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# Biofiltration of H<sub>2</sub>S in air - experimental comparisons of original packing materials and modeling

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## Abstract

The treatment of hydrogen sulfide using a biofilter packed with expanded schist and topped with a layer of a synthetic nutritional material (UP20) was examined at a constant H<sub>2</sub>S concentration (100 ppmv). The impact of the empty bed residence time (EBRT) on process performances was clearly underlined by varying the polluted air flow from 4 to 20 m<sup>3</sup> h<sup>-1</sup> corresponding to a variation in the EBRT from 63 to 13 s. Complete H<sub>2</sub>S degradation was observed when the EBRT was higher than 51 s. Experimental data collected at various EBRTs (13 – 63 s) were fitted using the Ottengraf model equations. The  $\alpha_{\text{lump}}$  parameter value was found to be 26.4 g<sup>1/2</sup> m<sup>-3/2</sup> h<sup>-1</sup>. This single parameter, which enables the performance of the biofilter as a whole to be characterized whatever its composition (mixture or layers of different packing materials) and whatever the EBRT, is a powerful tool to compare packing materials and to design such bioreactors. The  $\alpha_{\text{lump}}$  value characterizing the performances of expanded schist coupled with a thin layer of UP20 was higher than the  $\alpha_{\text{lump}}$  values obtained for other packing materials (natural or synthetic) reported in previous studies.

**Key words:** Biofilters; Modeling; Diffusion-Reaction; Waste Treatment; Hydrogen sulfide; Odors.

## Nomenclature

A: Specific area ( $\text{m}^2_{\text{biofilm}} \text{m}^{-3}_{\text{packing material}}$ )

C: Gas concentration ( $\text{g m}^{-3}_{\text{gas}}$ )

$C_L$ : Pollutant concentration in the biofilm ( $\text{g m}^{-3}_{\text{biofilm}}$ )

d: Diameter (m)

D: Diffusion coefficient ( $\text{m}^2_{\text{biofilm}} \text{s}^{-1}$ )

EBRT: Empty Bed Residence Time (s);  $\text{EBRT} = V / Q_v$

EC: Elimination Capacity ( $\text{g}_{\text{H}_2\text{S}} \text{m}^{-3}_{\text{packing material}} \text{s}^{-1}$ );  $\text{EC} = (Q_v / V) (C_{\text{in}} - C_{\text{out}})$

H: Height (m)

k: Zero order reaction rate constant ( $\text{g m}^{-3}_{\text{biofilm}} \text{s}^{-1}$ )

LR: Loading Rate ( $\text{g}_{\text{H}_2\text{S}} \text{m}^{-3}_{\text{packing material}} \text{s}^{-1}$ );  $\text{LR} = (Q_v C_{\text{in}} / V)$

m: Partition coefficient (-)

$Q_v$ : Gas flow rate ( $\text{m}^3_{\text{gas}} \text{s}^{-1}$ )

R: Reaction rate constant ( $\text{g m}^{-3}_{\text{packing material}} \text{s}^{-1}$ );  $R = k A \delta$

RE: Removal Efficiency (%);  $\text{RE} = 100 (C_{\text{in}} - C_{\text{out}}) / C_{\text{in}}$

U: Superficial gas velocity ( $\text{m}_{\text{gas}} \text{s}^{-1}$ )

V: Bed volume of packing material ( $\text{m}^3_{\text{packing material}}$ )

x: Length coordinate (m)

### *Greek letters*

$\alpha_{\text{lump}}$ : Lump parameter ( $\text{g}^{1/2} \text{m}^{-3/2}_{\text{packing material}} \text{s}^{-1}$ ) (Ottengraf's equations)

$\delta$ : Total biofilm thickness (m)

$\varepsilon$ : Porosity of the packing material (-)

$\phi$ : Thiele modulus (-)

$\lambda$ : Effective biofilm thickness (m)

$\sigma$ : Dimensionless length coordinate in the biofilm ( $= x/\delta$ )

### *Subscripts*

Crit: Critical

in: Inlet

out: Outlet

## 1. Introduction

Hydrogen sulfide (H<sub>2</sub>S) is a hazardous, toxic air pollutant. It is a colorless, corrosive and flammable gas. H<sub>2</sub>S can be problematic due to its unpleasant smell and low odor threshold. It is emitted from many industrial activities such as petroleum refining, leather, waste or wastewater treatments, food processing, anaerobic treatment of paper and pulp manufacturing. Conventionally, different processes have been used to remove H<sub>2</sub>S from waste gas streams involving chemical and physical methods. For some years, the focus has shifted toward using biofiltration. This process presents an attractive technology for treating pollutants from air due to its effectiveness, low energy consumption and minimal by-product generation. The gas stream flows through the filter bed. Pollutants are then transferred from the gas phase to the biofilm, where they are metabolized by microorganisms. The by-products of the complete biodegradation of air pollutants are CO<sub>2</sub>, water, and microbial biomass. In the case of H<sub>2</sub>S biodegradation, sulfur oxidizing bacteria (SOB) are responsible for removing H<sub>2</sub>S in aerobic conditions. For their maintenance and growth, SOB use H<sub>2</sub>S as a source of energy and CO<sub>2</sub> as the main source of carbon [1,2]. Bacteria from the genus *Thiobacillus* are responsible for the oxidation of H<sub>2</sub>S to sulfate and/or elemental sulfur according to the operating conditions [3,4]. The biofiltration of H<sub>2</sub>S is well documented (Table 1). As this table shows, a variety of packing materials are used and biofiltration performances are disparate. These packing materials include: (i) organic materials such as soil, peat, compost and pine bark [3,5–10] and different forms of activated carbons [11,12]; (ii) inorganic materials like pozzolan, expanded schist and lava rock [13–15]; (iii) synthetic media such as a patented biofilter medium (Biosorbens™) developed by Shareefdeen [16,17]. Nonetheless, recent studies highlighted that biofilters filled with expanded schist topped with a layer of synthetic nutritional material (UP20) were very efficient for removing high loading rates of H<sub>2</sub>S [15,18,19]. The good mechanical behavior of the expanded schist (low pressure drop) and the ability of biofilters to oxidize H<sub>2</sub>S under extreme acidic conditions for a long period confirmed the advantage of using expanded schist coupled with UP20 for industrial applications [14,15,18,20]. In biofiltration, the empty bed residence time (EBRT) is the key parameter influencing biofilter performances. Usually, EBRTs from 20 to 60 s are applied to remove H<sub>2</sub>S from air [6,16,21] but higher and lower values are reported in the literature. As illustrated in Table 1, the EBRT may be significantly different from one study to another (from 2 to 120 s, i.e. almost two orders of magnitude). Consequently, it is very difficult to compare the performance of different packing materials on the basis of the Elimination Capacity (EC in g m<sup>-3</sup> h<sup>-1</sup>) measured at different EBRTs and the Removal Efficiency (RE), which can be other than 100%. The literature results presented in Table 1 clearly illustrate that the comparison of biofilter performances is difficult. For instance, is it possible to compare the performance of peat reported by Oyarzun *et al.* [3] (EC = 14.8 g m<sup>-3</sup> h<sup>-1</sup> at EBRT = 120 s and RE = 100%) with that of the biofilter medium Biosorbens™ reported by Shareefdeen [16] (EC = 6 g m<sup>-3</sup> h<sup>-1</sup> at EBRT = 30 s and RE = 99%)? To overcome this problem, it would be better to base the comparison of the performances of packing materials on mathematical models. Several models have been proposed to predict the performances of biofilters and to improve biofilter design [22,23]. One of the earliest steady-state biofiltration models was developed by Ottengraf and Van den Oever [24]. Because of its mathematical simplicity, this model has been widely used for biofiltration [3,25,26]. Therefore, the objective of this work was to show

that a single parameter (called  $\alpha_{\text{lump}}$ ) derived from the Ottengraf model equations can be used as a simple tool to compare the performances of different carrier materials used in H<sub>2</sub>S biofiltration whatever the configuration of the biofilter and whatever the EBRT. The Ottengraf model was therefore applied (i) to determine the  $\alpha_{\text{lump}}$  parameter experimentally in order to evaluate the performance of a biofilter filled with expanded schist topped with a layer of synthetic nutritional material (UP20) and (ii) to compare this latter with the performance of packing materials reported in the literature. To achieve these objectives, the ability of the biofilter to oxidize H<sub>2</sub>S at different EBRTs should be determined beforehand. Therefore, this paper presents a brief description of the mathematical model used and details the experimental study carried out.

**Table 1.** Examples of recent biofiltration results reported in the literature on the treatment of gas polluted by H<sub>2</sub>S.

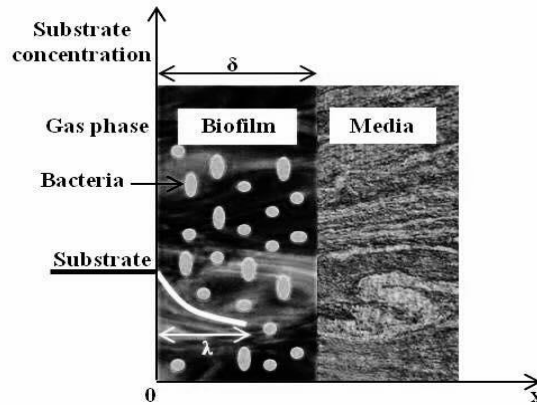
| Packing material               | EBRT (s)       | Elimination Capacity EC (g m <sup>-3</sup> h <sup>-1</sup> ) | Removal Efficiency RE (%) | Reference |
|--------------------------------|----------------|--|---------------------------|-----------|
| Compost                        | 45             | 64   | 100                       | [27]      |
| Peat                           | 120            | 14.8   | 100                       | [3]       |
| Peat                           | 60             | 65.9   | 90                        | [28]      |
| Peat                           | 57             | 25.5   | 50                        | [21]      |
| Fibrous peat                   | 29             | 5.7  | 90                        | [29]      |
| Peat moss                      | 20<br>40       |  | 85<br>90                  | [30]      |
| Pig manure + sawdust           | 24             | 5<br>46  | 97<br>83                  | [31]      |
| Pig manure + sawdust (ABONLIR) | 13.5<br>27     | 20<br>45   | 90<br>90                  | [32]      |
| Sugarcane bagasse              | 49             | 73   |                           | [7]       |
| Coconut fiber                  | 49             | 68   |                           | [7]       |
| Pine bark                      | 57             | 10   | 69                        | [19]      |
| Sapwood                        | 57             | 8  | 50                        | [21]      |
| Woodchips                      | 20<br>40       |  | 90<br>95                  | [30]      |
| Activated carbon               | 2 - 21         | 181  | 94                        | [33]      |
| Granular activated carbon      | 20<br>40       |  | 96<br>100                 | [30]      |
| Granular activated carbon      | 240            | 125  | 98                        | [34]      |
| Pellet activated carbon        | 2              | 181  | 94                        | [12]      |
| Exhausted carbon               | 4 - 25         | Up to $\approx$ 40   | 94 - 99                   | [35]      |
| Synthetic medium (UP20)        | 57             | 10   | 93                        | [19]      |
| Synthetic medium               | 20             |  | 99                        | [36]      |
| Synthetic medium (BIOSORBENS™) | 30             | 6  | 99                        | [16]      |
| Peat + UP20 (mixed)            | 57             | 25.5   | 80                        | [21]      |
| Expanded schist                | 16             | 30   | 100                       | [14]      |
| Expanded schist + UP20         | 16<br>24<br>35 | 20<br>25<br>36   | 90<br>90<br>100           | [15]      |
| Pozzolan + UP20 (layers)       | 57             | 10   | 74                        | [19]      |
| Porous ceramic                 | 6              | 160  | 96                        | [37]      |
| Ceramic                        | 20<br>40       |  | 80<br>83                  | [30]      |

|   |    |      |    |      |
|---|----|------|----|------|
| Dried activated sludge                                      | 30 | 44.3 | 82 | [38] |
| Mixed dried activated sludge with rice husk silica          | 30 | 52.3 | 97 | [38] |
| Polyurethane foam   | 80 | 56.6 | 95 | [39] |
| Polyurethane foam   | 49 | 66   |    | [7]  |
| Biomedium encapsulated by Na-alginate and polyvinyl alcohol | 51 | 6    | 99 | [40] |

## 2. Ottengraf model equations

In order to describe the mechanisms of transfer and biodegradation in the biofilter (Fig. 1), Ottengraf and Van den Oever [24] proposed a simple model based on the theoretical model built by Jennings *et al.* [41]. The hypotheses are as follows:

- Biodegradation occurs in a biofilm considered to be water.
- Biofilm thickness is small compared to the packing material diameter.
- Biomass concentration is homogeneous in the reactor.
- Gas phase is ideal.
- Gas phase is a plug flow.
- Mass transfer resistance in the gas phase is negligible.
- Regime is at steady-state.
- Equilibrium occurs at the gas-biofilm interface.



**Fig. 1.** Substrate concentration profile in the biofilm: diffusional regime.

Moreover, Ottengraf and Van den Oever considered that the reaction rate constant of the substrate elimination in the biofilm is of zero-order in the pollutant concentration, which assumes a very low value of the Michaelis-Menten constant in the Monod equation [24]. Zero-order kinetics are encountered at high concentrations of  $H_2S$ , which is generally the case in laboratory experiments. With these assumptions, the concentration of a nutrient component inside the biofilm ( $C_L$ ) is described using the differential equation:

$$D \frac{d^2 C_L}{dx^2} - k = 0 \quad (1)$$

with the boundary conditions:

$$x = 0; \quad C_L = \frac{c}{m} \quad (2)$$

$$x = \delta; \quad \frac{dC_L}{dx} = 0 \quad (3)$$

The solution of the differential equation is:

$$\frac{C_L}{C/m} = 1 + \frac{1}{2} \frac{\phi^2}{C/C_{in}} (\sigma^2 - 2\sigma) \quad (4)$$

with:

$$\phi = \delta \sqrt{\frac{m k}{D C_{in}}} \quad (5)$$

where  $\phi$  is the Thiele modulus,  $\sigma = x/\delta$  is the dimensionless length coordinate in the biofilm, and  $m = (C/C_L)$  is the gas-liquid partition coefficient obtained from Henry's law. Two situations expected to be common in biofilters were then considered by Ottengraf and Van den Oever [24]: zero-order kinetics with biological limitation and zero-order kinetics with diffusional limitation.

Considering the reaction-limited regime, the conversion rate of the pollutant is controlled by the reaction rate and Ottengraf and Van den Oever [24] calculated that the pollutant conversion is:

$$\frac{C_{out}}{C_{in}} = 1 - \frac{A k \delta}{C_{in}} \frac{H}{U} \quad (6)$$

Considering the diffusion-limited regime, the substrate concentration in the biofilm is high and the diffusion into the biofilm is a limiting factor. Hence, the substrate cannot reach the bacteria, which leads to the presence of some inactive biofilm zones. An active biofilm thickness  $\lambda$  is defined, less than the biofilm thickness  $\delta$  (Figure 1). In this case, the concentration profile provided by the Ottengraf model is given by the following equation:

$$\frac{C_{out}}{C_{in}} = \left( 1 - \frac{A H}{U} \sqrt{\frac{k \cdot D}{2 \cdot m}} \sqrt{\frac{1}{C_{in}}} \right)^2 \quad (7)$$

Taking into account that  $H/U = EBRT$ , Eq. (7) can be rewritten as:

$$\frac{C_{out}}{C_{in}} = \left( 1 - A \sqrt{\frac{k \cdot D}{2 \cdot m}} \sqrt{\frac{1}{C_{in}}} EBRT \right)^2 \quad (8)$$

From Eq. (8), Deshusses and Shareefdeen [23] included a parameter in this model (called  $\alpha_{lump}$ ) that can be expressed as follows:

$$\alpha_{lump} = A \sqrt{\frac{k \cdot D}{2 \cdot m}} \quad (9)$$

Subsequently, the concentration profile given by Eq. (7) can be written as:

$$\sqrt{\frac{C_{out}}{C_{in}}} = 1 - EBRT \alpha_{lump} \sqrt{\frac{1}{C_{in}}} \quad (10)$$

It should be kept in mind that in Eq. (9),  $k$  represents the reaction rate per unit of volume of the biofilm [24], while  $A$  is the surface area of the biofilm developed on the volume of packing material. As a result, the unit of the parameter  $\alpha_{lump}$  ( $\text{g}^{1/2} \text{m}^{3/2} \text{biofilm} \text{m}^{-3} \text{packing material} \text{s}^{-1}$ ) is difficult to understand from a physical point view. Moreover, it should be noted that this unit of the parameter  $\alpha_{lump}$  is not expressed in [23]. Shareefdeen suggested expressing it in ( $\text{s}^{-1}$ ). In this case, the pollutant gas concentration and the EBRT can be expressed in (ppmv) and (s), respectively [17,36]. In another publication [42], this author suggested expressing the pollutant gas concentration in ( $\text{kg} \text{m}^{-3}$ ), the EBRT in (s) and  $\alpha_{lump}$  in ( $\text{s}^{-1}$ ). However, in the latter case, the unit of the parameter  $\alpha_{lump}$  should be ( $\text{kg}^{1/2} \text{m}^{-3/2} \text{s}^{-1}$ ). Consequently, although the parameter  $\alpha_{lump}$  could be a suitable tool to compare the performance of the packing materials used in biofiltration, the units used in Eq. (10) to calculate this parameter should be considered with caution.

Using the definitions of the Loading Rate (LR) and the Elimination Capacity (EC) usually employed in studies devoted to biofiltration, Eq. (10) can be easily written as follows:

$$EC = LR \left( 1 - \left( 1 - \alpha_{lump} \sqrt{\frac{EBRT}{LR}} \right)^2 \right) \quad (11)$$

In Eq. (11), the unit of the parameter  $\alpha_{lump}$  is ( $\text{kg}^{1/2} \text{m}^{-3/2} \text{packing material} \text{h}^{-1}$ ) and thus, by determining EC and LR,  $\alpha_{lump}$  can be calculated in order to compare different packing materials for  $\text{H}_2\text{S}$  biofiltration as shown in [21,43]. In other words, by using EC and LR in ( $\text{g} \text{m}^{-3} \text{packing material} \text{h}^{-1}$ ) and EBRT in (h), the determination of  $\alpha_{lump}$  from Eq. (11) is unambiguous. Although some assumptions of the Ottengraf model are questionable, it has been validated by different authors in various conditions with good agreement between experimental data and calculated values [22]. Moreover, an analytical solution such as Eq. (11) is more suitable than a numerical solution for data analysis and biofilter design.

### 3. Materials and methods

#### 3.1. Packing material

Expanded schist, a natural inorganic support (Granulex Company; France; [www.granulex.fr](http://www.granulex.fr)) was used as packing material (Figure 2). Several studies carried out in laboratory-scale biofilters showed that this material is efficient for removing  $\text{H}_2\text{S}$  [14,15,18]. Moreover, its excellent mechanical behavior, which results in low pressure drops over a long period, is suitable for long-term industrial applications. The composition of this medium was determined using an Energy Dispersive X-ray Fluorescence Spectrometer (EDX-800HS,

Shimadzu Company). Its composition is as follows: SiO<sub>2</sub> (55.3%), Al<sub>2</sub>O<sub>3</sub> (20.2%), Fe<sub>2</sub>O<sub>3</sub> (13.3%) and K<sub>2</sub>O (5.1%). The characteristics of the pieces of expanded schist used in this study are given in Table 2. The specific surface area was measured by a Micromeritics AutoPore IV 9500 mercury porosimeter.

In order to provide nutrients for the expanded schist, a synthetic nutritional material (UP20) was manufactured in our laboratory. This is a cylindrical-shaped extruded material (7 mm in diameter and 15 mm in length) whose formulation has been described previously [44]. UP20 contains urea phosphate (CH<sub>4</sub>N<sub>2</sub>O, H<sub>3</sub>PO<sub>4</sub>), calcium carbonate (CaCO<sub>3</sub>) (C/N/P molar ratio: 100/10/5) and an organic binder (20% in mass) from the Elotex company. The Elotex binder contains ethylene and vinyl acetate. It has been shown that the combination of expanded schist and UP20 can be successfully used to remove high concentrations of H<sub>2</sub>S, up to 360 ppmv [20].

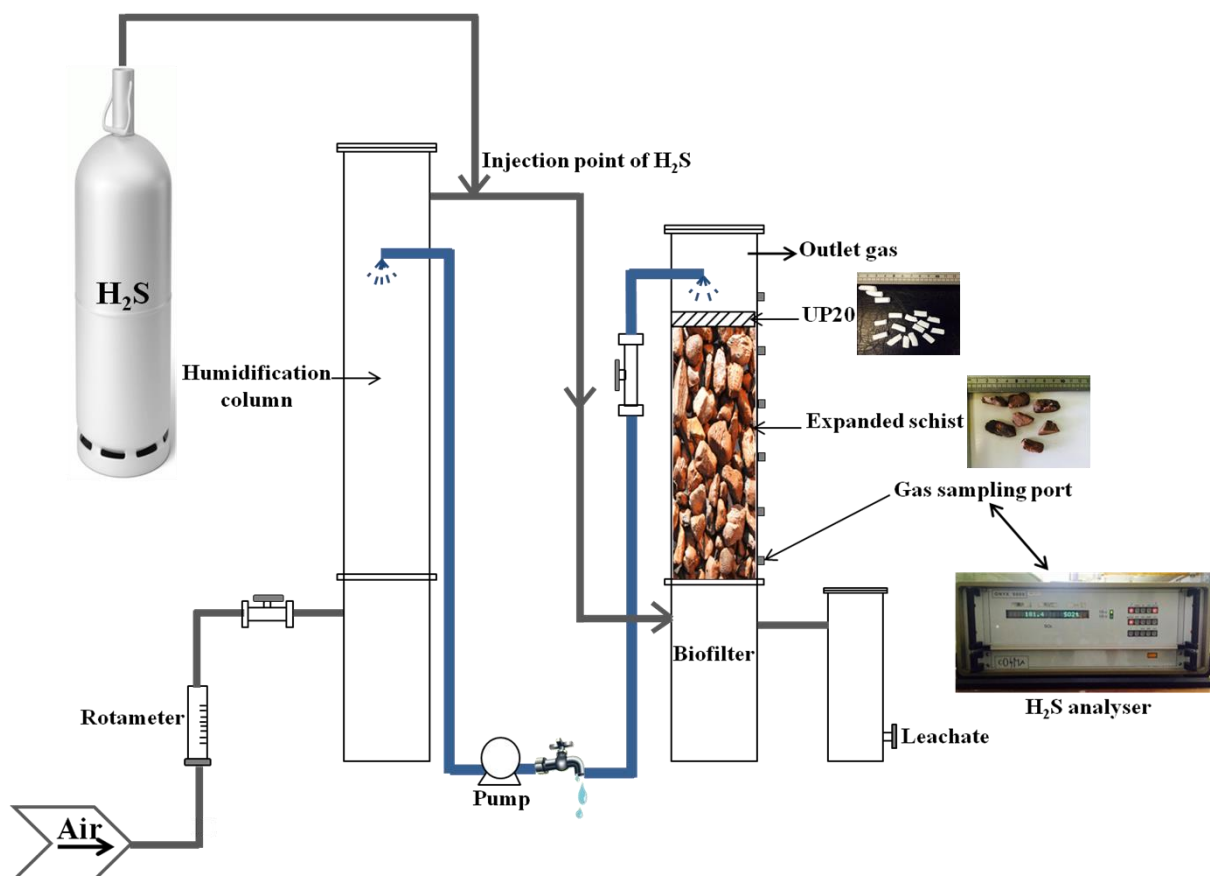
**Table 2.** Characteristics of (i) expanded schist used in this study; (ii) patented medium (Biosorbens™) developed by Shareefdeen *et al.* [16].

|   | <b>Expanded schist</b> | <b>(Biosorbens™)</b> |
|---|------------------------|----------------------|
| Bulk density (kg m <sup>-3</sup> )                      | 667                    | 650                  |
| Particle size (mm)                                      | 8 - 14                 | 5 - 25               |
| Specific surface area (m <sup>2</sup> g <sup>-1</sup> ) | 50.1                   | 40.9                 |
| pH  | ≈ 8                    | ≈ 8                  |

### 3.2. Experimental set-up

The laboratory-scale system used for the biofiltration of air loaded with H<sub>2</sub>S is shown in Figure 2. It consisted of a PVC column with an internal diameter of 300 mm. The biofilter was filled with 70 L of expanded schist (1 m height) topped with a layer of UP20 (2 cm thickness). The bed of packing material was inoculated with 5 L of a diluted solution of activated sludge (about 50 mg of dry sewage sludge per liter) from a domestic wastewater treatment plant (Tougas, Nantes, France). The initial biomass came from this activated sludge and was not previously acclimatized to H<sub>2</sub>S treatment.

As observed in Figure 2, a clean airflow was first humidified through a column (internal diameter 200 mm) packed with Hiflow rings (packing height = 1.50 m). Then, a stream of H<sub>2</sub>S (99.7% purity) controlled by a mass flow controller (Model 5850S, Brooks Instrument, Hatfield, USA) was diluted in the atmospheric air at the outlet of the humidification column. The polluted air was then introduced at the bottom of the biofilter (upward load flow mode). To maintain optimal bed humidity, a watering system was implemented at the top of the biofilter. Tap water was sprayed once a day at a constant flow of 0.8 L min<sup>-1</sup> for 15 minutes, which corresponds to a water flow rate of 12 L day<sup>-1</sup>. To measure H<sub>2</sub>S concentrations along the column, the biofilter was equipped with sampling ports located at the inlet and outlet of the biofilter, and at 10, 30, 50, 70, 90 and 100 cm from the biofilter bottom (Figure 2). The H<sub>2</sub>S concentration was measured with an Onyx 5220 device (measurement accuracy ± 1%) from the Cosma Environment SA Company (Passy, France).



**Fig. 2.** Schematic diagram of the experimental pilot-scale biofilter.

### 3.3. Operating conditions

At the beginning of the experiment, the biofilter was fed continuously with 40 ppmv of H<sub>2</sub>S for three weeks. The polluted air flow rate was 4 m<sup>3</sup> h<sup>-1</sup> corresponding to an EBRT of 63 s (LR = 3.2 g m<sup>-3</sup> h<sup>-1</sup>). This period represented the acclimatization of the microbial communities to the biofilter environment. Nutrients were supplied by the layer of UP20 placed on top of the expanded schist. At the end of the acclimatization period, the H<sub>2</sub>S concentration was increased from 40 ppmv to 100 ppmv. The impact of EBRT on the degradation of H<sub>2</sub>S was then studied by varying the polluted air flow from 4 to 20 m<sup>3</sup> h<sup>-1</sup> corresponding to an EBRT range of 63 to 13 s (Table 3). The concentration of H<sub>2</sub>S was maintained at a constant value (100 ppmv) throughout the duration of the study.

**Table 3.** Operating conditions ( $\text{H}_2\text{S}$  concentration was maintained at a constant value of 100 ppmv).

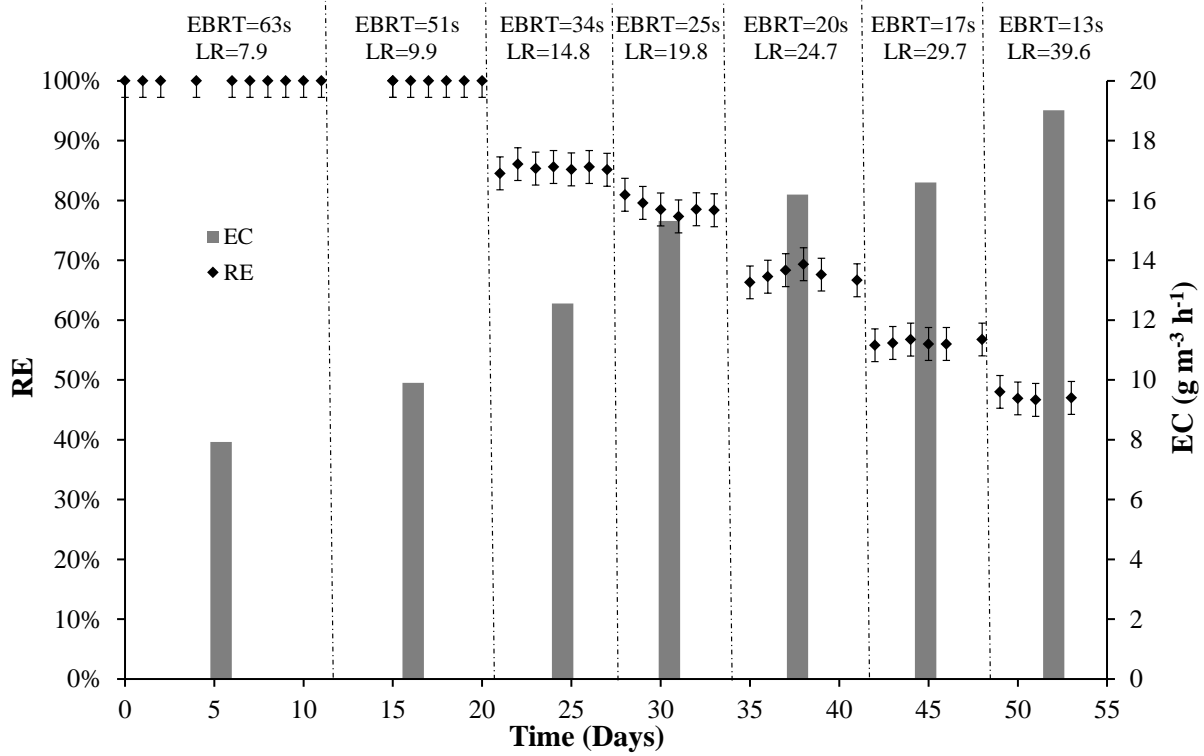
| Phase | Duration<br>(days) | $Q_v$<br>( $\text{m}^3 \text{h}^{-1}$ ) | EBRT<br>(s) | LR<br>( $\text{g m}^{-3} \text{h}^{-1}$ ) |
|-------|--------------------|---|-------------|---|
| 1     | 11                 | 4                                       | 63          | 7.9                                       |
| 2     | 10                 | 5                                       | 51          | 9.9                                       |
| 3     | 6                  | 7.5                                     | 34          | 14.8                                      |
| 4     | 7                  | 10                                      | 25          | 19.8                                      |
| 5     | 8                  | 12.5                                    | 20          | 24.7                                      |
| 6     | 6                  | 15                                      | 17          | 29.7                                      |
| 7     | 5                  | 20                                      | 13          | 39.6                                      |

## 4. Results and discussion

### 4.1. Effect of Empty Bed Residence Time (EBRT) on biofilter performances

Empty bed residence time (EBRT) is one of the most important operating factors that affect the performances of the biofilter. As shown in Figure 3,  $\text{H}_2\text{S}$  was totally removed with an EBRT greater than 51 s. By decreasing the EBRT, the removal efficiency decreased; the lowest RE value (47%) was obtained for an EBRT of 13 s. This result, widely reported in the literature devoted to biofiltration [8,15,45], is due to the increase in the gas superficial velocity in the bed material leading to a mass transfer limitation between the gas phase and the liquid phase. Using expanded schist coupled with UP20 to remove  $\text{H}_2\text{S}$  from waste air, Romero Hernandez *et al.* [15] showed that by increasing the EBRT from 16 to 35 s, the performances improved by about 35% in terms of elimination capacity. For a constant  $\text{H}_2\text{S}$  concentration of 100 ppmv, the elimination capacity is also defined as a function of the loading rate in Figure 3. At EBRT = 63 s, the elimination capacity (EC) was  $7.9 \text{ g m}^{-3} \text{ h}^{-1}$  (equivalent to RE = 100%). The biofilter achieved an RE higher than 80% up to LR =  $14.8 \text{ g m}^{-3} \text{ h}^{-1}$  (EBRT = 34 s), whereas at EBRT = 13 s, the EC value was  $19 \text{ g m}^{-3} \text{ h}^{-1}$  at an LR of  $39.6 \text{ g m}^{-3} \text{ h}^{-1}$  but the RE was only close to 45%.

These elimination capacity values are of the same order of magnitude as results reported in recent publications (Table 1). However, it should be noted that these results cannot be directly compared because the inlet concentrations and EBRTs are different. Moreover, no relationship between the properties of the packing materials and biofilter performances can be proposed. As can be observed in Table 1,  $\text{H}_2\text{S}$  elimination capacities can usually reach  $60 - 70 \text{ g m}^{-3} \text{ h}^{-1}$ , but higher EC values can be obtained for materials based on activated carbon. However, such material is too expensive to be retained for full-scale applications [46].



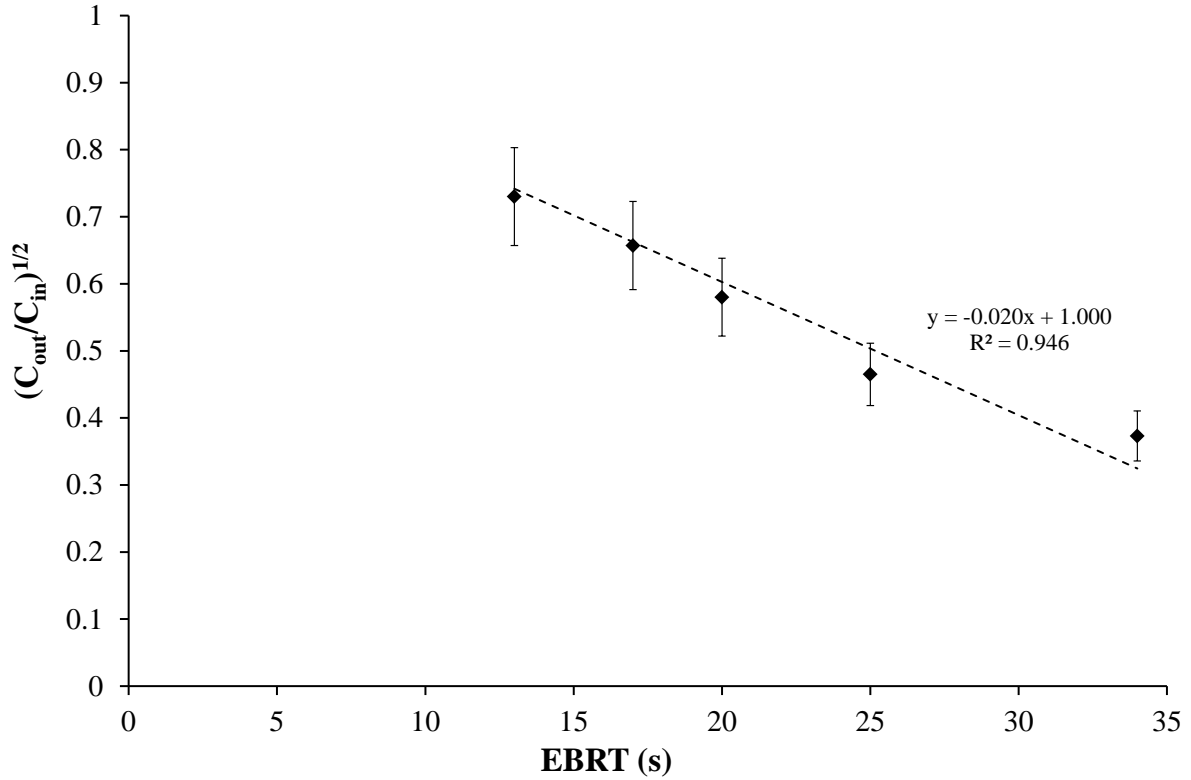
**Fig. 3.** EBRT influence on biofilter performance (inlet  $\text{H}_2\text{S}$  concentration = 100 ppmv). LR unit:  $\text{g m}^{-3} \text{h}^{-1}$ .

#### 4.2. Determination of the $\alpha_{\text{lump}}$ parameter

In order to compare the performance of expanded schist coupled with UP20 with that of other materials reported in the literature, the units used to determine  $\alpha_{\text{lump}}$  have to be selected carefully. Two cases are now considered.

Firstly, the experimental results can be directly compared with the values reported by Shareefdeen for two patented media [36,42]. From Eq. (10), the parameter  $\alpha_{\text{lump}}$  was deduced from the curve  $(C_{\text{out}}/C_{\text{in}})^{1/2}$  versus EBRT (Figure 4) in which the pollutant gas concentration and the EBRT were expressed in (ppmv) and (s), respectively. The value of  $\alpha_{\text{lump}}$  calculated from the slope of the curve was  $0.20 \pm 0.02 \text{ s}^{-1}$  according to the units used. This value is of the order of magnitude as the value reported for the patented medium (Biosorbents<sup>TM</sup>) used by Shareefdeen [17] ( $\alpha_{\text{lump}} = 0.18 \text{ s}^{-1}$ ). This medium, whose properties given in Table 2 are close to those of expanded schist, was demonstrated as efficient for removing  $\text{H}_2\text{S}$  concentrations up to 40 ppmv ( $\text{LR} = 6.7 \text{ g m}^{-3} \text{h}^{-1}$ ) at an EBRT of 30 s [16]. Using another synthetic medium (consisting of a hydrophilic nucleus, hydrophobic coatings, metallic powder, micro-organisms, nutrients and acid; US Patent Pub. No. 2205/0084949 A1), Shareefdeen reported  $\alpha_{\text{lump}} = 0.204 \text{ s}^{-1}$ . This medium gave complete  $\text{H}_2\text{S}$  removal ( $\text{RE} > 99\%$ ) at an EBRT of 20 s for  $\text{H}_2\text{S}$  concentrations of 50 ppmv [36]. By comparing  $\alpha_{\text{lump}}$  values, these results highlight that a natural material, such as expanded schist coupled with a small amount of synthetic nutritional material UP20, is equivalent to synthetic media specifically developed for  $\text{H}_2\text{S}$  biofiltration. Moreover, due to its ability to treat high  $\text{H}_2\text{S}$  loading rates and its good mechanical behavior over a long period (no attrition, no bed compaction and low pressure

drops), expanded schist should be preferred for industrial applications. According to the definition given in Eq. (9), the parameter  $\alpha_{\text{lump}}$  depends on: (i) A: the specific surface area of biofilm covering the packing material; (ii) k: the reaction rate constant; (iii) D: the diffusion coefficient of H<sub>2</sub>S in the biofilm; and (iv) m: the partition coefficient of H<sub>2</sub>S between the gas and liquid phases. Since the studies of Shareefdeen [17,36] and the present work are based on the biofiltration of H<sub>2</sub>S in air, it can legitimately be assumed that they involve the same values of the diffusion coefficient and the partition coefficient. In other words, the  $\alpha_{\text{lump}}$  values differ only on account of the values of A and k ( $\alpha_{\text{lump}} \sim A k^{1/2}$ ). As the characteristics of the expanded schist used in this study are roughly similar to those of the patented medium (Biosorbens™) used in [17] in terms of specific surface area (Table 2), it can be concluded that the biofilms developed on both materials should present similarities in terms of microbial ecosystems and biodegradation kinetics. The results of the present study and those of Shareefdeen can be compared with the data reported in Oyarzun *et al.* [3]. Using a biofiltration system filled with peat as a solid support inoculated with *Thiobacillus thioparus*, these authors studied the performance of a biofilter treating H<sub>2</sub>S at an inlet concentration of 257 ppmv for three different EBRTs (26, 52 and 120 s). From their data, it is possible to calculate that the value of the  $\alpha_{\text{lump}}$  parameter for peat was 0.12 s<sup>-1</sup>. This is 40% lower than the value obtained for the expanded schist coupled with UP20. As  $\alpha_{\text{lump}} \sim A k^{1/2}$ , this result may seem surprising at first sight. However, it should be noted that the intrinsic value of the specific surface area must be considered cautiously because the whole surface developed by the solid is not necessarily available for the development of the biofilm. Thus, whereas the specific surface area of peat is 705 m<sup>2</sup> g<sup>-1</sup> [21], i.e. 14 times higher than that of schist (Table 2), the value of the  $\alpha_{\text{lump}}$  parameter is lower for peat than for schist, which cannot be solely due to different values of k. Although the physical properties of the packing material are important, this result highlights that the overall properties of the packed bed have to be taken into account. The calculation of the specific surface area of biofilm covering the packing material is given below.



**Fig. 4.** Predicting the biofiltration process according to the Ottengraf model (Eq. 10).

Secondly, the parameter  $\alpha_{lump}$  can be calculated from Eq. (11) by drawing the graph  $(1-EC/LR)^{1/2}$  versus  $(EBRT/LR)^{1/2}$ . The line is similar to that reported in Figure 4, but the value of  $\alpha_{lump}$  corresponding to the slope of the line was  $26.4 \pm 2.6 \text{ g}^{1/2} \text{ m}^{-3/2} \text{ h}^{-1}$  according to the units used in this case (EC and LR in  $(\text{g m}^{-3} \text{ packing material h}^{-1})$  and EBRT in (h)). This method is useful because the  $\alpha_{lump}$  value determined in this case enables the performances of the biofilter as a whole to be characterized whatever its composition (mixture or layers of different packing materials) and whatever the EBRT. Consequently, the value of  $\alpha_{lump}$  can be compared with data reported in the literature (Table 4). It appears that the result obtained for the biofilter filled with expanded schist topped with a thin layer of UP20 is significantly higher than the results obtained for other packing materials, even those recorded with peat, considered the reference for  $\text{H}_2\text{S}$  biofiltration.

**Table 4.** Values of the  $\alpha_{lump}$  parameter reported in [21]

| Packing material                                  | $\alpha_{lump} (\text{g}^{1/2} \text{ m}^{-3/2} \text{ h}^{-1})$ |
|---|--|
| Sapwood   | 7.9  |
| Sapwood-UP20 (2 layers)                           | 8.4  |
| Pine bark   | 11.4   |
| Pozzolan-UP20                                     | 11.9   |
| Peat  | 15.3   |
| Peat-UP20 (2 layers)                              | 15.9   |
| Peat-UP20 (mixed)                                 | 21.3   |
| Expanded schist-UP20 (2 layers) <i>This study</i> | $26.4 \pm 2.6$   |

The physical properties of the expanded schist, i.e. suitable particle size, void fraction and surface available for biomass attachment, largely explain this result. The specific surface area of biofilm covering the packing material can be estimated from Eq. (9). From the literature data, the diffusion coefficient of H<sub>2</sub>S in the biofilm is  $D = 5.796 \cdot 10^{-6} \text{ m}^2 \text{ h}^{-1}$  and the partition coefficient of H<sub>2</sub>S between the gas and the liquid phase is  $m = 0.387$  [47]. Based on the experimental EC values given in Figure 3 (from 10 to 20  $\text{g m}^{-3} \text{ packing material h}^{-1}$ ), the reaction rate per unit of volume of the biofilm  $k$  can be calculated using the relationship given in Ottengraf and Van Den Oever [24]:

$$k = \frac{EC}{(1-\varepsilon) \left[ 1 - \left( 1 - \frac{2\delta}{d} \right)^3 \right]} \quad (12)$$

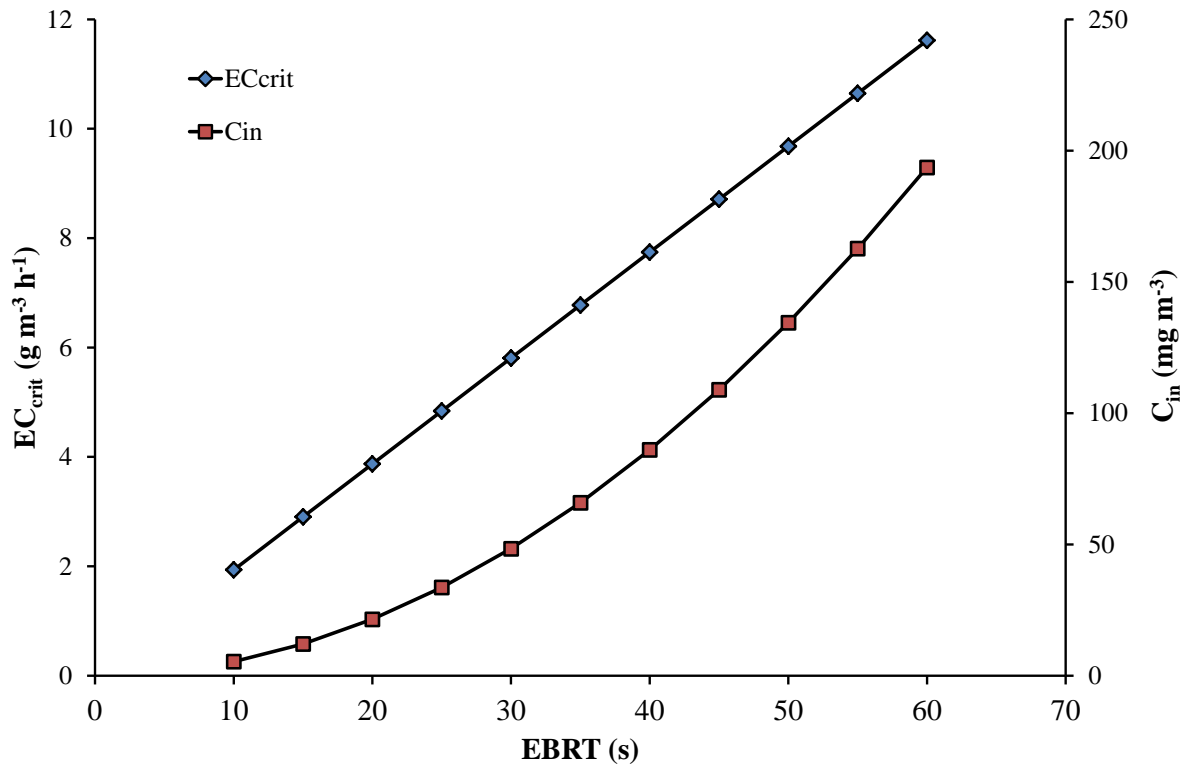
The porosity  $\varepsilon$  of the packed bed composed of pieces of expanded schist with an average particle diameter of 10 mm was determined to be 0.41 during the biofiltration of H<sub>2</sub>S [14]. Assuming a biofilm thickness of around 100  $\mu\text{m}$ , which seems very reasonable, the  $k$  values range from 300 to 580  $\text{g m}^{-3} \text{ biofilm h}^{-1}$  and hence the values of the specific surface area of the biofilm are from 600 to 400  $\text{m}^2 \text{ biofilm m}^{-3} \text{ packing material}$ , respectively. These results are consistent with the estimated specific surface area of round particles, 10 mm in diameter [48]. However, although such values are of the order of magnitude of the expected results, the Ottengraf model is not adapted to obtain accurate values of biofilm specific surface area. Other models based on pressure drop measurements could be used to refine the results [14,48,49]. Nonetheless, it can be concluded that the Ottengraf model, through the determination of  $\alpha_{\text{lump}}$ , is a powerful tool to describe simply the operation of a highly complex system such as a biofilter. Moreover, it can be used to design such bioreactors. For instance, from the  $\alpha_{\text{lump}}$  value, the critical value of the elimination capacity ( $EC_{\text{crit}}$ ) can be determined using Eq. (13) deduced from Eq. (11). This parameter corresponds to the value at which the loading rates exceed the elimination capacities of the biofilter, generating removal efficiencies less than 100%.

$$EC_{\text{crit}} = \alpha_{\text{lump}}^2 EBRT \quad (13)$$

The determination of the inlet concentration corresponding to  $EC_{\text{crit}}$  is obtained using Eq. (14) deduced from Eq. (10):

$$C_{\text{in}} = \alpha_{\text{lump}}^2 EBRT^2 \quad (14)$$

Figure 5 gives an overview of the ability of a biofilter filled with expanded schist completed with a thin layer of UP20 to treat efficiently ( $RE = 100\%$ ) a waste gas loaded with H<sub>2</sub>S. As can be observed, for an EBRT higher than 30 s, the biofilter can treat inlet gas concentrations higher than 50  $\text{mg m}^{-3}$ , which are higher than those encountered in many industrial applications. Consequently, this result usefully completes the data previously reported about biofiltration using expanded schist as the packing material [15,18,20].



**Fig. 5.** Determination of the critical value of the elimination capacity ( $EC_{crit}$ ) according to EBRT for a biofilter filled with expanded schist completed with a thin layer of UP20. Determination of the corresponding inlet gas concentration ( $H_2S$ ).

## 5. Conclusion

The biological removal of  $H_2S$  in waste gas at a constant concentration (100 ppmv) was carried out using a biofilter packed with expanded schist and topped with a layer of a synthetic nutritional material (UP20). The effect of EBRT on the removal efficiencies of hydrogen sulfide was underlined. At  $EBRT > 51$  s,  $H_2S$  was totally removed, whereas when the EBRT decreased, the removal efficiency decreased. From the EBRT results, the Ottengraf model equations were used to evaluate the performance of a biofilter. The parameter  $\alpha_{lump}$  deduced from the Ottengraf model was used to compare the performance of expanded schist coupled with UP20 with that of other materials reported in the literature. In this study, the value of  $\alpha_{lump}$  for expanded schist coupled with UP20 was found to be  $26.4 \text{ g}^{1/2} \text{ m}^{-3/2} \text{ h}^{-1}$ . This value, which enables the performance of the biofilter as a whole to be characterized whatever its composition (mixture or layers of different packing materials) and whatever the EBRT, is higher than those obtained for other packing materials (natural or synthetic) reported in the literature. Finally, the  $\alpha_{lump}$  parameter, determined from Eq. (11) using EC and LR in ( $\text{g m}^{-3} \text{ packing material h}^{-1}$ ) and EBRT in (h), appears to be a powerful tool to compare unambiguously the packing materials used in biofiltration.

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