CONCENTRATION, SOURCES AND RISK ASSESSMENT OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) IN SEWAGE SLUDGE FROM XUCHANG CITY, CHINA

CHEN, J.[1] – YAN, H.[1*] – DU, J. M.[2,3]

[1]*College of Urban and Environment, Xuchang University, Xuchang 461000, China*

[2]*School of Environment, China University of Geosciences, Wuhan 430074, China*

[3]*Faculty of Sciences, University of Waterloo, Waterloo, Canada*

[*]*Corresponding author*
e-mail: yanhuichj08@163.com

(Received 29th Jan 2024; accepted 4th Jun 2024)

Abstract. Polycyclic aromatic hydrocarbons (PAHs) are persistent organic pollutants in the urban environment, because of their hydrophobic and lipophilic character, PAHs is an easily enriched in sludge, which is by-product of sewage treatment and widely used in cultivated land. Xuchang city is an important grain production area in China, the amount of sludge for agricultural utilization in Xuchang city is gradually increasing during recent years. To investigate the concentrations, potential sources and ecological risk of PAHs in sludge of Xuchang City, dewatered sludge samples from 10 large-scale sewage treatment plants were collected and analyzed. The results showed that the total concentration of PAHs varied from 1835 to 7188 μg/kg, with a mean value of 4362 μg/kg. The average value of PAHs concentration met the limit of China's agricultural sludge control standard, while the total concentrations of PAHs from HY, YC and RH exceeded the control standard and cannot be directly used for agricultural utilization. PAHs mainly contain 4~6 rings species, while 2~3 rings species occur rarely. The source tracing results showed that vehicle exhaust, industrial sources, coal and wood combustion are the main sources of PAHs in sludge. The ecological risk assessment showed that the toxic equivalent concentration TEQ_{BaP16} range from 75.5 to 760.2 μg/kg and $TEQ_{BaPcancer}$ range from 74.6 to 758.5 μg/kg. $TEQ_{BaPcancer}$ accounts for up to 98% of TEQ_{BaP16}, suggesting that carcinogenic PAHs species are the main contributors to the risk of PAHs. TEQ_{BaP10} in this study was almost 8 times higher than that specified value in the agricultural soil standard. The result of risk assessment suggests that PAHs in sludge of Xuchang City pose serious ecological and carcinogenic risk. Therefore, sludge must be conducted harmless treatment before agricultural utilization.
Keywords: *BaP, toxic equivalent factor, carcinogenic effects, sewage sludge, PAHs*

Introduction

Sludge is an inevitable solid by-product of sewage treatment, which contains many different types of hazardous substances, including heavy metals and pathogenic bacteria, organic pollutants and could release a foul odor (Xue et al., 2019; Liu et al., 2022). The production of sludge in China was over 80 million tons in 2018 (Hu et al., 2020), safe and effective disposal of such a large amount of waste is needed to prevent them from harming environment and human health (Bian et al., 2021). At present, the treatment methods of sludge mainly include landfill, incineration and soil utilization (Liu et al., 2021). The landfills have been completely banned in China, the incineration may produce fly ash, dioxins and other substances that may cause secondary pollution. On the other hand, sludge contains numerous nitrogen, phosphorus, kalium and organic matter, which are also considered as a renewable energy sources (Fathy et al., 2023). Therefore, the soil utilization was encouraged to be a great potential disposal methods of sludge (Vrom, 1994; Han et al., 2019; Tomczyk et al., 2020; Li et al., 2021). However, due to stringent environmental protection standards, the pollutants that contains in sludge should be fully recognized and assessed before its agricultural utilization (Man et al., 2017).

Among various organic pollutants in sludge, polycyclic aromatic hydrocarbons (PAHs) are the most widely distributed persistent organic pollutants (Cheng et al., 2007), PAHs reach the wastewater treatment plants by sewage systems or wet-dry deposition from the atmosphere (Manoli and Samara, 1999; Stevens et al., 2003; Blanchard et al., 2004; Chen et al., 2022). PAHs are easily enriched in sludge because of their hydrophobic and lipophilic. At present, 16 PAHs species have been listed as priority pollutants by the U.S. Environmental Protection Agency, the PAHs content of sludge in different cities varies greatly, generally ranging from 1000-30000 μg/kg (Paulsrud et al., 2000; Zhang et al., 2008). Some PAHs species have strong carcinogenicity and mutagenicity, and are easy to be bioaccumulated (Mojiri et al., 2019). Therefore, the research on the concentration of PAHs, ecological risk and how to removal of PAHs from sludge has received considerable attention (Cai et al., 2007; Guo et al., 2019; Zhang et al., 2019; Gencosman et al., 2021).

In order to gain a comprehensive understanding of PAHs characteristics in sludge of Xuchang City, sludge samples were collected from 10 large-scale sewage plants, the concentration, sources and risks assessment were analyzed and evaluated, the objective of this study was to provide reference basis for reasonable disposal and resource utilization of sludge in Xuchang city.

Materials and methods

Study area

Xuchang city (113°03′~114°19′E, 33°16′~34°24′N) is located at the central part of China, and has a temperate monsoonal climate, with an annual average temperature of 14.6 ℃ and an average annual precipitation of 727 mm. The geomorphic type is mainly alluvial plains, soil is composed of alluvium material that riched in nutrients and organic matter. Such superior natural conditions make Xuchang city an important grain-producing area, particularly of high-quality wheat and corn in China. Therefore, the quality of Xuchang’s cultivated land is crucial to ensure food security. In recent years, the sludge production of Xuchang city has increased year by year, which reached 177400 tons in 2020 (Xuchang Ecological Environment Bureau, 2021). Xuchang city is also facing the safety utilization problem of sludge.

Sludge sample collection

Dewatered sludge samples were collected from 10 major wastewater treatment plants (WWTPs) in Xuchang City, labelled RBK, HY, TN, QY, RH, ZH, YWH, YH, YC and YHB, respectively (*Fig. 1*). Each sample is about 2 kg. The sample is sealed in a polythene bag and brought back to the laboratory. Sludge samples are pre frozen at -20℃ and vacuum freeze-dried at -50℃, and then screened at 100 mesh for standby.

Sample analysis

The pressurized fluid extraction method was used for sample extraction. About 1.00 g sludge sample that add appropriate amount of diatomite was stirred evenly and ground into fine particles, then transferred into the pressurized fluid extraction tank, decafluorobiphenyl was added before extraction. After the sample was concentrated to

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

about 1 mL, 5 mL of n-hexane was added and concentrated to about 1 mL. This process was repeated for 3 times to convert the solvent to n-hexane. The solid-phase extraction column (filled with magnesium silicate) was used as the purification column, and fixed on the solid-phase extraction device, the purification column was washed by 4 mL dichloromethane, and balanced for 5 min with 10 mL n-hexane. The effluent was discard, and the concentrated sample was transferred into the column before the solution dries. The concentration vessel was washed by 3 mL n-hexane for 3 times, and the washing solution transferred into the column. The column was eluted with 10 mL dichloromethane-n- hexane mixture for 5 min, and the eluent was concentrated to about 1 mL, then 3 mL acetonitrile was added and concentrated to less than 1 mL to convert the solvent to acetonitrile, finally, fix the volume to 1 mL and sealed for analysis. Standards are the EPA 610 PAHs mix, which contains 16 species of PAHs. They are naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluor), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF) benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DbahA), benzo[ghi]perylene (BghiP) and indeno[1,2,3-cd] pyrene (IP). The concentration of PAHs was determined by Agilent 1260 infinity II high performance liquid chromatograph, which equipped with UV and fluorescene detector. The chromatographic column is Zorbax eclipse PAH chromatographic column, which with particle size of 5 μm, column length of 250 mm and inner diameter of 4.6 mm. The column flow rate is 1 mL / min and temperature is 35℃, and the injection volume is 10 μL.

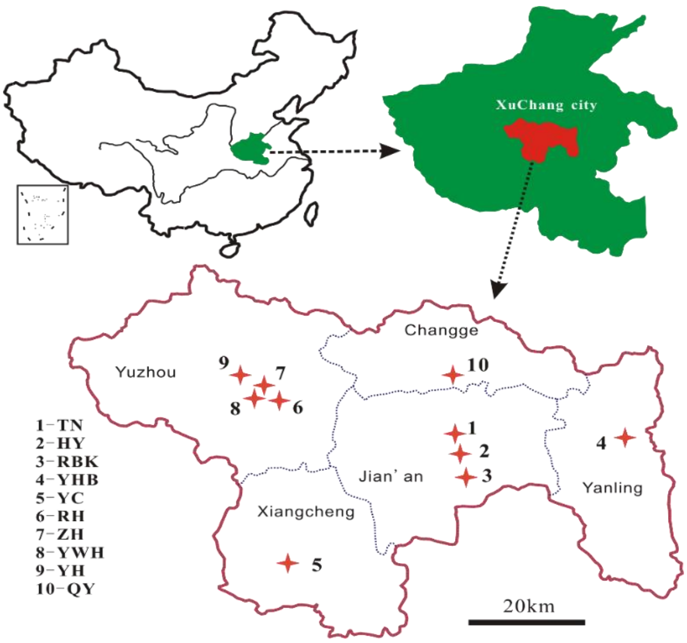

***Figure 1.** Location map of the 10 sampling WWTPS*

The procedure described above was checked by blank analyses, replicate analyses and spiked recovery rate. No PAHs species were detected in the blank experiment (the blank sample was prepared in the same steps that quartz sand was used to replace the sludge). The relative deviation of replicated experimental data was ±15%, and the recovery rates ranged from 75 to 119%.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

Risk assessment of PAHs in sewage sludge

Risk of PAHs in sludge was estimated by calculated total BaP toxic equivalent concentration (TEQ_{BaP16}), as BaP is considered as a good index for total PAH carcinogenicity. In order to calculate the TEQ_{BaP} for each individual PAH species, it requires the use of the toxic equivalent factor.

The following formula is used to calculate BaP toxic equivalent concentration:

$$TEQ_{BaP16} = \sum PAHs_i \times TEF_i \qquad \text{(Eq.1)}$$

where i represent each individual PAH species, TEF_i is the i species relative to BaP carcinogenic potency (Tsai et al., 2004).

Results

Concentration of PAHs in sewage sludge in Xuchang City

The ranges of total concentrations of PAHs were from 1835 to 7188 μg/kg, with a mean value of 4362 μg/kg (*Table 1*). The lowest and highest level of total PAHs was found in the sample collected from YWH (1835 μg/kg) and RH (7188 μg/kg), respectively (*Table 1*). The ranks of total concentrations of PAHs from the 10 WWTPS ranked in the order of RH > HY > YC > QY > TN > ZH > YHB > RBK > YH > YWH > YWH.

Table 1. *Concentrations of PAHs in the sewage sludge from 10 WWTPS (μg/kg)*

PAHs	TN	HY	RBK	YHB	YC	RH	ZH	YWH	YH	QY
Nap	ND	59.4	ND	ND	ND	ND	ND	ND	ND	ND
Acy	ND	ND	ND	549	608	627	ND	ND	ND	240
Flu	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Ace	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Phe	372	279	234	77	380	359	325	177	131	270
Ant	ND	179	ND	ND	ND	ND	ND	ND	ND	ND
Fluor	1469	1987	1313	306	1098	187	1045	771	299	278
Pyr	ND	217	ND	ND	ND	479	ND	ND	ND	ND
Chry*	198	110	229	128	372	411	150	157	ND	829
BaA*	106	ND	78.5	51	192	57.8	ND	ND	ND	178
BbF*	378	1411	434	410	413	998	191	324	ND	684
BkF*	422	1281	874	2289	1818	3425	2384	406	1455	763
BaP*	89.9	ND	ND	ND	256	175	ND	ND	ND	414
BghiP*	773	793	246	ND	456	232	ND	ND	ND	646
IP*	218	ND	96.8	ND	137	120	ND	ND	ND	259
DbahA*	212	59.9	79.5	ND	138	117	ND	ND	ND	184
∑PAHs	4238	6376	3585	3810	5868	7188	4095	1835	1885	4745
∑PAHscancer	2397	3655	2038	2878	3782	5536	2725	887	1455	3957

ND represent under detection limit; * indicates a carcinogenic species

The total concentrations of 8 carcinogenic PAHs species were from 887 to 5536 μg/kg, with a mean value of 2931 μg/kg. The percentage of 8 carcinogenic PAHs species in total PAHs were from 48 to 83%, with a mean value of 66%, which may indicate that there is a certain risk of carcinogenesis in Xuchang city’s sewage sludge.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

Proportion of PAHs components with different rings

Different PAHs components have different toxicity and carcinogenic effects. Generally, low ring PAHs, such as 2- and 3-rings have strong acute toxicity, while high ring PAHs, such as 4-, 5-, 6-rings have severe carcinogenicity and mutagenicity, and easy to be bioaccumulated (Cheng et al., 2007). Proportion of PAHs species with different rings are presented in *Figure 2*. It was found that in this study, the percent of 2-, 3-rings PAHs were from 7 to 17%, with a mean value of 11%, the percent of 4-, 5-, 6-rings PAHs were from 83 to 93%, with an average value of 89%. This indicated that high rings PAHs species are the main components. This is mainly attributed to the sources and treatment process of sewage from different WWTPS and may also be partly related to the volatility of components. 2- and 3-rings PAHs species with small molecular weight mainly exist in gaseous form and are easy to volatilize and decompose, while 4-, 5-, 6-rings PAHs species with large molecular weight mainly exist in granular form and are not easy to migrate, it can also be seen from the lower detection rate of 2- and 3-rings PAHs than that of high rings.

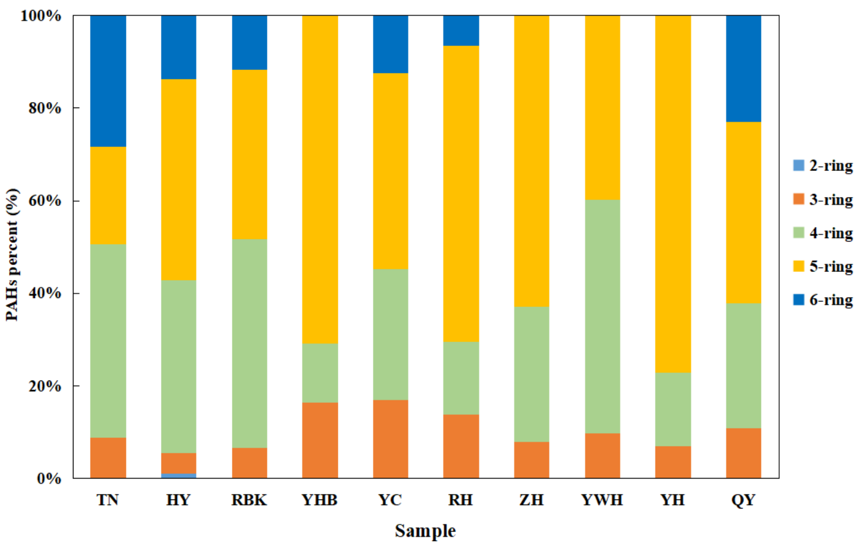

Figure 2. *Percentage PAHs of different rings species in sludge*

Discussion

Concentrations of PAHs in sewage sludge from 10 WWTPS

As can be seen from *Table 1*, the ranges of total concentrations of PAHs were from 1835 to 7188 μg/kg, with an average value of 4362 μg/kg, which is lower than that in Beijing, Wuxi and Paris, but higher than that in Guangzhou, which is basically equivalent to that in domestic cities, such as Dalian, Shanghai, Qingdao and Guiyang (*Table 2*). The average concentration of PAHs in sludge of Xuchang city is lower than control standards of pollutants in sludge for agricultural use of China. The standard stipulates that the total concentrations of PAHs of Class A sludge and Class B sludge should be less than 5000 μg/kg and 6000 μg/kg, respectively. Class A sludge refers to the sludge that can be used as farmland, garden and grassland, and Class B sludge can be used as garden, grassland and farmland without edible crops. On the average, it seems that the sludge in Xuchang city can be directly used as agricultural fertilizer, but the total concentrations of

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

PAHs of HY, YC and RH sewage treatment plants exceed 5000 μg/kg, RH even exceeds 6000 μg/kg, which means that the sludge of those plants cannot be directly used for agriculture.

Table 2. *PAHs concentrations in sludge at home and abroad (μg/kg)*

City	Concentrations	Reference
Beijing	2470 ~ 25900	Dai et al., 2007
Wuxi	12180 ~ 31780	Zhang et al., 2008
Paris	14000 ~ 31000	Blanchard et a., 2004
Guangzhou	1110 ~ 1570	Yu et al., 2013
Dalian	2080 ~ 5380	Yu et al., 2013
Shanghai	1156 ~ 6803	Fang et al., 2008
Qingdao	1970 ~ 6580	Zhai et al., 2011
Guiyang	1899 ~ 4265	Yang et al., 2015
Xuchang	1835 ~ 7188	This study

There are remarkable differences in total concentrations of PAHs among sewage treatment plants, the highest value that collected from RH (7188 μg/kg) almost 4 times higher than the lowest value that collected from YWH (1835 μg/kg). This difference is mainly attributed to the sources and treatment process of sewage of different WWTPs. The 10 WWTPs in this study mainly uses same treatment, which is anaerobic anaerobic oxidizing (A2/O) system and Orbal oxidation ditch process. Therefore, the difference levels in PAHs concentration caused by sewage treatment process is very small, mainly due to the source of sewage. It is found that the total concentrations of PAHs in sludge from TN, HY, YC, RH, ZH and QY, which are mainly treating industrial sewage are significantly higher than that from RBK, YHB, YWH, YH, which are mainly treating domestic sewage.

RH and YC are located in Yuzhou county and Xiangcheng county, respectively. These counties are rich in coal resources. The incomplete combustion of coal and other mineral resources are the main sources of PAHs in the environment, which led to the highest PAHs content in RH and YC sludge. The wastewater treated by TN and HY are mainly the wastewater of wig industry. The dyestuff and colourant, which enriched in PAHs will be used during decolorization, dyeing and washing of wig, which should be the reason for the high content of PAHs in TN and HY sludge. The PAHs content of RBK, YHB, YWH and YH are relatively lower since these WWTPs mainly deals with domestic sewage.

Sources of PAHs in sewage sludge in Xuchang City

The sources of PAHs in sludge are complex. The relative abundance of different ring PAHs is usually used to infer whether they come from combustion or oil pollution. 4~6 rings PAHs (high molecular weight, HMW) mainly come from high-temperature combustion of fossil fuels, and 2~3 rings PAHs (low molecular weight, LMW) come from oil pollution (Fernandes et al., 1997). When LMW/HMW>1, PAHs mainly come from oil pollution, while LMW/HMW<1, PAHs mainly come from high temperature combustion of fossil fuels (Budzinski et al., 1997). In this study, LMW/HMW range from 0.07 to 0.20, indicated that PAHs mainly come from fossil fuel combustion.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

The ratios of individual PAHs species, such as BghiP/BaP, IP/(IP+BghiP), BaA/(BaA+Chry), Fluor/(Fluor+Pry) are also frequently employed as diagnostic tools to identify the origin of PAHs (Yunker et al., 2002; Cheng et al., 2007; Manoli and Samara, 2008). The diagnostic ratios calculated in this study were compared with those potentia source emissions (*Table 3*). The mean value of BghiP/BaP (1.50) indicates PAHs in this study mainly from vehicle exhaust and coal combustion, while vehicle exhaust are considered to be main sources from the mean value of IP/(IP+BghiP) (0.45) and BaA/(BaA+Chry) (0.40). Nevertheless, the mean value of Fluor/(Fluor + Pry) (0.83) indicates PAHs from coal and wood combustion. It can be seen that the results of source judgment are different when using different diagnostic ratios. In fact, the diagnostic ratios are often difficult to discriminate between sources as the value of different sources are often overlap. Moreover, the diagnostic ratios can be altered due to the reactivity of some PAHs species under certain environmental conditions and media.

Table 3. *Diagnostic ratios and potential source emissions*

	Vehicle exhaust	**Coal combustion**	**Industrial sources**	**Wood combustion**	**This study**
BghiP/BaP	1.2-3.3	0.15-1.11	0.02-0.06		1.50
IP/(IP+BghiP)	0.21-0.70		0.36-0.57	0.62	0.45
BaA/(BaA+Chry)	0.22-0.64	0.50-0.55	0.23-0.89	0.43	0.40
Fluor/(Fluor+Pry)	0.4-0.5	>0.5		>0.5	0.83

Data after Manoli and Samara (2008) and Yunker et al. (2002)

However, despite these shortcomings, the main sources can still be determined by application of multiple ratios. Double diagnostic ratio was used to determine the source more reliable (*Fig. 3*). As can be seen from *Figure 3*, Fluor/(Fluor+Pry) of sludge of 9 WWTPs is over 0.5, only RH is less than 0.4 (0.28), IP/(IP+BghiP) of sludge of 5 WWTPs is between 0.2 and 0.4, and other 5 WWTPs of IP/(IP+BghiP) is 0 (IP+ BGHIP) (IP was under the detected limit value). In summary, coal and wood combustion, oil combustion are the mainly sources of PAHs in Xuchang sludge.

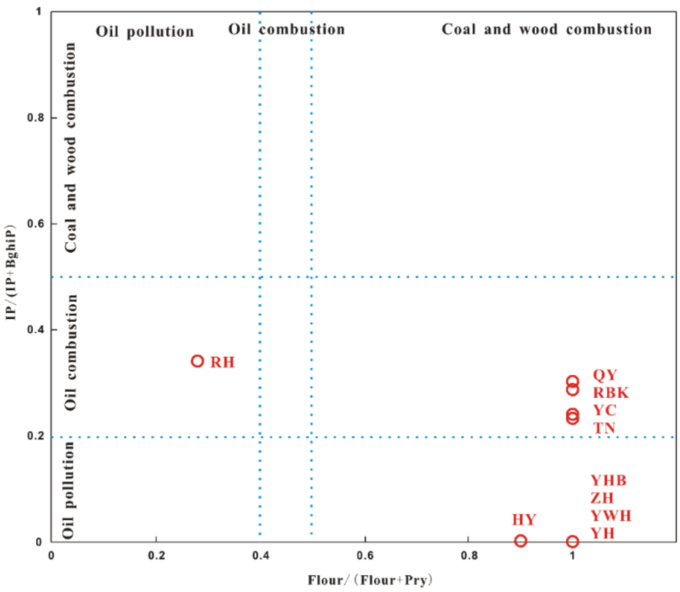

Figure 3. *Double diagnostic ratio of PAHs in sludge from 10 WWTPS*

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

Risk assessment of PAHs in sewage sludge in Xuchang City

BaP is considered as a good index for total PAH carcinogenicity. However, BaP is easily degraded in the presence of sunlight and oxidants and has a very short half-life (Mastral et al., 2003). Due to degradation, BaP concentration alone does not give a good indication of the hazard posed by all the PAHs (WHO/IPCS, 1998). In this study, the detection rate of BaP was only 40%, the PAHs' carcinogenic character could be underestimated if this compound alone is taken as the representative of carcinogenicity.

In this study, risk of PAHs in sludge was estimated by calculated total BaP toxic equivalent concentration (TEQ_{BaP16}). In order to calculate the TEQ_{BaP} for each individual PAH species, it requires the use of the toxic equivalent factor (TEF) of the given species relative to BaP carcinogenic potency (*Table 4*) (Tsai et al., 2004). The *formula (1)* was used to calculate BaP toxic equivalent concentration.

Table 4. *The toxic equivalent factor (TEF) of PAH species*

PAHs	TEF	PAHs	TEF	PAHs	TEF	PAHs	TEF
Nap	0.001	Phe	0.001	Chry	0.01	BaP	1
Acy	0.001	Ant	0.01	BaA	0.1	BghiP	0.01
Flu	0.001	Fluor	0.001	BbF	0.1	IP	0.1
Ace	0.001	Pyr	0.001	BkF	0.1	DbahA	1

The values of TEQ_{BaP16} in this study range from 75.5 to 760.2 μg/kg, with a mean value of 398.4 μg/kg (*Figure 3*). The rank of 10 WWTPS were QY>RH>YC>TN>HY>YHB>ZH>RBK>YH>YWH. $TEQ_{BaPcancer}$ represent toxic equivalent concentration of 8 carcinogenic PAHs species, which were from 74.6 to 758.5 μg/kg, with a mean value of 396.8 μg/kg, $TEQ_{BaPcancer}$ accounts for up to 98% of TEQ_{BaP16}, indicated that 8 carcinogenic PAHs species are the biggest contributor to the risk of PAHs (*Fig. 4*).

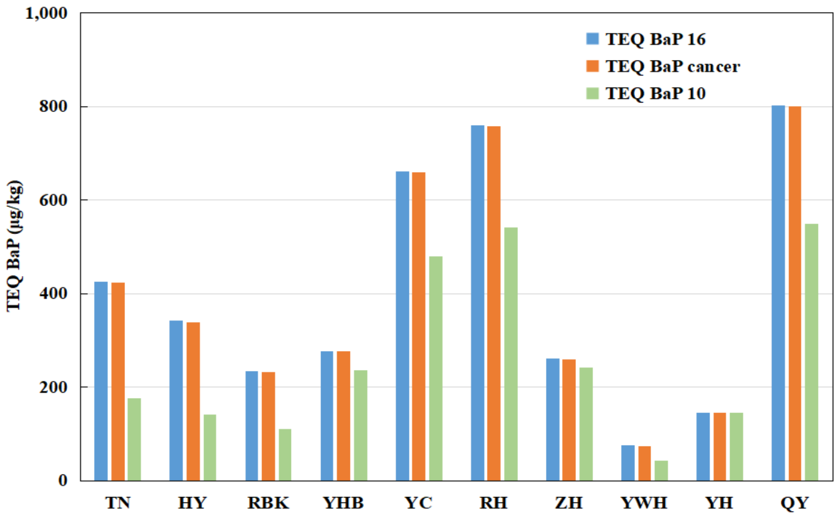

Figure 4. *$TEQ_{BaP\ 16}$, $TEQ_{BaP\ cancer}$ and $TEQ_{BaP\ 10}$ of PAHs in sludge from 10 WWTPS*

Considering that there is no recognized evaluation standards for the ecological risk of PAHs in sludge, TEQ_{BaP10} standard were used here for risk assessment (Vrom, 1994). TEQ_{BaP10} represent toxic equivalent concentration of 10 PAHs species (Nap, Phe, Ant,

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

Flour, BaA, Chry, BkF, BaP, IP and BghiP) defined in Dutch agricultural soil quality standard. In this study, TEQ_{BaP10} were from 43.1 to 549.3 μg/kg, with a mean value of 266.7 μg/kg, the average value of TEQ_{BaP10} almost 8 times higher than the agricultural soil standard (33 μg/kg) (*Fig. 3*) (Vrom, 1994). The result of risk assessment suggests that PAHs in sludge of Xuchang City have serious ecological and carcinogenic risk. Therefore, the quantity of sludge in agricultural utilization must be controlled or conduct harmless treatment before used.

Conclusions

The total concentration of PAHs in Xuchang's sludge from 1835 to 7188 μg/kg, with a mean value of 4362 μg/kg, the average value of PAHs concentration met the limit of China's agricultural sludge control standard. However, PAHs concentrations from HY, YC and RH exceeded the control standard, which means that the sludge from those plants cannot be directly used for agricultural utilization.

The source tracing results showed that vehicle exhaust, industrial sources, coal and wood combustion are the main sources of PAHs in Xuchang sludge. The ecological risk assessment showed that the toxic equivalent concentration TEQ_{BaP16} and $TEQ_{BaPcancer}$ range from 75.5 to 760.2 μg/kg and 74.6 to 758.5 μg/kg, respectively. $TEQ_{BaPcancer}$ accounts for up to 98% of TEQ_{BaP16}, suggest that carcinogenic PAHs species are the main contributors to the risk of PAHs. TEQ_{BaP10} almost 8 times higher than that specified value in the relevant agricultural soil standard. The result of risk assessment suggests that PAHs in sludge of Xuchang City have serious ecological and carcinogenic risk. Therefore, sludge must be conducted harmless treatment before agricultural utilization.

Acknowledgments. The authors would like to express their grateful thanks to Dr Chen Mi for sample preparation and laboratory assistance. This work was funded by the National Natural Science Foundation of China (42173062).

REFERENCES

[1] Bian, C., Ge, D., Wang, G., Dong, Y., Yuan, H. (2021): Enhancement of waste activated sludge dewaterability by ultrasound-activated persulfate oxidation: operation condition, sludge properties mechanisms. – Chemosphere 262: 128385.

[2] Blanchard, M., Teil, M. J., Ollivon, D., Legenti, J., Chevreuil, M. (2004): Polycyclic aromatic hydrocarbons polychlorobiphenyls in wastewaters sewage sludge from the Paris area (France). – Enviro Res. 95: 184-197.

[3] Budzinski, H., Jones, I., Bellocq, J., Piérard, C., Garrigues, P. H. (1997): Evaluation of sediment contamination by polycyclic aromatic hydrocarbons in the Gironde Estuary. – Mar Chem. 58: 85-97.

[4] Cai, Q. Y., Mo, C. H., Wu, Q. T., Zeng, Q. Y., Katsoyiannis, A., Férard, J. F. (2007): Bioremediation of polycyclic aromatic hydrocarbons (PAHs) contaminated sewage sludge by different composting processes. – J Hazard Mater 142: 535-542.

[5] Chen, W., Peng, B., Huang, H., Kuang, Y., Qian, Z., Zhu, W., Liu, W., Zhang, Y., Liao, Y., Zhao, X. (2022): Distribution Potential Sources of OCPs PAHs in Waters from the Danshui River Basin in Yichang, China. – Int. J. Environ. Res. Public Health. 19: 263. https://doi.org/10.3390/ijerph19010263.

[6] Cheng, J., Yuan, T., Qian, W., Zhao, W., Xie, H., Ma, Y., Ma, J., Wang, W. H. (2007): PM10-bound polycyclic aromatic hydrocarbons (PAHs) cancer risk estimation in the

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639

atmosphere surrounding an industrial area of shanghai, China. – Water Air and Soil Pollut. 183: 437-446.
[7] Dai, J. Y., Xu, M. Q., Chen, J., Yang, X. P., Ke, Z. S. (2007): PCDD/F, PAH heavy metals in the sewage sludge from six wastewater treatment plants in Beijing, China. – Chemosphere 66: 353-361.
[8] Fang, H. L., Chen, L., Peng, X. L., Zhou, X., Zhou, X., Lin, L., Nan, P. (2008): Distribution characteristics of polynuclear aromatic hydrocarbons in sludge from major municipal sewage treatment plants in Shanghai. – Acta Pedologica Sinica 45: 1164-1169. (In Chinese with English Abstract).
[9] Fathy, D., Wagreich, M., Fathi, E., Ahmed, M. S., Leila, M., Sami, M. (2023): Maastrichtian Anoxia and Its Influence on Organic Matter and Trace Metal Patterns in the Southern Tethys Realm of Egypt during Greenhouse Variability. – ACS Omega 22: 19603-19612.
[10] Fernes, M. B., Sicre, M. A., Boireau, A., Tronczynski, J. (1997): Polyaromatic hydrocarbon (PAH) distributions in the seine river its estuary. – Mar Pollut Bull. 34: 857-867.
[11] Gencosman, B. C., Sanli, G. E. (2021): Prediction of polycyclic aromatic hydrocarbons (pahs) removal from wastewater treatment sludge using machine learning methods. – Water, Air, and Soil Pollut. 232: 1-17.
[12] Guo, Y., Rene, E. R., Wang, J., Ma, W. (2020): Biodegradation of polyaromatic hydrocarbons the influence of environmental factors during the co-composting of sewage sludge green forest waste. – Bioresource Technol 297: 122434. https://doi.org/10.1016/j.biortech.2019.122434.
[13] Han, J., Liang, Y., Zhao, B., Wang, Y., Xing, F., Qin, L. (2019): Polycyclic aromatic hydrocarbon (PAHs) geographical distribution in China their source, risk assessment analysis. – Environ. Pollut. 251: 312-327.
[14] Hu, Y., Xia, Y., Maio, F. D., Yu, F., Yu, W. (2020): Investigation of polycyclic aromatic hydrocarbons (PAHs) formed in three-phase products from the pyrolysis of various wastewater sewage sludge. – J Hazard Mater. 389: 122045.
[15] Li, W., Zhu, N., Yuan, H., Shen, Y. (2021): Influence of sludge organic matter on elimination of polycyclic aromatic hydrocarbons (PAHs) from waste activated sludge by ozonation: controversy over aromatic compounds. – Sci. Total Environ. DOI:10.1016/j.scitotenv. 2021.149232.
[16] Liu, H. D., Xu, G. R., Li, G. B. (2021): Pyrolysis characteristic kinetic analysis of sewage sludge using model-free master plots methods. – Process Saf Environ. 149: 48-55.
[17] Liu, J., Lin, L., Wang, K., Ding, R., Xie, Z., Zhang, P. (2022): Concentrations Species of Mercury in Municipal Sludge of Selected Chinese Cities Potential Mercury Emissions from Sludge Treatment Disposal. – Front. Environ. Sci. 10: 895075. doi: 10.3389/fenvs. 2022.895075.
[18] Man, Y. B., Chow, K. L., Cheng, Z., Mo, W. Y., Chan, Y. H., Lam, J. C. W., Lau, F. T. K., Fung, W. C., Wong, M. H. (2017): Profiles removal efficiency of polycyclic aromatic hydrocarbons by two different types of sewage treatment plants in Hong Kong. – J. Environ. Sci. 53: 196-206. (In Chinese with English Abstract).
[19] Manoli, E., Samara, C. (1999): Occurrence Mass balance of polycyclic aromatic hydrocarbons in the Thessaloniki sewage treatment plant. – J Environ Qual. 28: 176-187.
[20] Manoli, E., Samara, C. (2008): The removal of polycyclic aromatic hydrocarbons in the wastewater treatment process: experimental calculations model predictions. – Environ Pollut. 151: 477-485.
[21] Mastral, A. M., López, J. M., Callén, M. S., García, T., Murillo, R., Navarro, M. V. (2003): Spatial temporal PAH concentrations in Zaragoza, Spain. – Sc. Total Environ. 307: 111-124.
[22] Mojiri, A., Zhou, J. L., Ohashi, A., Ozaki, N., Kindaichi, T. (2019): Comprehensive review of polycyclic aromatic hydrocarbons in water sources, their effects treatments. – Sci. Total Environ. https://doi.org/10.1016 /j.scitotenv.2019.133971.

[23] Paulsrud, B., Wien, A., Nedland, K. T. (2000): A survey of toxic organics in Norwegian sewage sludge, compost and manure. – Norwegian Water Technology Centre ASOSLO: 51-60.

[24] Stevens, J. L., Northcott, G. L., Stern, G. A., Tomy, G. T., Jones, K. C. (2003): PAHs, PCBs, PCNs, organochlorine pesticides, synthetic musks, polychlorinated n-alkanes in U.K. sewage sludge: survey results implications. – Environ Sci Technol. 37: 462-467.

[25] Tomczyk, B., Siatecka, A., Jdruchniewicz, K., Sochacka, A., Oleszczuk, P. (2020): Polycyclic aromatic hydrocarbons (PAHs) persistence, bioavailability toxicity in sewage sludge- or sewage sludge-derived biochar-amended soil. – Sci Total Environ 747: 141123.

[26] Tsai, P. J., Shih, T. S., Chen, H. L., Lee, W. J., Lai, C. H., Liou, S. H. (2004): Assessing predicting the exposures of polycyclic aromatic hydrocarbons (PAHs) their carcinogenic potencies from vehicle engine exhausts to highway toll station workers. – Atmos Environ. 38: 333-343.

[27] Vrom. (1994): Environmental quality objectives in the Netherlands: A review of Environmental quality objectives their policy framework in the Netherlands. – The Hague: Ministry of Housing, Spatial Planning Environment.

[28] WHO/IPCS. (1998): Environmental Health Criteria 202: selected non-heterocyclic polycyclic aromatic hydrocarbons. – WHO, Geneva.

[29] Xuchang Ecological Environment Bureau. (2021): Bulletin of environmental prevention control of solid waste pollution in Xuchang City in 2020.

[30] Xue, Y., Wang, C., Hu, Z., Zhou, Y., Xiao, Y., Wang, T. (2019): Pyrolysis of sewage sludge by electromagnetic induction: biochar properties application in adsorption removal of Pb (II), Cd (II) from aqueous solution. – Waste Manage. 89: 48-56.

[31] Yang, S. B., Li, J., Zhang, C. H., Wu, Y. G., Yan, C. L. (2015): Distribution, sources risk assessment of polycyclic aromatic hydrocarbons (PAHs) in sewage sludge from Guiyang City. – Chinese J. Ecol. 34: 1675-1681. (In Chinese with English Abstract).

[32] Yu, Y. X., Chen, H., Wang, X. M., Han, J. B., Tao, P. (2013): Pollution characteristics of organic contaminant in sludge from wastewater treatment plants sludge ocean dumping disposal in China. – Mar Environ Sci. 32: 652-656. (In Chinese with English Abstract).

[33] Yunker, M. B., Macdonald, R. W., Vingarzan, R., Mitchell, R. H., Goyette, D., Sylvestre, S. (2002): PAHs in the Fraser river basin: Acritical appraisal of PAHs ratios as indicators of PAH source composition. – Org. Geochem. 33: 489-515.

[34] Zhai, J., Tian, W., Liu, K. (2011): Quantitative assessment of polycyclic aromatic hydrocarbons in sewage sludge from wastewater treatment plants in Qingdao, China. – Environ Monit Assess. 180: 303-311.

[35] Zhang, X. Y., Zhou, L. X., Cui, C. H., Zheng, X. X. (2008): Content and its major influencing factors of Polycyclic Aromatic Hydrocarbons (PAHs) in Municipal Sewage Sludge of Jiangsu Province. – Environ Sci. 29: 2271-2276. (In Chinese with English Abstract).

[36] Zhang, X., Yu, T., Li, X., Yao, J., Liu, W., Chang, S., Chen, Y. (2019): The fate enhanced removal of polycyclic aromatic hydrocarbons in wastewater sludge treatment system: a review. – Critical Reviews in Environm. Sci. Technol. 49: 1425-1475.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/2204_36293639