

Development of a Chemical Fate Model for Trickling Filters Based on the Activated Sludge Model *SimpleTreat*: A Case Study for LAS

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A new chemical fate model for trickling filter wastewater treatment plants was developed, using the same principles as the steady-state nonequilibrium activated sludge model *SimpleTreat*, in combination with an existing biofilm model. To test the new model in detail, a pilot-scale (200 L) trickling filter was built and operated. Using this setup, removal of the surfactant LAS was measured under different well-characterized operating conditions. The new model could be fitted to data on LAS removal in the pilot-scale filter as well as in two full-scale domestic treatment plants. The same biodegradation rate coefficient (derived from activated sludge data) could be used in all cases, except for one laboratory experiment with higher LAS influent concentrations. It is concluded that the effect of high influent concentrations on the biodegradation rate should be further investigated.

Introduction

In the last 20 years, several models which describe the fate and behavior of individual organic compounds in biofilms have been formulated (e.g. refs 1–6). Specific applications to trickling filter systems are given in e.g. refs 6–8.

In this paper, the development of a new approach to chemical fate modeling in trickling filters is presented, based on the *SIMPLEBOX* method (9) combined with the existing steady-state biofilm diffusion/biodegradation model of Melcer *et al.* (6). In *SIMPLEBOX*, chemical fate is calculated under steady-state nonequilibrium conditions, using a mass balance between several completely mixed boxes (cf. Level III models after Mackay and Paterson (10)). Previously, this method was applied in the activated sludge chemical fate model *SimpleTreat* (11, 12), which is used as a standard in European Union environmental risk assessment.

The biodegradation and sorption aspects of the newly developed fate model were tested using experimental data for the surfactant LAS (Linear Alkylbenzene Sulfonate), which were obtained in a well-characterized pilot-scale trickling filter setup.

The model was also applied to predict the fate of LAS in two full-scale trickling filter plants in Yorkshire (UK), for which

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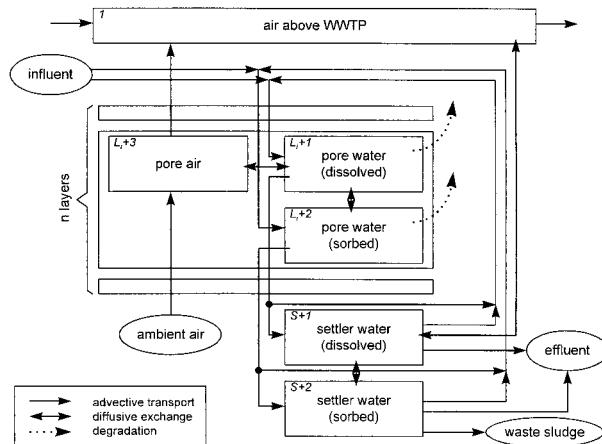


FIGURE 1. Model scheme of a trickling filter plant.

a detailed monitoring study was performed by Holt *et al.* (13) in the framework of the GREAT-ER project (Geography-referenced Regional Exposure Assessment Tool for European Rivers (14)).

Chemical Fate Model Formulation

Process Description. The modeled trickling filter configuration was limited to the combination of a filtration unit coupled with a secondary settler. Chemicals are present in the dissolved phase and sorbed to suspended solids; through ad-/desorption there may be an interchange between these phases. The dissolved chemical can diffuse into the biofilm. Suspended solids with sorbed chemicals may be filtered out of the water; chemicals associated with biofilm solids may be released in the sloughing process. Finally, (bio)degradation may take place inside the biofilm and in the water.

Systems Analysis and Segmentation. The trickling filter system was subdivided into a number of completely mixed boxes (Figure 1). The first segmentation was between the filter unit and the settler. The settler was divided further into a dissolved and a sorbed phase. The filter itself was split up into multiple horizontal layers (cf. tanks-in-series). For this study, 5 horizontal layers were used. Each of these was subdivided into two compartments: pore water–dissolved phase and pore water–sorbed phase. Downward transport of chemicals through the filter unit occurs between the pore water of the different layers. Within a layer, exchange processes take place between the sorbed and dissolved phases.

Biodegradation mainly occurs by biofilm activity. Continuous chemical diffusion from the pore water into the biofilm is only possible if a concentration gradient is maintained by biodegradation in the biofilm. These coupled processes are dealt with by the steady-state biofilm model of Melcer *et al.* (6). In this model, the calculated chemical diffusion flux from the pore water into the biofilm is expressed as chemical removal out of the water phase. Hence, the biofilm compartment need not be represented as an extra box. Note that this modeling approach was also successfully applied to predict the fate of biodegradable chemicals in river biofilms (15).

To simplify the model, it was assumed that filtration and release (by sloughing) of the sorbed chemical balance each other (resulting in a zero net effect), and chemical sorption equilibria within the biofilm were not considered.

Next to the between-layer transport, there is water and solids transport by means of the effluent recycles to the top

filter layer. Influent (dissolved and sorbed phase) enters the plant into the top filter layer, and effluent (dissolved and sorbed phase) leaves the plant via the settler as well as waste sludge (sorbed phase only).

Model Equations. Nontrickling filter specific equations were taken as such from *SimpleTreat* version 3.0 (12). For brevity, no model equations are mentioned in this paper but are instead included in the Supporting Information.

Advective transport of water and solids between layers (both expressed as volumetric fluxes) are calculated from a simple flow balance, taking into account the recycle flows. Settling is expressed by a fixed solids removal efficiency.

Between the pore water's dissolved phase and the biofilm, chemicals are exchanged by diffusion. This was not described as such but was considered together with biodegradation as a lumped chemical elimination from the pore water. For the calculation of diffusive exchanges, the fugacity approach is applied. Expressions for fugacities, diffusion coefficients, and kinetics were taken directly from *SimpleTreat*, after Mackay and Paterson (16). For trickling filter solids, the same sorption kinetics were assumed as those used for activated sludge in *SimpleTreat*.

Biodegradation in the dissolved phase of the water compartment was expressed as the sum of two first-order rates: biodegradation by suspended biomass and disappearance of the chemical into the biofilm. In the sorbed phase, only the suspended biomass activity is taken into account. For suspended biodegradation, "double" first-order kinetics (both in active biomass and in chemical concentration) are used. A bioavailability correction factor can be applied for the sorbed phase (e.g. ref 34). Melcer et al. (6) developed a biofilm model which predicts the mass flux of a chemical from the bulk water into the biofilm per unit of interfacial area. This process is driven by diffusion and biodegradation. In ref 6, an analytical solution for the chemical mass flux into the biofilm is given. This was converted to a first-order elimination rate coefficient, which could further be used in the *SIMPLEBOX* approach. In the presented model, the biofilm/water interfacial area has to be known. The actual biofilm/water interfacial area is determined by the surface area of the irregular biofilm surface. As no simple or universal estimation methods for this interfacial area exist, in this paper a default correction (of a factor 2) was used to convert carrier material surface to biofilm surface (cf. ref 15).

Pilot-Scale Trickling Filter

To verify the model's predictions, a pilot-scale trickling filter plant was constructed. In this pilot plant, the removal of the surfactant LAS was determined under different well-characterized operating conditions.

Development of a Pilot-Scale Trickling Filter Plant. The filter's design was focused on practical feasibility and on the possibility to easily quantify and control all relevant operating parameters and conditions. Its dimensions were chosen to represent a cylindrical core taken from a full-scale unit (cross-sectional area = 0.118 m², volume = 0.213 m³). A polystyrene carrier material was used (specific surface area = 220 m²/m³ or 0.0225 m² per unit, density = 61 kg/m³, porosity of 96%, plastic volume per unit = 5 mL). Metcalf and Eddy (23) advise to apply a high rate loading with plastic filter media. A volumetric loading rate $B_v = 0.6 \text{ kgBOD}/\text{m}^3 \cdot \text{d}$ and a hydraulic surface loading rate B_A of 15 m³/m²·d (= 0.625 m/h) were selected. The recycle ratio was 3.5. This is higher than recommended (23, 24), but it was retained to ensure proper wetting of the filter material and a uniform spatial distribution of the influent over the filter's surface. The settler diameter was fixed at 0.25 m (maximal overflow velocity = 1.5 m/h). Based on ref 25, a settler height of 0.75 m was chosen. In domestic trickling filters, air flow (by natural ventilation, chimney effect) is poorly quantified. An upward

TABLE 1. Experimental Conditions in Pilot-Scale Trickling Filter

| | series A | series B | series C | series D |
|-------------------------------|-----------|-----------|-----------|------------|
| influent flow rate (L/min) | 0.29 | 0.29 | 0.29 | 0.29 |
| filter unit flow rate (L/min) | 1.30 | 0.44 | 1.30 | 1.30 |
| recycle ratio (-) | 3.5 | 0.5 | 3.5 | 3.5 |
| short recycle ratio (-) | 3.5 | 0.5 | 1.75 | 0 |
| long recycle ratio (-) | 0 | 0 | 1.75 | 3.5 |
| influent LAS (mg/L) (measd) | 5.3 ± 2.1 | 3.5 ± 0.5 | 4.2 ± 0.9 | 15.2 ± 4.5 |
| no. of LAS measurements | 9 | 4 | 7 | 5 |
| total biofilm thickness (μm) | 100–500 | 500–700 | 500–700 | 550–750 |

air flow through the filter of 10 L/min was chosen based on ref 6. By an electronic balance (modified Mettler-Toledo Spider 1S-150, max. capacity 150 kg, accuracy 10 g) the filter unit's mass was continuously monitored, to determine hydraulic and biofilm characteristics. As influent to the trickling filter plant, a synthetic sewage was used (after ref 26), with a COD:N:P ratio of 462:43:9 = 100:9.3:2.

Hydraulic and Biomass Characterization. *Hydraulics.* A NaCl (conductivity) tracer test was conducted before the startup of the biological experiments (i.e. in the absence of biofilm). The mean HRT (at a flow of 1.3 L/min) was 6.35 min. In the presence of biofilm, a similar tracer test was conducted using the fluorescent agent Thioflavine-S (as NaCl may disrupt biological activity). The mean tracer retention time at a flow of 1.3 L/min was 38.6 min, which is a factor 6 longer than without biofilm. This can be explained by diffusion and eventually adsorption of the tracer substance into the biofilm (e.g. ref 27)—hence, the tracer retention time in the presence of biofilm was not representative of the hydraulic retention time.

Based on the measurements with the electronic balance, the total water mass in the presence of biofilm was 8.27 kg (at 1.3 L/min) or 7.59 kg (at 0.5 L/min). From this, the mean HRT in the presence of biofilm was estimated to be 6.36 min (at 1.3 L/min) or 15.18 min (at 0.5 L/min). Without biofilm (at 1.3 L/min) this was 5.12 min, which is in line with the tracer test. The calculated fraction of water in the filter pores (with biofilm) was 4% (1.3 L/min) or 3.7% (0.5 L/min).

Biofilm Characterization. Biofilm density was approximately 40 g/L (which is a typical value, e.g. ref 6). The total biofilm thickness was determined from the mass of the filter unit, assuming a wet density of 1 kg/L and a total carrier surface area within the filter of 46.86 m². During the first month of operation, the average biofilm thickness evolved from 50 to 100 μm. The next month it quickly increased to more than 900 μm. Feeding the filter with only tap water for 1 week reduced biofilm thickness to range between 500 and 750 μm during the next 2 months. These measurements corresponded well with additional thickness measurements based on the wet or dry mass of individual carriers.

For LAS biodegradation, the presence of oxygen is needed (e.g. ref 28). However, it may be that only the upper layer of a biofilm is aerobic (e.g. ref 29). In the above, the total biofilm thickness was determined, including both active and inactive biomass. Hence, these measurements are not representative of the active biofilm thickness.

Experimental Methods. *Steady-State LAS Removal Experiments.* Four experimental series were conducted (Table 1). In A the operating conditions were high-rate, while for B the recycle flow was set much lower. Series C was identical to A. In D the influent LAS concentration was 3–4 times higher than in the other experimental series. Under steady-state conditions, LAS concentrations were measured at four locations: (1) pilot plant influent; (2) filter inflow; (3) filter outflow; and (4) final effluent. Based on this information, LAS removal was calculated over the entire pilot plant (final effluent versus influent) and single-pass (filter outflow versus filter inflow).

TABLE 2. Overview of LAS Measurements and Removals

| | series A | series B | series C | series D |
|---|----------|----------|----------|----------|
| LAS Measurements (% Relative to Influent Concentrations) | | | | |
| (1) influent (total) | 100 | 100 | 100 | 100 |
| (2) filter inflow (total) | | | | |
| measured | 74 ± 20 | 77 ± 20 | 61 ± 14 | 38 ± 14 |
| calculated | 64 ± 17 | 83 ± 6 | 56 ± 14 | 34 ± 8 |
| (3) filter outflow (total) | 54 ± 22 | 50 ± 17 | 44 ± 18 | 15 ± 11 |
| (4) effluent (total) | 51 ± 17 | 52 ± 24 | 44 ± 17 | 15 ± 10 |
| (4) effluent (dissolved) | 45 ± 16 | 41 ± 18 | 36 ± 14 | 7 ± 5 |
| LAS Removal (%) | | | | |
| total WWTP | 49 ± 17 | 48 ± 24 | 56 ± 17 | 90 ± 3 |
| single-pass ^a | 19 ± 14 | 41 ± 16 | 25 ± 15 | 58 ± 19 |

^a Based on calculated concentrations for the filter inflow.

Biological Adaptation to Different LAS Concentrations. The evolution of single-pass LAS removal over the filter unit was regularly monitored over a 40 day period, during which the influent LAS concentration was step-changed from 8 to 25 mg/L after 5 days. This experiment was conducted to examine the (dynamic) adaptation of the filter microbiology to higher LAS concentrations.

Analytical. LAS was measured by the aspecific Azure-A method for anionic surfactants (30). This was applicable because no other anionic surfactants than LAS were present in the influent. LAS samples were preserved by adding formaldehyde to 1%.

Results and Discussion

Pilot-Scale Trickling Filter LAS Removal Experiments.

Steady-State LAS Removal. An overview of the LAS measurements and removal efficiencies is given in Table 2. The measurements in the filter inflow reservoir (2) were considered unreliable, as sampling (here as well as at all other locations) disrupted the hydraulic balance in this tank. Instead, calculated concentrations for this reservoir (from the mass balance of the influent and recycle flows) were used to derive the single-pass (filter outflow versus inflow) LAS removal. No significant difference was found between LAS removal in the first three experimental series (Tukey test, $\alpha = 0.05$). With the lower recycle ratio (B), single-pass removal was much higher than with the high recycle (A, C), but in terms of total LAS removal this effect was neutralized by the lower number of passes through the filter. Removal in Series D (with higher influent LAS concentrations) was significantly higher than in the other series (Tukey test, $\alpha = 0.05$). The single-pass removal efficiency was also clearly higher. Moreover, the LAS influent concentrations did not appear to influence the effluent concentrations (no significant difference between the effluents of the four experimental series: Tukey test, $\alpha = 0.05$). The measurements of series D were made after the higher LAS influent concentrations had been applied for one month. This could suggest that the amount of competent biomass (capable of biodegrading LAS) may have adapted itself to the higher LAS concentrations in the influent. Similar observations have been described for activated sludge (33, 34). On the other hand, this effect might also be due to the occurrence of different kinetics at higher LAS concentrations.

LAS removal in A, B, and C was not significantly different, whereas the total biofilm thickness in B and C was much higher than in A. Furthermore, in D LAS removal was clearly higher than in B and C, but the biofilm thickness was similar. From these observations, it can be derived that the total biofilm thickness apparently had no effect on LAS removal. Hence, the active biofilm thickness must have been similar in the four series, which confirms that it was much smaller than the total thickness. A plausible value can be estimated

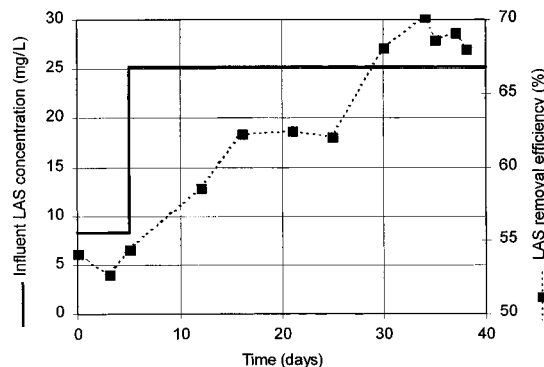


FIGURE 2. Adaptation of LAS removal to higher influent concentrations.

TABLE 3. LAS Parameters

| | | |
|--------------------------|--|---|
| K_d | sorption partitioning coefficient | 2000 L/kg |
| H | Henry's law constant | $1 \times 10^{-6} \text{ Pa}\cdot\text{m}^3\cdot\text{mol}^{-1}$ |
| K_b | biodegradation rate coefficient | $0.278 \times 10^{-6} (\text{g}/\text{m}^3)^{-1}\cdot\text{s}^{-1}$ |
| α_{sorbed} | sorbed phase biodegradation correction | 1 (-) |
| D_i | diffusion coefficient in water | $55 \times 10^{-6} \text{ m}^2/\text{s}$ |

as the smallest total thickness which occurred: between 100 and 200 μm .

Biological Adaptation to Higher LAS Concentrations. The results of the dynamic adaptation experiment are shown in Figure 2. At an influent level of 8 mg/L, single-pass LAS removal was on average 54%. After the LAS concentration in the influent had been tripled, the removal gradually increased, reaching a new equilibrium after ca. 4 weeks (on average 69%). This increase in removal efficiency followed a linear pattern ($R^2 = 0.915$).

These observations confirm the hypothesis of biological adaptation to higher LAS levels in the influent. This indicates that for LAS, specific "double" first-order biodegradation kinetics are not universally valid and are only applicable within a specific concentration range. Further research is needed to fully quantify the degree and the kinetics of adaptation and to include these into the biofilm biodegradation model.

Model Application and Testing for LAS. The chemical properties used to model LAS are given in Table 3. The sorption coefficient and Henry's law constant were taken from Cowan et al. (35). Because of its low sensitivity, an average from ref 6 (not specifically for LAS) was taken for the diffusion coefficient. The sorbed phase biodegradation correction factor was set to 1 (cf. refs 35 and 36). The "double" first-order biodegradation rate coefficient was calculated from the first-order rate coefficient typically used for biodegradation by suspended biomass in activated sludge (3 h^{-1} , with mixed liquor $SS = 3000 \text{ mg}/\text{L}$, e.g. in ref 11).

An overview of default parameter values, which are not specific to the modeled filter units nor to the chemical LAS, is presented in the Supporting Information.

Pilot-Scale Trickling Filter. Plant operating parameters were known. Hydraulic and biofilm characteristics were measured or estimated (active biofilm thickness = 150 μm , laminar water layer thickness = 100 μm , biofilm/water interfacial area = 200 m^2/m^3). As all required parameters were available, it was possible to apply the model without any calibration based on the LAS elimination measurements. The uncalibrated predictions for the series A, B, and C were within one standard deviation of the mean measured removals, both for total and single-pass removal. The relative deviation was at the most 20%. For series D, on the other hand, a much larger deviation was found: over 30% underestimation

TABLE 4. Full-Scale Trickling Filter Plant Dimensions

| | Gargrave | Dowley Gap |
|--|----------|------------|
| filter surface area (m ²) | 538 | 13130 |
| filter volume (m ³) | 1236.5 | 25130 |
| settler surface area (m ²) | 112.5 | 1016 |
| settler volume (m ³) | 360 | 2830 |

of the total removal and over 60% underestimation for the single-pass.

Next to the uncalibrated simulations, the single-pass removal predictions for the different series were fitted to the measurements by tuning the fraction of water in the filter (and hence also the HRT). The calibrated HRTs in series A, B, and C still corresponded very well with the measurements. The calibrated HRT for series D was unrealistic: it was a factor 3 higher than expected. To fit the model to series D using realistic hydraulics, it was necessary to increase the biodegradation constant by a factor 3.5, which indicates that LAS removal kinetics may be faster at higher influent concentrations. This is a shortcoming of the trickling filter fate model as it is presented here: it can only deal with this situation by recalibration of the biodegradation rate coefficient. Hence, it is recommended to focus further research on the effect of influent concentrations on LAS biodegradation rates and on the effect of biological adaptation to different concentrations on these rates.

Full-Scale Sewage Treatment Plants. In the low-rate domestic sewage treatment plants of Gargrave and Dowley Gap (Yorkshire, UK), LAS removal over the trickling filter (excluding primary treatment) was measured by Holt et al. (13). Gargrave is a very small plant, treating the sewage of 1450 people, while Dowley Gap serves 30,150 people. Flows and LAS elimination were measured in both treatment plants (13). On average, LAS was removed for 88.4% in Gargrave and for 83.0% in Dowley Gap. Plant dimensions are given in Table 4. No effluent recycles are applied in these facilities. Data on trickling filter suspended solids were not available; instead a default value of 200 mg/L was assumed, together with a secondary settler solids removal efficiency of 90%. For both plants, the bed material consisted of rock, with a porosity of 50% and a specific surface area of approximately 100 m²/m³. The biofilm/water interfacial area was assumed to be twice that area. For the stagnant water zone thickness and the biofilm density, defaults from ref 6 were used. An active biofilm thickness of 150 μm was chosen.

The fraction of water in the filter was the only parameter used for calibration. Two calibrations were performed: one based on 1 day's measurement in Gargrave and one based on 1 day's measurement in Dowley Gap. The calibrated values (9.4% and 6.6%) correspond with a total water layer thickness of 300–500 μm and a mean HRT of 5.6 h (Gargrave) or 3.4 h (Dowley Gap). Subsequently, these calibrations were applied to all other monitored situations. When the model calibration was based on 1 day's measurement in Gargrave, LAS removal in the same plant during the next day was over-predicted by 6%. This could be due to the very high removal found in the calibration case (92.4%), which may have been exceptional and possibly not representative of steady-state. For Dowley Gap, the calibration based on 1 day could be extrapolated more reliably to the other three measurement days: the deviation of predicted removal was at the most 3.5%. Extrapolating the Gargrave calibration to Dowley Gap resulted in an overestimation of LAS removal by 8 to 12%. The Dowley Gap calibration caused a (smaller) underestimation of removal in Gargrave by 3 to 6%. As mentioned above, the higher predictive error with the Gargrave calibration could be due to the nontypical high removal found in the calibration case.

Concluding Remarks

Plant-specific calibrations produced more accurate predictions than the extrapolation of another plant's calibration or the use of noncalibrated parameters. However, if a relative deviation in the order of 10% (at the most 20% for the noncalibrated parameters used to model the pilot-scale plant) is deemed acceptable, such a plant-specific calibration appeared to be not strictly required.

Note that in this work, only the biodegradation and sorption aspects of the model were confronted with measurements. It is stressed that to test the relevance and validity of the volatilization aspects, further research, using other chemicals, is needed.

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Supporting Information Available

Box numbering of Figure 1, *SIMPLEBOX* approach, default parameter values, and nomenclature. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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