The Distribution and Temporal Change of Mercury in a Sewage Treatment Plant

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Abstract — In order to understand the distribution characteristics and temporal change of mercury species in a sewage treatment plant,sewage and sludge samples were collected from influent and effluent of the Second Sewage Treatment Plant of Jiaozuo for 10 months.Atomic Fluorescence method was used to detect mercury.Total mercury (THg) and methylmercury (MeHg) were found the average concentrations of 2.00×103ng/L and 16.2ng/L.THg in grit chamber mainly exists with granule state.THg were positively correlated to TSS (P=0.003). MeHg and dissolved MeHg of influent were found at average concentrations of 7.5ng/L and 0.59ng/L.Inffluent THg and MeHg concentrations have correlation with TSS concentrations (P=0.003, P=0.035).Dissolved MeHg was increases with the temperature rising. THg and Dissolved THg of effluent were found at the average concentrations of 10.6ng/L and 2.2ng/L. Mean daily removals of THg and dissolved THg at the plant were 98.9% and 65%. The ratio (Dissolved THg/THg) has increased in effluent.MeHg and dissolved MeHg of effluent were found that the average concentrations of 0.33ng/L and 0.1ng/L. THg and MeHg contents of the effluent met the requirement of "Urban sewage treatment plant pollutant discharge standard" (GB18918-2002). Mean daily removals of MeHg and dissolved MeHg were 93.5% and 70.7%. MeHg removal efficiency is higher than dissolved MeHg removal efficiency and the MeHg removal efficiency is lower than the THg removal efficiency.The pressure-filter sludge ((3.94±0.53) ×103ng/g) was greater than the national standard for soil environmental quality of grade iii.The average concentrations of MeHg was 6.35±0.84ng/g.The ratio of MeHg accounted for THg was minimal(less than 1%). Using the Index of geoaccumulation and potential ecological risk index method to analysis THg in pressure-filter sludge.The result of indicated that THg in pressure-filter sludge have high potential ecological risk.Thus the sludge might not be applied to farm land after suitable treatments.

Keywords-THg;Dissolved THg;MeHg;Dissolved MeHg; Pressure-filter sludge

I. INTRODUCTION

Mercury is one of the highly toxic heavy metal elements that cause environmental pollution.In the world,more than 10000 tons of mercury is mined every year,and about 5000 tons were lost[1],which has leaded the content of mercury in the ambient medium to nearly three times more than the industrial revolution,and caused serious Environment pollution.The inorganic mercury in the environment,through the action of biological and non-biological methylation,can thansform into more poisonous methyl mercury.It would cause harm to human health through the food chain enter human body[2].

With the speeding up of urbanization process and the ability of sewage treatment improving, urban sewage treatment plants become the main collection of people production and living pollutants.By the end of 2013,sewage treatment capacity is about 1.47 × 108 m3/day, processing power is about 6.8×106 m3/day more than the end of 2012[3].According to statistics,the process of urban sewage treatment would produce city sludge,whose volume was 0.02% of the whole volume of sewage[4]. In our country, by the end of September 2011,the annual production of wet discharge sludge (moisture content 80%) has reached 34.8 million tons [5].

Domestic and International studies have found that inorganic and methyl mercury may generally exists in the urban sewage,the processing of sewage treatment plant effluent and sludge in the city.At present a domestic study of mercury content in municipal sewage of Beijing found that the total mercury levels between 0.21～2.23μg/L[6].The research of sludge dewatering of Jianxi district sewage treatment plant by Ge Xiaoyan[7].They found that the mean concent of sludge samples of mercury (6.57 ug/g) is higher than the national average (3.18 ug/g). MeHg is very limited to study each operating unit of urban sewage treatment plants and sludge in China.The research of Gilmour and Bloom for a sewage treatment plant in Pennsylvania shown that the contents of THg in the wastewater was 156g/day, after processing, about 95% of the THg exists in sludge, only 6～7g/day THg into the receiving waters.At the same time, the secondary treatment effluent and sludge were

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detected in MeHg,the concentration of them were 1.01～3.15 ng/L,4.03 ～ 5.69 ng/L,1.6 ～ 5.2ng/g[8].The analyzation of three life sewage treatment plant of Canada from Bodaly,the concentration of THg in sewage for 2～160 ng/L, MeHg concentration was 0.5～4.3ng/L,the THg concentration of procesed sewage reduced to 3～14 ng/L,MeHg concentration reduced to 0.1 ～ 0.4 ng/L.The temperature has an effect on concentration of MeHg[9].In view of this,Sewage and sludge of the second sewage treatment plant of Jiaozuo were researched,studied the distribution characteristics and temporal change of mercury species in a sewage treatment plant. The results of the study for the prevention of urban sewage in the inorganic mercury and MeHg pollution,assessment of sewage and sludge discharge and disposal of the environmental and for health risks provide scientific basis.

II. MATERIAL AND METHODS

A. Sample collection and pretreatment

The second sewage treatment plant of Jiaozuo where is located in the eastern of Jiaozuo Harvest Road,it has a surface area of 8.09×104 m2.it provides high quality treatment of wastewater for 300,000 people and received most of domestic wastewater from the east of Shanyang district, Macun district, JiaoNan new district and the south of Shahe district.In addition to open channel(Wengjian River and Li River.) input.One part of processed sewage was reused in the plant,the rest of the processed sewage released into the New River. The second sewage treatment plant of Jiaozuo has used improved oxidation ditch process.The whole process system is composed of four parts:pretreatment system,biochemical treatment system.sludge treatment system and disinfection system(Fig .1-1.).

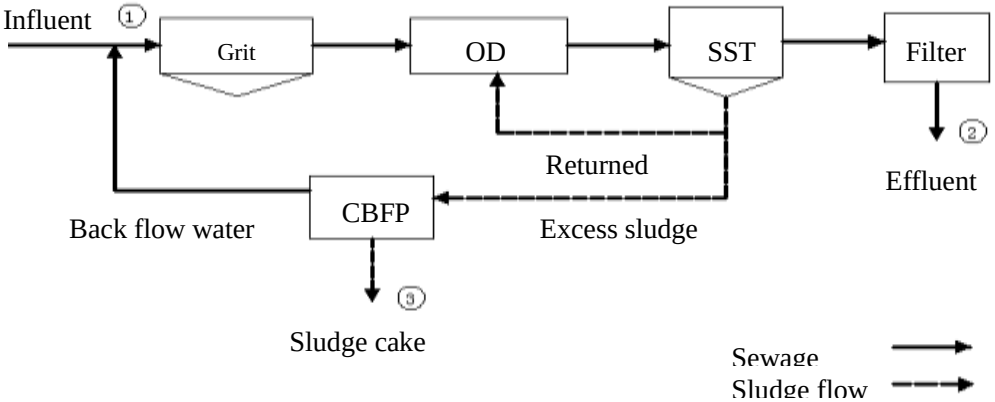

Figure1-1.Schematic Diagram of the Second Sewage Treatment Palant of Jiaozuo

Whole water grab samples were collected in raw sewage at the influent ① , the effluent ② and the press filter sludge ③ for THg and MeHg analysis in the Sewage treatment plant for ten months(November,2014～October,2015, Do not include January and February,2015).Samples were collected each month 10 between 8:00 AM to 18:00 PM and collected every two hours.All samples were collected in acid-washed Teflon bottles,double bagged and put in clean cooler ice,transported them to the laboratory.THg and MeHg samples that were preserved by the addition of concentrated trace-metal grade HCl(5%) acid stored refrigerated,while those for MeHg analysis were frozen.Filtered and unfiltered samples were split for determination of THg and MeHg. Approximately 500 mL of each sample was filtered under vacuum using an acid-cleaned disposable filter unit (0.45μm, cellulose membrane).Sludge samples were freezed and dried,grinded and sieved through 100 mesh sieve, and then sealed cryopreservation.

B. The analysis method of the sample

1) The analysis method of THg

Samples were oxidized used BrCl to release Hg from particles and organic complexes,then pre-reduced using hydroxylamine hydrochloride,prior to quantification by tin chloride(SnCl2) reduction. Sewage Samples were analyzed using standard techniques for Hg analysis by cold vapor atomic fluorescence spectrometry (CVAFS).This method is outlined in USEPA Method 1631.Detection limit was 0.5ng/L[10].

Sewage sludge samples were analyzed for MeHg by DMA80 mercury analyzer.Before determined the standard and sample,using 100mL,0.5～1mol/L HNO3 to repeat analysis,until absorbance fluctuation was less than 0.0030 and ran the blank quartz plate.

2) *The analysis method of MeHg*

Sewage Samples were digested with KBr/H2SO4 solution(2mL) and CuSO4 solution(1mL),extracted with CH2Cl2 and reverse extracted in water with water bath(45℃)[11].After reverse extracted,samples were ethylated with sodium tetraethylborate solution,followed by purging of volatile methylethyl Hg onto Tenax carbotrap,and then separation by isothermal gas chromatography and CVAFS detecting MeHg.Detection limit was 0.06 ng/L.

Sludge MeHg sample detection method is the same with the method of detection for Sewage Samples.Detection limit was 0.12 ng/g.

Frequent calibration standard checks and procedural blank analyses helped assure analytical accuracy.Analytical results were as follows: ERM-CC580,certified value =132±3mg/kg(THg), certified value =75.5±4ug/kg(MeHg).Analytical accuracy was checked by multiple daily analyses of each of three different certified reference,mean RSD were 10%(THg) and 15%(MeHg).The average recovery of standard material is 83%～106% for the analysis of THg and MeHg.

THg contents in the grit chamber influent were analyzed(Fig .3-1.). It was found that the contents of THg ((0.23±0.02 ～ 9.73±0.82 ） ×103 ng/L) were highly variable,giving average concentrations of 2.00×103ng/L.THg concentrations in raw sewage varied widely,from2～150 ng/L[9].The THg concentrations in the second sewage treatment plant of Jiaozuo were far above THg concentrations in sewage treatment plant of Winnipeg.Main reasons for this difference may be due to the different inffluent qulity and the different groundwater seepage.Inffluent THg concentrations has positivly correlation with TSS concentrations

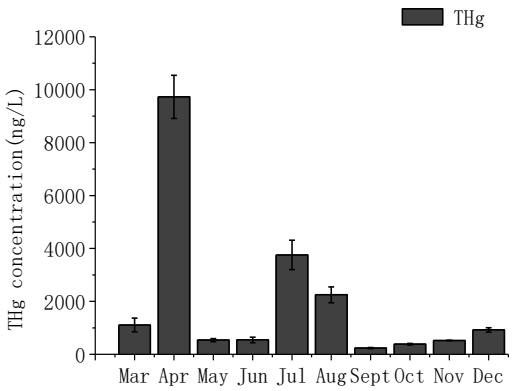

Figure 3-1 .Influent THg Concentration

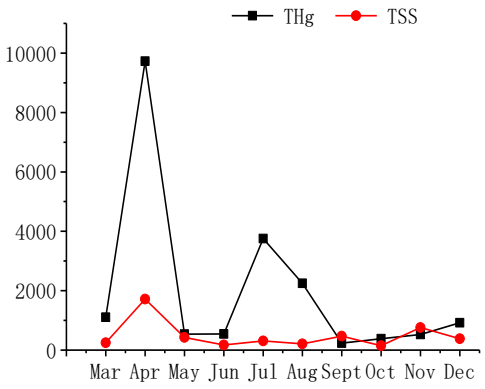

Figure 3-2 The Relationship Between Influent THg

III. RESULTS AND DISCUSSION

A. Variation of mercury in raw sewage influent

1) THg variation in raw sewage influent

It was found that the contents of dissolved THg （ 2.32±0.28 ～ 124±20.2ng/L ） were highly variable(Fig .3-3),giving average concentrations of 16.2ng/L. Dissolved THg gave the highest content in August and gave lowest content in November.The ratio(Dissolved THg/THg) was relatively low(0.04%～5.5%)(Fig .3-4),giving average concentrations of 1.2%.THg in grit chamber mainly exists with granule state.It was due to the suspended solids in the water and sediment has strong adsorption to the

(P=0.003)(Fig .3-2.).THg gave the highest concentration in April and gave lowest concentration in September(in grit chamber).So THg concentrations will be eventually rsduced with TSS removal in equipment.

mercury,leading to mercury in water migration to sediments.Concentrations of influent dissovled THg also spanned limited ranges 0.68–1.47 ng/L for dissolved THg from St.Paul Minnesota wastewater treatment plant[12].This result much less than what we determined dissolved THg contents,but giving average concentrations of 36% was higher than the ratio of dissolved THg in our sewage treatment plant.Main reasons for this difference may be due to the different influence sewage quality.

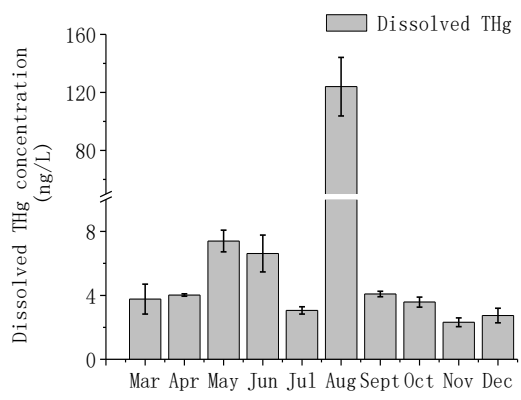

Figure3-3.Influent DissolvedTHg Concentration Variation in Grit Chamber

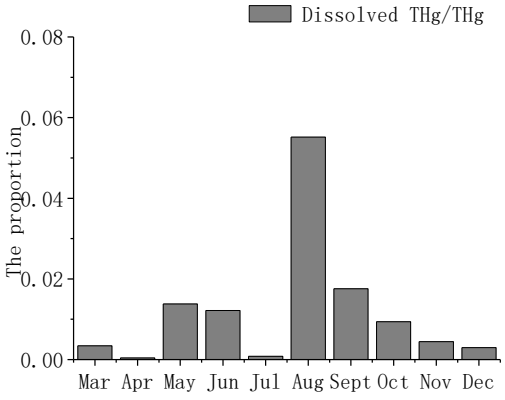

Figure3-4. The Rtio Dissolved THg Represented

2) MeHg variation in raw sewage influent

It was found that the contents of TMeHg（1.46±0～22.6±2.86ng/L ） were highly variable(Fig .3-5),giving average concentrations of 7.5ng/L.Inffluent TMeHg concentrations has correlation with TSS concentrations (P=0.032)(Fig .3-6). THg gave the highest content in April and gave lowest content in November in grit chamber sludge.It is simlar with the concentrations variation of MeHg in Onondaga County Metropolitan Wastewater Treatment Plant[13].

The contents of dissolved MeHg were 0.11±0.01～2.17±0.13ng/L,giving average concentrations of 0.59ng/L.The concentrations of dissolved TMeHg was higher during warmer months(may to August) than colder moths(September to December).MeHg is an anaerobic process and largely affected with sulfate reducing bacteria[14]. MeHg might increase with the temperature rising[15].Higher temperatures make it easier for MeHg in sewage pipe.It has the same phenomenon that the concentrations of influence MeHg increase with the temperature rising in a sewage treatment plant in New York[16].The ratio(Dissolved MeHg/MeHg) was 5%～28%,on average concentrations was 11.6%(Fig .3-6),it shows that MeHg in grit chamber mainly exists with granule state.The ratio (MeHg/Hg) was 0.23%～1.7%,on average concentrations was 11.6%(Fig .3-7), THg was the main form of mercury in grit chamber influente.It was found that the ratio(Dissolved MeHg/ Dissolved THg)）were highly variable(1.8%～28%), giving average ratio of 10%.

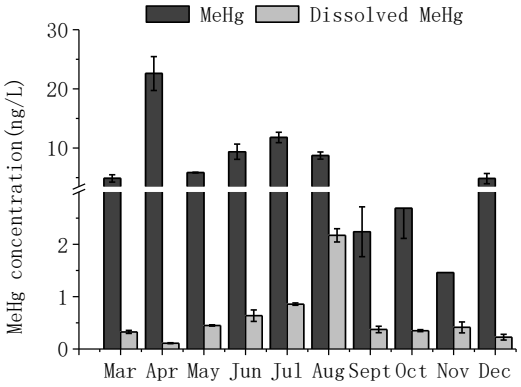

Figure 3-5.Influent MeHg Concentration

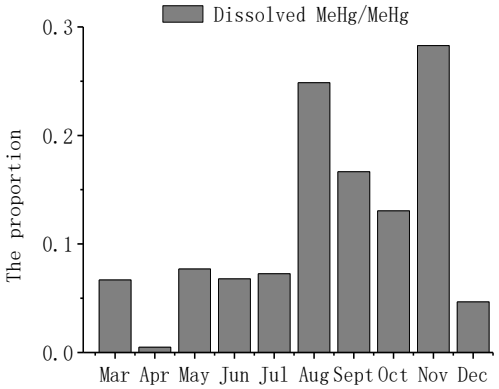

Figure3-6.The Ratio Dissolved MeHg Depresented of

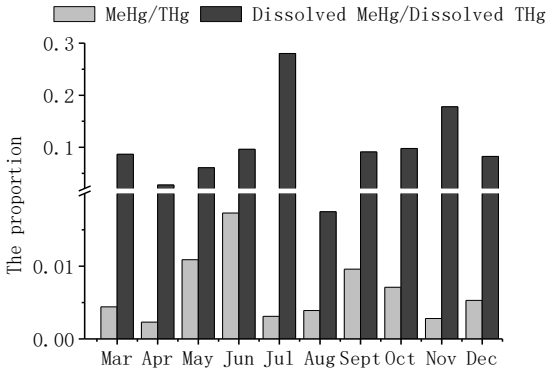

Figure3-7.The Ratio MeHg Represented of

B. Variation of mercury in effluent

1) THg variation in effluent

It was found that the contents of final effluent THg(1.78±0.22 ～ 43.3±0.67ng/L)were highly variable(Fig .3-8),giving average concentrations of 10.6ng/L.THg contents of the final effluent met the requirement of "Urban sewage treatment plant pollutant discharge standard" (GB18918-2002)(1ug/L)[17].Compared with the grit chamber influent,THg reduced about 96.1% ~ 99.9%,the average removal rate of 98.9%.It was similar with sewage treatment plant in St. Paul Minnesota(removal rate of 99%)[12],but slightly more than removal rate of wastewater treatment plant in Winnipeg[9].Probably the difference of influent sewage quality or the differences of processing treatment effect.It shows that The second sewage treatment plant of Jiaozuo have high removal efficiency for THg.

The contents of dissolved THg were 0.35±0.02～8.03±0.09ng/L,giving average concentrations of 2.2ng/L(final effluent). Compared with the grit chamber influent,THg reduced about 28.1% ~ 93.5%,the average removal rate of 65%.The fraction of dissolved THg accounted for 1.3%～81% of THg in final effluent and higher than grit chamber influent (Fig .3-9).In the sewage treatment process,it appears the phenomenon that Mercury ions may from the form of particle state to dissolved.It is similar with wastewater treatment plant in St. Paul Minnesota [12].

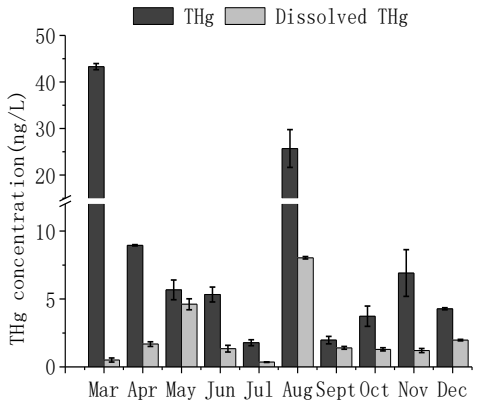

Figure 3-8.Effluent THg and Dissolved THg

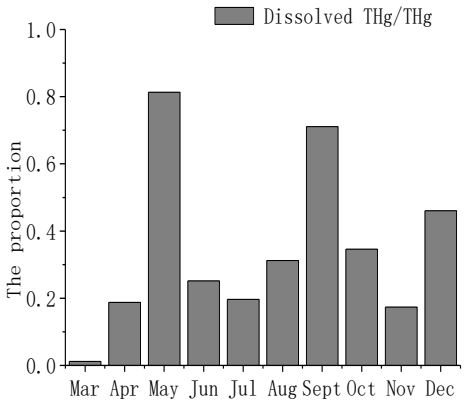

Figure 3-9. The Ratio Dissolved THg Represented

2) MeHg variation in effluent

MeHg content in the final effluent(0.13±0.01 ～ 1.11±0.26ng/L)met the requirement of "Urban sewage treatment plant pollutant discharge standard" (GB18918-2002)(10ng/L)(Fig .3-10).81.0%～99.4% MeHg was removed form the raw sewage by the current sewage treatment processes.The average removal rate(93.5%) is slightly lower than removal rate of MeHg in st. Paul Minnesota sewage treatment plant(97%)[12].

The content of dissolved MeHg were 0.02±0.002～ 0.19±0.03ng/L,giving average concentrations of 0.1ng/L in final effluente. 11.2%～95.2% dissolved MeHg was removed form the raw sewage by the current sewage treatment processes,the average removal rate of 70.7%.

It was found that the ratio (dissolved MeHg/MeHg)were highly variable (Fig .3-11),giving average ratio of 46%.The ratio(Dissolved MeHg/MeHg) increased to (10～88)% in the effluente. It was due to the removal efficiency of MeHg higher than dissolved MeHg removal efficiency.MeHg represented(0.6～20)% of THg in final effente,on average of7%(Fig .3-12), while it is higher than the ratio (Dissolved MeHg/THg) in inffluente.It shows that the removal efficiency of MeHg lower than THg removal efficiency,and final effluent mainly as form of THg.It was found that the ratio (dissolved MeHg/dissolved THg)were highly variable (1.3%～54%),giving average ratio of 11%(Fig .3-12).

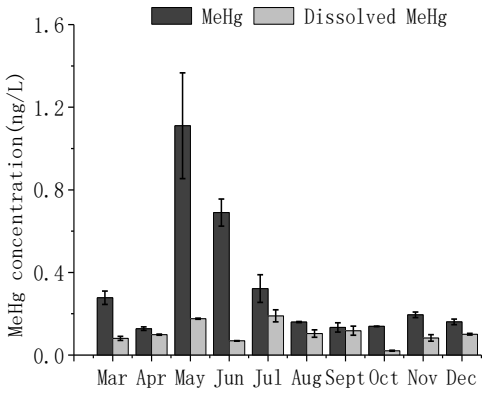

Figure3-10.Effluent MeHg Concentration Variation

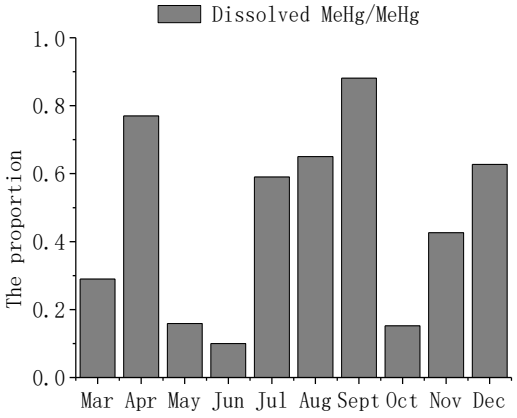

Figure3-11.The Ratio Dissolved MeHg Represented

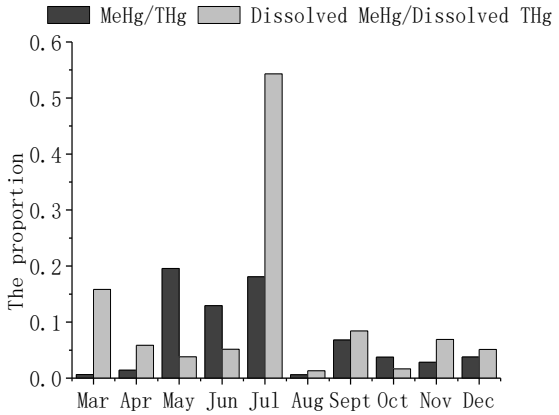

Figure3-12. The Ratio MeHg Represented

C. *Variation of mercury in the sludge*

The content of THg in the press filter sludge was between (2.02±0.29) mg/kg and (6.88±1.42) mg/kg, the average of (3.94±0.53)mg/kg (Fig .4-1),it is higher than the national standard for soil environmental quality of gradeⅢ（PH>6.5,Hg concent≤1.5mg/kg[18].The content of THg in press filter sludge of the second sewage treatment plant of Jiaozuo is more greater than St.Paul Minnesota sewage wastewater treatment plant[12],probably different influent THg content.

The content of MeHg in the filter sludge was between

3.76±0.613ng/g and 10.4±2.13ng/g,the average of 6.35±0.84ng/g.It reached the lowest in April and highest in March.MeHg accounted for 0.08% ~ 0.30% of THg (Fig .4-2).

D. Potential ecological risk assessment

Potential ecological risks of Hg in sludge mainly exist in the utilization of the sludge, especially in the process of land use. Geo-accumulation index method proposed and Hakanson potential ecological risk index method were used to assess the ecological risk of press filter sludge.

Geo-accumulation index method is also called Mull index, which is developed in the late 1960 s in Europe,It is widely used in the study of heavy metal pollution in sediment and other material degree of quantitative indicators, for its formula is as follow:

$$Igeo=\log2[Cn/(k\times Bn)] \quad (1)$$

$$Bn=0.025 \quad (2)$$

Cn represents heavy metal content in the sediment.Bn represents the sediments of the element geochemical background values(This evaluation element background values selected the corresponding soil environmental background values of Henan provinc)[20].K represents the coefficient accounted for the differences of rocks around may cause changes in the background value(It is generally 1.5,used to characterize the sedimentary characteristics,rock geological and other effects).Evaluation of heavy metal pollution must be considered in addition to the man-made pollution factors,environmental geochemical background values,also should be considered natural diagenesis that may cause factors of background value change.The accumulation exponential method noticed these factors, and made up for the inadequacy of other evaluation methods[21].

Hakanson put forward potential ecological risk index method,according to characteristics of heavy metal properties and environmental behavior,which is from the point of view of sedimentology proposed to evaluate heavy metals pollution in sediments.This method not only considered sediment heavy metal content,the ecological effect,environmental effect of heavy metals and toxicology,with comparable,equivalence attribute evaluation index classification method, and quantitative separated potential ecological harm degree.It is used more widely and more advanced method[22-23].The computation formula of potential ecological risk index is as follows:

$$Eri=TriCn/Bn \quad (3)$$

Cn represents heavy metal content in the sediment.Bn represents the sediments of the element geochemical background values.Tri stands for a single pollutant toxicity response parameters, which reflects the relationship of heavy metal among in water phase,the sedimentary solid phase and the biological phase. Hakanson developing the standardization of the toxic response coefficient of Hg is 40.Geo-accumulation index and ecological risk degree were shown in TABLE I.

TABLE I. GEO-ACCUMULATION INDEX AND ECOLOGICAL RISK DEGREE

I_{geo}	Pollution Level	Pollution Degree	E_r^1	Risk Degree
$I_{geo}\leq 0$	0	clean	$E_r^1<40$	低
$0<I_{geo}<1$	1	Slight pollution	$40\leq E_r^1<80$	中等
$2<I_{geo}<3$	3	Moderate pollution	$80\leq E_r^1<160$	较高
$4<I_{geo}<5$	5	Heavy pollution	$160\leq E_r^1<320$	高
$I_{geo}>5$	6	Serious pollution	$E_r^1\geq 320$	极高

In practice,Only in combination with the evaluation of regional soil, sediment and the characteristic of Hg,evaluation purpose,appropriate evaluation method should be used.Lu fengjuan [24]used geo-accumulation index method and Hakanson potential ecological risk index method,it quantitatively determined the mercury pollution in fiver sediments and potential ecological risks,through the analysis of mercury contents in 29 river bottom sampling points in Jiading District .Shanghai.Guo fuxing[25] used Hakanson potential ecological risk index method to study distribution patterns of Hg in surface sediments of the Yellow Sea. The potential ecological evaluation proves that surface sediment in the major sites were in the slight ecological risk condition,but that of some sites were in moderate ecological risk condition. Level of potential ecological risk for Hg is medium.

Geo-accumulation index of mercury in press filter sludge is between 5.75 and 7.52 in the second sewage treatment plant of Jiaozuo,average of 6.72.The pollution level is 6,it belongs to the serious pollution.potential ecological risk index of mercury in press filter sludge is between 3232 and 11008,average of 6304,with very high risk(TABLE II).In view of this,presss filter sludge has very high ecological risk and should not be used for farming without effective treatment.

TABLE II. ASSESSMENT OF THE ECOLOGICAL RISK OF HG IN PRESS FILTER SLUDGE

	T_r^1	B_n（mg/kg）	C_n(mg/kg)	I_{geo}	Pollution Degree	E_r^1	Degree of Risk
Mar	40	0.025	6.88	7.52	Serious pollution	11008	Very high
Apr	40	0.025	4.50	6.91	Serious pollution	7200	Very high
May	40	0.025	3.56	6.67	Serious pollution	5690	Very high
Jun	40	0.025	2.50	6.06	Serious pollution	4006	Very high
Jul	40	0.025	2.02	5.75	Serious pollution	3232	Very high
Aug	40	0.025	3.17	6.40	Serious pollution	5072	Very high
Sept	40	0.025	3.97	6.73	Serious pollution	6352	Very high
Oct	40	0.025	4.22	6.81	Serious pollution	6752	Very high
Nov	40	0.025	3.09	6.36	Serious pollution	4944	Very high
Dec	40	0.025	5.53	7.20	Serious pollution	8848	Very high
average	40	0.025	3.94	6.72	Serious pollution	6304	Very high

IV. CONCLUSION

This study showed that the concentrations of THg and dissolved THg ((0.23±0.02～9.73±0.82）×103ng/L and 2.32±0.28～124±20.2ng/L) were highly variable in raw sewage with a change of time.The average of 2.00×103ng/L and 16.2ng/L.Dissolved THg only represented(0.04～5.5)% of THg in influent.THg in grit chamber mainly exists with granule state. The concentrations of MeHg and dissolved MeHg(1.46±0 ～ 22.6±2.86ng/L and 0.11±0.01 ～ 2.17±0.13ng/L) were highly variable in raw sewage,average of 7.5ng/L and 0.59ng/L.Influent THg and MeHg were correlated to TSS(P=0.003,P=0.032).Dissolved MeHg content was increased with the temperature rising.THg was the main form of mercury in grit chamber influente.

Concentrations of THg and dissolved THg(1.78±0.22～43.3±0.67ng/L and 0.35±0.02～8.03±0.09ng/L) were highly variable in final effluent,average of 10.6ng/L and 2.2ng/L.THg contents of the final effluent met the requirement of "Urban sewage treatment plant pollutant discharge standard" (GB18918-2002)(1ug/L).96.1% ～ 99.9% THg and 28.1%～93.5% dissolved THg were removed from the raw sewage by the current sewage treatment processes.The second sewage treatment plant of Jiaozuo have high removal efficiency for THg.Concentrations of MeHg and dissolved MeHg(0.13±0.01 ～ 1.11±0.26ng/L and 0.02±0.002 ～ 0.19±0.03ng/L), average of 0.33ng/L and 0.1ng/L.MeHg content in the final effluent met the requirement of "Urban sewage treatment plant pollutant discharge standard" (GB18918-2002)(10ng/L).81.0% ～ 99.4% MeHg and 11.2%～95.2% dissolved MeHg were removed from the raw sewage by the current sewage treatment processes.The ratio(Dissolved MeHg/MeHg) increased to (10～88)% and MeHg represented(0.6～20)% of THg in final effluent,while it is higher than the ratio (Dissolved MeHg/THg) in inffluent.

Using the Index of geoaccumulation and potential ecological risk index method to analysis THg in pressure-filter sludge.The result of indicated that THg in pressure-filter sludge have high potential ecological risk.Thus the sludge might not be applied to farm land after suitable treatments.

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