

# Content of Polycyclic Aromatic Hydrocarbons in Soil Fertilized with Composted Municipal Waste

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## Abstract

The object of our study is soil fertilized with a variety of compost produced from unsorted municipal waste maturing in heaps for different lengths of time, and with green waste compost. Municipal waste composts were applied at rates of 10 and 30 g·kg<sup>-1</sup> soil, whereas green waste compost was introduced to soil as 10 g·kg<sup>-1</sup> soil. Processes that take place during compost maturation have resulted in an increased concentration of polycyclic aromatic hydrocarbons. More PAHs were determined in municipal waste composts than in green waste compost. Fertilization of soil with municipal waste composts increased the content of PAHs in soil, but in most cases the natural concentration of these compounds was not exceeded. However, the total PAHs more than 200 µg·kg<sup>-1</sup> d.m. soil, which is above the natural content, were observed in the objects where the highest rates of one- and three-month-old composts were applied. In the objects fertilized with green waste compost, total PAHs (as well as the levels of particular compounds) were close to the values recorded for the control object.

**Keywords:** compost, municipal solid waste, PAHs, soil

## Introduction

Our civilization has largely contributed to environmental pollution caused by a variety of substances, including polycyclic aromatic hydrocarbons. They are chemical compounds composed of benzene rings characterized by a relatively long half-life and harmful effect on the natural environment. The actual adversity of these compounds depends on the number and type of bonds between benzene rings [1-3]. Although PAHs are not listed in the Stockholm Convention as substances whose production should be limited, they appear in the registry of persistent organic pollutants prepared as part of the United Nations Environment Program (UNEP). These compounds are also among the most frequently examined persistent organic pollutants (POPs) and often considered as model compounds for this group of contaminants [4].

Polycyclic aromatic hydrocarbons are hardly soluble in water, but are well soluble in fats. As a result, they are highly capable of accumulating in live organisms. Many PAHs possess strong carcinogenic, mutagenic, and toxic properties. Therefore, their presence in the environment, and in food and water particularly, is unwanted [5-7].

The primary source of emissions of PAHs to atmospheric air is incomplete combustion of fossil fuels and wood. But PAHs can also originate from other sources, such as motor vehicles, coal and oil furnaces, tarmac production, tobacco smoke, waste dumps, and sewage sludge [8-17]. The presence of these compounds is detected in all parts of the natural environment, although they tend to accumulate mostly in soil.

PAHs can permeate plants in many ways, including soil fertilization with alternative fertilizers produced from municipal solid waste or sewage sludge [18-20]. Thus, the present study has been undertaken to determine the risk of PAH contamination caused by fertilizing soil with unsorted municipal soil waste and green waste compost.

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## Experimental Procedures

Soil fertilized with compost produced from unsorted municipal waste using the MUT-DANO technology and green waste compost produced under natural conditions was examined.

The municipal solid waste for composting originated from an area where the share of biodegradable waste (kitchen waste, paper) in the morphological composition of waste is over 50%. The material for production of urban green waste compost was comprised of leaves, grass, and shredded branches from landscape work in the town of Suwałki.

Composting municipal waste with the aid of the MUT-DANO technology is carried out in a mixed system that consists of homogenization and preliminary initiation of the composting process carried out for 24-48 hours in a biostabilizer, followed by static composting on a composting field. The waste that enters the biostabilizer is previously purified mechanically by some fractions, e.g. large-size waste, paper, scrap iron, or plastic waste. The waste fills up 2/3 of the biostabilizer's capacity. In the chamber of the biostabilizer, waste homogenization takes place, after which the waste is heated up to 45°C and subjected to hygienization and inoculation with microorganisms that carry out biological degradation of organic matter. The biostabilizer ends with a drum sieve, where the material selected for further composting undergoes two-stage sifting, after which it is transported to a conveyor separator where small, hard particles (gravel, pieces of glass, or ceramics) are removed. Further stages of compost production take place in a composting yard. Compost production usually goes on for 1.5 to 6 months.

A pot experiment was performed in a greenhouse belonging to the University of Warmia and Mazury in Olsztyn. Polyethylene Kick-Brauckman pots were filled with 10 kg of proper brown soil, of the grain size distribution of light loamy sand and the reaction  $\text{pH}_{\text{KCl}}=5.5$ , collected from humus horizon  $A_p$ . The physicochemical properties of the soil are specified in our previous paper [21]. The soil moisture content in the pots was maintained at 60% of field water capacity. The soil was fertilized with municipal waste compost that had matured in heaps for 1, 3, and 6 months, or with green waste compost that had matured for 6 months. Municipal waste compost was introduced to soil at rates of 10 and 30  $\text{g}\cdot\text{kg}^{-1}$  soil, whereas green waste compost was added at a dose of 10  $\text{g}\cdot\text{kg}^{-1}$  soil.

Samples of the composts applied in the experiment were taken from three different piles that were left to mature for different periods of time. This procedure was adapted for organizational reasons as it was impossible to obtain compost samples identical in initial composition but different in maturity.

Prior to the experiment, the composts and soil were analyzed to determine the content of total nitrogen, organic carbon, and 16 polycyclic aromatic hydrocarbons listed by the US Environmental Protection Agency. The PAHs determined in the experimental material are: indeno(1,2,3-cd)pyrene, benzo(ghi)perylene, benzo(a)pyrene,

benzo(b)fluoranthene, dibenzo(ah)anthracene, benzo(k)fluoranthene, benzo(a)anthracene, chrysene, fluoranthene, pyrene, acenaphthalene, acenaphthene, fluorene, phenanthrene, anthracene, and naphthalene. The same hydrocarbons were determined in soil after harvest in the first year following the application of the composts.

Concentrations of PAHs were determined using high performance liquid chromatography (HPLC) with UV detection in a ThermoSeparation Product-type liquid chromatographer. For this purpose, samples of the waste and soil weighing 20 g were extracted twice with dichloromethane ( $2\times 40\text{ cm}^3$ ) in an ultrasonic bath (Sonic 3, Polsonic) for 1x45 min and 1x30 min. The extract was centrifuged and evaporated to dryness. The residue was dissolved in a mixture of acetonitrile:water (1:1) ( $3\text{ cm}^3$ ) and purified via extraction to solid phase (SPE, Solid Phase Extraction) using  $C_{18}$  columns ( $3\text{ cm}^3$ , 500 mg) (JT Baker-Mallinckrodt). Each column was conditioned with one portion of methanol ( $1\times 3\text{ cm}^3$ ), followed by two portions of a mixture of water: 2-propanol (9:1) ( $2\times 3\text{ cm}^3$ ). The resulting extract was supplied to the column. Once it was dried, the column was rinsed with one portion ( $1\times 3\text{ cm}^3$ ) of a mixture of methanol:water (1:1) in order to remove any additional impurities, after which it was dried again and the PAHs were washed off with one portion ( $1\times 3\text{ cm}^3$ ) of acetonitrile. At all the stages of purification, the flow of the solvents was set at approximately  $1\text{ cm}^3\text{ min}^{-1}$ .

Quantitative and qualitative determinations of particular PAHs were performed on a liquid chromatographer (ThermoSeparation Product) at a wavelength of 254 nm. A mixture of acetonitrile:water (80:20) at 31°C was taken as the movable phase. The flow of the movable phase was set at  $1\text{ cm}^3\text{ min}^{-1}$ . For the separation of PAHs, a Sherisorb PAH column (Schambeck SFD GmbH) was used. All concentrations of polycyclic aromatic hydrocarbons were expressed as the dry mass of a sample dried at 105°C. Each sample was analyzed in three replicates and the ultimate result was an arithmetic mean of the three determinations.

The statistical computations were conducted using analysis of variance for one-factor experiments, and the least significant differences (LSD) were established at a level of significance  $\alpha=0.05$ .

## Results and Discussion

Properties of composts changed during maturation. The content of organic carbon fell by over 12%, while the content of total nitrogen rose by nearly 50% (Table 1). As a result, the C:N ratio decreased from 19.20 to 11.16. The value of reaction measured in  $\text{H}_2\text{O}$  ranged from 6.94 to 7.07, while that in  $1\text{ mol dm}^{-3}\text{ KCl}$  was within 6.34 and 6.87. Composts which matured longer had a higher pH compared to fresher ones. The results produced by this experiment are close to those reported by other authors [22]. Green waste compost contained nearly half the amount of organic carbon and total nitrogen found in six-month compost produced from unsorted municipal solid waste, while the C:N ratios in both composts were similar (Table 1).

Table 1. Content of organic carbon, total nitrogen and pH of composts.

Parameter		Duration of municipal solid waste compost maturation in piles			Green waste compost
		1 month	3 months	6 months	
Organic C (%)		12.10	11.30	10.60	5.50
Total N (%)		0.63	0.87	0.95	0.46
C:N		19.20	12.98	11.16	11.96
pH in	1 mol dm <sup>-3</sup> KCl	6.34	6.87	6.75	6.70
	H <sub>2</sub> O	6.94	7.06	7.07	7.27

Table 2. Content of PAHs in composts ( $\mu\text{g kg}^{-1}$  d.m.).

PAHs	Duration of municipal solid waste compost maturation in piles			Green waste compost	LSD <sub>p=0.05</sub>
	1 month	3 months	6 months		
Indeno(1,2,3- cd)pyrene	108.7	135.0	121.9	65.3	15.7
Benzo(ghi)perylene	130.5	170.0	172.3	92.4	27.7
Benzo(a)pyrene	131.8	137.1	154.0	103.9	24.1
Benzo(b)fluoranthene	92.7	191.2	217.8	76.5	26.1
Dibenzo(ah)anthracene	nd*	nd	nd	nd	-
Benzo(k)fluoranthene	nd	nd	nd	nd	-
Benz(a)anthracene	58.5	85.5	74.9	52.3	13.1
Chrysene	49.0	55.5	62.1	28.7	9.9
Fluoranthene	30.1	23.4	34.1	7.1	5.9
Pyrene	103.5	97.1	98.6	109.9	19.4
Acenaphthalene	39.5	50.7	61.8	16.8	10.7
Acenaphthene	21.2	21.3	33.7	32.4	5.1
Fluorene	96.5	101.5	110.4	68.4	16.8
Phenanthrene	102.9	112.7	64.4	25.4	17.5
Anthracene	26.2	26.1	21.4	6.3	3.6
Naphthalene	nd*	nd	nd	nd	-
$\Sigma$ PAHs	991.1	1,207.1	1,227.4	685.4	-

nd\*– not detected

The composts tested in our experiment contained various amounts of PAHs. Compost maturation raised the sum of the 16 analyzed PAHs from 991.1 in the one-month compost to 1,227.4  $\mu\text{g kg}^{-1}$  d.m. in the six-month compost. The green waste compost contained half the amount of these compounds compared to the solid waste compost maturing for the same length of time (Table 2). Similar results were reported by Lazzari et al. [23], who demonstrated that modifications in the concentrations of PAHs while composting municipal waste are strictly related to the maturation stage. During the biooxidation phase, PAH concentrations decline, only to rise in later stages of compost maturation.

Composts based on solid waste contained the highest amounts of benzo(b)fluoranthene, benzo(ghi)perylene,

benzo(a)pyrene, and indeno(1,2,3-cd)pyrene. The highest levels of these PAHs were determined in the six-month compost. Much lower concentrations were found for chrysene as well as acenaphthalene and anthracene.

There are no established permissible threshold levels of PAHs for municipal waste composts, therefore the appropriate values are taken by comparison with the thresholds set up for sewage sludge. The indicator compound for total PAHs is benzo(a)pyrene [24]. In the analyzed composts, the value of this compound ranged within 131.8 and 154  $\mu\text{g kg}^{-1}$  d.m., thus being much smaller than the values cited as permissible for sewage sludges.

The three-month-long maturation period of municipal solid waste composts caused the highest increase in the

Table 3. Correlation coefficients between the duration of compost maturation and content of some PAHs in municipal solid waste composts.

PAHs				
Benzo(a)pyrene	Benzo(b)fluoranthene	Indeno(1,2,3- cd)pyrene	Benzo(a)anthracene	Chrysene
0.16	0.76*	0.14	0.50	0.24

\* – correlation significant at  $p=0.05$ .

content of benzo(b)fluoranthene and benzo(ghi)perylene. Green waste compost contained the highest amounts of benzo(a)pyrene and benzo(ghi)perylene, but the lowest concentration of chrysene. The levels of the other PAHs were similar in both composts.

In either type of composts, no presence of dibenzo(ah)anthracene, benzo(k)fluoranthene, or naphthalene was detected. According to Brändli et al. [25], PAHs of a low rather than a high number of rings are mainly degraded during composting. High-ringed PAHs are accumulated, which may be stimulated by the reduction of composted mass (40-60%). In another study, it has been demonstrated that composting did not have any considerable influence on modifications of PAHs, and the concentration of compounds of high molecular weight remained stable [26].

The coefficients of the correlation between the duration of compost maturation and the content of some PAHs revealed that composting had a significant effect on the level of benzo(b)fluoranthene. The values of the other correlation coefficients were statistically non-significant (Table 3).

Déportes et al. [27] claim that the concentration of PAHs in municipal waste compost varies over a wide range, from 1 to 250 ppm, and for particular compounds the content range is from 0.0006 to 49.3 ppm. Such a broad range of concentrations of PAHs in municipal waste compost, conditioned by their origin, has been confirmed by the results cited by other authors [28-29]. Compost produced from waste derived from small towns contained twice as many PAHs as compost made of biowaste. A similar relationship has also been observed in this study.

Other researchers [25] have also demonstrated that, in general, compost made of household waste contained more PAHs, except for naphthalene, than composted green waste. The average content of the 16 analyzed PAHs in green waste composts was  $1,715 \mu\text{g}\cdot\text{kg}^{-1}$  d.m., as compared to  $1,915 \mu\text{g}\cdot\text{kg}^{-1}$  d.m. in compost made of household waste.

Prolonged composting of waste resulted in lower content of pyrene and anthracene (depressed by ca 5 and 22.4%, respectively, in six- versus one-month compost). Regarding the other polycyclic aromatic hydrocarbons (fluoranthene, acenaphthalene, acenaphthene and fluorene), there was an increase in their concentrations, which was positively correlated with the maturation time of composts. However, the increase in the content of PAHs in mature composts versus one-month compost was significant only in the case of acenaphthalene and acenaphthene. Three-month compost contained the highest amount of phenanthrene and prolonged maturation resulted in a significant decline in the concentration of this compound.

The results prove that even at the highest concentrations of PAHs, as determined in the oldest composts, their permissible level ( $3,000 \mu\text{g}\cdot\text{kg}^{-1}$  d.m.), established by the Biowaste Directive, published by the European Commission, has never been exceeded [30]. Thus, it can be concluded that application of the analyzed composts for fertilization should not pose a threat, such as pollution of the natural environment with persistent organic substances, e.g. polycyclic aromatic hydrocarbons.

The analyzed composts made of municipal solid waste raised the content of PAHs in soil compared to the unfertilized treatment. Differentiation of the content of PAHs in soil was the result of the type of composts introduced to soil and the applied rates. Higher average concentrations of these compounds were found in soil enriched with the one-month compost than with more mature composts. The content of the sum of the 16 PAHs in soil fertilized with composted urban green waste was similar to the presence of these compounds determined in the control treatment soil (Table 4).

The content of particular PAHs in soil was varied. Both fertilization rates of municipal waste compost, irrespective of their maturity stage, caused an increase in the content of benzo(a)anthracene, chrysene, fluoranthene, pyrene, acenaphthene, fluorene, phenanthrene, and anthracene with respect to the control treatment. In most cases, the increase was demonstrably larger when the higher fertilization rate was applied (Table 4). Independently from the type of applied compost, the content of acenaphthalene was lower, relative to the control treatment, in the soil from the treatments fertilized with the lower fertilization rate.

In addition, concentrations of particular PAHs in soil were evidently influenced by the maturity stage of composts. The average content of most of PAHs in soil fertilized with the youngest compost was higher than the content determined in soil fertilized with older composts. The lower rate ( $10 \text{ g}\cdot\text{kg}^{-1}$  soil) of the one-month compost caused a small increase in the content of indeno(1,2,3-cd)pyrene, benzo(b)fluoranthene, fluoranthene, pyrene, acenaphthene, phenanthrene, and naphthalene. The same rate of the three-month compost decreased the content of indeno(1,2,3-cd)pyrene, benzo(ghi)perylene, benzo(a)pyrene, benzo(b)fluoranthene, and acenaphthalene below the levels determined in the control treatment. In that case, the content of the other PAHs was similar to the concentrations determined in the unfertilized soil. In turn, the lower rate of the six-month compost raised the content of all PAHs except acenaphthalene.

Table 4. PAH content in soil ( $\mu\text{g}\cdot\text{kg}^{-1}$  d.m.).

Dose of compost ( $\text{g kg}^{-1}$ )	PAHs														$\Sigma$ PAHs
	Indeno(1,2,3-cd)pyrene	Benzo(g,h,i)perylene	Benzo(a)pyrene	Benzo(b)fluoranthene	Benzo(a)anthracene	Chrysene	Fluoranthene	Pyrene	Acenaphthalene	Acenaphthene	Fluorene	Phenanthrene	Anthracene	Naphthalene	
0	8.9	13.7	15.5	11.2	4.2	3.3	6.6	7.5	9.7	4.5	0	2.4	1.6	3.9	93.0
One-month-old municipal solid waste compost															
10	10.5	13.7	16.0	13.2	5.2	4.6	9.0	9.6	7.1	7.0	1.0	5.1	1.8	6.4	110.2
30	22.0	29.4	30.5	25.1	16.7	14.2	26.6	33.4	12.7	13.6	1.8	15.6	6.0	6.7	254.3
Three-month-old municipal solid waste compost															
10	8.4	11.0	13.5	9.8	4.8	4.0	7.1	8.1	5.9	4.1	0.4	3.3	1.7	3.8	85.9
30	22.5	28.1	31.2	25.0	13.6	11.4	20.7	22.5	9.3	10.0	1.4	11.3	3.0	9.4	219.4
Six-month-old municipal solid waste compost															
10	14.3	16.4	23.2	19.5	8.0	7.3	11.5	15.3	7.6	5.5	0.9	3.6	2.5	3.8	139.4
30	13.2	14.4	21.6	18.7	8.6	8.0	20.2	18.3	7.7	14.3	1.7	9.4	2.1	9.5	167.7
Green waste compost															
10	10.1	12.5	13.8	10.6	5.1	4.4	7.7	8.1	6.6	5.7	0.7	2.4	1.2	3.8	92.7

The effect attributable to the maturity of composts was different when the higher rate of fertilizers was applied. For less mature composts (one- and three-month old), application of the higher rate led to a several-fold increase in most PAHs in soil. In contrast, when the six-month compost was introduced to soil in the higher dose, only the levels of benzo(a)anthracene and chrysene rose slightly, whereas the concentrations of fluoranthene, pyrene, acenaphthene, phenanthrene, and naphthalene grew considerably in comparison to the values determined in the treatments receiving the lower rate of this compost. As regards the remaining PAHs, the influence of both rates of this compost was similar.

The compost made of green waste produced when trimming green areas in a town did not have any significant effect on modifications in the content of particular PAHs in soil, as most of them were found to be at a similar or lower level than determined in the control treatment. There were only two exceptions: acenaphthene and fluorene, whose concentrations were similar to those found in soil fertilized with municipal solid waste compost, maturing for the same time and applied at the same rate as green waste compost (Table 4).

The above results prove that fertilization with less mature composts may lead to soil contamination with PAHs. Hence, when using composted waste one must take its age into account. The rate at which polycyclic aromatic hydrocarbons metabolize depends on the physicochemical

properties of soil, which condition the development of soil microorganisms. Soil's sorptive properties are another important factor that affects this metabolism. Improvement of soil reaction, by liming for example, accelerates decomposition of PAHs in soil, thus the de-acidifying role of composts is emphasized [31-33]. Kästner and Mahro [34] claim that the content of organic matter plays an important role in degradation of polycyclic aromatic hydrocarbons. They add to it certain groups of microorganisms, although it is difficult to assess which of the above factors is more important. In the study performed by Haderlein et al. [37], the content of pyrene in soil collected from a municipal landfill and then composted decreased by barely 3% after 15 days of composting. However, when this soil was enriched with organic matter (alfalfa and maple leaves), the mineralization of this compound proceeded over 50% faster than in soil without such a supplement.

Apart from the applied fertilization, the content of PAHs in soil was also conditioned by the type of soil. Higher levels of these compounds were found in loamy than in sandy soils [38].

Many authors [39-41] report that nearly 90% of PAHs present in the environment can be found in soil, and their content is positively correlated with the extent of industrialization of a given region. More PAHs in soil tend to be accumulated in urban areas, where the dominant polycyclic aromatic hydrocarbons are phenanthrene, benzo(a)pyrene, and naphthalene [42]. In areas less affected by emissions



from anthropogenic sources, the content of PAHs in soil can be a few hundred-fold higher than in unpolluted regions.

Depending on the content of the sum of PAHs, there are six degrees of soil contamination with these compounds, based on the assumption that their natural concentration is  $200 \mu\text{g kg}^{-1}$  d.m. [43, 44]. Considering the above criterion, our own study enables us to conclude that the content of PAHs in soil fertilized with municipal waste composts, in most of the examined combinations, was within the permissible limit. A small excess above the natural content of PAHs in soil was observed in the soil sampled from two treatments, i.e. soil fertilized with the one-month compost, where total PAH was  $254.3 \text{ kg}^{-1}$  d.m., and with the three-month compost, where it reached  $219.4 \text{ kg}^{-1}$  d.m. In both cases, this effect was produced by the higher fertilization rate. Although these concentrations only slightly exceeded the natural content, they imply that the soil from both treatments should be classified as having an elevated PAH content.

### Conclusions

As a result of the maturation process, composts contain higher quantities of polycyclic aromatic hydrocarbons. Municipal solid waste composts contained more of these compounds than green waste compost. Neither of the two types of composts contained dibenzo(ah)anthracene, benzo(k)fluoranthene, or naphthalene. The one-month compost produced from municipal waste had the highest concentration of anthracene and pyrene, while the three-month compost was the richest in indeno(1,2,3-cd)pyrene, benzo(a)anthracene, and phenanthrene and the concentration of the other PAHs was the highest in the six-month compost.

Fertilization of soil with municipal waste composts raised the soil content of PAHs, although in most cases the natural level of these compounds was not exceeded. A small excess of the PAH sum, above  $200 \mu\text{g kg}^{-1}$  d.m. (which is considered natural concentration) occurred in the treatments fertilized with the highest rate of composts maturing for one and three months. In the treatments enriched with urban green waste compost, the total content of PAHs was similar to that in the control.

The results enable us to conclude that the threat of soil pollution with PAHs caused by the application of composts produced from municipal solid waste or urban green waste is very small. It is, however, of utmost importance to determine safe rates of composts, which would take into account the levels of these noxious compounds in substances added to soil and in the fertilized substrate. Much attention should be paid to the conditions under which such composts are to be used when crops for human food or animal fodder are grown.

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