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Verification studies of a simplified model for the removal of dichloromethane from waste gases using a biological trickling filter (part II)

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Verification studies of a simplified model for the removal of dichloromethane from waste gases using a biological trickling filter (Part II) *

R. M. M. Diks and **S. P. P. Ottengraf,** Eindhoven

4 Experimental results and discussion

4.1 Transient state behaviour and biological stability

If a biological trickling filter is applied in practice, operational stability must be guaranteed for many years, while fluctuations of the elimination performance and thus transient states have to be avoided or at least minimized. In practice as well as in the laboratory investigations it is very important to know the time scale of these transient states and the processes which are responsible. The start-up period of the lab-scale trickling filter was therefore followed closely.

This start-up of the filter system took place by inoculating the liquid phase with about 10 litre of a suspension of Hyphomicrobium sp. GJ21, that had been cultivated in batch, in a 5 mM dichloromethane containing medium [26]. The micro-organisms were allowed to immobilize on the packing material forming a biolayer by growth, by recirculating the liquid phase through the trickling filter system.

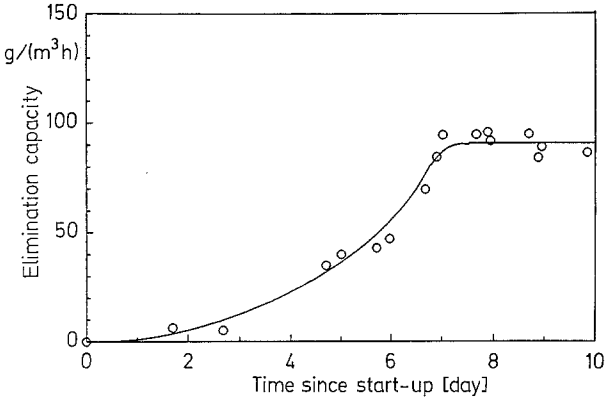

Fig. 4. The elimination capacity versus time during the start-up period; $u_l = 6$ m/h; $u_g = 80$ m/h; $C_{go} = 4$ g/m^3

During this period the system was operated countercurrently at a superficial liquid velocity of 6 m/h, while the dichloromethane concentration amounted to 4 g/m^3 at a superficial gas velocity of 80 m/h and a temperature of 30 °C.

Figure 4 shows the elimination capacity versus time, as determined during this period. It can be recognized that after about three days the filter activity started to increase rapidly, until it levelled off at an elimination capacity of 90 g/m^3 h after about seven days.

A similar recovery time of the filter performance as shown in Fig. 4 has also been observed after the biological activity

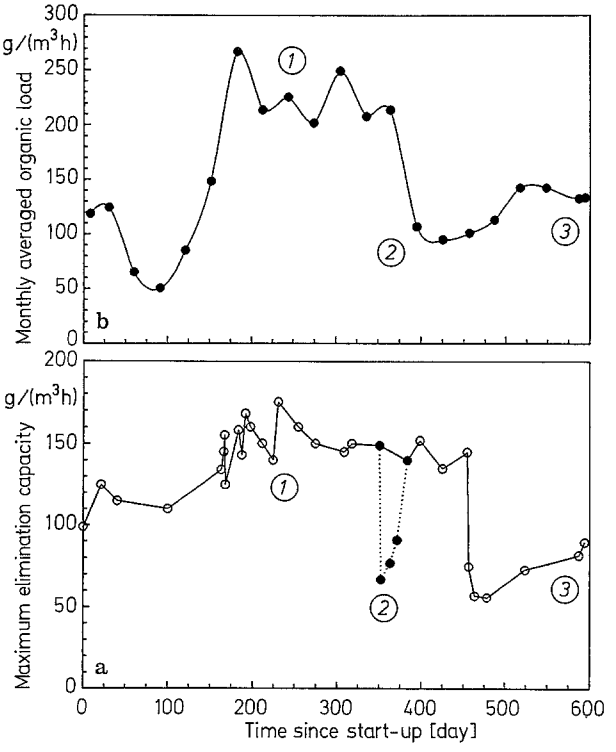

Fig. 5. a The maximum elimination capacity EC_{max}, determined at high organic load, plotted versus time. **b** Monthly averaged organic load (o) for the total period of operation

* This paper presents the experimental results of an investigation to the biological removal of dichloromethane in a trickling filter. The theoretical analysis of the system has been treated in Part I (published in Vol. 6, No. 3)

decreased down to low values, due to severe pH disturbances, which is illustrated by (2) in Fig. 5a. In this figure the maximum elimination capacity of the trickling filter, being the potential elimination performance of the system, is plotted versus time.

A sudden decrease of the EC_{max} resulted after a pH > 10 ((2) in Fig. 5a), while the monthly averaged organic load (Fig. 5b) was maintained at 220 g/(m^3 · h). It is shown that a total recovery of the elimination capacity took place within three weeks.

During a period of nearly two years the filter system was subjected to many different conditions in order to evaluate the effect of the various process parameters e.g. the inlet concentration, gas- and liquid flow rates, temperature etc.

However, from (1) in Fig. 5a and b (day 190–380) it can be seen that a stable trickling filter performance can be achieved as the EC_{max} remains constant ($\approx$150 g/(m^3 · h)), while a constant average organic load is maintained (220 g/(m^3 · h). This can only be explained by the existence of a balance between biomass accumulation by growth and biomass removal by decay and sloughing, resulting in a constant hold-up of active biomass, and thus a constant maximum elimination capacity.

From the foregoing it will be clear that the biology of the system is quite uneffected by short-term fluctuations in the organic load, which is very important in view of the dynamic behaviour of the filter system at briefly changing process conditions.

It can thus be concluded that the assumption of steady-state is valid in experiments which require a changing organic load, if the total period of time consumed only amounts to several days. On a longer term a steady-state will exist if a constant average organic load is maintained.

4.2 The trickling filter performance at standard conditions

As stated above the filter system was subjected to many different conditions during the period of operation. A reference set of process conditions was thus required in order to investigate the stability of the biological system by comparing the elimination capacity in the long run at this reference level. After preliminary experiments these reference conditions were set at a superficial gas- and liquid velocity of 160 m/h resp. 3.6 m/h, a temperature of 20 °C and an inlet gas concentration of 1.8 g/m^3. In case no experiments were carried out, the trickling filter was operated at these conditions.

The elimination capacity thus recorded will be referred to as the standard elimination capacity, which is shown in Fig. 6. It appears to be rather constant during the total period of operation, which affirms a stable trickling filter performance. It can also be concluded that this elimination capacity is quite independent of the effects introduced by whatever experiment carried out in the system.

In order to evaluate the physical and kinetic process parameters presented in Table 1, the elimination capacity of the system was determined regularly as a function of the inlet gas concentration in short term experiments (several days) at various process conditions. In the following this path is referred to as the performance curve.

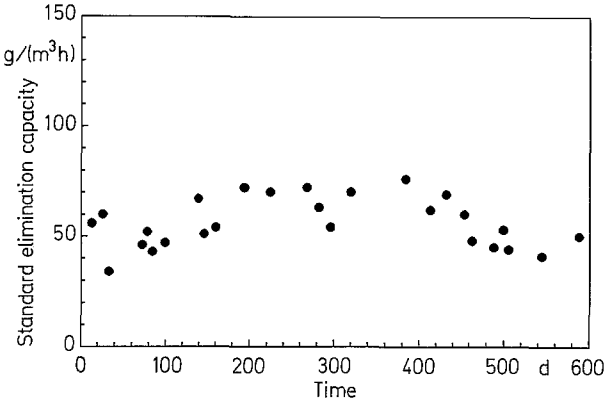

Fig. 6. The elimination capacity at standard conditions (•) plotted versus time; $u_g = 160$ m/h; $u_l = 3.6$ m/h; $C_{go} = 1.8$ g/m^3

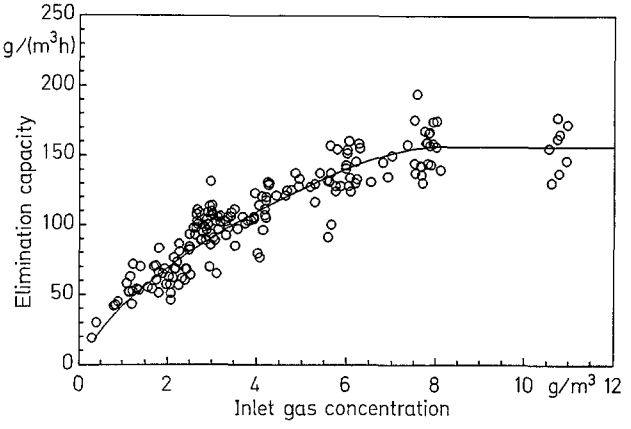

Fig. 7. The dichloromethane elimination capacity versus the inlet gas concentration at $u_l = 3.6$ m/h; $u_g = 160$ m/h; $EC_{max} = 157$ g/(m^3 · h); $C_{lcr} = 45$ g/m^3; data (o); Theoretical-model (solid line)

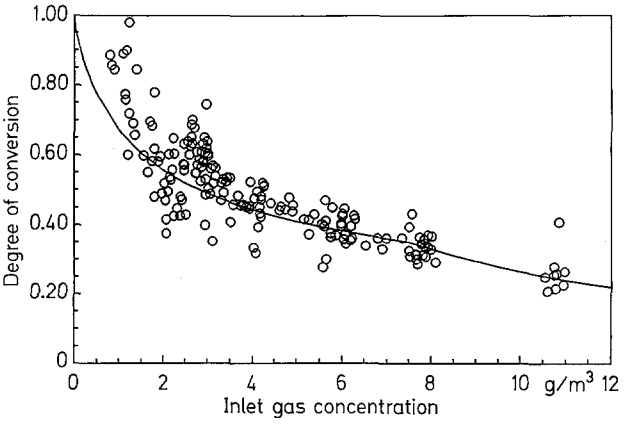

Fig. 8. Degree of conversion versus the inlet gas concentration of dichloromethane at standard conditions. $EC_{max} = 157$ g/(m^3 · h); $C_{lcr} = 45$ g/m^3. Experimental data (o); UCM-model (solid line)

In Fig. 7 the standard performance curve is shown, which is composed of all measurements performed at standard superficial gas- and liquid velocity. In Fig. 8 the degree of conversion calculated from these results is plotted versus the inlet concentration.

The experimental data in Fig. 7 show that the elimination capacity has a fairly time-independent relation to the inlet concentration. In this context it should be emphasized that the experimental data have been obtained during a period of operation of the filter system of nearly two years. The UCM model was used to calculate the EC_{max}, $C_{l\,cr}$ and N_{og} by numerical fitting the experimental data, using SAS statistics on a PC. The resulting parameter values, presented in Fig. 7 and Table 4, were used to calculate the theoretical performance curve according to the counter-current flow model. The solid line in Fig. 7 represents the theoretical performance curve of both models, as the differences between both theoretical performance curves are only minimal, which has already been pointed out in part I.

The computational procedure also showed that the number of transfer units as fitted by SAS was very high ($\gg 5$). From Eq. (A.4) it can be seen that no mass-transfer limitations are present for $N_{og} > 5$ (error $< 1\%$). The high values of N_{og} thus found indicate that SAS was not able to determine the exact value of N_{og} using this data set, but it can be concluded that the gas- and liquid mass-transfer resistance is very low or even negligible. This indicates that the biological process inside the biofilm is the rate-limiting process, while the gas- and liquid phase are close to equilibrium.

This was checked by measuring the axial concentration profiles in both phases. A representative example of the results is shown in Fig. 9, which shows indeed that both phases are close to equilibrium.

In these latter experiments the number of transfer units was determined by fitting the experimentally determined concentration profiles. However, fitting the concentration profiles which are calculated according to the counter-current flow model at $u_g = 160$ m/h and $u_l = 3.6$ m/h, it was concluded that the N_{og} number amounted to about 4, which confirms a very low mass-transfer resistance. From N_{og} values found in various situations it followed that in comparison with the N_{og} number calculated according to the correlations for mass transfer of Onda [35], a correction of about a factor two seems necessary in order to apply the Onda-correlations for the description of the mass-transfer in the trickling filter.

From the performance curve in Fig. 7, it can further be seen that the experimentally determined value of EC_{max} was much lower than can be expected beforehand on basis of intrinsic parameters. As EC_{max} is defined as $K_0 \cdot \delta \cdot a_w$, its magnitude can be estimated from literature data [25, 34–37]. With $\mu_{max} = 2.6\,d^{-1}$, $X_b = 75\,kg/m^3$ and $Y = 0.17\,g\,TSS/g\,DCM$, a zeroth order reaction constant $K_0 = 4.8 \cdot 10^4$ g/(m^3 · h) (defined per unit of biolayer volume) is calculated. Using $\delta \approx 250$ μm [37, 42] and $a_w = 158\ m^{-1}$ [35] it follows for the maximum elimination capacity for a fully active biolayer of solely Hyphomicrobium sp. to amount to $1.9 \cdot 10^3$ g/(m^3 · h).

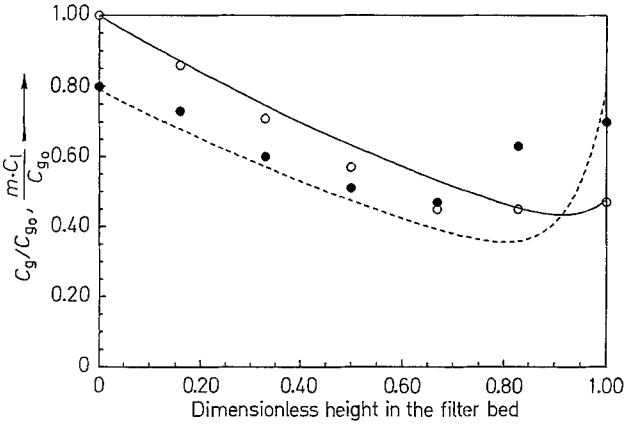

Fig. 9. The axial gas- (o) and liquid (•) concentration profiles, for counter-current flow at $C_{go} = 2.3$ g/m^3; $EC_{max} = 157$ g/(m^3 · h); $C_{l\,cr} = 45$ g/m^3; $u_l = 3.6$ m/h; $u_g = 160$ m/h; $N_{og} = 4$

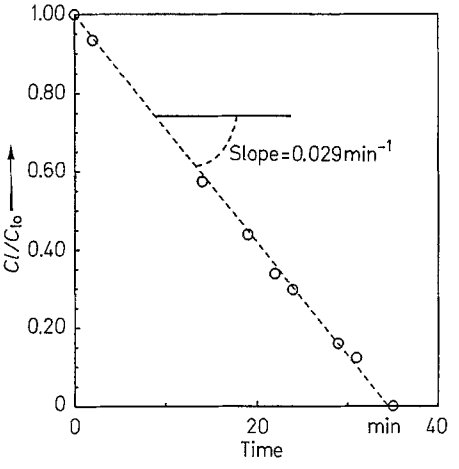

Fig. 10. The determination of the specific activity from a substrate depletion curve. $C_{lo} = 1$ mM; $X_s = 1.85\,kg/m^3$; $R_s = \text{slope} \cdot C_{lo} \cdot M_{DCM}/X_s = 0.08$ g DCM/(g TSS · h)

As this value is more than ten times the experimental EC_{max}, the literature data were evaluated, and it was concluded that K_0 was the most doubtful parameter. Therefore, biomass was taken from the filter bed, and its activity was determined in batch experiments. For this purpose biomass present in the recirculated water phase as well as biomass immobilized on the packing particles was separated and suspended in a buffered medium [26].

The specific activity of the biomass, defined as the amount of substrate degraded per unit of biomass and time, was determined by following the substrate depletion with time in 100 cm^3 of the buffered suspension, containing 1 mM dichloromethane.

In Fig. 10 such a depletion curve is shown, which normally is a straight line, as growth is negligible in comparison with the amount of biomass initially present. From this figure a specific activity can be calculated of $R_s = 0.08$ g DCM/(g TSS · h), a value which has been found several times during the total period of operation.

As this value is about a factor of eight lower than the specific activity of the growing pure culture of Hyphomicrobium sp (0.64 g/(g · h)), calculated as the ratio of μ_{max} and Y, it indicates that only about 12% of the biomass present in the trickling filter actually degrades dichloromethane. The remaining and major part of the biofilm obviously consists of inactive material and secondary organisms, as suggested in literature [40, 41]. Microbiological investigations of the biomass revealed, apart from the dichloromethane-degrading micro-organism, several other strains of bacteria, and a multitude of higher organisms like flagellates, ciliates, nematodes etc.

From $R_s = 0.08$ g DCM/(g TSS · h) a K_0 value of $6 \cdot 10^3$ g/m^3 h can now be estimated and the EC_{max} thus amounts to 237 g/(m^3 · h), which is of the same magnitude as the experimentally determined value in the trickling filter.

Although the use of kinetic data determined in batch investigations for a pure culture is often advocated [29], it can be concluded from the foregoing that it is a riskful procedure to estimate volumetric degradation rates for a dichloromethane degrading biofilm in a trickling filter system, due to the accumulation of inactive biomass and secondary organisms.

4.3 *Co-current versus counter-current flow*

As indicated by the numerical analysis of the models, only small differences exist between the trickling filter performance at co-current and counter-current flow throughout the parameter range of practical interest.

In order to verify this experimentally, the trickling filter performance curves were determined at several values of u_l and u_g for both modes of operation. In Fig. 11 the results are given for $u_g = 200$ m/h and $u_l = 10.8$ m/h.

It will be clear from this figure that a comparable filter performance is reached for both modes of operation, which is confirmed by the values of EC_{max} and $C_{l\,cr}$ presented in Table 3, which also lists the results of a similar experiment performed at $u_l = 3.6$ m/h.

It can be seen that the values of EC_{max} at $u_l = 3.6$ m/h presented in Table 3 are much lower than those presented in Fig. 7 at the same liquid flow rate. However, the present experiments were carried out at (3) in Fig. 5a and b (day 500–600). These figures show that the maximum elimination capacity in this period, which had been decreased strongly after a pH disturbance (day 450), recovered to only 90 g/(m^3 · h) as the average organic load was reduced at that time ($\approx$125 g/(m^3 · h)).

As illustrated by Fig. 12, considerable axial concentration gradients still exist, which was also observed in case of a counter-current flow (Fig. 9). Nevertheless, none of the two processes clearly results in a better performance. As already presented, this is probably caused by the recirculation of the liquid phase which has a smoothing effect on the axial liquid concentration gradients. Moreover, as the biofilm effectiveness factor only depends on the square root of the liquid concentration according to Eq. (7b), little net effect results in the elimination capacity of the trickling filter, whether operated at co-current or counter-current flow.

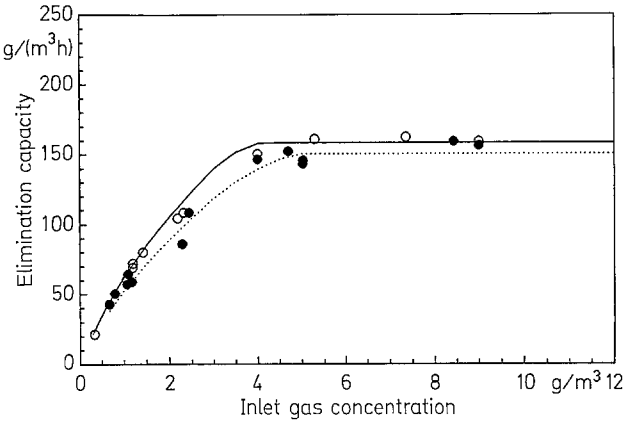

Fig. 11. The trickling filter performance at co-current (o; ——) and counter-current flow (●; · · · ·); $u_l = 10.8$ m/h and $u_g = 200$ m/h; $EC_{max} = 155$ g/(m^3 · h); $C_{l\,cr} = 19$ g/m^3

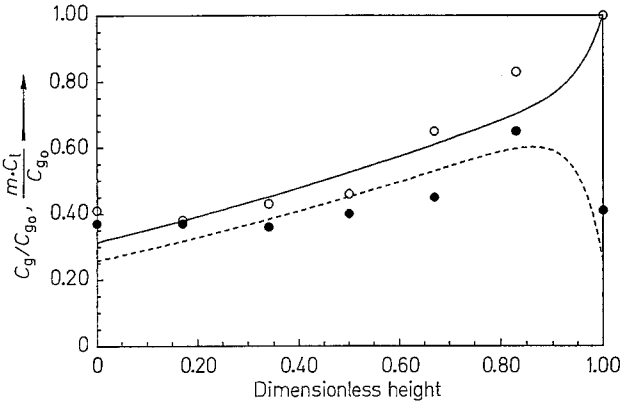

Fig. 12. Axial gas (o; ——) and liqid (●; · · · ·) concentration profiles for co-current flow at $C_{g_o} = 2.3$ g/m^3; $EC_{max} = 158$ g/(m^3 · h); $C_{l\,cr} = 17$ g/m^3; $u_l = 10.8$ m/h; $u_g = 200$ m/h

Table 3. The influence of the relative flow direction of the gas phase on the trickling filter performance curve at different liquid rates ($u_g = 200$ m/h)

	u_l [m/h]	EC_{max} [g/(m^3 · h)]	$C_{l\,cr}$ [g/m^3]
Counter-current	3.6	93	13
	10.8	151	20
Co-current	3.6	80	22
	10.8	158	17

4.4 *The influence of the superficial gas velocity*

From a process engineering point of view the gas velocity is expected to have a considerable effect on the system as it acts highly upon its mean residence time. In order to verify this

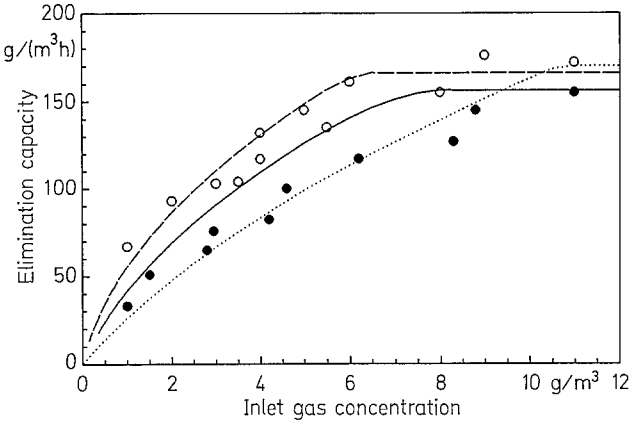

Fig. 13. The trickling filter performance data versus the inlet concentration determined at $u_g = 80$ m/h (•), 320 m/h (o), and the UCM-curves at 80 m/h (dotted line), and 320 m/h (dashed line)

Table 4. Influence of the superficial gas velocity on the trickling filter performance at $u_l = 3.6$ m/h

u_g [m/h]	EC_{max} [g/(m³ · h)]	$C_{l\,cr}$ [g/m³]	N_{og}
80	175	45	$\gg 5$
160	157	45	$\gg 5$
320	167	44	$\gg 5$

influence, experiments were carried out in which the trickling filter performance curves were determined at different superficial gas velocities. Counter-current flow was applied as not stated otherwise.

In Fig. 13 the measured trickling filter elimination capacity is plotted in relation to the inlet gas concentration for $u_g = 80$ m/h and $u_g = 320$ m/h, as well as the performance curves according to the theoretical model. The solid line in Fig. 13 represents the standard elimination performance, which has already been shown in Fig. 7. The parameter values which were found by fitting the UCM-model to the experimental data with SAS are presented in Table 4.

From this figure it can be seen that the elimination capacity increases at a higher superficial gas velocity for inlet concentrations smaller than about 11 g/m³. The figure also shows that the critical inlet gas phase concentration, indicating the transition from the diffusion- to the reaction limited regime in the trickling filter concerned, decreases from about 11 g/m³ at 80 m/h to 7.5 g/m³ at 160 m/h and 6.3 g/m³ at 320 m/h. As small fluctuations of the elimination capacity are known to exist in a continuously operating trickling filter, the EC_{max} values found at each superficial gas velocity can be regarded constant. The values of EC_{max} and $C_{l\,cr}$, which characterize the activity and thickness of the biofilm in the system, are both listed in Table 5.

It must be recognized that the critical inlet gas phase concentration can not be directly compared to $C_{l\,cr}$ using the Henry-coefficient, as the $C_{g_{ocr}}$ is a critical inlet concentration of the total system, while $C_{l\,cr}$ refers to the level of the liquid concentration, which has to be exceeded anywhere in the system, in order to reach the reaction limited regime. From Eq. (A.6) the relation between $C_{g_{ocr}}$ and $C_{l\,cr}$ can be derived using $C_{go} = C_{g\,cr}$ at $\eta = 1$. It follows:

$$C_{og\,cr} = \frac{EC_{max}\,H}{u_g(1-\exp\{^{-N_{og}}\})} + m\,C_{l\,cr}. \tag{13}$$

The SAS fitting procedure of the experimental data presented in Fig. 13 again revealed high N_{og} values ($\gg 5$). The observed increase of the elimination capacity at elevated gas flow rates and at any inlet concentration < 11 g/m³ can be explained by the experimentally encountered decrease of the degree of conversion. The lower degree of conversion results as the organic load increases linearly to the gas velocity at constant inlet concentration, while the elimination capacity increases less than linearly according to Eqs. (A.6) and (A.7). Consequently, the average gas phase concentration in the system increases at higher gas flow rates and constant C_{go}. As a higher average liquid concentration in this situation results, the biofilm efficiency and thus the elimination capacity also increases. Theoretically this can be derived from Eq. (A.6), in which the K_1^* value decreases, while K_2^* remains constant at a higher gas velocity. This results in an increased dependence of η on K_2^* and thus higher η values. At high inlet concentrations ($\gtrsim 11$ g/m³) the filter system performance is only limited by the rate of the biological reaction, as the biolayer is fully penetrated with substrate. Therefore, an increased average gas phase concentration in the system at a higher gas flow rate does not increase the elimination capacity.

As dichloromethane is only slightly soluble in water, the rate of mass-transfer has frequently been considered as the rate-limiting step of the overall process of dichloromethane removal from waste gases [20, 21]. Moreover, the application of a trickling filter is questioned for poorly soluble substrates [17].

However, not only the standard performance of the lab-scale trickling filter (Fig. 7) indicated that gas-liquid mass transfer resistance was negligible, but it can also be concluded from these experiments that in the trickling filter bed the elimination rate of dichloromethane is not limited by gas-liquid mass-transfer resistance even at low concentrations, which indicates that gas- and liquid phases will be close to equilibrium. The elimination rate can be well described by the counter-current flow model, as well as the UCM-model. Due to its simplicity the UCM model is advocated for practical application.

4.5 The influence of the superficial liquid velocity

Due to the recirculation of the liquid phase in the trickling filter system, not only the hydrochloric acid produced can be removed from the filter bed, but it is also possible to control

the physiological conditions in the liquid phase. Apart from the optimal conditions for the biological reaction the liquid flow may also influence the system performance by e.g. the formation and the thickness of the biofilm, and thus the maximum elimination performance which can be reached.

The overall influence of the superficial liquid velocity on the elimination capacity of the system was therefore investigated for u_l values ranging from 1.8 to 10.8 m/h.

The response of the system to an instantaneous increase of the liquid flow rate from 3.6 m/h to 7.2 m/h and thereupon to 10.8 m/h at a constant inlet gas concentration of 2.3 g/m^3 was investigated. The result in Fig. 14 shows that this increase is quickly followed by a rise of the elimination capacity.

It can also be observed that the increase of the liquid flow rate from 3.6 to 7.2 m/h almost yields a doubled elimination capacity, while an increase from 7.2 m/h to 10.8 m/h only results in a slight improvement of the degradation rate. These results may probably be explained by the existence of a "Randomly Wetted Area".

From a static point of view one would expect the existence of an active biofilm only on the continuously wetted area in the filter bed, which is kept active by the continuous supply of substrate and the simultaneous removal of the acids produced. On the non-wetted area no biofilm is expected to grow.

This would imply that an instantaneous increase of the liquid flow rate will initially only result in an increased interfacial area for mass-transfer, but no increased elimination rate as no biofilm (area) is added. However, as this increased elimination capacity was yet observed, it must be concluded that an increased amount of biomass already present was involved at higher liquid rates.

The phenomenon can be understood by a more dynamic character of the wetting process. Correlations from literature for the degree of wetting as suggested by e.g. Onda [35], only predict an average value for the specific wetted area, but they do not provide any information about the total area which becomes wetted "now and then" (i.e. a randomly wetted area). As the removal of the acids and thus the local pH is dependent on the wetting frequency of such spots, the biomass may survive the resulting low pH values at places where this wetting frequency exceeds certain values. These spots only contribute to a minor extend to the total elimination rate at lower liquid flow rates.

Thus the randomly wetted area contains biomass with a potential activity at higher liquid rates, as this area will partly be turned into a continuously wetted surface. Consequently, the total amount of biomass involved in the elimination increases as the optimal pH value is restored.

In a second series of experiments, the performance curve of the trickling filter was investigated at different superficial liquid velocities in order to model its influence theoretically. The experiments were carried out sufficiently long in order to allow the development of a biofilm on the total wetted area involved at each liquid flow rate.

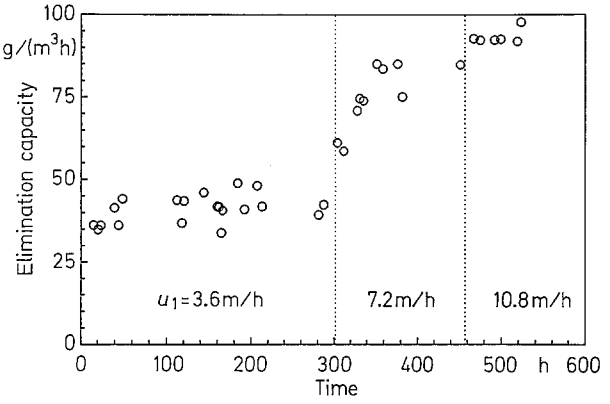

Fig. 14. The effect of the superficial liquid velocity on the elimination capacity at $C_{go}=2.3$ g/m^3 and $u_g=200$ m/h

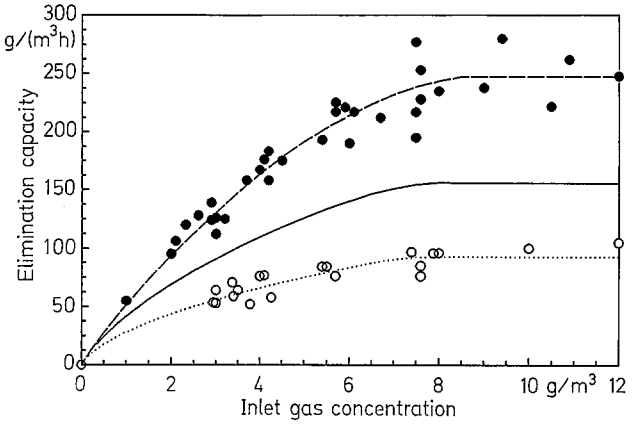

Fig. 15. The elimination capacity curves at a superficial liquid velocity of 1.8 m/h (o; dotted line) and 7.2 m/h (•; dashed line) and 3.6 m/h (solid line) at $u_g=160$ m/h

Table 5. Influence of the superficial liquid velocity on the maximum elimination capacity at $u_g=160$ m/h

u_l [m/h]	EC_{max} [g/(m^3 · h)]	C_{lcr} [g/m^3]	N_{og}
1.8	94	55	$\gg 5$
3.6	157	45	$\gg 5$
7.2	241	27	$\gg 5$
10.8	271	24	$\gg 5$

The results of the measurements at 1.8 m/h and 7.2 m/h are presented in Fig. 15, which gives the experimental data as well as the theoretical curves at these liquid velocities. The solid line in Fig. 15 represents the behaviour of the system at standard conditions already shown in Fig. 7. The resulting parameter values are listed in Table 5.

It will be clear that the liquid flow rate strongly influences the elimination capacity in the whole range of the inlet gas phase concentration. This is again explained by the dependence of the specific wetted area (a_w) on the superficial liquid

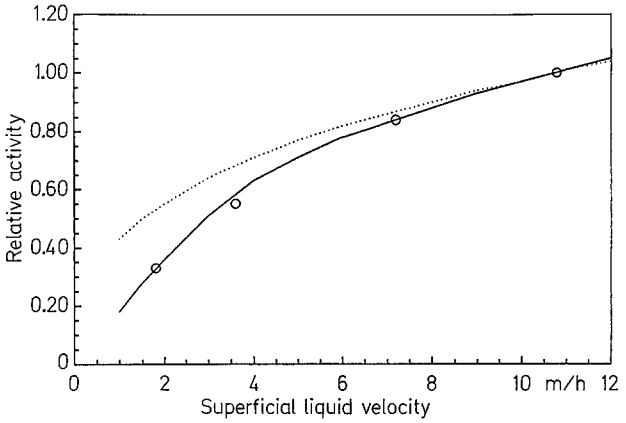

Fig. 16. The influence of the liquid velocity on the relative activity according to the relative wetted area (dotted line), the axial pH gradient (solid line) and experimental data of relative EC_{max} (o)

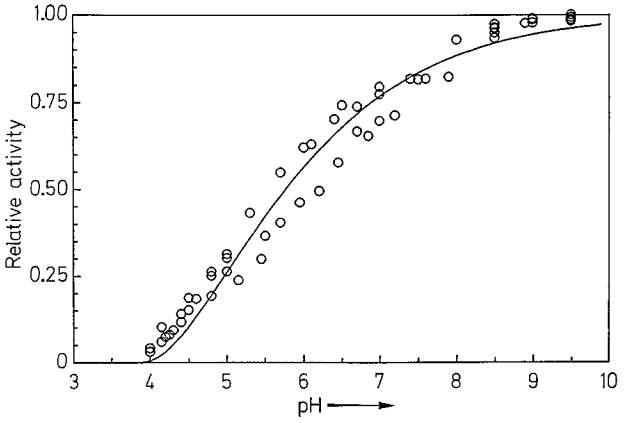

Fig. 17. The relative activity of the trickling filter biomass versus the pH in the liquid phase

velocity, as the elimination capacity is related to the wetted area by $K_0 \cdot \delta \cdot a_w \cdot \eta$. As for $\eta = 1$ the system is reaction limited, i.e. the elimination capacity is not influenced by the gas phase concentration, the influence of u_l on a_w can be estimated from the maximum elimination capacities reached at different liquid flow rates.

Figure 16 shows the effect of the liquid rate on the relative maximum elimination capacity (relative activity), which is defined as the ratio of EC_{max} at any superficial liquid rate and EC_{max} at 10.8 m/h, the latter being the maximum liquid rate applied.

As the EC_{max} is thought to be proportional to the degree of wetting, the relative maximum elimination capacity can be calculated according to the relation of e.g. Onda [35]. This is represented in Fig. 16 by the dotted line. From this figure it can be concluded that at lower superficial liquid velocities the relative EC_{max} is much more reduced than expected on basis of the relative degrees of wetting.

The existence of an axial pH gradient is thought to be responsible for this phenomenon, as the hydrochloric acid produced in the biofilm accumulates in the flow direction of the liquid phase. This axial pH gradient is influenced by the acid production rate, but also by the amount of dissolved buffering compounds e.g. phosphates, ammonium and carbonates. The phosphates and the ammonium are added to the system as inorganic nutrients, while carbonates are formed by the neutralization of the carbonic acid produced in the substrate degradation. The dependence of the rate of the biological reaction on the local pH value has been determined for the trickling filter biomass in batch investigations in our laboratory. In Fig. 17 the relative biological activity is plotted versus the pH-value in the liquid phase. It is shown that a maximum activity is reached at about pH = 9, while the activity reduces to about zero at pH = 4.

Table 6. The experimental maximum elimination capacity, relative to the EC_{max} at $u_l = 10.8$ m/h, compared to the relative degree of wetting (Onda) with and without a correction for the axial pH gradient

u_l [m/h]	$\left(\frac{EC_{max}}{EC_{max,\,10.8}}\right)_{exp}$	$\left(\frac{a_w}{a_{w,\,10.8}}\right)_{Onda}$	Relative activity after pH and a_w correction
1.8	0.33	0.53	0.35
3.6	0.55	0.69	0.58
7.2	0.84	0.87	0.89
10.8	1	1	1

Using the buffer concentrations and the pH-activity curve, the axial pH gradient as well as the resulting EC_{max} can be calculated in dependence of the liquid flow rate. This overall effect is represented by the solid line in Fig. 16, while Table 6 lists the experimental and theoretical data.

The experimental relative EC_{max} showed a good agreement with the calculations, as did the experimental pH values in the liquid draining from the filter bed. At the maximum elimination capacity, the pH in the liquid phase dropped from an initial value of 8 down to 4–4.5 at 1.8 m/h, while at 3.6 m/h a value of 5.5–6 was reached. At $u_l = 10.8$ m/h no pH drop was observed, which confirms the applicability of the EC_{max} reached at this fluid flow rate as a reference level.

From Table 5, which lists the data of the experiments carried out at different liquid flow rates, it can be recognized that at higher liquid velocities a decreasing value for the critical liquid concentration is calculated. This can be explained by a decrease of the effective biofilm thickness due to sloughing as a result of an increased sheer stress by the liquid film. Indeed higher biomass concentrations in the liquid phase were obtained experimentally, however, much lower than could be expected on basis of the $C_{l\,cr}$ values. Hence, the filter bed apparently also acts as a mechanical filter for suspended solids. A redistribution of the sloughed biomass over the wetted area in the filterbed has taken place, which positively influences the elimination capacity in the diffusion limited regime.

From this observation it can be concluded that the loss of biomass due to sloughing and washing out by the drain of

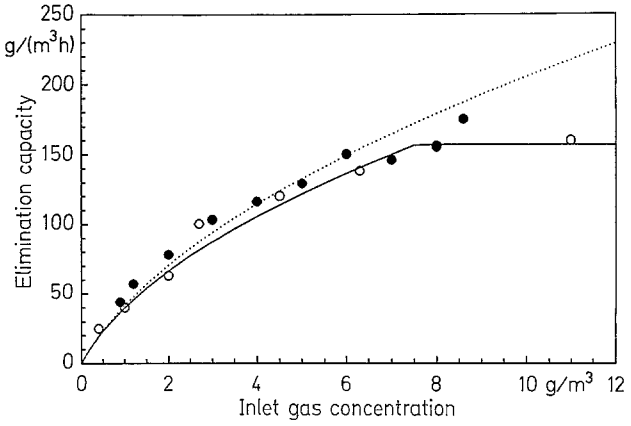

Fig. 18. The performance of a counter-currently operated trickling filter at 20 °C (o; ——) and 30 °C (●; · · · ·). The curves are calculated according to the theoretical model

liquid from the system is thus strongly reduced by the recirculation of the liquid phase and the mechanical filtering capacity of the packed bed.

4.6 The influence of the temperature on the trickling filter performance

If applied in practice, trickling filters will normally be subjected to temperature fluctuations as a result of e.g. a changing temperature of the inlet gas, the evaporation of water from the system, a change of temperature of the surroundings, and heat generated by the recirculation of the liquid phase.

In order to investigate the influence of the temperature on the elimination capacity of a trickling filter system experiments were performed at 20 °C and 30 °C at a liquid- and gas flow rate of 3.6 m/h and 160 m/h respectively, and at changing inlet gas concentrations.

From the experimental data shown in Fig. 18, it can be concluded that the temperature hardly affects the overall performance of the system up to a gas inlet concentration of ca. 8 g/m^3.

This result may be surprising as it is well known that the temperature effects the biological reaction rate to a great extent. From batch growth experiments in our laboratory an activation energy of 50 kJ/mol was determined for the degradation of dichloromethane by Hyphomicrobium sp. GJ 21 [25]. This value is very common for microbial growth and substrate degradation rates, which typically amount to 50–75 kJ/mol [5, 45].

From the value of the activation energy given, an increase of the reation rate K_0 and hence of the number of reaction units N_r and the critical liquid concentration $C^*_{l\,cr}$ by a factor two can be estimated according to Arrhenius' law for a temperature rise from 20 °C to 30 °C. It can be shown that the effect of a temperature increase in the range indicated on the acceleration of the diffusion rate inside the biofilm is very small, while an increase of the gas-liquid mass-transfer coefficient can be neglected.

Table 7. The influence of a rise in temperature from 20 °C to 30 °C on the most important biological and physical parameters

Kinetic data	20 °C	30 °C
EC_{max} [g/(m^3 · h)]	157	309
$C_{l\,cr}$ [g/m^3]	44	87
m	0.11	0.16
N_{og}	>5	>5

The fact that the overall performance of the system in the diffusion limited range ($C_{go} < 8$ g/m^3) is nevertheless not affected by a temperature rise can be explained by a decrease of the mass-transfer rate from the gas to the liquid phase as described by Eqs. (8) and (9). Due to an increase of the distribution coefficient m at higher temperatures, the equilibrium liquid concentration of dichloromethane decreases, which reduces considerably the driving force for mass-transfer between the mobile phases.

According to Leighton and Calo [34] an increase of the distribution coefficient can be calculated from 0.11 at 20 °C to 0.16 at 30 °C.

The counterbalancing effect of a temperature rise on the microbial reaction rate and the gas-liquid distribution coefficient has been confirmed by the calculated curves at 20 °C and 30 °C according to the counter-current flow model and the UCM-model presented in the Appendix. The parameter values used in these calculations are listed in Table 7. The theoretical curves shown in Fig. 18 are in good agreement with the experimental results.

5 Conclusions

The results from the present investigation concerning the removal of dichloromethane from waste gases in a biological trickling filter show that a stable performance can be achieved on a long term, while the start-up of the system is rather quick.

In the trickling filter described, the gas-liquid mass-transfer resistance appeared to be very low. Hence, a simplified steady-state model was developed, which could very well describe the elimination performance of the trickling filter at various conditions. The trickling filter performance of the system presented in this paper, which appeared to be independent of the liquid temperature, can be characterized by a maximum dichloromethane elimination capacity of 157 g/(m^3 · h) and a critical liquid concentration of 45 g/m^3 at a superficial liquid rate of 3.6 m/h.

The theoretical model also showed that the degree of conversion in a trickling filter can be described as a function of the total superficial gas contact time. Not only does this imply that the height and diameter of a system can freely be chosen at a constant reactor volume, but it also appeared that the performance of a series of successive trickling filters is hardly affected by the number of successive stages, each with

its own recirculation system, as long as a total gas contact time is maintained constant.

In order to design a trickling filter without pilot plant experiments, knowledge is not only required about the intrinsic kinetic growth parameters, but also of the characteristics of the biofilm to be formed. From the present investigation it appears that the application of the intrinsic growth parameters can be a very riskful procedure as e.g. inactive biomass may accumulate, resulting in a lowered specific biomass activity. For the biomass in the trickling filter degrading dichloromethane a specific activity of 0.08 g DCM/(g TSS · h) was found, while the pure culture showed a specific activity of 0.64 g DCM/(g TSS · h).

Nevertheless the practical application of a trickling filter for dichloromethane removal is very promising, while the UCM model can be easily used to predict the trickling filter performance.

This model can be generally applied if the assumptions presented are met, provided that the relevant model parameters for the removal of other compounds by other microorganisms are known from a reliable estimation or rather from pilot plant data.

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