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CONTROL OF LANDFILL GASES EMISSION WITH PARTICULAR EMPHASIS ON BTEX

DEGRADACJA GAZÓW SKŁADOWISKOWYCH ZE SZCZEGÓLNYM UWZGLĘDNIENIEM BTEX

Abstract: Landfilling is the most popular way for waste disposal and has been widely applied globally. A large quantity of volatile organic compounds (VOCs) is released from landfills. Among them, BTEX (benzene, toluene ethylbenzene and xylene) is a major group of pollutants, which have now become a cause for concern worldwide because of their toxic properties. For this reason, strict regulations have come into force which induce researchers to find methods to reduce their emissions. This article contains descriptions of several aerobic metabolic pathways for the degradation of BTEX, which are provided by two enzymatic systems (dioxxygenases and monooxygenases). Special attention was paid to biofiltration - a method for improving the efficiency of treatment of BTEX released from landfills.

Keywords: landfilling, biogas, BTEX degradation, biofiltration, ortho-cleavage pathway, meta-cleavage pathway

Introduction

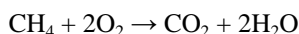
European Union policy pays great attention to sustainable development ie balancing economic development with quality of environment and quality of life in such a way that intra- and intergenerational justice should be assured. Most resources will be exhausted in the foreseeable future and emissions of greenhouse gases accelerate climate change. In according to Lindzen [1] climate change would not be so catastrophic. Nevertheless careful, more balanced management future development of our civilisation is of great importance [2-6]. Among these problems, greenhouse gas emissions have attracted most interest [1, 7, 8]. One major source of greenhouse gas emissions, particularly of methane, is landfill gas.

Landfilling is the most widely adopted practice for municipal solid waste disposal because it is the most economical waste management strategy currently available. Approximately 75% of municipal waste is composed of biodegradable organic materials. Different types of materials decompose at different rates. Food waste is hydrolyzed at the fastest rate among all substrates. Paper, cardboard, wood, textiles are slowly degraded while

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plastic and rubber are non-degradable [9, 10]. In the landfills, gaseous products are formed which are harmful to the environment. The most burdensome of these is methane, which content ranges from 48 to 65% (vol/vol) [11]. Landfills are estimated to contribute 11÷32 Tg CH₄/year into the atmosphere [12, 13]. In situations where the CH₄ concentration is lower than 30% an economical and ecologically applicable way to remove it is via biological oxidation using methanotrophic bacteria [12, 14]. A simplified scheme for the reaction by which methanotrophs oxidise CH₄ is as follows [15, 16]:



Besides methane (Table 1), landfill gas also contains other harmful substances such as non-methane organic compounds detrimental to human health and psychological comfort. According to the EPA report [17] 39% of *Non-methane Organic Compounds* (NMOC) produced in landfills are VOCs. In the group of VOCs emitted from landfills most common are: *benzene*, *toluene*, *ethylbenzene* and *xylene*s (*o*-, *m*-, *p*-xylene) (BTEX). It has long been known that exposure to BTEX can lead to numerous health problems including cancer, nervous irritation or damage to the central nervous system [13].

Table 1

The composition of gases released from landfills [18-20]

The main components of gases emitted from landfills			
Compound	Percent by volume	Compound	Percent by volume
Methane	45÷60	NMOCs	0.01÷0.6
Carbon dioxide	40÷60	(nonmethane organic compounds)	
Nitrogen	2÷5	Sulfides	0÷1
Oxygen	0.1÷1	Hydrogen	0÷0.2
Ammonia	0.1÷1		
Trace constituents of the gases emitted from landfills			
Compound	Concentration range [mg/m ³]	Compound	Concentration range [mg/m ³]
Alkanes		Alkenes	
Propane	< 0.1÷1.0	Butadiene	< 0.1÷20
Butane	< 0.1÷90	Butene	< 0.1÷90
Pentane	1.8÷105	Pentadiene	< 0.1÷0.4
Cycloalkanes		Cycloalkenes	
Cyclopentane	< 0.2÷6.7	Limonene	2.1÷240
Cyclohexane	< 0.5÷103	Other terpenes	14.3÷311
Methylcyclopentane	< 0.1÷79		
Halogenated compounds		Aromatic hydrocarbons	
Chloromethane	< 0.1÷1	Benzene	0.4÷114
Chlorofluoromethane	< 0.1÷10	Styrene	< 0.1÷7
Dichloromethane	< 0.1÷190	Xylenes	34÷470
Chloroform	< 0.1÷0.8		
Chlorobenzene	< 0.1÷2.1		
Esters		Organic compounds containing sulphur	
Ethyl acetate	< 0.1÷64	Carbon sulphide	< 0.1÷1
Methyl butyrate	< 0.1÷15	Carbon disulphide	< 0.1÷2
Ethyl propionate	< 0.1÷136	Methyl mercaptan	< 0.1÷87
Alcohols		Ethers	
Metanol	< 0.1÷210	Dimethylether	0.02÷2
Etanol	< 0.1÷810	Methylethylether	< 0.1÷2
Butanol	< 0.1÷19	Diethylether	0.1÷12

One promising method for removal of both methane and NMOC is biological oxidation.

Biological oxidation has many advantages, such as the possibility of methane oxidation in a wide range of concentrations and the possibility of oxidation of other harmful pollutants. Researchers [21-29] have described various methods for methane removal from landfills.

Microbial decomposition

There are a number of microorganisms, which degrade hydrocarbons. Biodegradation of aromatic hydrocarbons with the participation of bacteria consists of five phases:

- absorption of the hydrocarbon molecule to the cell by diffusion or active transport,
- preparation of the substrate to break the ring,
- cleavage of the aromatic ring,
- converting a compound formed after the ring cleavage to amphibiotic intermediates,
- inclusion of intermediates into central metabolic routes.

Most hydrocarbons are biodegradable under both aerobic and anaerobic conditions [30].

Aerobic degradation

Aerobic biodegradation of aromatic hydrocarbons goes through three key steps: hydroxylation of the aromatic ring, its subsequent cleavage and oxidation to aliphatic compounds to Krebs cycle intermediates and then to CO_2 and H_2O [31]. Key enzymes in the aerobic degradation of aromatic hydrocarbons are oxygenases (di- and monooxygenases). Products of hydroxylation of the ring and its further oxidation are catechol and **protocatechuic** acid. The reaction is catalyzed by dehydrogenases. These compounds (catechol and **protocatechuate**) are the substrates of ring-cleavage enzymes. There are two principle routes by which this can occur:

- *ortho*-cleavage (the intradiol position), or
- *meta*-cleavage (the estradiol position)

In the *ortho*-cleavage, the aromatic ring (either in protocatechuate or in catechol) may be opened by cleaving the bond between two neighbouring carbon atoms carrying hydroxyls. If the opening of the aromatic ring involves other bonds taking a *meta*-position, it is termed *meta*-cleavage.

Catechol is either oxidized in a reaction catalyzed by catechol-1,2-oxygenase which is an *ortho* pathway, or is oxidized in a reaction catalyzed by 2,3-dioxygenase, the *meta* pathway to 2-hydroxymuconic semialdehyde. The products of the final steps of the *ortho*-cleavage pathway are succinic acid, acetyl-CoA, while further transformation of *meta*-cleavage leads to acetaldehyde and pyruvate.

Protocatechuate is metabolized by a homologous set of enzymes: protocatechuate 3,4-dioxygenase and protocatechuate 4,5-dioxygenase [30, 32].

Most of bacteria known as good hydrocarbon degraders are chemo-organotrophic species able to use hydrocarbons as a source of carbon and energy. The genetic potential and certain environmental factors such as temperature, pH, nitrogen and phosphorus sources seem to determine the rate and the extent of degradation [34].

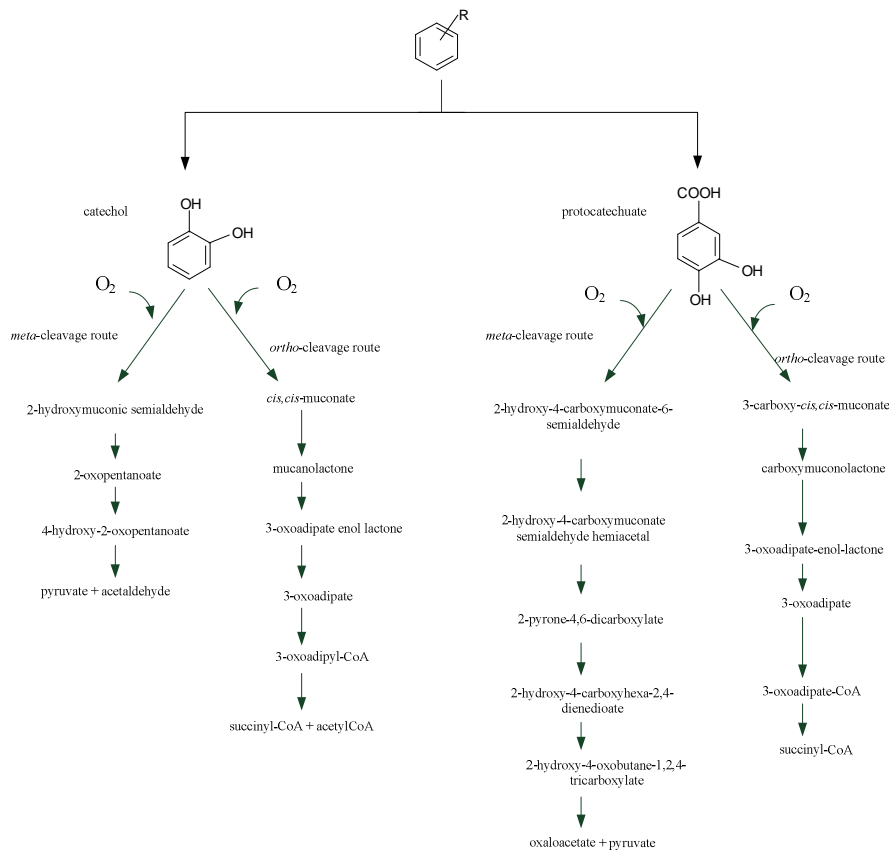


Fig. 1. The biodegradative routes of hydrocarbons [30, 31, 33]

Table 2

BTEX degrading microorganisms - aerobic biodegradation characteristics [35]

Substrate	Microorganism	Comments	References
BTEX (benzene, toluene, ethylbenzene, <i>o</i> -, <i>m</i> - and <i>p</i> -xylene)	<i>Phanerochaete chrysosporium</i>	significant effect of temperature on degradation rate	[36]
	<i>Thermus aquaticus</i> ATCC 25104 and <i>Thermus</i> sp. ATCC 27978	BTEX degradation at 60 and 70°C (10÷40%)	[37]
	<i>Rhodococcus rhodochrous</i>	<i>o</i> -xylene enhanced degradation and ethyl benzene as a potential inhibitor; interaction between different combinations of BTEX components	[38]
	<i>Cladophialophora</i> sp. strain T1	toluene and ethyl benzene as growth substrates, cometabolism of <i>o</i> - and <i>m</i> -xylene	[39]

Substrate	Microorganism	Comments	References
benzene, toluene, <i>p</i> -xylene	<i>Pseudomonas</i> sp. strain CFS-215; <i>Arthrobacter</i> sp. strain HCB	benzene dependent degradation of toluene and <i>p</i> -xylene; enhanced degradation of benzene and <i>p</i> -xylene in the presence of toluene	[40]
	<i>Pseudomonas</i> sp. B1; <i>Pseudomonas</i> sp. X1	cometabolic removal of <i>p</i> -xylene and benzene-inhibition kinetics	[41]
	<i>Pseudomonas</i> strain PP01	cometabolic removal of <i>p</i> -xylene	[42]
benzene, toluene	<i>Pseudomonas</i> sp. D8	degradation of benzene and toluene in 30°C and pH = 7, effect of nitrate, sulfate, and phosphate on degradation	[43]
benzene, toluene	<i>Trametes versicolor</i>	single substrate biodegradation characteristics	[44]
toluene	<i>Paecilomyces variotii</i> and <i>Exophiala oligosperma</i>	effect of temperature and pH on the degradation	[45]

Anaerobic biodegradation

During the past two decades anaerobic biodegradation of aromatic pollutants has also been a subject of extensive research. Most microorganisms are not capable of complete degradation of mononuclear aromatic compounds, but they catalyze the anaerobic biotransformation of independent reactions of these compounds. During these changes, the aromatic ring does not undergo fission, and only ring substituents are converted. Degradation of mononuclear aromatic compounds under anaerobic conditions takes place by benzoyl-CoA, which is converted into acetyl-CoA. Since aromatics are more reduced than the products formed, microorganisms capable of degrading these compounds must be able to dispose of the reducing equivalents generated. Degradation of aromatic hydrocarbons has been reported for denitrifying, iron- or sulphate-reducing bacteria as well as photosynthetic and fermenting bacteria [46].

Anaerobic degradation of hydrocarbons is a two-phase process. In the first stage, the aromatic substrates are converted to one of a few central intermediates (usually benzoyl-CoA), which are suitable for reductive dearomatisation. In the second phase, degradation of aromatics is initiated by the reduction of these intermediates to acyclic compounds, which are then further degraded to acetyl-CoA and CO₂ [46, 47].

Method used for landfill gas removal

Currently used methods for removal of mainly methane from landfills are bio-covers, bio-windows, biofilters, temporary and daily-covers. The most popular are biofilters and biocovers. IPCC recommends biofilters and biocovers as reference methods for mitigation of greenhouse gas emission from landfills. The devices should be commercialized by 2030 [28, 56, 57]. Taking into account that the volatile organic compounds, including BTEX are also oxidized in the aerobic microbial processes, it seemed appropriate to use previously developed methods for removal of methane and these compounds.

As previously mentioned, the most effective methods for removal of methane from landfill gas are biofilters and biocovers. Although the literature on the removal of BTEX from landfills using biocovers is very limited there is much research on BTEX removal by biofiltration (Table 3).

Table 3

BTEX degrading microorganisms - anaerobic biodegradation characteristics [35]

Substrate	Microorganism	Comments	References
benzene	<i>Geobacter</i> spp.	oxidize benzene in Fe(II)-reducing conditions	[48, 49]
	<i>Desulfobacterium</i> spp.	benzene mineralization into carbon dioxide in 5 days	
toluene	<i>G. metallireducens</i>	first pure culture for toluene oxidation	[50, 51]
	<i>Azoarcus</i> spp.	oxidize toluene in nitrate-reducing conditions	
	<i>Thauera</i> spp.		
ethyl benzene	<i>Thauera</i> related	denitrifying bacteria completely mineralize methylbenzene	[52, 53]
xylene	<i>D. acetonicum</i> related	mineralizes <i>o</i> - and <i>m</i> -xylene	[53-55]

Biofiltration

Biofiltration is an effective and inexpensive technology for the treatment of volatile, toxic air pollutants emitted from landfills. Due to its low investment and operating costs, high efficiency (> 99%), safe operating conditions and low energy consumption, biofiltration is one of the most effective methods for the removal of gases emitted from landfills. It generates no undesirable by-products and converts many organic and inorganic compounds into harmless oxidation products [58-60].

During biofiltration, landfill gases percolate through the biofilter filled with porous material and are biologically degraded by microorganisms which are immobilised on packing materials. Acclimation of microorganisms to the newly introduced volatile gases takes between one and two weeks before biodegradation occurs. The packing material within the biofilter (which serves as a habitat and a supplier of nutrients and water) can be composed of biomaterials such as compost, peat, wood bark to more amorphous materials such as charcoal, sand, volcanic ash, porous clay and polystyrene mixture [61, 62]. The selection of suitable materials is one of the key issues in constructing biofilters. First, they should have a high surface area since they must provide sufficient porosity for gas exchange. An appropriate surface area also creates habitats for a large number of microorganisms [63, 64]. When using natural materials, a significant advantage is the wide diversity of microorganisms that inhabit them - in the immediate proximity to the bacteria there are also a number of fungi and actinomycetes active in the degradation of hazardous substances.

Table 4

Maximum elimination capacity and removal efficiency of BTEX [65]

Contaminants	Biofilter medium	Maximum elimination capacity [g/m ³ /h]	Average removal efficiency [%]	References
BTX	compost/perlite	50÷60	not available	[66]
BTX	compost	not available	83÷97	[67]
BTX	compost	108	not available	[68]
BTEX	compost	not available	> 90	[69]
BTEX	compost	not available	> 80	[70]
BTEX	compost	5.3	85%	[65]
BTEX	compost/active carbon	not available	90%	[71]

Conclusions

Microbial decomposition is the best method to degrade greenhouse gases emitted from landfills. The compounds removed are used by microorganisms for growth and as an energy

source. The use of a microbial biofilter is reliable, highly efficient and easy to operate and maintain.

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DEGRADACJA GAZÓW ZE SKŁADOWISK ZE SZCZEGÓLNYM UWZGLĘDNIENIEM BTEX

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Abstrakt: Składowiska odpadów są najtańszą i najprostszą metodą ich zagospodarowania. Ze względu na uwalnianie z nich znacznych ilości *lotnych związków organicznych* (VOC) składowiska stanowią zagrożenie dla czystości powietrza, wód gruntowych oraz gleb. W grupie VOC uwalnianych ze składowisk odpadów najbardziej powszechne i jednocześnie bardzo toksyczne są BTEX (*benzen, toluen, etylobenzen, ksyleny*). Z tego też względu ważne jest poszukiwanie efektywnych metod eliminacji tych związków ze składowisk odpadów. W artykule przedstawiono obecny stan wiedzy odnoszący się do degradacji BTEX przez mikroorganizmy oraz opis metod stosowanych do usuwania BTEX ze składowisk odpadów ze szczególnym uwzględnieniem procesu biofiltracji.

Słowa kluczowe: składowiska odpadów, biogaz, degradacja BTEX, biofiltracja