP of Zürich-Werdhölzli (Fig. 8). Both aerobic and anaerobic degradation were found to be insignificant. Thus, adsorption to sludge is the most important process for organotins during wastewater treatment. Therefore, over 90% of the influent were removed by sorption and remained in the digested sludge. It should be noted, however, that the WWTP studied had a high performance and contained a step for suspended solids removal due to filtration, which is usually not met by other plants. For instance, elimination of TBT was estimated in the range of 70–80% in other Swiss plants. In digested sludges of 25 WWTPs the average concentration of mono-, di-, and tributyltin in 1995 were 0.5, 1.5, and 1.1 mg/kg dry weight, respectively [23].

Although regulation of TBT in anti-fouling took place, this did not result in a decrease of the contamination of digested sludge. The levels found probably represent a more general contamination pattern in Switzerland and other industrialized countries. The ecotoxicological consequences of soil amendment by sludge contaminated by these compounds for agricultural land is unknown and should be investigated.

Implications for Practical Environmental Protection and Outlook

Studies of the mass flow of organic compounds in wastewater treatment plants indicate that their fate is determined not only by microbial transformation, but also by physicochemical processes. A better knowledge of the physicochemical and biological processes is needed in order to assess the fate of trace compounds during wastewater treatment. However, it should be considered that even state-of-the-art analytical methods have allowed only to identify a minor fraction of organic contaminants in the effluents of wastewater treatment.

- Hydrophilic compounds enter the biological stage almost completely and must, therefore, be easily degradable under aerobic conditions.
- Part of organic compounds with lipophilic and amphiphilic properties enter the anaerobic sludge treatment directly. Therefore, they should be degradable under aerobic and anaerobic conditions in order to prevent accumulation in digested sewage sludges. Such properties must be postulated in particular for high-volume chemicals which are used directly in water.

- In Switzerland ca. 50% of the digested sludges are applied to agricultural soils. For many organic trace pollutants that are not rapidly biodegradable under anaerobic conditions, the soil becomes an important sink. It is extremely difficult to reliably assess the long-term effect of persistent contaminants on the soil ecosystem. Consequently, measures at the source should be taken to avoid inputs of these chemicals into the environment.

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