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Effects of gas flow rate and inlet concentration on xylene vapors biofiltration performance

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Abstract

According to previous literature works on biofiltration of mixtures of aromatic compounds vapors, xylene is the most resistant to biodegradation in biofilters. However, only limited information is available about the application of biofiltration to the removal of xylene vapors as the sole contaminant in the air stream. In the present work, biofiltration of xylene vapors has been investigated on a laboratory scale biofilter packed with a new filter material composed essentially of peat mixed with structuring and conditioning agents and initially inoculated with a microbial consortium. Three various gas flow rates, i.e. 0.4, 0.7 and 1 $m^3\,h^{-1}$, were tested for xylene inlet concentration ranging from 0.2 to 4 $g\,m^{-3}$. The biofilter proved to be highly efficient in the removal of xylene at a gas flow rate of 0.4 $m^3\,h^{-1}$ corresponding to a gas residence time of 157 s. For all the tested inlet concentrations, both the removal efficiency and the elimination capacity decreased for high gas flow rates. For all the tested gas flow rates, a decrease in the elimination capacity was noticed for high xylene inlet concentration. The follow-up of carbon dioxide concentration profile through the biofilter revealed that the mass ratio of carbon dioxide produced to the xylene removed was approximately 2.5/1, which confirms complete degradation of xylene if one considers the fraction of the consumed organic carbon used for the microbial growth.

Keywords: Biofiltration; Peat; Xylene isomers; Gas flow rate; Concentration; Carbon dioxide production

1. Introduction

Air biofiltration has been practiced since the early decades of the current century. However, this process has lost its popularity for many decades due to various operating problems that prevailed over its numerous advantages. As a response to increasingly stringent regulations for volatile organic compounds (VOCs) emissions during the last decades, increasing research efforts were deployed for the development of technologies that are both efficient and cost effective for the treatment of relatively dilute air emissions. The treatment of such emissions using conventional technologies such as incineration or adsorption is particularly expensive and energy exhausting. In this context, due to its competitive cost and its environmentally safe aspect, the biofiltration technology gained the interest of many researchers and industrialists. Owing to the recent progress in the knowledge of various aspects of this process, air biofiltration is now a well established and reliable technology widely used for air pollution control. For dilute air emissions, in comparison with the traditional physical and chemical technologies, biofiltration has many advantages including a negligible energy consumption, low investment and operating costs and the absence of environmental nuisances such as the transfer of pollutants to another phase or the release of hazardous or toxic wastes [1–3].

As for all the biological pollution control technologies, biofiltration is based on the natural ability of a specific microbial population to degrade the target pollutants. For the treatment of VOCs emissions, the organic pollutants are aerobically degraded by aerobic heterotrophic microbial species. Generally, a biofilter consists of a fixed bed of porous solid particles on which microorganisms are immobilized and form a liquid biofilm. The air stream is forced through the filter bed and the pollutant concentration gradient between the gas phase and the wet biolayer causes the transfer of pollutants to the biofilm in which they are subsequently biodegraded to water, carbon dioxide and biomass. Various natural materials and industrial biological residues such as peat, compost and wood bark are frequently used as filtering material. Such materials offer many advantages

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including their availability at low prices, a rich variety of indigenous microbial species content and a propitious biological medium for microbial growth and activity especially for their nutrients supply [2,4–8]. However, biofilters packed with these materials may be subject to operating difficulties such as compaction or channeling [9,10]. Recently, some synthetic materials including ceramic, glass, polystyrene, perlite, activated carbon, sandstone and clay are widely used as the main constituents of the filter bed or mixed with biological residues. Such materials, even if more expensive, offer the advantages of excellent mechanical and physical properties which permit to significantly increase the useful life of the filter bed [3,8,11–14].

In the present paper, the biofiltration of xylene vapors was investigated using a new filter material consisting of small balls composed of peat moss (70% w/w) mixed with two structuring and conditioning agents (30% w/w). The same filter material has been successfully used for toluene biofiltration [5], and this bed has shown advantages on other materials such as: suitable size of particles to prevent blockage due to microbial growth, long term useful life, strong buffering capacity and a pH range of 7.2–7.7 which is propitious for the growth of bacteria. Xylene isomers have been listed as hazardous and toxic atmospheric contaminants under CAAA (1990) [15]. Xylene is a major constituents of gasoline. Also, it is used as a solvent in many production industries including the painting and varnishing industries. In Canada, xylene vapor emissions were estimated in 1990 to be 96 kt/year. 58% of these emissions were attributed to the use of xylene as solvent, while xylene vehicles emissions constituted 39% of these emissions [16]. Only few studies focused on the performance of biofiltration in the removal of xylene as the sole pollutant in the air flow [17–19]. Baltzis and de la Cruz [17] studied the biofiltration of *para*-xylene and *meta*-xylene vapors and concluded that only *meta*-xylene could be removed in the biofilter. However, biofiltration of mixtures of benzene, toluene, xylene (BTX) and other hydrocarbon vapors has been investigated by many researchers in previously published work [6,20–23]. Most of these studies revealed that the removal of xylene was always less efficient compared to the other pollutants of the gas streams [18,23]. Questions arose as to whether the removal of xylene could only be achieved in the presence of other aromatic pollutants such as toluene, i.e. by cometabolism. Having shown in a previous work [18] that the biofiltration of xylene vapors as the sole contaminant in the air streams was successfully achieved on a filter bed previously used for the removal of toluene vapors, the present research focused on the use of a new filter material for the removal of xylene vapors. The objective of the present paper is to describe the effects of varying the xylene inlet concentration and the gas flow rate on the biofilter performance and some operating measurable parameters, such as temperature and carbon dioxide production. The results discussed here were obtained during 1 year of operation of the biofilter. Presentation of the daily measured performance of the biofilter during all the operating period is beyond the scope of this paper. Only the most pertinent results are presented and discussed. The impressive results reported on xylene vapors biofiltration and the particularly significant effect of the gas flow rate and the inlet concentration for this relatively recalcitrant pollutant contribute to extend our knowledge on the limitations of application of this technology to the treatment of xylene emissions and help in describing the effect of xylene concentration on the biodegradation kinetic rate.

2. Materials and methods

The main components of the experimental set-up used for the air biofiltration tests are shown in Fig. 1. The biofilter consisted of a vertical three stages cylinder made of rigid Plexiglas, with an inner diameter of 0.15 m and a total height of 1.5 m. It was filled with previously humidified, and conditioned peat balls to a total height of 1 m, evenly divided into three identical sections. The filter material at each section was supported on a sieve plate that ensured homogeneous distribution of gas flow over the cross section of the filter bed. The inflowing gas was supplied tangentially at the base of the column, at a height of 0.15 m under the first section of the filter bed, in order to evenly distribute the gas before contacting the filter material. To avoid drying effects of the filter bed by the air flow, the inlet gas was previously humidified in a separate humidification tower to more than 95% of relative humidity. In order to generate the contaminated air stream, a small fraction of air flow was saturated with xylene vapors and mixed with the air flow at the exit of the humidification column. Flow rates of the main fraction of the air stream and the fraction saturated with xylene vapors were metered by previously calibrated gas flowmeters in order to obtain the desired xylene inlet concentration and gas flow rate through the biofilter. The contaminated air at the inlet contained a mixture of xylene isomers at the following proportions: 19% (v/v) of *meta*-xylene, 65% (v/v) of *para*-xylene and 16% (v/v) of *ortho*-xylene. The contaminated airstream was in contact only with glass, Teflon and Plexiglas. A nutrient solution was periodically pumped over the filter material in order to maintain adequate moisture of the support media and supply the microbial population with additional nutrients. The nutrient solution consisted of an aqueous solution containing KH_2PO_4, NH_4NO_3, $(NH_4)_2SO_4$, NH_4HCO_3 and some trace nutrients such as $ZnCl_2$, $FeCl_3$, $CuCl_2$, $MnCl_2$, $Na_2B_4O_7$, $(NH_4)_6Mo_7O_{24}$ and $CoNO_3$. The filter bed was irrigated daily with 1 l of the nutrient solution evenly distributed at the top of each of the three sections of the filter bed.

At the start-up, the filter bed was inoculated by a specific activated consortium of microbial species. This inoculum was prepared from a consortium supplied by GSI Environnement Inc. (Enviro. biotech. microbial line products, EVB-110). The consortium consisted essentially of specific microbial aerobic and facultative anaerobic species.

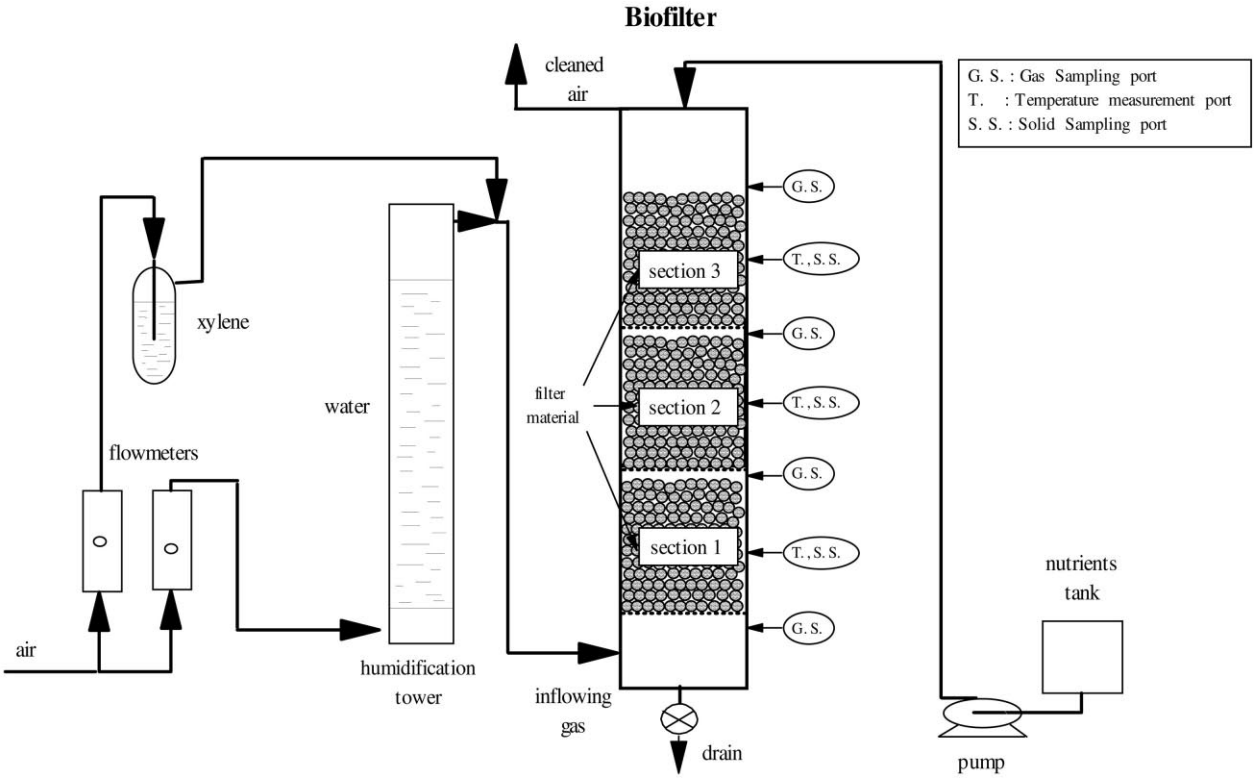

Fig. 1. Biofiltration unit.

The filtering medium temperature was measured daily by three type T thermocouples located at the mid level of each section of the filter bed. These thermocouples were connected to a digital temperature monogram (Omega DP 465). Three gas sampling ports were installed at the exit of each section and served as well for pressure drop measurements by a differential pressure manometer (Air Flow Developments, Canada, LTD, model 4u.5). The gas samples were analyzed for their xylene concentration by a total hydrocarbon analyzer (model FIA-220, Horiba) equipped with a flame ionization detector and a Hewlett–Packard gas chromatograph model 5890, coupled to a mass spectrometer (GC/MS) and equipped with an HP5 (silicon 5%, 30 m × 0.25 mm × 0.25 mm film thickness) capillary column. These two equipments were provided with a sampling loop which permitted automatic and repeated gas sampling directly from the biofilter. The gas phase at each location in the biofilter was also analyzed for the carbon dioxide concentration using a CO_2 analyzer (Ultramat 22P, Siemens) equipped with a continuous sampling pump. The gas analysis equipments were calibrated daily prior to xylene concentration measurements. Clean air and air at various known xylene concentrations served as standards. Also, three solid sampling ports located at the mid level of each section of the filter bed permitted the occasional withdrawal of peat ball samples for moisture and bacterial count analysis. The moisture content of the filter material was measured by an infrared moisture determination balance (model AD4713, AND). The bacterial counts on the filter material samples were performed using standard culture methods.

3. Results and discussion

Biofiltration of xylene vapors emissions was carried out over a period of 1 year at various operating conditions. Three various gas flow rates were tested: 0.4, 0.7 and 1 $m^3 h^{-1}$, corresponding to empty bed gas residence time of 157, 90 and 63 s, respectively. The xylene inlet concentration was also varied in the range of 0.2–4 $g m^{-3}$. During the year of operation, each set of operating conditions (gas flow rate and xylene inlet concentration) was tested at various discontinuous periods of time to insure the accuracy of the experimental results.

The inoculation of the filter material helped in accelerating the establishment of an active microbial population in the filter bed since the biodegradation of xylene in the biofilter was observed few days after the start up. Also, the daily measurement of pressure drop through the filter bed revealed the excellent mechanical and physical properties of the filter material since the pressure drop rarely exceeded 1 mm H_2O/m of packing height. Occasional excess biomass accumulation caused an increase in the pressure drop to a maximum of 40 mm H_2O/m of packing height. This high pressure drop peaks were easily controlled by adequate irrigation of the filter bed which allowed the removal of the

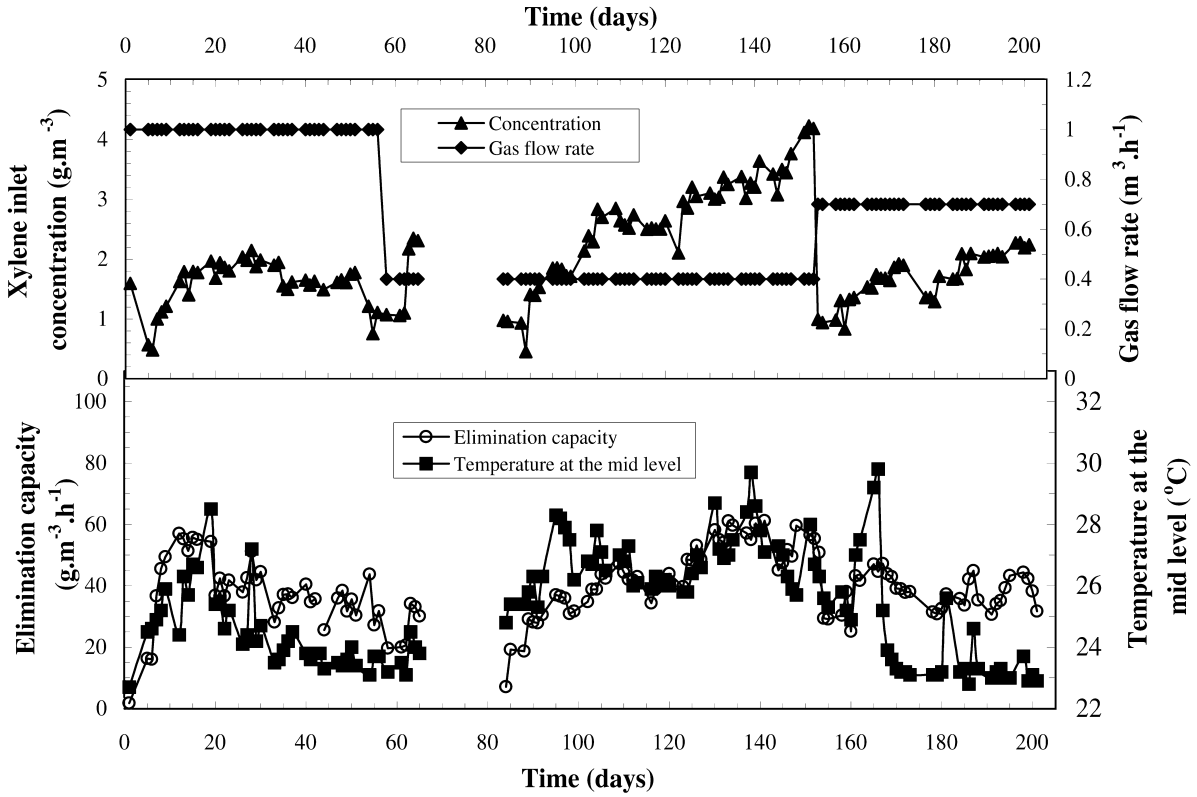

Fig. 2. Operating conditions (inlet xylene concentration and gas flow rate) vs. time; elimination capacity and temperature at the mid level vs. time.

excess of biomass. The follow-up of the moisture content of the filter material confirmed the adequacy of the occasional irrigation mode and the water retention capacity of the filter material since the moisture content of the filter material was successfully maintained around 60% (wet basis) which corresponds to the recommended filter material moisture for biofiltration processes [24,25].

The biofiltration performance is discussed in terms of the xylene inlet load (IL, $g\,m^{-3}\,h^{-1}$), the removal efficiency (X), the elimination capacity (EC, $g\,m^{-3}\,h^{-1}$) and the mass of carbon dioxide produced per units of filter medium volume and time, (PCO_2, $g\,m^{-3}\,h^{-1}$) which are evaluated using the following equations:

$$\mathrm{IL} = \frac{Q}{V} C_{g0}; \qquad X = \frac{C_{g0} - C_{gs}}{C_{g0}};$$

$$\mathrm{EC} = \frac{Q}{V}\left(C_{g0} - C_{gs}\right); \qquad \mathrm{PCO_2} = \frac{Q}{V}\left(C_{CO_2,s} - C_{CO_2,0}\right)$$

3.1. Temperature effect

In general, the temperature of the filter material increases along the filter bed from the bottom to the top. The same behavior has been reported in previous works [26]. This phenomenon is predictable since the metabolic biodegradation of the organic pollutants is an exothermic process. The energy released by these reactions causes the progressive rise of the gas temperature in the bioreactor, which in turn causes a positive temperature gradient in the filter bed from the gas inlet location to the outlet. Also, the lowest level of the bed is subject to cooling effect of the inlet gas which is at a temperature of 18°C.

During all the operation period, the temperature difference between two consecutive levels ranged from 1 to 3°C. Fig. 2 shows the temperature measured daily at the mid level of the biofilter and the corresponding total EC at the exit of the filter bed. Temperature variations at the mid level are very representative of the variation trends noticed in the other two levels.

The daily follow-up of the filter bed temperature variations clearly revealed a sensitive dependence between the temperature of the filter bed and its biofiltration performance. During the first 20 days following the start-up of the biofilter at a gas flow rate of $1\,m^3\,h^{-1}$, the temperature increased from 23 to 28°C. This increase was subsequent to the increase of EC. The decrease in EC from $60\,g\,m^{-3}\,h^{-1}$ to a mean value of $35\,g\,m^{-3}\,h^{-1}$ observed between Days 20 and 65 was also accompanied with a progressive decrease in the filter bed temperature to 23°C at the mid level. At a constant gas flow rate ($1\,m^3\,h^{-1}$), the daily fluctuations of the temperature of the filter bed generally were subsequent to the change in EC. During the second experimental phase with a gas flow rate of $0.4\,m^3\,h^{-1}$, the filter bed temperature was generally higher than the temperature measured in the first experimental period (with a gas flow rate of $1\,m^3\,h^{-1}$) and in the third experimental period (with a gas flow rate of

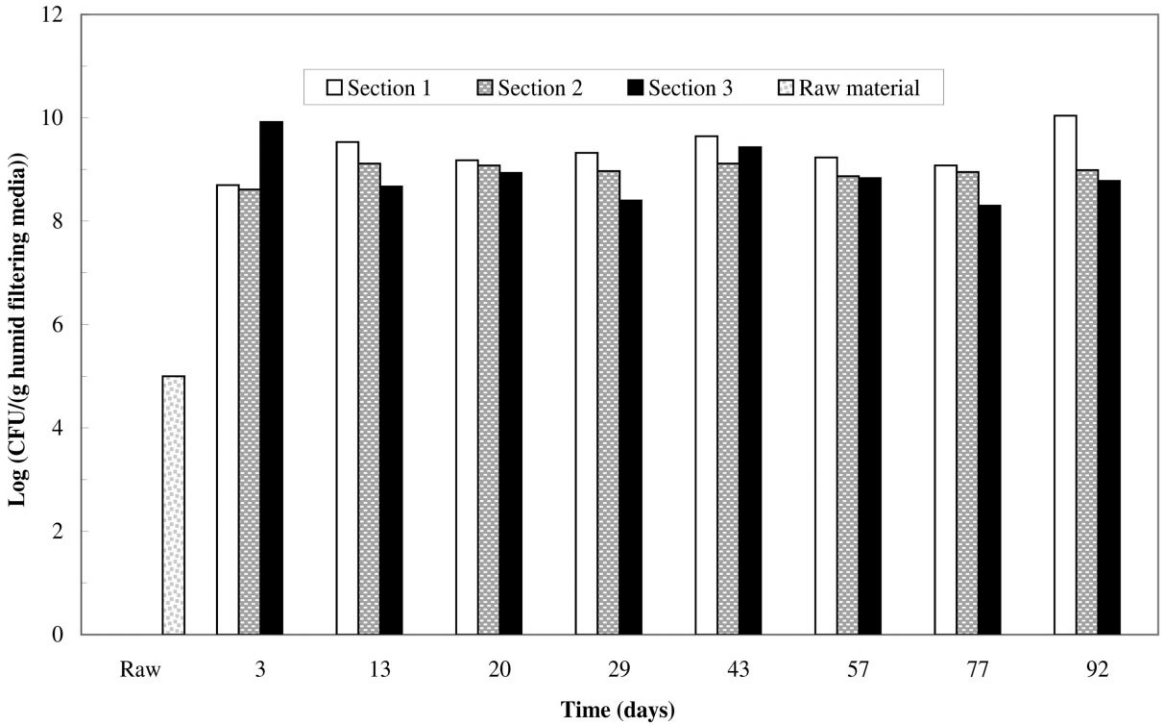

Fig. 3. Microbial count of heterotrophic aerobic bacteria at the three sections of the filter bed vs. time.

$0.7\,m^3\,h^{-1}$), even if the values of EC were almost similar, ranging for all flow rates between 10 and $60\,g\,m^{-3}\,h^{-1}$. In fact, the filter bed performances at a gas flow of $0.4\,m^3\,h^{-1}$ in terms of X, as will be discussed in a later section of this paper, was generally better than those obtained in the two other experimental periods. This behavior suggests a higher intensity of the metabolic microbial activity at that gas flow rate. Also, the cooling effect of the entering gas is less important at a smaller gas flow rate, which may partly explain higher bed temperature for smaller gas flow rates.

3.2. Bacterial count results

Aerobic heterotrophic bacteria counts in the filter material were regularly achieved for the follow-up of the microbial growth intensity inside the biofilter. Fig. 3 presents the aerobic heterotrophic bacteria counts in the raw non-inoculated filter material and in the samples withdrawn from each section of the filter bed versus time. These results show the attainment of a stable bacterial density, higher than the bacterial density measured in the raw material, few days after the start-up. During all the operating period, an average value between 10^8 and 10^{10} CFU/g of humid filtering material was reported for the various sections of the filter bed. However, except for the Day 3 after the start-up, the aerobic heterotrophic bacteria counts were always slightly higher in the first section of the filter bed, which can be related to the higher removal of xylene at this section in comparison with the two other sections. This behavior is probably due to the higher inlet xylene load at this section.

3.3. Gas flow rate and inlet xylene concentration effects

3.3.1. Removal efficiency and elimination capacity

The gas flow rate and the inlet pollutant concentration are the most important parameters in the biofiltration process. Both parameters quantify the amount of pollutant to be removed in the biofilter. The performance of a biofiltration unit is strongly dependant on the pollutants inlet concentration. Biofiltration has proven to be highly efficient for dilute air streams and even for more concentrated emissions of easily biodegradable pollutants. On the other hand, the biofiltration process is less proficient in the treatment of highly concentrated emissions of moderately or poorly biodegradable pollutants [27]. In fact, an increase in the inlet pollutant concentration enhances the transfer rate of the pollutant to the biofilm. However, high concentrations of some recalcitrant pollutants may produce inhibition effects on the metabolic activity of the microbial population [25]. Also, high inlet concentration in the air stream enhances the biomass production, which increases the volume of the biofilm surrounding the filter bed particles, decreases the porosity of the filter bed and causes restriction to the air flow and channeling in the filter bed [28,29]. On the other hand, for a specific filter bed volume, the gas flow rate is the key parameter for varying the gas residence time which should be long enough to permit the pollutant and oxygen transfer from the gas phase

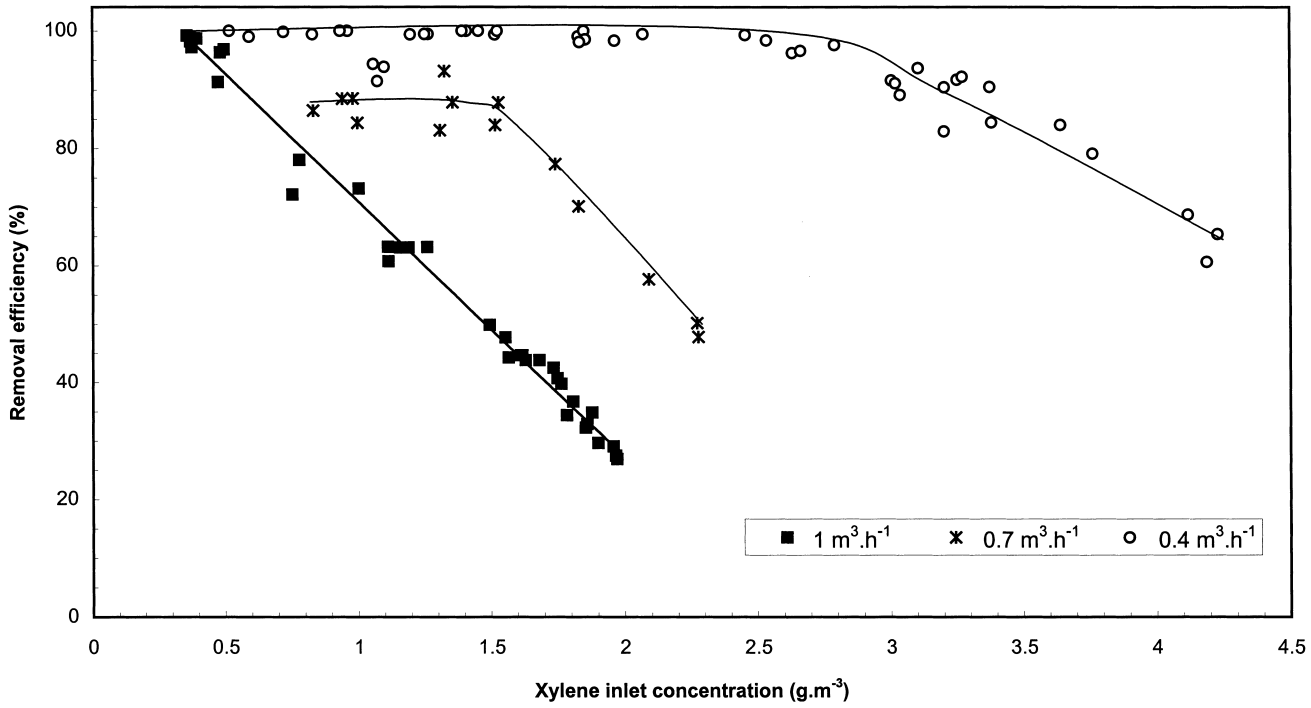

Fig. 4. Removal efficiency of xylene at the exit of the biofilter vs. xylene inlet concentration for various gas flow rates.

to the biofilm and their biodegradation by the immobilized microflora. Hence, the hydrodynamic behavior and the flow regime of the gas phase in the biofilter as well as the pollutant and oxygen transfer rate from the gas phase to the biofilm are conditioned by the gas flow rate and also, to some extent, by the inlet pollutant concentration.

In the present work, the combined effect of the xylene inlet concentration and the gas flow rate on the biofilter performance was investigated. Only the results obtained at steady state are discussed. Fig. 4 presents the removal efficiency of xylene versus the inlet xylene concentration for the various tested gas flow rates, i.e. 0.4, 0.7 and $1\,m^3\,h^{-1}$. Fig. 5 presents the elimination capacity as function of the inlet load for each gas flow rate. At a gas flow rate of $0.4\,m^3\,h^{-1}$, the removal of xylene is 100% for inlet concentrations ranging from 0.4 to $2.6\,g\,m^{-3}$ and decreases from 100 to 70% as the inlet concentration increases from 2.6 to $4.2\,g\,m^{-3}$ (Fig. 4). At this gas flow rate, for IL smaller than $70\,g\,m^{-3}\,h^{-1}$, corresponding to inlet concentrations smaller than $3.2\,g\,m^{-3}$, EC increases with IL (Fig. 5). For xylene IL between 70 and $85\,g\,m^{-3}\,h^{-1}$ (i.e. xylene inlet concentration between 3.2 and $3.8\,g\,m^{-3}$), EC is constant at the maximum value of $67\,g\,m^{-3}\,h^{-1}$. As IL is increased to $90\,g\,m^{-3}\,h^{-1}$, EC decreases to $55\,g\,m^{-3}\,h^{-1}$. At a gas flow rate of $0.7\,m^3\,h^{-1}$, X decreases from 88 to 45% for inlet xylene concentrations varying from 1 to $2.4\,g\,m^{-3}$ (Fig. 4). For this gas flow rate, Fig. 5 reveals a similar trend of variation of EC versus IL as for a gas flow rate of $0.4\,m^3\,h^{-1}$, except that the maximum EC for $0.7\,m^3\,h^{-1}$ is smaller, i.e. $52\,g\,m^{-3}\,h^{-1}$ for IL varying from 58 to $72\,g\,m^{-3}\,h^{-1}$. Similarly, at a gas flow rate of $1\,m^3\,h^{-1}$, the removal of xylene decreases from 100 to 25% for inlet concentrations ranging from 0.4 to $2\,g\,m^{-3}$. For loads smaller than $60\,g\,m^{-3}\,h^{-1}$, EC increases with IL to a maximum of $41\,g\,m^{-3}\,h^{-1}$. EC remains constant at its maximum value as IL increases between 60 and $95\,g\,m^{-3}\,h^{-1}$ and decreases for higher xylene loads. The analysis of the biofilter performance shows that both EC at constant IL and X for constant xylene inlet concentration decrease when the gas flow rate is increased. Higher gas flow rate decreases the contact time between the pollutant and the microbial population and consequently lowers the filter bed efficiency and EC. The general trend of the variation of EC versus the xylene IL for the various gas flow rates shows an increase in EC with increasing xylene IL to a certain value which depends on the gas flow rate. In these situations, the increase of the xylene inlet concentration enhanced the transfer rate of xylene from the gas phase to the biofilm so that more microorganisms participate to the biodegradation activity. This behavior can be described as a diffusion limitation regime. At the maximum EC, the entire active microbial population is involved in the biodegradation kinetics and the diffusion limitation does not occur for these operating conditions. As IL is increased above the upper limit of the diffusion limitation regime, EC first remains constant to its maximum value and then decreases for higher IL. Increase in the xylene inlet concentration above the maximum EC conditions causes a significant decrease in both X and EC. Such a behavior was not expected since previous research works [24,30–32] reported a constant EC with increasing pollutant IL in the absence of diffusion limitation, i.e. in the reaction limitation

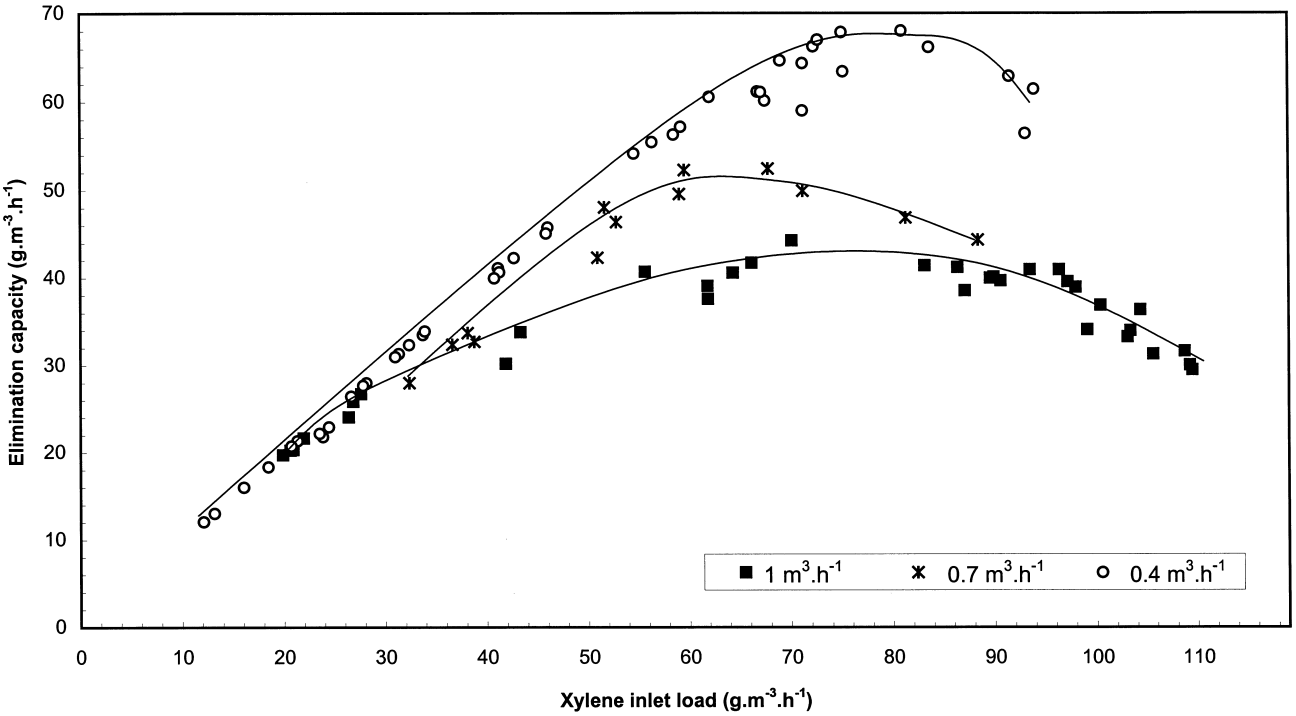

Fig. 5. Elimination capacity vs. xylene inlet load for various gas flow rates.

regime. In fact, the experimental results described in these works were adequately represented by a zero-order kinetics for the pollutant biodegradation rate, which leads to a constant quantity of pollutant degraded per unit of time and filter bed volume, i.e. elimination capacity. In the present experiments, the increase in the amount of xylene transferred to the biofilm enhanced by increasing the xylene inlet concentration above the maximum EC conditions seems to have an inhibition effect on the xylene biodegradation rate which causes a decrease in the amount of xylene biodegraded. This analysis reveals that the kinetics of xylene biodegradation in the biofilm is not likely to be zero-order and probably includes a term relating to inhibition at high xylene concentrations.

3.3.2. Carbon dioxide production

In the biofiltration process, the organic pollutants are aerobically degraded to water and carbon dioxide and used as the essential carbon source for the microbial growth. Hence, the profile of carbon dioxide concentration in the gas phase at the inlet and the outlet of the biofilter provides valuable information on the biofilter performance. A positive gradient of CO_2 concentration in the gas phase through the biofilter indicates that there is CO_2 production due the biodegradation of the organic pollutants. Fig. 6 shows the outlet carbon dioxide concentration versus the xylene inlet concentration for various gas flow rates. In all tested operating conditions, the outlet CO_2 concentration is always higher than the inlet CO_2 concentration ($0.72\,g\,m^{-3}$) indicating biodegradation of xylene in the biofilter. The concentration of CO_2 at the exit of the biofilter increases as the gas flow rate decreases. This result confirms the higher performance of the biofilter at smaller gas flow rates. For a gas flow rate of $0.4\,m^3\,h^{-1}$ and inlet xylene concentration smaller than $3.2\,g\,m^{-3}$, the outlet CO_2 concentration increases with increasing inlet xylene concentration to a maximum of $8\,g\,m^{-3}$ approximately. For higher inlet xylene concentrations, a slight decrease in the CO_2 concentration at the exit is observed. This behavior is in agreement with the observed variations of EC with the inlet xylene concentration at a gas flow rate of $0.4\,m^3\,h^{-1}$. The same remark is also valid for the results obtained with the gas flow rates 0.7 and $1\,m^3\,h^{-1}$. The variation of PCO_2 as function of EC for the various tested operating conditions of xylene inlet concentration and gas flow rates is presented in Fig. 7. In this figure, the mean experimental data lie reasonably around the line $y=2.5x$. This indicates that the ratio between PCO_2 and EC, i.e. the mass of CO_2 produced per mass of xylene removed, is on average equal to 2.5 for all tested conditions with a standard deviation of 0.5. In fact, this ratio should be 3.3 in the case of complete oxidation of xylene to water and carbon dioxide according to the following stoichiometric reaction:

$$C_8H_{10} + 10.5O_2 \rightarrow 8CO_2 + 5H_2O$$

However, in case of biodegradation of organic pollutants, a fraction of consumed organic carbon is used for the microbial growth according to the following metabolism:

$$\text{Organic pollutant} + \text{Oxygen} \rightarrow \text{Carbon dioxide} + \text{Water} + \text{Cellular material}$$

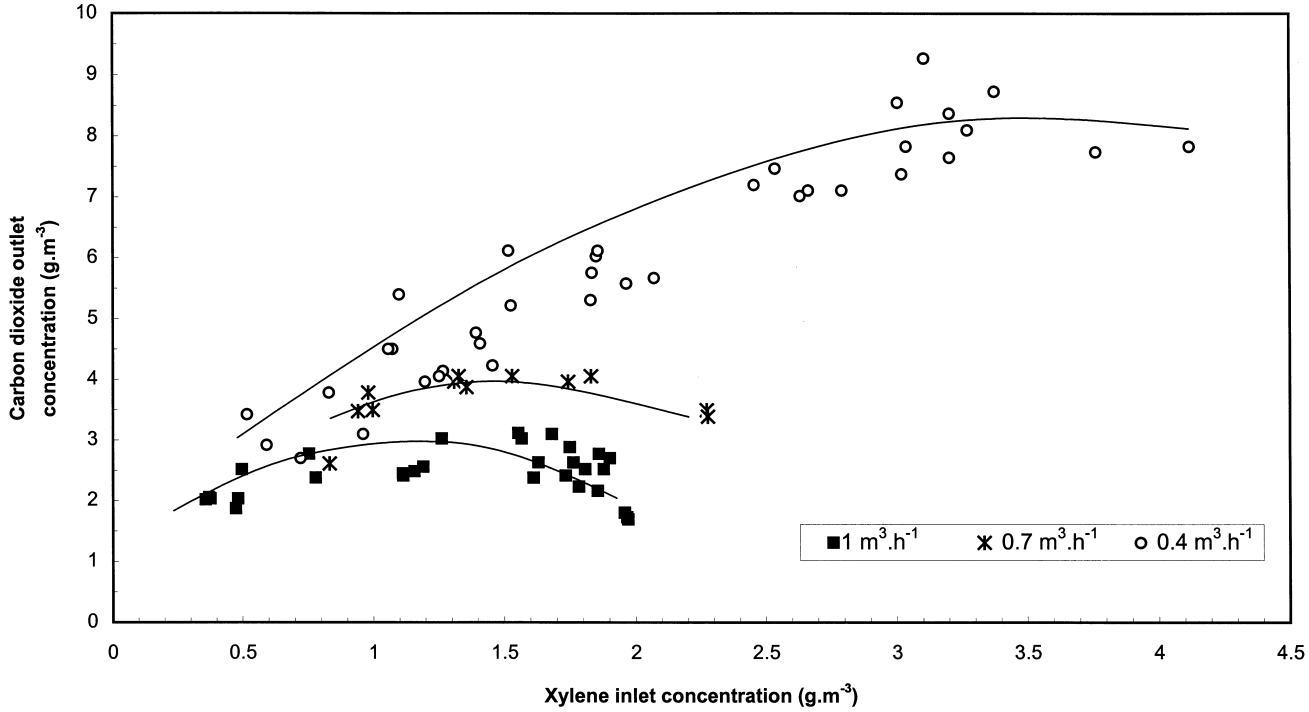

Fig. 6. Outlet carbon dioxide concentration vs. inlet xylene concentration for various gas flow rates.

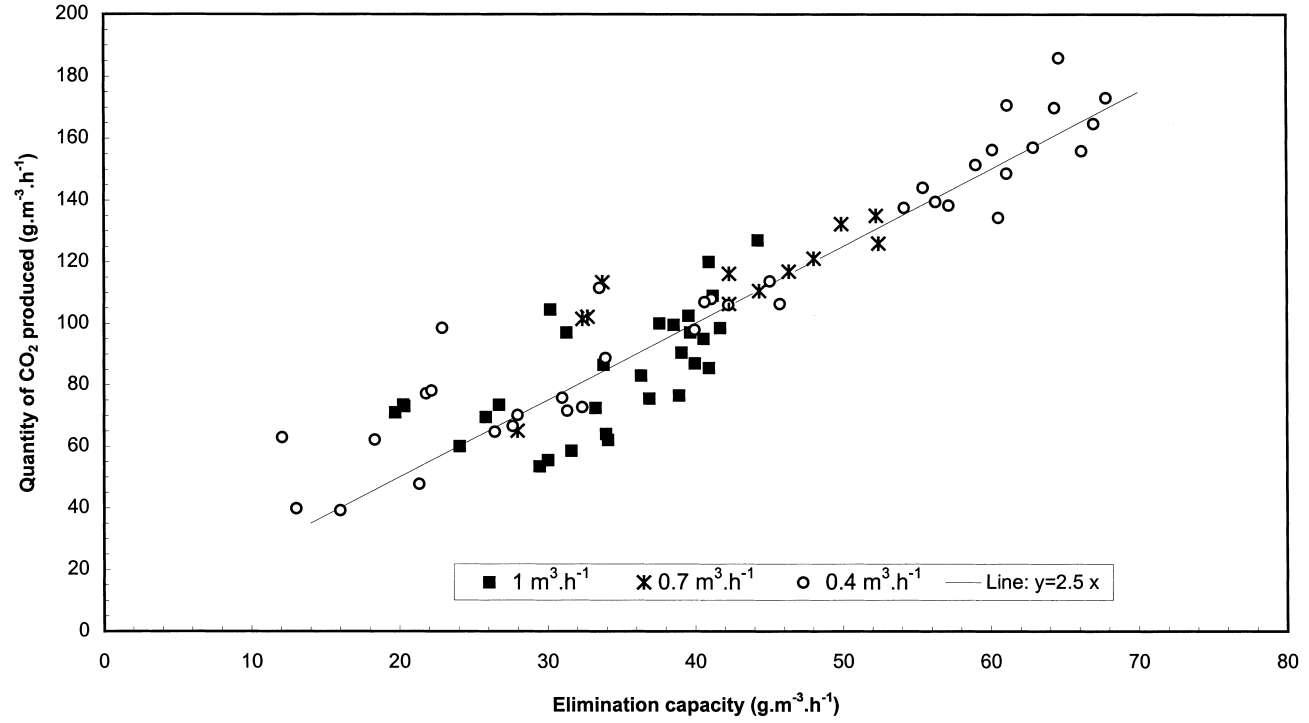

Fig. 7. Quantity of carbon dioxide produced at the exit of the biofilter vs. the elimination capacity.

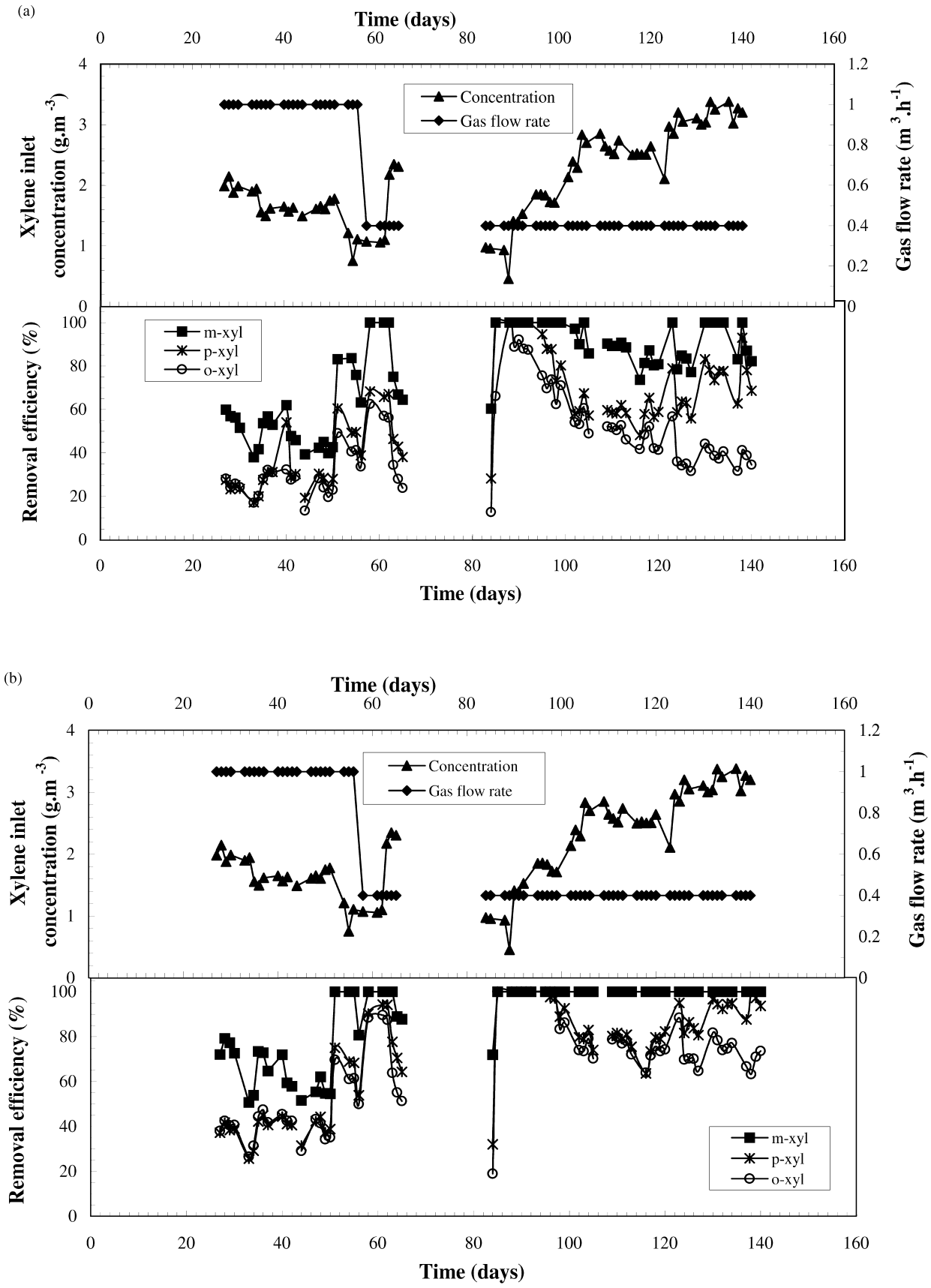

Fig. 8. Operating conditions (total xylene inlet concentration and gas flow rate) vs. time and individual removal efficiency of xylene isomers at the exit of: (a) the first level of the filter bed; (b) the biofilter.

This explains the observed deficit in CO_2 production in comparison with the case of complete chemical oxidation of xylene. In addition, in the biofiltration process, the biodegradation of pollutants occurs in the liquid phase (the wet biofilm), and the CO_2 produced may partly accumulate in biofilm as one of its solute species, HCO_3^-, H_2CO_3 or CO_3^{2-} which can cause a deficit in CO_2 in the gas phase. This may also partly explain the fluctuations of the experimental ratio. Interestingly however, the small difference between the experimental ratio and the ratio evaluated from the stoichiometric reaction of complete oxidation is evidence of the removal of xylene exclusively by aerobic degradation and eliminates any option like adsorption or incomplete oxidation of xylene in explaining the decrease of xylene concentration through the biofilter. Also, this analysis reveals that the follow up of the CO_2 concentration profile through the biofilter can be efficiently used for describing the biofilter performance.

3.4. Removal efficiency of xylene isomers

Fig. 8a and b show the individual removal efficiency of each isomer as function of time for a continuous period with various conditions of total inlet concentration and gas flow rate at the first section of the filter bed and the exit of the biofilter, respectively. Fig. 9a and b show the individual removal efficiencies of the xylene isomers at the exit of the biofilter versus the total inlet xylene concentration at a gas flow rate of 1 and $0.4\,m^3\,h^{-1}$, respectively. The increase of the total inlet concentration of xylene decreases *X* for all the three isomers. At a gas flow rate of $1\,m^3\,h^{-1}$, the difference between *X* for *para*-xylene and *ortho*-xylene is generally not very significant whereas the *meta*-xylene is more efficiently degraded (Fig. 8a, b and 9a). At a gas flow rate of $0.4\,m^3\,h^{-1}$, *X* for *ortho*-xylene was the lowest, followed by *para*-xylene and then *meta*-xylene (Fig. 8a, b and 9b). Owing to the low flow rate of $0.4\,m^3\,h^{-1}$, i.e. high empty bed residence time (157 s), *meta*-xylene was completely removed for all the tested inlet concentrations (Fig. 8b over the period of Days 80–140, and Fig. 9b). At higher gas flow rates, the overall removal efficiency generally decreases and so does the individual removal efficiency of all three xylene isomers. From the analysis of Fig. 9a and b the degrading microorganisms metabolize the xylene isomers preferentially in the order: *meta*-xylene, *para*-xylene and *ortho*-xylene. This order is established through the whole height of filter bed, beginning from the low level to the exit of the biofilter (Fig. 8a and b). These results demonstrate the varying affinity of the microbial species grown in the biofilter toward each isomer. A similar behavior was also reported in another study [18] of biofiltration of xylene vapors emissions at a pilot scale using a different filtering medium inoculated with a pure culture of four species of microorganisms. Also, a study by Baltzis and de la Cruz [17] revealed a higher maximum specific growth rate, estimated by flask experiments, on *meta*-xylene in comparison with *para*-xylene for a consortium capable of growing on all xylene isomers. The analysis of the previous works [17,18] and the present study confirms that among the xylene isomers, *meta*-xylene is naturally the most easily biodegradable while *ortho*-xylene is the less readily biodegraded.

4. Conclusion

Biofiltration of xylene vapors has been investigated, over a period of 1 year, in a laboratory scale up-flow biofilter packed with a new filter material. Various pollutant inlet concentrations and gas flow rates have been tested.The filter material exhibited good physical and mechanical properties as justified by the low pressure drop through the filter bed recorded during all the operation of the biofilter and the possibility of controlling the occasional excess biomass accumulation by adequate irrigation of filter material. A sensitive dependence between the temperature of the filter bed and the biofiltration performance was noticed. Higher filter bed temperature was recorded for tests at a gas flow rate of $0.4\,m^3\,h^{-1}$ for which the highest biofiltration performance, both in terms of elimination capacity and removal efficiency, was obtained. The bacterial count performed on filter material samples withdrawn regularly from the filter bed shows the establishment of a stable bacterial density only few days after the start-up. A slightly higher bacterial density was observed at the bottom section of the filter bed.

The experimental results revealed that the biofiltration performance both in terms of removal efficiency and elimination capacity was higher for smaller gas flow rates for all the tested xylene inlet concentrations. For all the tested conditions, the analysis of the individual removal efficiencies of the xylene isomers revealed that the order of the ease of biodegradation was *meta*-xylene followed by *para*-xylene, then *ortho*-xylene. For a gas flow rate of $0.4\,m^3\,h^{-1}$, complete removal of xylene was achieved for inlet concentrations up to $2.6\,g\,m^{-3}$. For all the gas flow rates, the elimination capacity is an increasing function of the inlet load for low xylene concentrations and reaches a maximum at an inlet concentration which depends on the gas flow rate. The maximum elimination capacity obtained were 67, 52 and $41\,g\,m^{-3}\,h^{-1}$ for gas flow rates of 0.4, 0.7 and $1\,m^3\,h^{-1}$, respectively. For inlet concentrations higher than the maximum elimination capacity conditions, the elimination capacity decreases. Such behavior which has never been reported in previous literature works, is evidence that the kinetics of xylene biodegradation by the active microbial population in the biofilm is not zero-order since high xylene concentration seems to produce inhibition effects on the microbial activity. The important data collected on carbon dioxide concentration profile through the biofilter revealed that the follow-up of the carbon dioxide production provides a reliable means for monitoring the biofiltration performance. For all the ex-

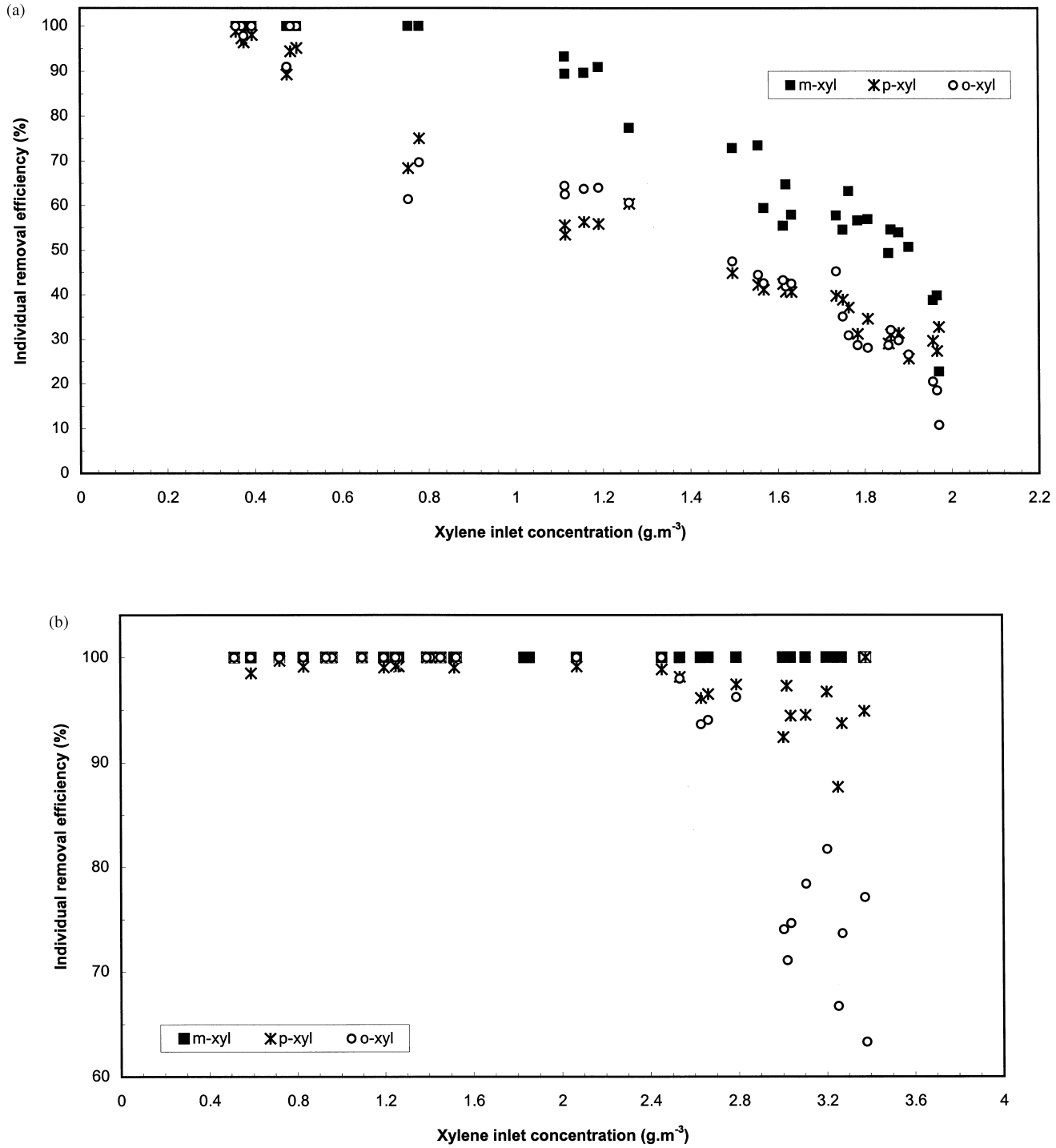

Fig. 9. Individual removal efficiency of xylene isomers at the exit of the biofilter vs. total xylene inlet concentration: (a) Q=1 m^3 h^{-1}; (b) Q=0.4 m^3 h^{-1}.

perimental conditions, the quantity of carbon dioxide produced was approximately 2.5 times the xylene elimination capacity. The stoichiometric ratio for complete oxidation of xylene being 3.3, the small deficit in carbon dioxide production can reasonably be attributed to the use of a fraction of consumed organic carbon for the microbial growth in the biofilm.

5. Nomenclature

CFU	colony forming units
C_{g0}	inlet concentration of the pollutant in the gas phase (g m^{-3})
C_{gs}	concentration of the pollutant in the gas phase at the exit (g m^{-3})

$C_{CO_2,0}$ inlet concentration of carbon dioxide in the gas phase ($g\,m^{-3}$)
$C_{CO_2,s}$ concentration of carbon dioxide in the gas phase at the exit ($g\,m^{-3}$)
EC elimination capacity of the pollutant ($g\,m^{-3}\,h^{-1}$)
IL inlet load of pollutant ($g\,m^{-3}\,h^{-1}$)
PCO_2 quantity of carbon dioxide produced in the biofilter ($g\,m^{-3}\,h^{-1}$)
Q gas flow rate ($m^3\,h^{-1}$)
V volume of the filter bed (m^3)
X removal efficiency of pollutant (dimensionless)

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