

AN INVESTIGATION ON THE EFFICIENCY OF AIR PURIFICATION USING A BIOFILTER WITH ACTIVATED BED OF DIFFERENT ORIGIN

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Recent studies in the area of biological air treatment in filters have addressed fundamental key issues, such as a biofilter bed of different origin composed of natural zeolite granules, foam cubes and wood chips. When foam and zeolite are mixed with wood chips to remove volatile organic compounds from the air, not only biological but also adsorption air purification methods are accomplished. The use of complex purification technologies helps to improve the efficiency of a filter as well as the bed service life of the filter bed. Investigations revealed that microorganisms prevailing in biological purification, can also reproduce themselves in biofilter beds of inorganic and synthetic origin composed of natural zeolite and foam. By cultivating associations of spontaneous microorganisms in the filter bed the dependencies of the purification efficiency of filter on the origin, concentration and filtration time of injected pollutants were determined. The highest purification efficiency was obtained when air polluted with acetone vapour was supplied to the equipment at 0.1 m/s of superficial gas velocity. When cleaning air from volatile organic compounds (acetone, toluene and butanol), under the initial pollutant concentration of ~ 100 mg/m³, the filter efficiency reached 95 %.

Keywords: biofilter, volatile organic compounds, biodegradation, zeolite, microorganisms

1. INTRODUCTION

Many organic materials used in many branches of industry, such as chemical, enamel and paint, oil processing and food industries escape into atmosphere in different ways. The most widely spread organic compounds are acetone, butanol, toluene, xylene and others. Due to volatile organic compounds emitted to the atmosphere as a result of human activity, photochemical oxidants are formed in large concentrations harmful to human health, damaging vegetation and the entire environment. Currently, one of the most promising air cleaning methods is biological air purification applying certain cultures of microorganisms. The use of this method is prospective when cultures of spontaneous microorganisms are cultivated in the biofilter bed. Biological air purification is cheap, efficient and produces no secondary pollutants (Alvarez-Hornos et al., 2007; Jantschak et al., 2004; Schwarz et al., 2001; Wani et al., 1997).

Microorganisms, oxidising hydrocarbons or pollutants, are the important group of organisms participating in the cycle of carbon metabolism. They are able to metabolise all organic and inorganic carbon compounds. Bacteria and micromycetes are the most significant organisms in this group. Bacteria are able of taking various hydrocarbons from the environment and have a characteristic feature of a short life cycle. The frequently detected *Arthrobacter* genera contain bacteria capable of oxidising

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hydrocarbons. Hydrocarbons are decomposed by more than 70 genera of microorganisms (Jankevičius and Liužinas, 2003; Malhautier et al., 2005). A large number of species able to decompose hydrocarbons were also identified in micromycetes. The most frequently detected species of micromycetes are of *Penicillium* genera (Amanullah et al., 2000; Jankevičius and Liužinas, 2003).

The basic element of biologic air purification equipment is a filtering medium as it is necessary to constitute a microorganism substrate and at the same time to provide them with the needful nutrients. In practice, the following biofilter beds of natural origin are used as filtering media: compost, peat, wood chips, barks and activated silt. Frequently, biofilter beds of artificial origin are composed of polyurethane, propylene, polythene, glass, ceramic balls and other materials. However, after some time all these materials disintegrate under the action of microorganisms (Aizpuru et al., 2003; Delhomenie et al., 2002; Deshusses et al., 1999; Szwast et al., 2012; Yamamoto et al., 2005; Zilli et al., 2001).

In order to increase the service life of biofilter beds and at the same time to increase purification efficiency of the equipment several purification methods could be combined – biological and adsorption ones. Since zeolite has a regular structure with pores of equal size and is characterised by a large internal area as well as thermal stability, it is commonly used in air purification as an adsorbent. Having mixed wood chips with zeolite the biofilter bed service life will be increased and its sorptive properties will be improved. Cultures of spontaneous microorganisms should be able to develop not only in wood chips but also in zeolite of inorganic origin. Microorganisms, accumulated in a biofilm formed on the zeolite surface, will decompose organic compounds accumulated in the zeolite pores. To maintain sorptive properties of the biofilter bed, wood chips can be mixed with materials of a larger sorption surface, such as foam. Thus, such a biofilter bed should have better sorptive properties, low density, low price and large area for air purification (Aizpuru et al., 2003; Cheng et al., 2006; Tymczyna et al., 2004; Yamamoto et al., 2005).

In the recent papers of Dorado et al. (2008, 2010, 2012a, b, c) the authors provided more information on the performance of biofilters packed with different hybrid packing material covered by a thin layer of compost and inoculated with activated sludge from a municipal WWTP and applied them to purify air containing toluene vapour. Dorado et al. (2012a) found that biomass accumulation in a biofilter was enhanced by the high toluene load, particle size of the material, and its spatial distribution in the bed enabled to collect information on biomass growth. The results of their experiments demonstrated that high loads of toluene can be treated by a non-specific consortium in biofiltration. They concluded that it is necessary to control excessive biomass accumulation avoiding reactor clogging to ensure long-term operation. Moreover, they demonstrated that the biofilter is able to recover rapidly the activity after several weeks of starvation.

In their next paper (Dorado et al., 2012b) the authors described a dynamic model describing volatile organic compound abatement and the corresponding biomass accumulation. The mathematical model was based on detailed mass balances which included the main processes involved in the system: advection, absorption, adsorption, diffusion, biodegradation and biomass growth. The model was calibrated and validated using experimental data obtained from a biofilter packed with clay pellets during its operation from inoculation to clogging. The model was satisfactorily validated during the operation of the biofilter under different process conditions.

The major conclusions presented in their third paper of Dorado et al. (2012c) concerned sorption capacity of the material and biomass growth on the support, resulting in biodegradation becoming more decisive in the performance.

The present paper is a contribution to evaluation of the process of air purification from the various gaseous pollutants using a biofilter consisting of different beds (natural zeolite, foam and wood chips).

2. INVESTIGATION PROCEDURE

Investigations were carried out using a biofilter prototype (Fig. 1). To ensure even distribution of airflow and humidity throughout the layer and to reduce aerodynamic resistance of the bed, the filter was equipped with five cassettes separated from each other by metal meshes. Each layer of the bed, except for a mixture of wood, barks and zeolite, was 0.85 m long, 0.65 m wide and 0.15 m high. After filling the cassettes with the filter materials the bed was activated by maintaining its temperature at 30 °C, biomedium acidity at $pH = 7$ and different concentrations of biogenic elements. In order to improve adaptation of spontaneous microorganisms in the biomedium, the air polluted with acetone vapour was injected to the equipment. Thus, microorganisms were provided with the required amount of oxygen and carbon elements (Baltrėnas et al., 2010).

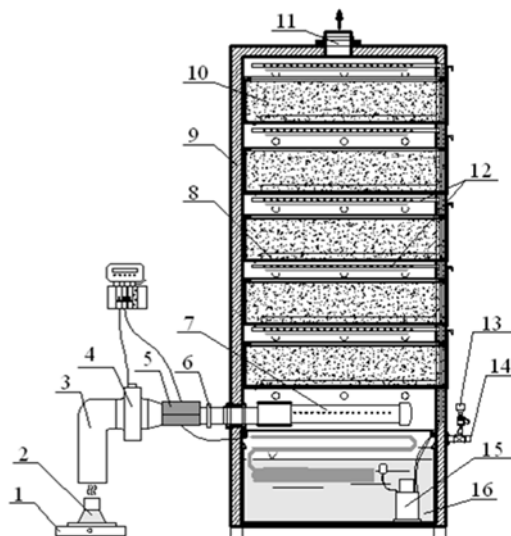


Fig. 1. Biofilter stand: 1 – electric oven, 2 – flask with a pollutant, 3 – inlet air duct, 4 – ventilator, 5 – channelised air heater, 6 – airflow control valve, 7 – collector, 8 – mesh, 9 – wall of biofilter, 10 – filter bed, 11 – outlet air duct, 12 – water sprayers, 13 – biogenic elements supply pipe, 14 – water supply pipe, 15 – water pump, 16 – water tank

A solution of mineral salts made of: K_2HPO_4 – 1 g, KCl – 0.5 g, $MgSO_4 \cdot 7H_2O$ – 0.5 g, $FeSO_4 \cdot 7H_2O$ – 0.1 g, $NaNO_3$ – 0.90 g, water – 1000 g. was supplied to the biomedium to ensure the growth of microorganisms. The salt solution was poured into a water tank and spread over each bed layer.

To obtain a higher efficiency of purification, the filter was loaded with the bed composed of zeolite granules, foam cubes, wood chips and barks. The lowermost cassette of biofilter was loaded with a bed composed of wood chips and barks mixed with 10–15 mm granules of natural zeolite. The well-known reasons for good adsorptive properties of zeolites are their high porosity. An average zeolite becomes saturated with pollutants after 4 days. However, these contaminants easily eliminate living microorganisms in the biofilter bed of wood and barks. To maintain homogeneity of the biofilter beds the mixing ratio of chips, barks and granules by volume was the same (1:1:1), and the height of the layer amounted to 100 mm. The second cassette from the bottom was loaded with 10–15 mm wood chips and barks mixed with 20×20×20 mm foam cubes. The mixing ratio of chips, barks and cubes by volume was also the same. The third, the fourth and the fifth cassette of biofilter was loaded with the bed of 10–15 mm wood chips and barks. The height of the bed layer in all four cassettes was 150 mm.

Before starting the biofilter, the bed was moistened by water sprayers installed over each layer. Water saturated with biogenic elements was supplied to the sprayers by a pump installed in the excess water tank. Operation of the water pump was controlled by a timer mounted at the biofilter control panel switching water pump every hour for 8 seconds. During experimental run the operation time of the

pump was regulated in order to maintain 75% humidity in the bed. To maintain humidity of the entire bed volume (0.387 m^3), about 7 dm^3 per day of water was spread over the beds.

To provide a uniform airflow and to make the excess water flow into the excess water tank, installed at the bottom of the filter, the layers of the biomedium were separated from each other by metal meshes. Humidity in the bed was controlled by a weighting method. Before sampling, the weighing bottles and their caps were dried for 1 hour at $105 \text{ }^\circ\text{C}$ in a drying cabinet, then, cooled down in a desiccator. The desiccated weighting bottles with their caps were weighted by means analytical balance.

The required temperature of the biomedium was maintained with the help of a channelised air heater, installed in an inlet of the air duct, which heated air supplied to the biofilter up to $30 \text{ }^\circ\text{C}$. A uniform temperature in the biofilter was kept by a temperature sensor which was regulated by a controller installed in the filter. The temperature in biofilter was maintained not only by a channelised air heater but also by the heating cable running under each layer of the bed. If the ventilator stopped working a heating cable kept uniform temperature of $30 \text{ }^\circ\text{C}$ in the biofilter.

Air polluted with acetone vapour of different concentrations was fed to the bed providing energy to the microorganisms. Different pollutant concentrations were obtained by diluting acetone with water and heating the solution on an electric stove. The biofilter bed was activated by means of supplying the airflow polluted with acetone vapour. Vapour concentration of the pollutant amounted to ca. 20 mg/m^3 . The pollutant was fed to the equipment 4 times per day for a period of 15 minutes. Later, by reducing the degree of pollutant dilution the concentration of organic compound was increased every 2 days by 20 mg/m^3 , and the duration of acetone supply was extended to 1 hour. Activation of the bed lasted for 3 weeks. To ensure uniform distribution of airflow and pollutant concentration over the entire bed area, an airflow distribution collector was installed at the lower part of the filter.

After activating the bed, air polluted with the acetone vapour was again supplied to the equipment. The concentration of acetone vapour at the inlet to the biofilter (before five bed layers) was maintained at about 100 mg/m^3 . Keeping a uniform airflow at 0.1 m/s superficial gas velocity, the air samples were taken to determine pollutant concentration. The air samples were taken through the sampling ports with screw-caps erected before and after each cassette. The air sample from the air duct was sucked through a stainless steel tube ($d = 5 \text{ mm}$, $l = 30 \text{ cm}$) into a clean 0.25 l gas pipette at the gas flow rate of 0.25 l/min . The sucking lasted 5 minutes. Upon the completion of sucking, the tips of pipette were hermetically closed with glass plugs and the hoses were also pressed with Mohr clamps. The samples were tested on the very day they were taken. Pollutant concentration was determined by means of a gas chromatograph SRI. The following testing parameters were set in chromatograph: nitrogen gas flow rate – 30 ml/min , hydrogen gas flow rate – 30 ml/min , air flow rate – 200 ml/min , column thermostat temperature – $100 \pm 2^\circ\text{C}$, evaporator's temperature – $200 \pm 5^\circ\text{C}$, detector's temperature – $200 \pm 5^\circ\text{C}$. After collecting air samples, the air flow rate supplied to the equipment was increased up to 0.2 m/s , and testing was repeated once again. The airflow was controlled with the regulated airflow valve. The testing was repeated by gradually increasing the flow of the supplied air up to 0.3 , 0.4 and 0.5 m/s . Gas flow rate and temperature of the airflow injected through the bed were measured by the meter Testo 400. Measurements were taken through the ports installed at the side of the biofilter.

To determine the dependence of biological air purification equipment on the concentration of injected pollutant, the acetone concentration injected into the equipment was increased up to 205 mg/m^3 . Pollutant concentration was increased by reducing dilution with water ratio and increasing the heating temperature of the electric stove. Then, having increased the initial concentration of acetone vapour up to 305 mg/m^3 , investigations were repeated. Upon the completion of experimental investigation with acetone, the equipment was purged for 3 hours with clean, unpolluted air enabling desorption of acetone vapour from the beds. Later on, the same investigation was repeated with other pollutants, i.e. butanol and toluene. In order to determine a dependence of bed purification efficiency on the height of the bed layer, pollutant concentrations were measured before and after each cassette of the biofilter.

When determining pollutant concentrations air samples were collected at the intended sampling points, and each measurement was repeated 3 times. Following the experience of earlier investigations carried out by other authors the present experimental studies lasted 1 year (Choi and Oh, 2002; Dorado et al., 2010; Gracy et al., 2006; Jorio et al., 2009; Trusek-Holownia et al., 2012; Vaiškūnaitė et al., 2005, 2008, 2011).

3. EXPERIMENTAL RESULTS AND DISCUSSION

The data on air purification efficiency achieved in the biofilter were collected after 2 months of experiments. The results of these investigations are presented in Figs. 2-6. Data in Fig. 2 reveal that acetone was best decomposed by microorganisms among the studied three pollutants. Acetone is entirely soluble in water, therefore, acetone vapours were more effectively adsorbed in a biofilm formed on the surface of all beds in the biofilter. As expected (Hwan & Tang, 1997; Morgado et al., 2004; Qi, 2005) the removal efficiency increased with the increasing number of cassettes (equivalent to the increasing bed height) asymptotically tending to 100%.

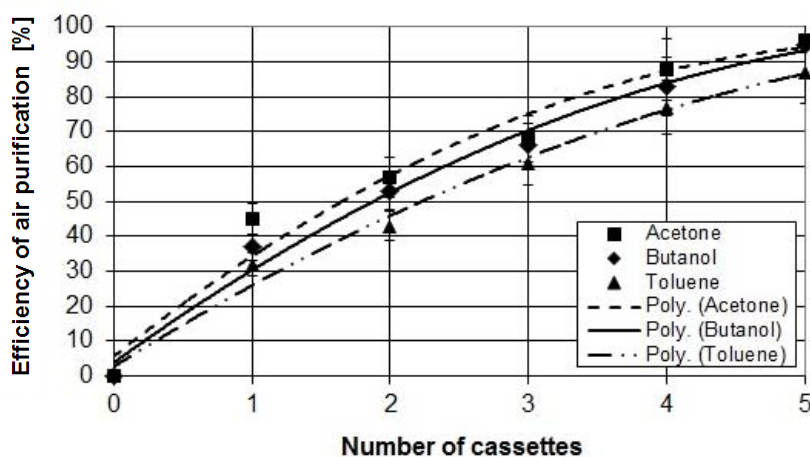


Fig. 2. Dependence of air purification efficiency of the biofilter on the number of cassettes; superficial gas velocity of the injected airflow - 0.1 m/s

The lowest (87%) air purification efficiency was obtained when the equipment was supplied with air polluted with toluene vapours. High (up to 95%) air purification efficiency was achieved for air polluted with butanol vapours. This effect could be explained by hydrophilic properties of the pollutant. Solubility of butanol in water determined in the course of investigations amounted to 3–5 g/100 ml of water. Besides, experimental investigation showed that microorganisms are better reproduced in those substrates which contain more dissolved biogenic elements. The last three bed layers, composed of wood chips and barks, were rather evenly filtering pollutants of different origins. Concentrations of the pollutants were considerably reduced in the second bed layer composed of the mixture of wood chips, barks and foam. After this layer, concentration of acetone vapour has decreased from 58 to 46 mg/m³. The decrease in pollutant concentration was influenced by high humidity of the bed which amounted to 85%, and the nutrients dissolved in water digested by microorganisms during metabolic process.

The effect of different concentrations of injected pollutant vapours on the efficiency of air purification by the biofilter was also determined. These investigations were carried out by injecting air polluted with volatile organic compounds into the equipment at a superficial gas velocity of 0.1 m/s. Under a high concentration of substrate – acetone vapours, the enzyme was saturated, i.e. substrate or molecules of the product always occupied its active centre. Therefore, with the increase in pollutant concentration the efficiency of air purification decreases.

The effect of pollutant concentration in the inlet air stream is shown in Fig. 3 for acetone vapour. The higher concentration of the pollutant the poorer is its removal efficiency. When the initial concentration of acetone vapour was 103 mg/m^3 , the filter efficiency reached 96 %, while after increasing the initial concentration to 305 mg/m^3 , the filter efficiency decreased to 80 % (Fig. 3).

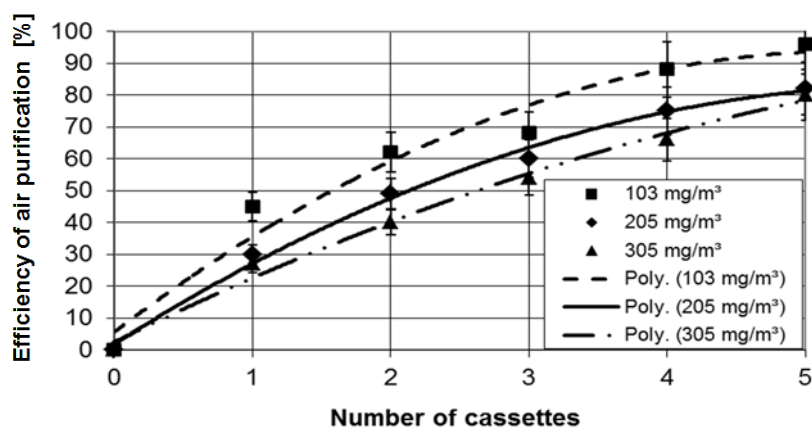


Fig. 3. Dependence of air purification efficiency of the biofilter on the number of cassettes under different initial concentration of acetone vapours

Similar results were obtained for butanol and toluene vapours (Figs. 4 and 5). When injecting air polluted with butanol of the initial pollutant concentration equal to 104 mg/m^3 , the filter efficiency after 5 bed layers attained 95%. This means that pollutant concentration decreases from 104 to ca. 10 mg/m^3 at the outlet from the biofilter. With increasing the initial concentration of the injected pollutant to 310 mg/m^3 , the filter efficiency decreased to only 78%, and the pollutant concentration at the outlet from the biofilter – to 68 mg/m^3 (Fig. 4). Lower efficiency of air purification from butanol can be explained by lower solubility of this pollutant in water.

The concentration of butanol was mostly decreased after the first filter cassette filled with activated bed of wood chips, barks and natural zeolite. After this bed layer the concentration of butanol vapour, under its initial concentration of 105 mg/m^3 , decreased to 63 mg/m^3 . Thus, hydrocarbon less soluble in water is better sorbed by the bed composed of wood chips, barks and zeolite granules. Butanol is trapped in zeolite granules, therefore, it remains in the activated bed for a longer time. In this way, the time of biochemical reactions is extended and degradation of butanol is improved, enhancing purification efficiency of the filter for this pollutant.

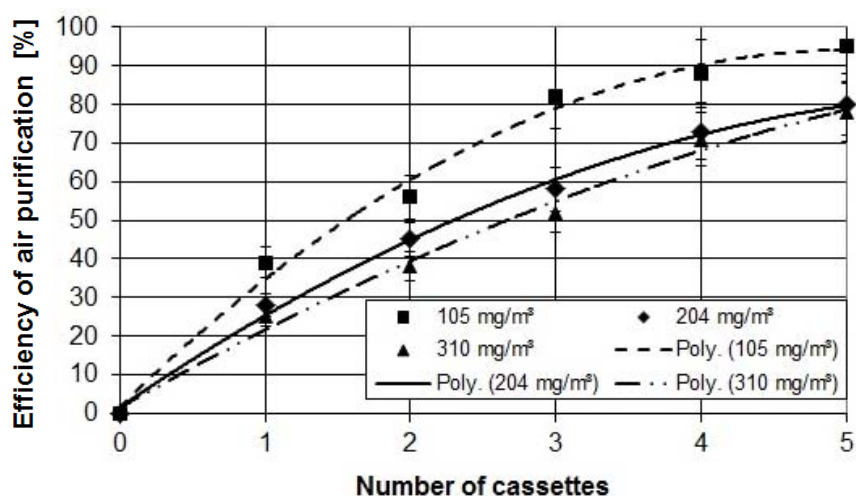


Fig. 4. Dependence of air purification efficiency of the biofilter on the number of cassettes under different initial concentration of butanol

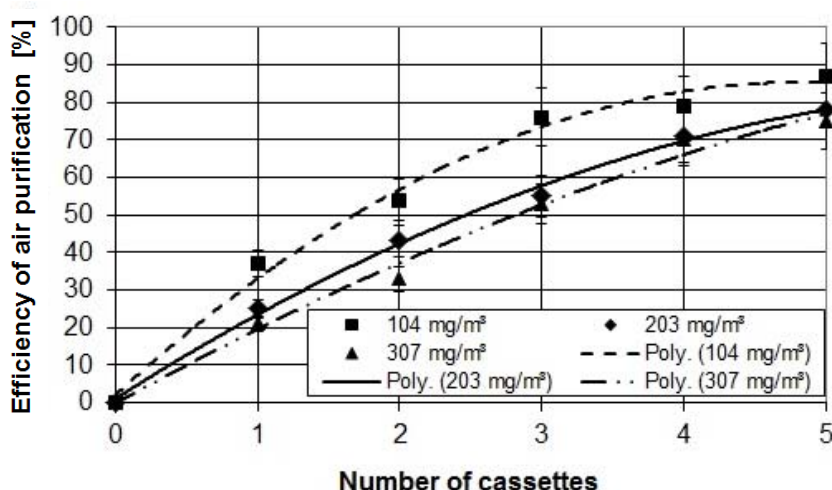


Fig. 5. Dependence of air purification efficiency of the biofilter on the number of cassettes under different initial concentration of toluene

As reported by Jantschak et al. (2004) biofiltration of organic compounds, abundantly formed in industry, such as acetone and butanol, enables reducing their vapour concentration from 900 to 15 mg/m³ (98% of removal efficiency). Qiang et al. (2006) achieved purification efficiency of xylene vapours in laboratory-scale biotrickling filters, filled with different ceramic materials not less than 95% for xylene inlet concentrations lower than 3000 mg/m³ and the air residence time in the filter between 17 and 85 s.

Though the data shown in Fig. 2 would indicate that the removal of toluene vapours in the biofilter was less effective compared to acetone or butanol pollutants, more careful examination of the data plotted in Figs. 3-5 leads to a conclusion that there are only minor differences in the removal efficiency between the studied compounds (within the experimental error).

The removal efficiency of toluene in biofilters has been studied by many researchers. In more recent papers (Alvarez-Hornos et al., 2007; Dorado et al. 2012a, b; Gracy et al., 2006; Hwang and Tang, 1997; Iliuta and Larachi, 2004; Jeong et al., 2006; Morgado et al., 2004; Wani et al., 1997) the reported removal efficiency for toluene or xylene reached 90% or more depending on the initial concentration and load of the pollutant, air residence time in biofilters and type of bed materials. High loads of toluene can be treated by a non-specific consortium in biofiltration (up to 1280 g m⁻³ h⁻¹ in the first 25 cm of the biofilter). However, it is necessary to control excessive biomass accumulation avoiding reactor clogging to ensure long-term operation.

The experimental data displayed in Figs. 2-5 can be approximated by either the second-order polynomial function, Eq. (1) or by Eq. (2).

$$E(x) = a_1x^2 + b_1x + c_1 \quad (1)$$

$$E(x) = a_2 [1 - \exp(-b_2x)] \quad (2)$$

Parameters a_1 , b_1 and c_1 in Eq. (1) as well as a_2 , b_2 in Eq. (2) are empirical constants estimated from each experimental data series and can be estimated from a non-linear regression analysis, while x denotes the number of filter cassettes. In contrast to the polynomial approximation the shape of function given by Eq. (2) is physically more adequate since with $x \rightarrow \infty$ the removal efficiency $E \rightarrow a$, while polynomial functions are applicable only within a certain range of the x values, and any extension beyond this range will most likely yield incorrect values of E . The quality of approximation may be assessed by comparing the values of the regression coefficient, R .

Table 1. Values of coefficients in Eqs. (1) and (2)

Acetone (Fig. 3)							
mg/m ³	<i>a</i> ₁	<i>b</i> ₁	<i>c</i> ₁	<i>R</i> ²	<i>a</i> ₂	<i>b</i> ₂	<i>R</i> ²
103	-3.070	32.940	5.663	0.9620	102.5	0.4666	0.9744
205	-2.328	27.45	1.907	0.9930	100.9	0.3265	0.9959
305	-1.183	21.008	2.231	0.9930	123.9	0.1955	0.9935
Butanol (Fig. 4)							
105	-3.6836	36.982	1.4880	0.9915	110.4	0.4064	0.9917
204	-2.0645	25.986	1.4500	0.9963	105.5	0.2845	0.9975
310	-1.1844	21.239	1.4715	0.9916	133.8	0.1748	0.9915
Toluene (Fig. 5)							
104	-3.5139	34.151	2.0159	0.9891	96.06	0.4569	0.9930
203	-2.4705	27.4930	1.0181	0.9786	92.82	0.3554	0.9807
307	-1.0030	20.543	-0.6030	0.9888	173.4	0.1183	0.9890
Fig. 2							
acetone	-2.7034	31.35	5.5389	0.9676	108.0	0.4035	0.9741
butanol	-2.1484	28.666	3.6149	0.9868	119.6	0.2978	0.9894
toluene	-1.5587	24.491	2.4842	0.9899	127.9	0.2220	0.9902

Examination of the values of this coefficient reveals that data approximation by means of Eq. (2) offers slightly better accuracy. However, the difference between them is within the experimental error.

At the increased concentrations of the pollutant vapours injected into the equipment, the filtration efficiency decreases since microorganisms have no sufficient time to completely decompose volatile organic compounds. Pollutants are best decomposed in the first bed made up of the activated mixture of wood chips, barks and zeolite. Such a decrease in the concentration of a pollutant is conditioned by the largest pollutant load entering the first layer of filter bed (Converti et al., 1997). Thus, an assumption could be made that the thickness of biofilm in this layer is the largest. The second biofilter layer, filled up with a mixture of wood chips, barks and foam cubes, was the best to sorb and decompose acetone and butanol. Since toluene is an organic material much less soluble in water, its concentration after this layer varied not as much as that of acetone.

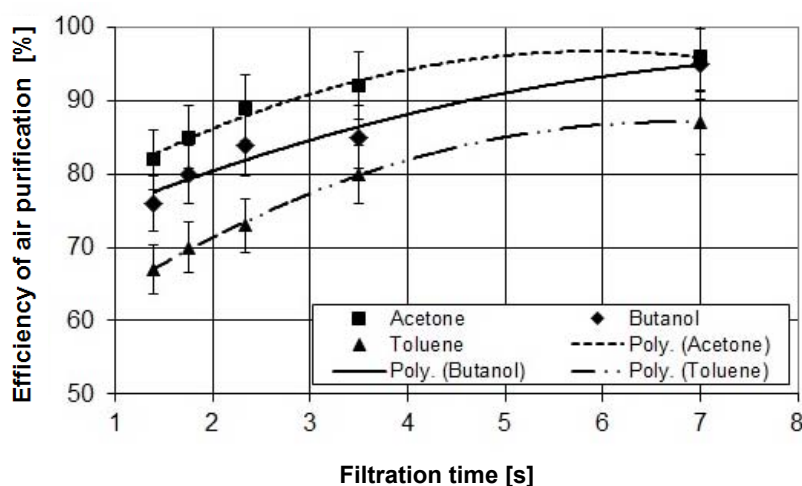


Fig. 6. Dependence of air purification efficiency of the biofilter on the empty bed filtration time (EBRT) when the concentration of injected pollutant is 104 ± 5 mg/m³

Removal efficiency of pollutants from cleaned air depends on gas residence time in biofilters (Alvarez-Hornos et al., 2007; Dorado et al. 2008, 2010, 2012a; Gracy et al., 2006; Hwang and Tang, 1997; Jeong et al., 2006; Qi, 2005; Yoon and Park, 2002; Zilli et al., 2001). The longer is the empty bed residence time (EBRT) of polluted gas the higher removal efficiency can be observed. The data given in Fig. 6 confirm this conclusion.

The gas residence time depends on the superficial gas velocity of airflow passed through the biofilter. The highest removal efficiency in the biofilter was obtained when passing polluted air through the biofilter at a superficial gas velocity of 0.1 m/s. Under this superficial gas velocity the filtration time of pollutants reached 7 s, during which the concentration of acetone vapour at the exit from the biofilter decreased to 96%, of butanol – 95%, and of toluene – 79%. The lowest 67% efficiency of the equipment was obtained when air polluted with toluene was filtered in the biofilter at the filtration time (EBRT) of 1.4 s. During 20–40 days of the experimental investigation, adsorption of pollutants by the biofilter bed decreased to 15% accompanied with an increasing air cleaning efficiency to 45%. After 65 days of experimental investigation, the amount of pollutants absorbed by the filter bed became equal to that removed by the oxidised process and the overall efficiency of air cleaning was about 70–95 %.

In order to improve the biofilter efficiency at high pollutant concentrations ($> 500 \text{ mg/m}^3$), it is necessary to increase the number of cassettes of the equipment or to reduce superficial gas velocity of the airflow supplied to the equipment. In this way, the time of biochemical reactions occurring in the filter will be increased.

4. CONCLUSIONS

- When combining biological and adsorptive methods of air purification, high removal efficiency of the studied air pollutants reaching up to 96 % can be achieved.
- The largest 96 % efficiency of the biofilter was achieved when cleaning air polluted with acetone at the inlet concentration of 103 mg/m^3 . Similar removal efficiency was also obtained for butanol. A high degree of acetone and butanol removal is conditioned by good solubility of the pollutant in water.
- From the results of present investigations it can be concluded there are only minor differences (within the experimental error) in the removal efficiency between the studied compounds.
- Pollutants were best sorbed by the first layer of biofilter filled with a mixture of zeolite granules and wood chips. The first layer of the bed is distinguished by good sorptive and biological properties.
- When increasing the concentration of a pollutant, the efficiency of air purification equipment decreases. Having increased the concentration of acetone vapours fed into the biofilter from 103 to 305 mg/m^3 , purification efficiency decreased from 96 to 80 %. Better removal efficiency in the biofilter can be expected at lower pollutant concentrations.

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