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# Biofiltration of methane in presence of ethylbenzene or xylene

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## ABSTRACT

Landfill gases (LFGs) are gaseous emissions containing several harmful compounds such as volatile organic compounds (VOCs), volatile inorganic compounds (VICs) along with methane (CH<sub>4</sub>) which is a powerful greenhouse gas. In the case of small or older landfills, disposal of LFG can be economically challenging, and a possible solution is biofiltration. Biofiltration of CH<sub>4</sub> in the presence of two VOCs was studied, ethylbenzene (EB) for biofilter 1 (BF1) and xylene (X) for biofilter 2 (BF2). Both biofilters were packed with inorganic packing materials and operated with an empty bed residence time (EBRT) of 4.5 min. Methane concentrations varied from 2000 to 10,000 ppmv along with 2 concentrations for both VOCs i.e., 200 and 500 ppmv. In the case of individual VOC's removal, the acclimation period of the microorganisms to EB and X were close to 30 days, indicating a similar adaptation period for X and EB. The addition of CH<sub>4</sub> under low inlet concentration of 2000 ppmv had minor effect on VOCs biodegradation such that the average VOC removal efficiencies (REs) remained above 85% for VOC inlet concentrations of 200 ppmv while the average CH<sub>4</sub>-RE were around 56% in both biofilters. When CH<sub>4</sub> inlet concentration increased up to 10,000 ppmv, inhibition became an issue for all VOC concentrations and dropped VOC and CH<sub>4</sub>-REs down to 80% of their original values. The type and concentration of the VOC played a key role in the intensity of the inhibition. In general, mass transfer was controlling the elimination of the three substrates.

### Keywords:

Biofiltration

Biotechnology

Inhibition

VOC

Greenhouse gas

Synergistic effect

Inorganic packing

## 1. Introduction

Methane (CH<sub>4</sub>) with a global warming potential (GWP) approximately 21–25 times higher than carbon dioxide (CO<sub>2</sub>), is the most important greenhouse gas (GHG) after CO<sub>2</sub> (Padrón et al., 2020). Estimations in 2020 indicate that an annual global emission around 570 million metric tonnes of CH<sub>4</sub> has been emitted worldwide from which 60% are from anthropogenic sources such as landfills, the oil and gas industries, sewers and wastewater treatment plants, coal mines, biogas production and the agricultural and livestock industries (IEA, 2020; Khabiri et al., 2021; MacKay et al., 2019). Methane concentration in the atmosphere is increasing by 0.6% (10 ppbv) each year (Padrón et al., 2020). Atmospheric CH<sub>4</sub> concentration was estimated around 1875 ppbv in 2019 (Schiermeier, 2020). The current CH<sub>4</sub> emission estimation indicates the possibility of an increase in global temperature above 3 °C by the end of this century. A reduction in emissions is therefore required

in order to meet the temperature increase limit of 1.5–2 °C indicated in Paris Agreement (COP21) (Nisbet et al., 2019).

Landfills are important sources of CH<sub>4</sub> emission due to anaerobic degradation of organic matters producing a biogas called landfill gas (LFG) with CH<sub>4</sub> concentrations up to 60% v/v (Padrón et al., 2020; Pecorini et al., 2020). It is estimated that 68 million metric tons of CH<sub>4</sub> were produced globally from waste management systems including landfills in 2019 (IEA, 2020). Along with CH<sub>4</sub>, other compounds can be found in LFG such as volatile organic compounds (VOCs) and volatile inorganic compounds (VICs) (Nikiema et al., 2007). Concentration of total VOCs in LFG can vary in the range of 40–1000 mg.m<sup>-3</sup> (Gong et al., 2019). Among VOCs, aromatic compounds such as xylene (X) and ethylbenzene (EB) are commonly present in LFGs, which can cause cardiorespiratory diseases (Borhani and Noorpoor, 2017; Ran et al., 2018). For example, Sevimoğlu and Tansel (2013) reported concentrations varying in the range of 10–80 mg.m<sup>-3</sup> and 20–50 mg.m<sup>-3</sup> for X and

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EB, respectively, in LFG.

For CH<sub>4</sub> concentration in LFG exceeding 60% v/v (Nikiema et al., 2007), LFG can be valorized for electricity production or heating (Ciula et al., 2018; Themelis and Ulloa, 2007). When CH<sub>4</sub> concentration is in the range of 35–60% v/v, valorization may not be economically sustainable, and LFG physico-chemical elimination techniques are preferred. Several LFG elimination techniques exist such as thermal and catalytic oxidation, but the most commonly used mitigation technique is flaring (Ménard et al., 2012). In case of concentrations below 3% v/v (the case of small-scale landfills or landfills older than 30 years), biotechnologies present several advantages over abovementioned conventional techniques for LFG treatment in term of feasibility, environmental aspect and economical sustainability (Malakar et al., 2017; Pecorini et al., 2020).

The most used biotechnology for CH<sub>4</sub> and VOC mitigation is biofiltration. This bioprocess relies on microorganisms to act as biocatalysts and transforming pollutants to CO<sub>2</sub>, water (H<sub>2</sub>O), biomass, etc. (Delhoménie and Heitz, 2005). The microorganisms are supported in a humidified packed bed (organic and/or inorganic or synthetic) forming a biofilm and are periodically supplied with a nutrient solution (NS) containing essential nutrients (Khabiri et al., 2020a).

Methane and VOC biofiltration have been intensively studied over the last two decades. Fjølsted et al. (2020) eliminated 60% of CH<sub>4</sub> from LFG with an inlet load (IL) of 9–66 g.m<sup>-3</sup>.h<sup>-1</sup> and an empty bed residence time (EBRT) in the range of 24–167 min in a compost packed biofilter (BF) in a landfill in Denmark. Lebrero et al. (2016) obtained a removal efficiency (RE) close to 90% at a CH<sub>4</sub>-IL of 40 g.m<sup>-3</sup>.h<sup>-1</sup> for a BF packed with compost and inoculated with a pure strain of *Graphium* sp. Cho et al. (2008) obtained an EB-RE of 92% in a rock-compost BF for ILs varying between 10 and 40 g.m<sup>-3</sup>.h<sup>-1</sup> and an EBRT of 62 s. Similar X-REs were obtained by Natarajan et al. (2014) in date palm tree bark packed BF for X concentrations between 0.1 and 3 g.m<sup>-3</sup> and an EBRT around 2 min.

The biofiltration of several components in a mixture from different chemical types and structures can be challenging due to the increased complexity of genetic and enzymatic regulation mechanisms in the co-degradation. For example, CH<sub>4</sub> was found to be inhibited by VOCs such as EB or X (Lee et al., 2011; Dobslaw and Ortlinghaus, 2020). Liao et al. (2018) indicated that interaction effects increased as much as the complexity of gas mixture elevated along with the microbial community changes according to the components of the gaseous mixture. Albanna et al. (2010) found that VOCs such as trichloroethane, trichloroethylene and dichloromethane act as uncompetitive inhibitors on CH<sub>4</sub> degradation by bonding to the enzyme-substrate complex and deactivating the enzymes.

Even though several studies of VOC elimination by biofiltration were reported in the literature, only a few of them treated CH<sub>4</sub> and VOCs simultaneously. Khabiri et al. (2020b) used an inorganic BF to treat a mixture of CH<sub>4</sub> (IL of 6–70 g.m<sup>-3</sup>.h<sup>-1</sup>) and styrene (IL of 9–32 g.m<sup>-3</sup>.h<sup>-1</sup>) at an air flow rate of 0.018 m<sup>3</sup>.h<sup>-1</sup>. A complete styrene elimination was observed (RE = 100%) while CH<sub>4</sub>-RE was between 43 and 80%. Zdeb and Lebiocka (2016) used a compost/expanded clay BF to treat a model LFG composed of CH<sub>4</sub>, benzene, toluene, EB and X (BTEX). Due to a long EBRT (1680 min), all pollutants were removed with REs above 90% at a total VOCs-IL of 0.005 g.m<sup>-3</sup>.h<sup>-1</sup> and CH<sub>4</sub>-IL of 12 g.m<sup>-3</sup>.h<sup>-1</sup>. Although the authors in the abovementioned studies usually indicated a negative effect of VOCs presence on CH<sub>4</sub> elimination, further studies on simultaneous biofiltration of CH<sub>4</sub> and VOCs are required to understand the interactions between the pollutants. Since LFG contains CH<sub>4</sub> along with multiple VOCs and VICs, the studies on LFG biofiltration should consider the interactions between CH<sub>4</sub> and VOCs as well as the possible inhibition that can be caused by one component on another.

Although a single pollutant biodegradation is usually affected by its biodegradability and mass transfer/kinetic limitations, the simultaneous biodegradation of several pollutants presents additional challenges in form of interaction effects (Cheng et al., 2016). The interaction among

CH<sub>4</sub> and VOCs can be either antagonistic, synergistic or neutral (Yang et al., 2018). In antagonistic interactions, the biodegradation of CH<sub>4</sub> is altered by the presence of VOCs that has a toxic or inhibitory effect. For example, styrene (Khabiri et al., 2020b), toluene and chlorobenzene (Ménard et al., 2012) were found to have an antagonistic effect on CH<sub>4</sub> biodegradation. On the contrary, by considering a mixture of CH<sub>4</sub> and VOCs, the enhancement of CH<sub>4</sub> biodegradation can be explained by co-metabolism phenomena, growth-benefit effect or solubility improvement of the substrate by the presence of a hydrophilic compound (Lamprea Pineda et al., 2021). Methane biodegradation was found to be strengthened in presence of dimethyl sulphide (DMS) (Kim et al., 2013). In neutral interactions, no significant change in biodegradation occurs despite the presence of several substrates. For example, the presence of benzene and toluene had no effect on CH<sub>4</sub> elimination (Kim et al., 2013).

It is hypothesized that EB or X tend to have an inhibitory effect on CH<sub>4</sub> elimination due to their toxicity and possible competitive inhibition for methanotrophic bacteria (the main microorganisms responsible for CH<sub>4</sub> biodegradation) (Lee et al., 2011). Methane might also have a negative effect on VOC elimination.

The aim of this study was to investigate the simultaneous biofiltration of CH<sub>4</sub> with a VOC as a binary mixture e.g., CH<sub>4</sub>/X or CH<sub>4</sub>/EB, in two identical BFs with inorganic packing materials at different concentrations of CH<sub>4</sub> and VOCs. The use of an inorganic packing material would allow a BF to have stable long-term performance but presents challenges like the absence of nutrients and indigenous microorganisms; the addition of inoculum and NS is therefore required (La et al., 2018). To the authors best knowledge, the elimination of mixtures of CH<sub>4</sub>/X or CH<sub>4</sub>/EB in BFs has never been studied. The first specific objective was to study the biofiltration of two individual VOCs (EB or X) under two levels of concentrations. The second specific objective was to study the biofiltration of both VOCs with CH<sub>4</sub> addition at different ILs varying between 19 and 95 g.m<sup>-3</sup>.h<sup>-1</sup> and to investigate the interactions between EB or X in presence of CH<sub>4</sub>. The possible inhibition of the VOCs on CH<sub>4</sub> elimination as well as the effect of high CH<sub>4</sub> concentrations on VOCs elimination were studied.

## 2. Materials and methods

### 2.1. Biofilters experimental set-up

Fig. 1 shows the experiments' set-up. Two (2) upflow BFs (BF1 and BF2) made of plexiglass with a volume of 0.018 m<sup>3</sup> (1 m height and 0.15 m diameter) were used for a period of 281 days for BF1 and 278 days for BF2. The BFs were composed of three beds (0.3 m height). The bottom bed was packed with an inorganic material with the following properties: an average diameter of 7.5 mm, a void fraction of 0.55, a density of 750 kg.m<sup>-3</sup> and a specific surface area of 470 m<sup>2</sup>.m<sup>-3</sup>. The properties of the middle and top beds packed with inorganic materials were: an average diameter of 5.0 mm, a void fraction of 0.43, a density of 1200 kg.m<sup>-3</sup> and a specific surface area of 1250 m<sup>2</sup>.m<sup>-3</sup>. The material with the highest void fraction was chosen to be in the bottom bed as it is more likely to have biomass accumulation (Ferdowsi et al., 2019). The composition of the materials cannot be indicated for confidentiality reasons.

Methane (Praxair Inc., Canada) flow rate was controlled by a mass flow controller (MFC) (Brooks, USA) and was injected at the bottom of the bioreactor along with humidified air and one VOC (EB for BF1 and X for BF2). Air was humidified by passing through a bubbler humidifier providing a relative humidity of 90%. Dry air was injected through a VOC bubbler filled with either EB (>99%, Sigma-Aldrich, USA) for BF1 or X (>99%, Fisher chemicals, USA) for BF2. Both humidified and dry air flows were controlled by rotameters (Brooks, USA).

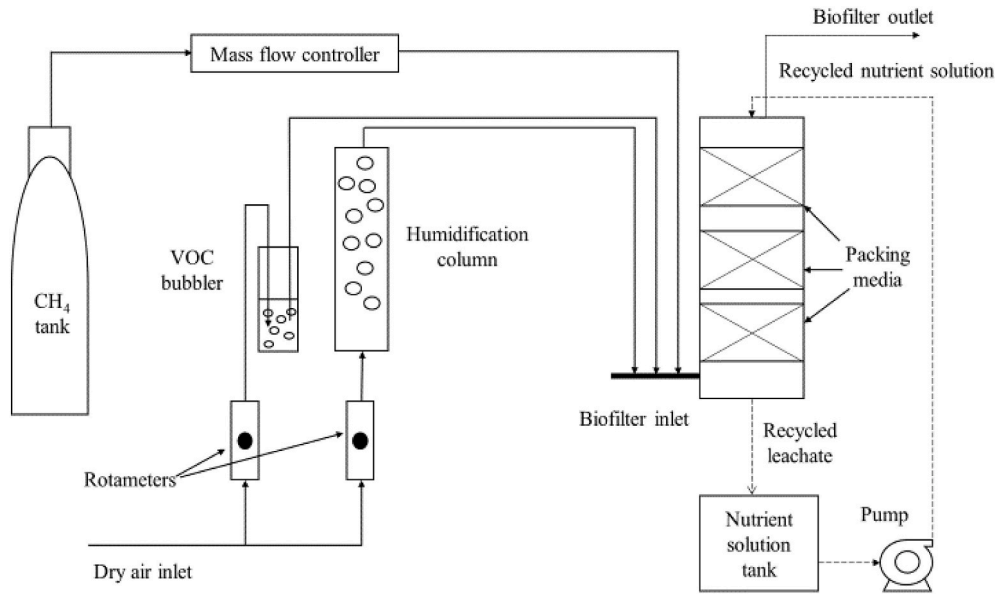


Fig. 1. Experimental set-up for biofiltration of CH<sub>4</sub> and VOCs.

## 2.2. Analytical tools

Gas samples were taken from the inlet and outlet of the BFs. A total hydrocarbon flame ionization analyzer equipped with a single continuous flame ionization detector (FIA 510, HORIBA, Japan) was used to analyze the concentrations of CH<sub>4</sub> and VOCs. A CO<sub>2</sub> gas analyzer (ULTRAMAT 22P, SIEMENS, Germany) was used to measure CO<sub>2</sub> concentrations in the BFs inlet and outlet and to monitor CO<sub>2</sub> production. Gas samples from the BFs were extracted by vacuum pumps and directly supplied to the hydrocarbon and CO<sub>2</sub> analyzers.

## 2.3. Performance parameters

The performance parameters used to evaluate the biofiltration process are the following:

$$\text{- Inlet load: IL} = C_{in} Q/V \text{ [g.m}^{-3}\text{.h}^{-1}\text{]} \quad (1)$$

$$\text{- Elimination capacity: EC} = (C_{in} - C_{out}) Q/V \text{ [g.m}^{-3}\text{.h}^{-1}\text{]} \quad (2)$$

$$\text{- Removal efficiency: RE} = 100 * (C_{in} - C_{out}) / C_{in} \text{ [%]} \quad (3)$$

$$\text{- CO}_2 \text{ production rate: PCO}_2 = (C(\text{CO}_2)_{out} - C(\text{CO}_2)_{in}) Q/V \text{ [g.m}^{-3}\text{.h}^{-1}\text{]} \quad (4)$$

Where Q is the gas flow rate (m<sup>3</sup>.h<sup>-1</sup>), V is the packed bed volume (m<sup>3</sup>) and C<sub>in</sub> and C<sub>out</sub> are the BF's inlet and outlet pollutant concentrations, respectively (g.m<sup>-3</sup>). C(CO<sub>2</sub>)<sub>in</sub> and C(CO<sub>2</sub>)<sub>out</sub> are the CO<sub>2</sub> concentrations in the BFs inlet and outlet, respectively (g.m<sup>-3</sup>).

## 2.4. Experimental methods and operating conditions

Five (5) liters of leachate from an active BF treating CH<sub>4</sub> (IL = 60 g.m<sup>-3</sup>.h<sup>-1</sup>, RE = 76%) were used to inoculate the packing materials that had been thoroughly washed with tap water prior to inoculation. The leachate was recirculated 3 times on the same day from the top to the bottom of the BF in order to ensure homogeneous distribution of microorganisms through the filter bed (Ferdowsi et al., 2016). The addition of NS was started a week later. The components and composition of the NS were the same as the one used by Khabiri et al. (2020a). The NS was then recycled in a 20\*10<sup>-3</sup> m<sup>3</sup> tank and fed to the BF from the top at a rate of 60\*10<sup>-3</sup> m<sup>3</sup>.h<sup>-1</sup> for 2 min every 2 days. The experiments were designed based on 8 consecutive phases and the NS tank was refreshed at the end of each phase. Gas flow rate was kept constant during the whole

experiments at 0.24 m<sup>3</sup>.h<sup>-1</sup> corresponding to an empty bed residence time (EBRT) of 4.5 min for both BFs.

At the beginning of the experiment, VOCs (EB for BF1 and X for BF2) were injected to the BFs along with humidified air at an IL of 13 g.m<sup>-3</sup>.h<sup>-1</sup> (Phase 1) corresponding to inlet concentration of 200 ppmv. After reaching a pseudo-steady state (constant BF performance), VOC-IL was increased to 32 g.m<sup>-3</sup>.h<sup>-1</sup> corresponding to inlet concentration of 500 ppmv (Phase 2). Once a pseudo-steady state was reached, CH<sub>4</sub> was added to the inlet gas at ILs of 19 (Phase 3), 57 (Phase 4), 95 (Phase 5) g.m<sup>-3</sup>.h<sup>-1</sup> corresponding to concentrations of 2000, 6000, 10,000 ppmv, respectively. The transition from one CH<sub>4</sub>-IL to another was only carried out after a pseudo-steady performance was obtained. The IL of VOCs was then returned to 13 g.m<sup>-3</sup>.h<sup>-1</sup> and CH<sub>4</sub>-ILs were changed again to 19 (Phase 6), 57 (Phase 7), 95 (Phase 8) g.m<sup>-3</sup>.h<sup>-1</sup>. Carbon dioxide concentrations were measured every 3 days. Table 1 presents the different phases of the experiments.

## 3. Results and discussion

### 3.1. Methane and ethylbenzene removal efficiencies for different inlet loads (BF1)

Fig. 2 presents CH<sub>4</sub>-REs and EB-REs for BF1 in different phases of experiments. In Phases 1 and 2, only EB was injected to BF1. Ethylbenzene-REs of 83±5 and 89±3% were obtained for ILs of 12.0±0.2 g.m<sup>-3</sup>.h<sup>-1</sup> (EB concentration of 200 ppmv) and 32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup> (EB concentration of 500 ppmv), respectively. Ethylbenzene-RE increase can be attributed to the development and growth of microorganisms

Table 1  
Operating conditions during the experiments.

Phase	VOC concentration (ppmv)	CH <sub>4</sub> concentration (ppmv)	Duration for BF1 (days)	Duration for BF2 (days)
1	200	0	37	37
2	500	0	45	39
3	500	2000	45	42
4	500	6000	15	27
5	500	10,000	54	57
6	200	2000	34	34
7	200	6000	27	18
8	200	10,000	24	15

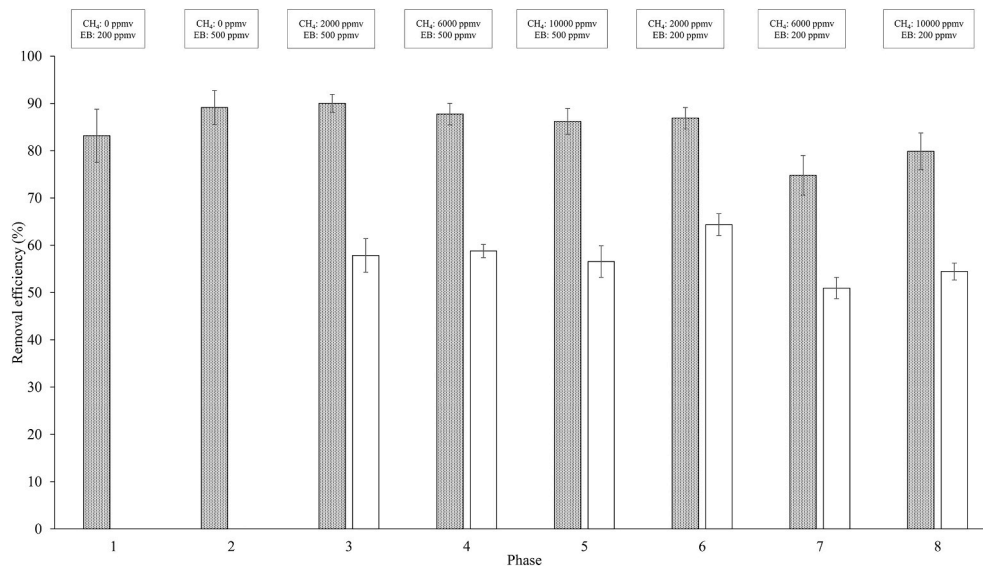


Fig. 2. Pollutants removal efficiencies for each phase in BF1 (▨for EB-RE and □for CH<sub>4</sub>-RE).

responsible for EB removal and/or the increase of EB bioavailability in the biofilm phase. Gallastegui et al. (2017) obtained a similar EB-RE of 90% during the acclimation period for an IL varying in the range of 4.5–16.1 g.m<sup>-3</sup>.h<sup>-1</sup> in a black slag packed BF at an EBRT of 3 min.

In Phase 3, CH<sub>4</sub> was added to BF1 at an IL of 19.0± 0.7 g.m<sup>-3</sup>.h<sup>-1</sup> (CH<sub>4</sub> concentration of 2000 ppmv). According to Fig. 2, CH<sub>4</sub>-RE reached 58±4% while EB-RE remained almost constant at 90± 2% (EB-IL was kept constant at 32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup>). Regarding CH<sub>4</sub> elimination as a sole pollutant, Hernández et al. (2015) obtained a higher CH<sub>4</sub>-RE of 62% for an IL of 18 g.m<sup>-3</sup>.h<sup>-1</sup> and an EBRT of 4.4 min in a tree bark BF. Gómez-Borraz et al. (2017) reported a CH<sub>4</sub>-RE of 62% for an EBRT of 19 min and a CH<sub>4</sub>-IL of 21 g.m<sup>-3</sup>.h<sup>-1</sup> in a compost BF. About the removal of CH<sub>4</sub> and a VOC in a mixture, Khabiri et al. (2020b) obtained a CH<sub>4</sub>-RE of 64% for CH<sub>4</sub>-IL of 20 g.m<sup>-3</sup>.h<sup>-1</sup> in the presence of styrene (styrene-IL = 32 g.m<sup>-3</sup>.h<sup>-1</sup>) in a BF packed with inorganic materials with an EBRT of 6 min. Methane-RE (58±4%) in Phase 3 was 8% lower than REs obtained for CH<sub>4</sub>-ILs close to 19 g.m<sup>-3</sup>.h<sup>-1</sup> in the abovementioned studies due to a possible inhibition for CH<sub>4</sub> biodegradation by EB presence or the shorter EBRT of 4.5 min. Lee et al. (2011) reported a slight inhibitory effect of EB on methanotrophic bacteria in a batch bioreactor as CH<sub>4</sub> oxidation rate dropped from 100 to 80% when EB was added to an enriched CH<sub>4</sub>-oxidizing microbial culture from landfill cover soil. Methane was injected at a concentration of 5000 ppmv along with 100–200 ppmv of EB. Incubation was carried out at 30 °C and at a shaking rate of 180 rpm.

In Phase 4, CH<sub>4</sub>-RE and EB-RE were 59±1 and 88± 2%, respectively when CH<sub>4</sub>-IL was increased to 54.0±2.0 g.m<sup>-3</sup>.h<sup>-1</sup> (CH<sub>4</sub> concentration of 6000 ppmv) while EB-IL remained unchanged at 32.0± 0.4 g.m<sup>-3</sup>.h<sup>-1</sup>. Methane-RE and EB-RE did not significantly differ from Phase 3 possibly due to the microorganisms' adaptation to higher CH<sub>4</sub>-ILs.

In Phase 5, BF1 tolerated the high CH<sub>4</sub> and EB concentrations (10,000 ppmv for CH<sub>4</sub> and 500 ppmv for EB) while being fed simultaneously. According to Fig. 2, CH<sub>4</sub>-RE and EB-RE stayed over 57% and 86% respectively for corresponding CH<sub>4</sub>-IL and EB-IL of 96± 2.0 and 32±0.4 g.m<sup>-3</sup>.h<sup>-1</sup>, respectively. Methane-RE (57±3%) was higher compared to REs reported in some previous studies despite the short EBRT of 4.5 min and the presence of EB in this study. Kim et al. (2014) obtained CH<sub>4</sub>-RE of 51% for an IL of 99.5 g.m<sup>-3</sup>.h<sup>-1</sup> with an EBRT of 20 min (more than 4 times higher than the EBRT used in the present study) in a perlite packed BF. Nikiema and Heitz (2009) reported that for the same CH<sub>4</sub> concentration (10,000 ppmv), CH<sub>4</sub>-RE was around 50% for an EBRT around 4.5 min.

For the following Phases 6–8, the EB inlet concentration returned to the lowest level of 200 ppmv while CH<sub>4</sub> inlet concentration varied from 2000 to 10,000 ppmv. In other words, groups of Phases (3 and 6), (4 and 7), and (5 and 8) were identical in terms of CH<sub>4</sub> inlet concentration, but different in terms of EB inlet concentration.

In Phase 6, CH<sub>4</sub>-IL and EB-IL decreased to 20.0±0.5 and 12.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>, respectively, which corresponds to the lowest values of CH<sub>4</sub> and EB inlet concentrations tested in this study. A comparison between Phases 3 and 6 shows higher CH<sub>4</sub>-RE of 64±2% in Phase 6 compared to 58±4% in Phase 3 for a similar CH<sub>4</sub>-IL and a lower EB-IL (32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup>) in Phase 3 compared to Phase 6 (12.0± 0.5 g.m<sup>-3</sup>.h<sup>-1</sup>). It is worth mentioning that CH<sub>4</sub> was introduced to BF1 in Phase 3 for the first time. Therefore, the CH<sub>4</sub> degrading community was probably more developed in Phase 6 compared to Phase 3. Also, the reduction of EB-IL in Phase 6 compared to Phase 3 possibly moderated the inhibitory effect of EB on CH<sub>4</sub> degradation (Albanna et al., 2010). Ethylbenzene-RE in Phase 6 (87±2% for an IL of 12.0± 0.5 g.m<sup>-3</sup>.h<sup>-1</sup>) was mildly lower than in Phase 3 (90±2% for an IL of 32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup>). This might be due to high CH<sub>4</sub>-IL during Phase 5 (96.0±2.0 g.m<sup>-3</sup>.h<sup>-1</sup>) which could have led CH<sub>4</sub> degraders to be dominant as a microbial culture at the end of Phase 5 and beginning of Phase 6. In contrast, due to the presence of EB as a single substrate, the EB degraders were likely dominant at the end of Phase 2 just before the beginning of Phase 3.

In Phase 7, CH<sub>4</sub>-IL was increased to 57.0±0.6 g.m<sup>-3</sup>.h<sup>-1</sup> while EB-IL was kept at 12.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>; the lowest REs of this study were obtained (51±2% for CH<sub>4</sub> and 75±4% for EB). Methane-RE and EB-RE both dropped since CH<sub>4</sub>-IL was 3 times higher than in Phase 6 which might cause inhibition for CH<sub>4</sub> and EB degradation. For a similar CH<sub>4</sub>-IL (54±2 g.m<sup>-3</sup>.h<sup>-1</sup>) and a higher EB-IL (32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup>), higher CH<sub>4</sub>-RE and EB-RE had been obtained (59±1 and 88±2%, respectively) in Phase 4 compared to Phase 7. The EB-RE drop in Phase 7 compared to Phase 4 could be attributed to the dominance of CH<sub>4</sub> degraders over EB degraders due to significantly higher CH<sub>4</sub>-IL compared to EB-IL. The synergistic effect of high level of EB concentration (500 ppmv) on CH<sub>4</sub> and EB biodegradations in Phase 4 might be the reason for higher CH<sub>4</sub> and EB-REs in Phase 4.

In Phase 8, a slight improvement in the performance of BF1 was observed for an increased CH<sub>4</sub>-IL of 99.0±4.2 g.m<sup>-3</sup>.h<sup>-1</sup> and at a constant EB-IL of 12.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>. Methane-RE and EB-RE were obtained as 54±2 and 80±4%, respectively. Methane and EB-REs in Phase 8 were lower than REs in Phase 5 (similar CH<sub>4</sub>-IL to Phase 5 but lower EB-IL of 12.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>). A decrease from 57±3 to 54±2% for CH<sub>4</sub>-RE and from 86±3 to 80±4% for EB-RE were observed in Phase 8

compared to Phase 5.

These results indicated that for high level of CH<sub>4</sub>-ILs in the range of 54–99 g.m<sup>-3</sup>.h<sup>-1</sup>, the presence of high EB-IL of 32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup> (Phases 4 and 5) led to higher CH<sub>4</sub>-REs compared to the presence of low EB-IL of 12.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup> (Phases 7 and 8). In other words, high level of EB concentration (500 ppmv) under high CH<sub>4</sub> inlet concentration (6000–10,000 ppmv) could support synergistic interactions. In this regard, high EB concentrations (500 ppmv) might have favored the development of bacterial communities who contributed to CH<sub>4</sub> biodegradation in addition to the VOC removal.

### 3.2. Simultaneous CH<sub>4</sub> and xylene biodegradation (BF2 in comparison with BF1)

Methane-RE and X-RE for BF2 in 8 Phases are presented in Fig. 3. In Phases 1 and 2 (Fig. 3), X-RE increased from 76±5 to 90±6% when X-ILs increased from 12.0±0.2 g.m<sup>-3</sup>.h<sup>-1</sup> (X inlet concentration of 200 ppmv) to 32.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup> (X inlet concentration of 500 ppmv). Li et al. (2020) obtained a similar X-RE of 90% for ILs of 20–30 g.m<sup>-3</sup>.h<sup>-1</sup> and an EBRT of 1.5 min compared to 4.5 min used in the current study. Gallastegui et al. (2017) also reported a X-RE of 86% for an IL of 16.5 g.m<sup>-3</sup>.h<sup>-1</sup> and an EBRT of 3 min in a BF packed with black slag.

According to Figs. 2 and 3, both BFs displayed an increasing VOC-RE trend over Phases 1 and 2. The percentage of VOC-RE improvement from Phase 1 to Phase 2 were 7 and 18% for BF1 (EB) and BF2 (X), respectively.

In Phase 3, CH<sub>4</sub> was added at an IL of 20±1 g.m<sup>-3</sup>.h<sup>-1</sup> under X-IL of 32.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>. A CH<sub>4</sub>-RE of 55±9% was obtained which was similar to CH<sub>4</sub>-RE in Phase 3 in BF1. The addition of CH<sub>4</sub> to BF2 in Phase 3 had no underline effect on the VOC removal and X-RE remained above 87%.

During Phase 4 in BF2, CH<sub>4</sub>-IL was increased by almost 3 times (54 ± 2 g.m<sup>-3</sup>.h<sup>-1</sup>) compared to Phase 3 under constant X-IL of 32.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>. Both CH<sub>4</sub>-RE and X-RE slightly increased to 57±2 and 90±2%, respectively. Methane-IL increase caused no inhibition since appropriate microorganisms for both X and CH<sub>4</sub> removal were probably developed and adapted to remove larger amounts of pollutants.

In Phase 5, CH<sub>4</sub>-IL was increased to 90±4 g.m<sup>-3</sup>.h<sup>-1</sup> while X-IL remained unchanged (32.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>). Unlike BF1, the CH<sub>4</sub>-IL increase had an inhibitory effect on both CH<sub>4</sub> and X biodegradation as REs decreased from 57±2 (Phase 4) to 49±2% (Phase 5) and from 90±2 (Phase 4) to 77±1% (Phase 5) for CH<sub>4</sub> and X, respectively. Lee et al. (2011) reported that X has more toxicity or inhibition effect on

methanotrophs than EB during tests on methanotrophs isolated from a landfill soil which could explain the CH<sub>4</sub>-RE decrease.

The addition of CH<sub>4</sub> at increasing ILs did not significantly affect EB conversion as mean EB-RE was 87±4% for Phases 3, 4 and 5. Unlike relatively constant EB-REs in BF1, X-RE slightly decreased from 87 (Phase 3) to 77% in Phase 5 (BF2) possibly due to CH<sub>4</sub> inhibitory effect on X since EB has been reported to be more biodegradable than X (Deeb and Alvarez-Cohen, 1999).

For identical CH<sub>4</sub> concentrations, the lower level of VOC concentration (200 ppmv in Phases 6, 7 and 8) resulted in lower VOC-RE (Figs. 2 and 3). For instance, EB-RE and X-RE decreased from 88 to 75% and from 90 to 75% respectively, when VOC inlet concentration dropped from 500 to 200 ppmv in Phase 7 compared to Phase 4 under constant CH<sub>4</sub> inlet concentration of 6000 ppmv.

Regarding CH<sub>4</sub> removal, the concentration of accompanying VOC played opposite roles in BF1 and BF2. According to Fig. 3, a 500 ppmv concentration of X (Phases 3, 4 and 5) resulted in lower CH<sub>4</sub>-REs (6% lower) compared to a X concentration of 200 ppmv while the EB high concentration (Fig. 2) had almost a mild synergistic effect on CH<sub>4</sub>-RE for CH<sub>4</sub> concentration ranging from 6000 to 10,000 ppmv.

### 3.3. Pollutants elimination capacities as a function of their inlet loads

Fig. 4 (a) presents CH<sub>4</sub> and EB-ECs for BF1 as a function of their ILs. Maximum CH<sub>4</sub>-EC was 54.0±2.9 g.m<sup>-3</sup>.h<sup>-1</sup> for a CH<sub>4</sub>-IL of 99.0±4.2 g.m<sup>-3</sup>.h<sup>-1</sup>. Elimination capacities were higher than those reported for the same CH<sub>4</sub>-IL range in inorganic packed BFs. Nikiema and Heitz (2010) obtained CH<sub>4</sub>-EC of 49.5 g.m<sup>-3</sup>.h<sup>-1</sup> for CH<sub>4</sub>-IL of 90 g.m<sup>-3</sup>.h<sup>-1</sup> (EBRT = 4.1 min). Vergara-Fernandez et al. (2020) reported a maximum CH<sub>4</sub>-EC of 42.2 g.m<sup>-3</sup>.h<sup>-1</sup> for CH<sub>4</sub>-ILs in the range of 300–400 g.m<sup>-3</sup>.h<sup>-1</sup>. Ethylbenzene-EC increased from 8.0±0.6 to 28±2 g.m<sup>-3</sup>.h<sup>-1</sup> for ILs of 12.0±0.2 and 32.0±0.4 g.m<sup>-3</sup>.h<sup>-1</sup>, respectively. Results were slightly higher than those of Gallastegui et al. (2013) who obtained EB-EC in the range of 8–20 g.m<sup>-3</sup>.h<sup>-1</sup> for ILs of 10–30 g.m<sup>-3</sup>.h<sup>-1</sup> in a black slag BF with EBRTs ranging from 100 to 200 s.

Methane and X-ECs as a function of ILs in BF2 are presented in Fig. 4 (b). The highest CH<sub>4</sub>-EC obtained (54±4 g.m<sup>-3</sup>.h<sup>-1</sup>) for an IL of 101±3 g.m<sup>-3</sup>.h<sup>-1</sup> was similar to the highest CH<sub>4</sub>-EC in BF1. Xylene-EC increased from 8.0±0.4 to 29.0±2.3 g.m<sup>-3</sup>.h<sup>-1</sup> for X-ILs of 9.0±0.6 to 32.0±0.5 g.m<sup>-3</sup>.h<sup>-1</sup>. The values were close to Natarajan et al. (2014) results who obtained X-EC of 9–29 g.m<sup>-3</sup>.h<sup>-1</sup> in a tree-bark BF with ILs varying between 3 and 40 g.m<sup>-3</sup>.h<sup>-1</sup>.

In Fig. 4, the deviation of CH<sub>4</sub>-EC versus CH<sub>4</sub>-IL from the 100%

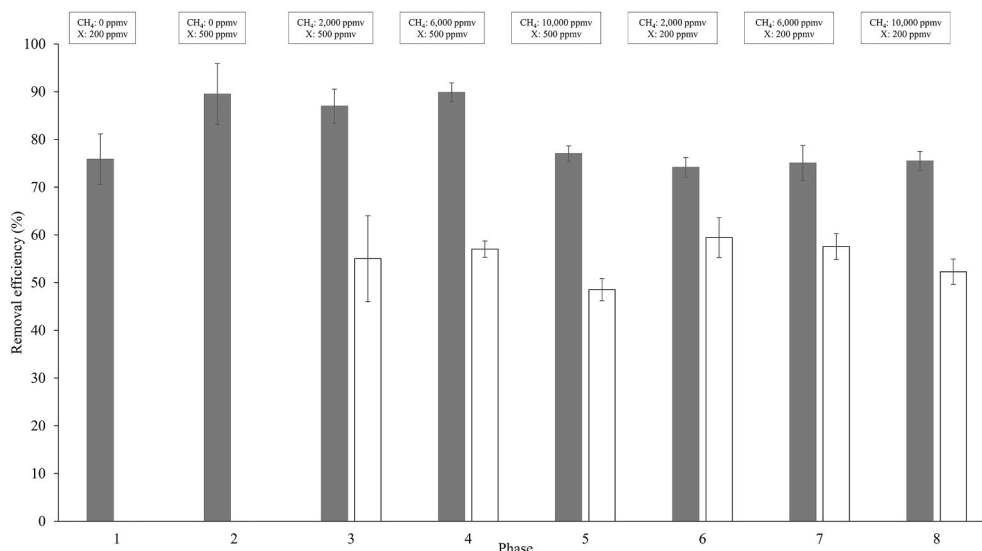
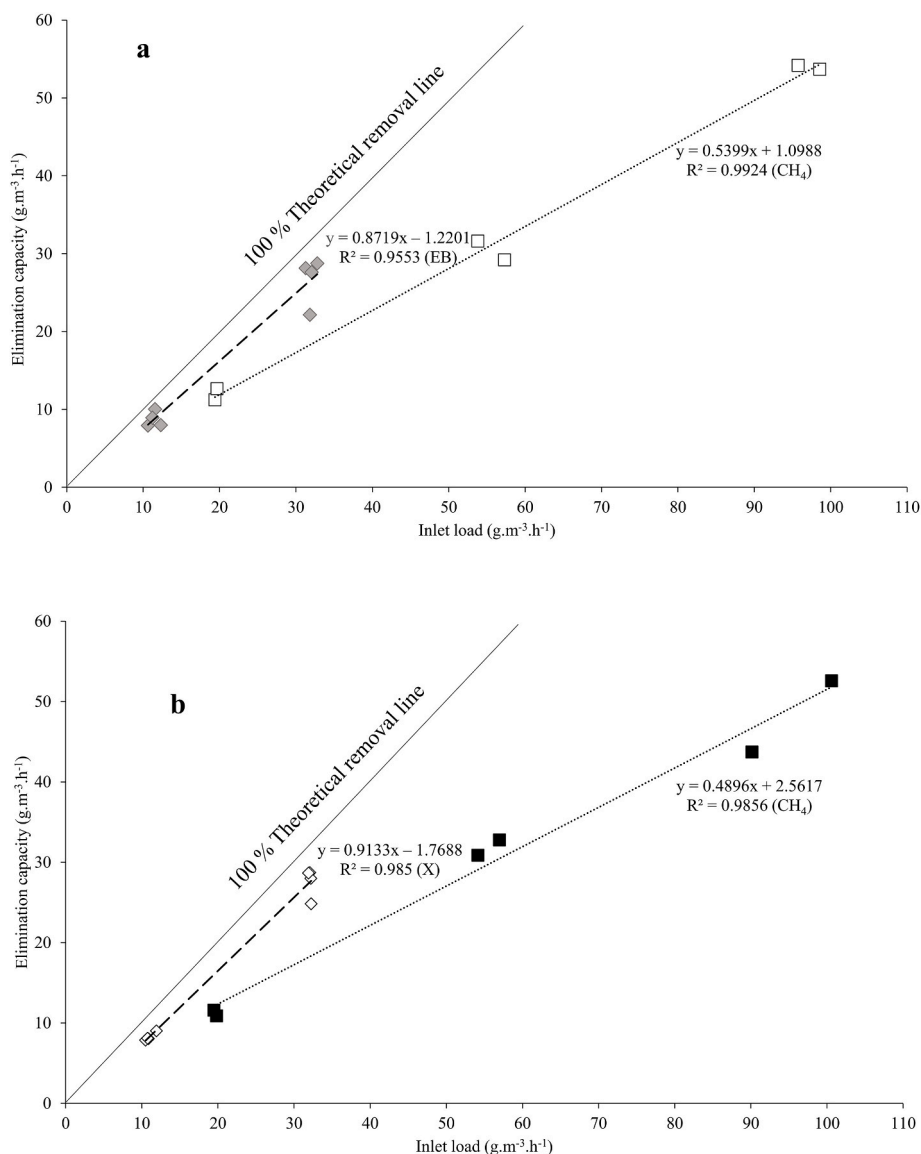


Fig. 3. Pollutants removal efficiencies for each phase in BF2 (■ for X-RE and □ for CH<sub>4</sub>-RE).



**Fig. 4.** Pollutants elimination capacity as a function of inlet load for (a) BF1 (□ for CH<sub>4</sub>-EC and ◆ for EB-EC) and (b) BF2 (■ for CH<sub>4</sub>-EC and ◇ for X-EC).

removal line was an indicator of incomplete conversion of CH<sub>4</sub> in both BF1 and BF2 (RE<100%). However, a high proportion of EB and X were successfully eliminated and the VOC-EC versus VOC-IL curves showed a least deviation from the 100% removal line due to higher solubility of VOCs in water compared to CH<sub>4</sub> (0.022 g.kg<sup>-1</sup> for CH<sub>4</sub> and 0.150 and 0.106 g.kg<sup>-1</sup> for EB and X, respectively at 20 °C, 1 atm) and more biodegradability of VOCs compared to CH<sub>4</sub> (Coquelet et al., 2008; ETB, 2021).

Elimination capacity as a function of IL for CH<sub>4</sub> (in BF1 and BF2) increased linearly. Therefore, the highest EC (EC<sub>max</sub>) for both BF1 and BF2 could not be reached and critical ILs could not be determined. The linear trend of CH<sub>4</sub>-EC as a function of CH<sub>4</sub>-IL in BF1 and BF2 indicates that CH<sub>4</sub> biodegradation was under mass transfer limitations (Ferdowsi et al., 2017).

### 3.4. Total carbon dioxide production as a function of total elimination capacity

Carbon dioxide production rate (PCO<sub>2</sub>) is an indicator of biodegradation activity. The values of EC and PCO<sub>2</sub> can be used to determine the

nature of biodegradation reactions (biomass production, etc.) occurring within the BF. The mass ratio PCO<sub>2</sub>/EC in case of theoretical oxidation (no oxidation by-products or biomass production) is 2.75 for CH<sub>4</sub> and 3.32 for EB or X as single pollutants. The theoretical PCO<sub>2</sub>/EC mass ratio in a BF treating CH<sub>4</sub> and VOC (EB or X) simultaneously, would therefore range between 2.75 and 3.32 depending on CH<sub>4</sub> and VOC concentrations in the mixture (Khabiri et al., 2020b).

Fig. 5 presents the total PCO<sub>2</sub> as a function of total EC (the summation of CH<sub>4</sub>-EC and EB or X-EC) for BF1 and BF2. Carbon dioxide production increased from 17±1 to 102±8 g.m<sup>-3</sup>.h<sup>-1</sup> for total EC in the range of 8.0±0.6–82±6 g.m<sup>-3</sup>.h<sup>-1</sup> in BF1 and from 14±2 to 79±8 g.m<sup>-3</sup>.h<sup>-1</sup> for total EC ranging from 9.0±0.6–69±4 g.m<sup>-3</sup>.h<sup>-1</sup> in BF2. A higher PCO<sub>2</sub> for BF1 (22% higher than in BF2) can be attributed to higher EC obtained in BF1 as the mixture EB + CH<sub>4</sub> was easier to biodegrade than X + CH<sub>4</sub>.

A linear relation between PCO<sub>2</sub> and EC variation was obtained with slopes of 1.02 and 0.94 for BF1 and BF2, respectively. The slopes show the PCO<sub>2</sub>/EC ratios (mass ratio), and the deviation from the theoretical mass ratios (lower than 2.75–3.32) indicates that microorganisms produced biomass and other by-products such as extracellular polymeric

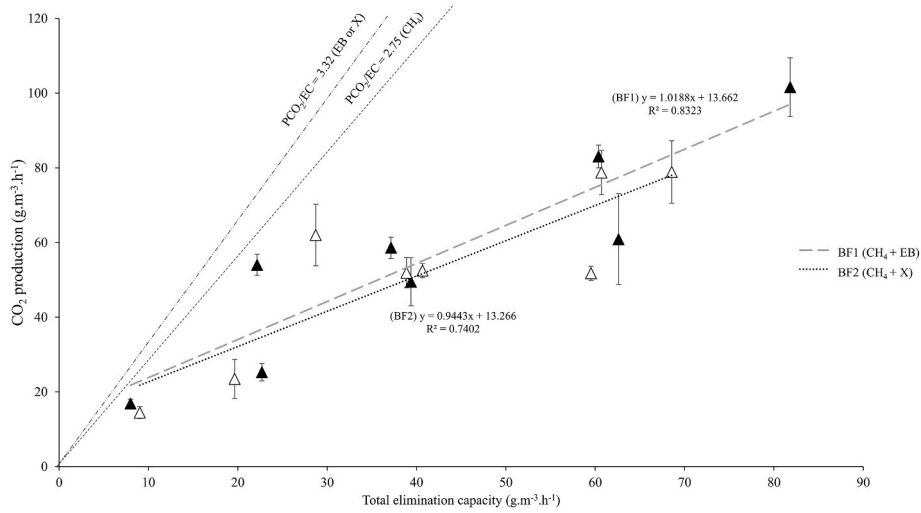


Fig. 5. Carbon dioxide production as a function of elimination capacity for BF1 and BF2 (▲ for BF1 and △ for BF2).

substances (EPS), catechol, formaldehyde, etc., along with CO<sub>2</sub> (Cruz-García et al., 2019; Khabiri et al., 2020b; Shahi et al., 2016). Since the slope for BF1 was 7% higher than for BF2, a higher portion of the

consumed carbon was converted to CO<sub>2</sub> compared to biomass.

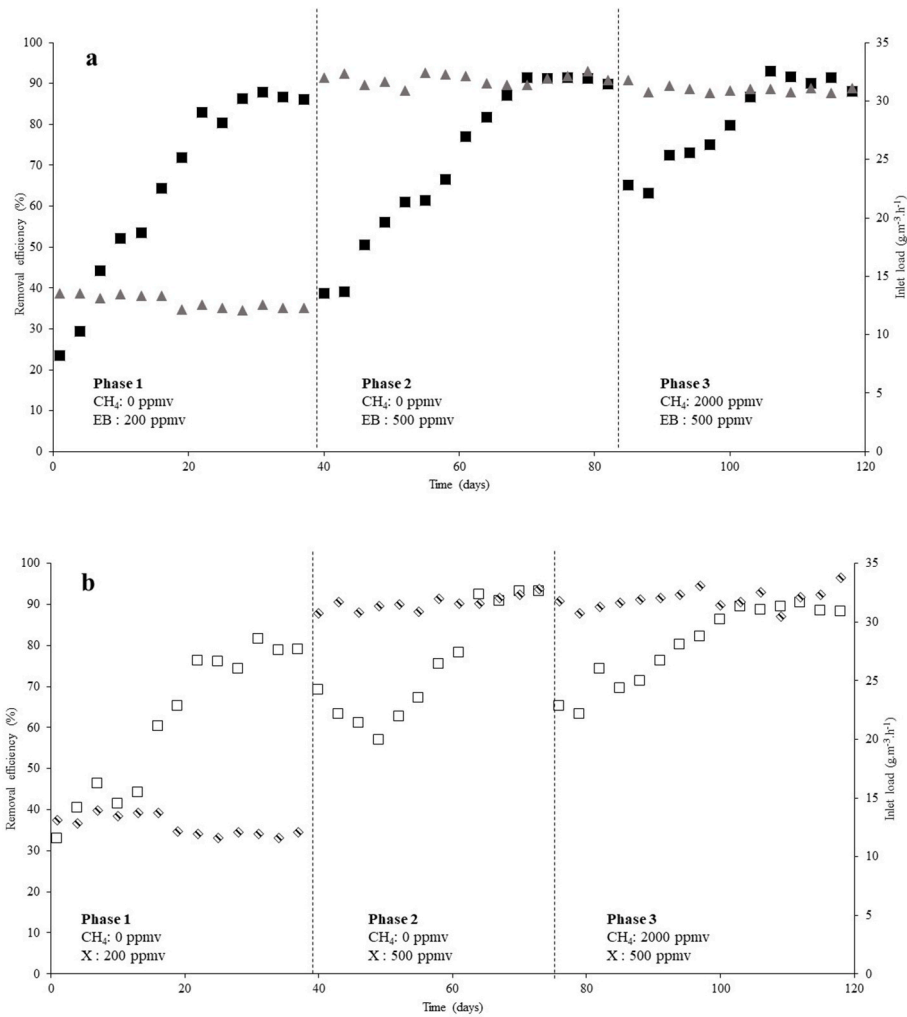


Fig. 6. Dynamic response to concentration increase and CH<sub>4</sub> addition (a) BF1 (■ for EB-RE and ◆ for EB-IL) and (b) BF2 (□ for X-RE and ◇ for X-IL).



### 3.5. VOC biofiltration dynamic response to concentration increase and CH<sub>4</sub> addition

Fig. 6 (a) presents the variations of EB-RE as a function of time during Phases 1, 2 and 3. These first three Phases were selected to investigate VOC elimination as a single pollutant and in a mixture with CH<sub>4</sub> introduced during Phase 3. The BF response to VOC concentration increase as well as CH<sub>4</sub> addition was monitored over time. Ethylbenzene-RE was 23% one day after inoculation when EB was injected at an IL of  $12.0 \pm 0.2 \text{ g.m}^{-3}.\text{h}^{-1}$ . Ethylbenzene-RE gradually increased from day 1 to day 30 (Phase 1) when it reached a pseudo-steady state (RE around 88%). The results indicated that the adaptation period was longer than usual for VOCs treatment in BFs. Garcia-Paña et al. (2008) obtained EB-RE close to 90% after about 5 days of inoculation with a fungi consortium for EB concentration around  $40 \text{ g.m}^{-3}$  and an EBRT of 1.7 min in a BF packed with vermiculite. Son and Striebig (2001) also reported an EB-RE of 99% at day 1 in a compost-based bed BF for EB-IL of  $6 \text{ g.m}^{-3}.\text{h}^{-1}$  and an EBRT of 2 min. The difference in the adaptation period might be due to the absence of indigenous microorganism community in the inorganic packing or lack of EB degraders in the initial inoculum as the inoculum used was a leachate from a BF treating only CH<sub>4</sub>.

Ethylbenzene-RE dropped sharply to 40% (decreased by 55%) after EB-IL was increased to  $32.0 \pm 0.4 \text{ g.m}^{-3}.\text{h}^{-1}$  (Phase 2). This RE reduction demonstrated that the increased EB-ILs were beyond the degradation capacity of microorganisms. Volckaert et al. (2013) reported an EB-RE decrease from 95 to 60% when EB-IL was increased from 12 to  $50 \text{ g.m}^{-3}.\text{h}^{-1}$  in BF packed with macadamia nutshells and with an EBRT of 90 s. Microorganisms in BF1 gradually adapted to EB-IL after 30 days and EB-RE returned to 91% similarly to the end of Phase 1.

According to Fig. 6 (a) when CH<sub>4</sub> was added at an IL of  $19.0 \pm 0.7 \text{ g.m}^{-3}.\text{h}^{-1}$  (Phase 3) at day 82, EB-RE dropped from 90 to 63% and then gradually increased to 93% after 20 days. Ethylbenzene-RE decrease after CH<sub>4</sub> addition was related to the required adaptation period to the increased substrates ILs and the presence of a new substrate (CH<sub>4</sub>) in biofilm.

Fig. 6 (b) presents X-RE during the first 3 phases in BF2. In Phase 1, when X was injected at an IL of  $12.0 \pm 0.2 \text{ g.m}^{-3}.\text{h}^{-1}$ , X-RE was 33% at the first day of the operation and gradually increased from day 1 to day 30 to reach a pseudo-steady state (X-RE of approximately 76%). Therefore, the acclimation period for BF1 and BF2 were similar. Singh et al. (2017) reported X-RE of 40% (IL =  $12 \text{ g.m}^{-3}.\text{h}^{-1}$ ) from first day and reached a pseudo-steady state condition after 20 days with a X-RE of 99% possibly due to a high EBRT of 59 min in a BF packed with wood charcoal.

In Phase 2, X-IL was increased to  $32.0 \pm 0.5 \text{ g.m}^{-3}.\text{h}^{-1}$  at day 40 and X-RE gradually decreased to reach 57% at day 49 (27% RE decrease). Then X-RE increased from 57 to 92% after 15 days. According to Fig. 6 (a) and (b), X-RE decrease after the X-IL increase between Phase 1 and Phase 2 was smooth and less sudden compared to EB-RE. The microorganisms responsible for X removal were likely more resistant to a sudden increase of substrate IL compared to EB's microorganisms. However, a higher RE was obtained for EB possibly due to higher biodegradability of EB compared to X (Natarajan et al., 2017).

At day 76, CH<sub>4</sub> was injected at an IL of  $20 \pm 1 \text{ g.m}^{-3}.\text{h}^{-1}$  in Phase 3. The X-RE sharply dropped to 65% and then gradually increased to 89% after approximately 30 days. BF2 behaved similarly in terms of RE fluctuation following CH<sub>4</sub> addition as BF1 during Phase 3.

## 4. Conclusion

Two identical inorganic-based bed biofilters were employed to eliminate either EB or X as VOC solely and in the presence of CH<sub>4</sub> under the same operating conditions and range of concentration variations. According to the chemical structure similarities between the target VOCs such as number of carbon and aromaticity, the behavior of the biofilters were similar in many aspects. Both biofilters successfully

adapted to the elimination of the single VOCs and VOC-REs higher than 90% for VOC-ILs up to  $32 \text{ g.m}^{-3}.\text{h}^{-1}$  were obtained. Also, the addition of CH<sub>4</sub> could be tolerated in both biofilters owing to the initial CH<sub>4</sub>-degrading rich inoculum in the original leachate used for inoculation. However, certain substrate characteristics differences such as biodegradability, inhibitory effects and toxicity for microbial cultures resulted in some different behaviors in terms of performance in both biofilters. For CH<sub>4</sub> concentration of 2000 ppmv, both biofilters reached CH<sub>4</sub>-REs around 56% under VOC concentration of 200 ppmv. The CO<sub>2</sub> production rate increase when EB or X were involved in the presence of CH<sub>4</sub> confirmed the microbial development and activity during the operation in both biofilters.

## Credit author statement

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## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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