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淮南潘一采煤塌陷区土壤-底泥中多氯联苯 (PCBs) 分布特征

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摘 要: 采煤塌陷水域因煤炭开采后, 地表沉陷积水而成, 其原来的陆生环境逐渐演变为水生环境, 沉陷前的土壤变为沉陷后水体的底泥, 土壤中的污染物也随之发生迁移转换。为了研究 PCBs 在淮南潘一塌陷区土壤和底泥沉积物中的含量和分布特征, 在塌陷区不同采样点采集 22 个柱状样品, 将处理好的样品在 GC/MS 上分析, 结果表明: 该塌陷区土壤和底泥沉积物中 14 种 PCB 残留总量范围为 1 581 ~ 5 510 ng/kg 和 3 278 ~ 5 973 ng/kg, 平均值为 2 447 ng/kg 和 4 339 ng/kg (干重)。0 ~ 20 cm 范围内, PCB138 未检出, 其他 13 种 PCB 含量在不同深度处于同一污染水平。当探讨 PCBs 含量随土壤和底泥深度变化规律时, 对每种多氯联苯进行分析, 能更好的总结出 PCBs 污染物在纵向的分布规律。

关键词: 采煤塌陷区; 底泥沉积物; PCBs; 垂向分布

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Distribution of PCBs in soil and sediment of subsidence water in Panji No. 1 coal mine of Huainan

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Abstract: The coal mining subsidence water occurs because of surface subsidence after coal mining. The terrestrial environment gradually evolved into an aquatic environment at the same time, and the soil evolved into substrate sludge. Therefore, the contaminants in the soil migrated and changed along with the evolution. In order to study the distribution of PCBs in the soil and sediment of Huainan Panji No. 1 coal mine subsidence water, 22 soil and sediment cores were collected in various locations, and the prepared samples were analyzed on GC/MS. Analysis results show that: the residual concentration of total 14 PCBs in soil and sediment are 1 581 - 5 510 ng/kg and 3 278 - 5 973 ng/kg, with average value 2 447 ng/kg and 4 339 ng/kg (dry weight) respectively. PCB138 is not detected, and the content of other 13 kinds PCB in different depths are at the same level. When studying the PCBs variation in soil and sediment, the analysis of each kind of PCB can better summarize the distribution law of PCBs in longitudinal direction.

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Key words: subsidence area; sediment deposition; PCBs; vertical distribution

多氯联苯(PCBs)难溶于水,具有亲脂性,它们其中一些被称为二噁英类化合物,二噁英类物质中最毒者的毒性相当于氰化钾的1 000倍以上,即使低浓度时也会对生物体造成伤害,具有潜在的致癌性。在1940s到1970s,人类生产了大量的PCBs^[1],期间中国生产了10 000 t PCBs产品,占全球总生产量的1%^[2]。中国主要生产1号PCBs和2号PCBs,1号PCBs以三氯联苯为主,主要用于生产变压器和电容器;2号PCBs以五氯联苯为主,主要作为油漆添加剂^[3]。PCBs被广泛应用于工业和商业领域,变压器油、电容器、液压油、油漆和印刷油墨均含有PCBs,人类的生产活动也会带来PCBs污染^[4-5]。PCBs迁移过程十分复杂,可以通过大气沉降^[6-7]、鸟类迁徙^[8-10]、和洋流^[11]等方式带到很远的地区。PCBs经过长距离的迁移之后有93.1%进入土壤,3.5%进入海洋,4.1%转移进入沉积物^[12-13],最近的一些研究结果^[14-18]表明,底泥沉积物是PCBs等污染物的源和汇。

国内外专家学者对河流、湖泊、海湾和近海沉积物中PCBs有大量的研究^[19-25],但对采煤塌陷区土壤和采煤塌陷水体底泥中多氯联苯等POPs的研究极少。潘一塌陷水体为该区最大的塌陷水体,它是由陆生生态环境演变而来,由于形成年限较短,水体的沉积物不是过去历史时期的沉积,而是由周围农田土壤的演变而形成的,仍保留着农田土壤的污染特征。该塌陷水体的特殊性在于,它是因村庄和农田受煤炭开采活动的影响,地表塌陷形成的,既受到煤炭开采活动和居民活动的影响,再者塌陷水体与泥河相连,又受到来自矿业废水的污染。因此研究该塌陷水体PCBs的含量特征和分布规律,对研究其他采煤塌陷区的PCBs等POPs污染具有代表性和重要意义。

1 材料与方法

1.1 样品采集

样品采集所用的棕色玻璃瓶以及实验过程中用到的烧杯、移液管等玻璃器皿均通过碱液(6 g 氢氧化钠溶于6 mL水中再加50 mL乙醇)润洗,并用自来水冲洗干净,再用蒸馏水润洗3遍,超纯水润洗1~2遍,置于烘箱中烘干备用。本研究采用专用柱状底泥采样器和专用柱状土壤采样器进行样品采集,将采集的20 cm柱状底泥和土壤样,每5 cm分1层,然后装进棕色玻璃瓶中,并用锡箔纸封口。土壤采样点和底泥采样点共计22个,采样点用便携式GPS进行

定位,采样点布设如图1所示,采样时间为2014年8月,采样后立即送回实验室,-20℃保存至分析。

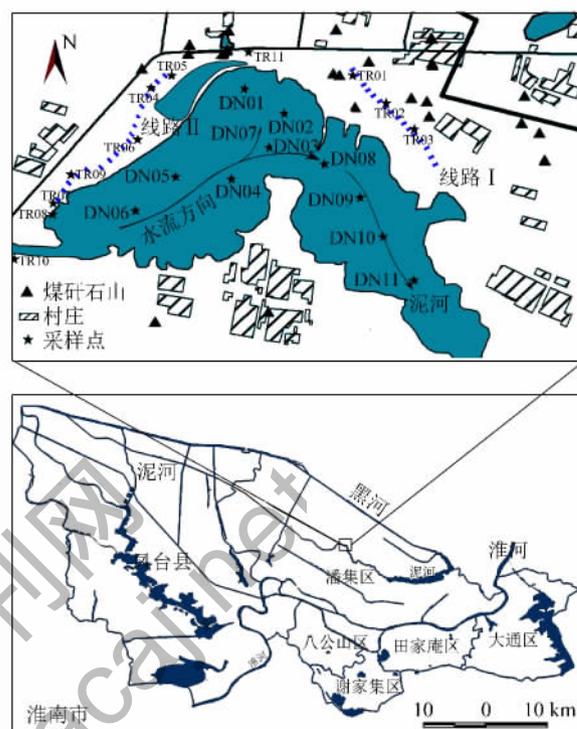


图1 采样点示意

Fig. 1 Sampling stations

1.2 试剂与材料

有机溶剂正己烷(*n*-Hexane)和二氯甲烷(Dichloromethane)均为色谱纯,无水硫酸钠(分析纯),使用之前,在超声振荡器中,用正己烷浸泡超声30 min,室温下冷却备用,硅胶(试剂纯,100~200目)在超声振荡器中,用正己烷浸泡超声30 min,移植蒸发皿,于马弗炉中灼烧(130℃)16 h,冷却至室温储存备用,中性氧化铝在超声振荡器中,用正己烷浸泡,超声震荡30 min,移入蒸发皿,在马弗炉中450℃灼烧12 h,自然冷却后移入广口密封瓶中,放入干燥器中保存待用。14种多氯联苯目标化合物混合标准液,回收率指示物标准试剂为PCB65,内标为PCB204,混合标准液、回收率指示物和内标均购自美国标准物质研究中心。

1.3 样品提取

土样经过冷冻干燥研磨并过200目筛子,准确称取10 g土样,然后置于索氏提取器,加入PCB65做回收率指示物,用200 mL体积比为1:1的二氯甲烷/正己烷混合溶剂于60℃水浴中提取24 h。提取液经无水硫酸钠过滤后,旋转蒸发至2~3 mL,过硅胶氧化铝层析柱净化,层析柱(内径1.5 cm,长30 cm)由

下往上依次装入无水硫酸钠、中性氧化铝和硅胶,它们的体积比为1:1:2。最后用50 mL体积比为7:3的正己烷/二氯甲烷混合液淋洗出PCBs,再次旋蒸至1~2 mL,最后经高纯氮吹脱至0.5 mL,转移到进样瓶中,加内标PCB204并定容至1 mL。

1.4 样品的分析

PCBs的分析在PE公司生产的GC/MS上进行,色谱柱为PE clarus Elite-5 MS;色谱柱升温程序为柱温80℃保持1 min,然后以30℃/min的速度升温至160℃保持1 min,再以3℃/min的速度升温至265℃保持1 min。样品定性分析采用标样的各色谱峰保留时间对实际样品中的PCBs进行定性,并通过GC/MS验证,采用内标曲线法定量。

1.5 质量控制与质量保证

实验过程中采用美国环保署(US EPA)推荐的质量控制方法,所有数据经过严格的质量控制,每次进样前都进行空白对比,空白样品中未能检出目标物。16种混合标样的基质加标回收率在74.4%~110.28%,回收率指示物PCB65在底泥样品中的回收率在70.14%~136.98%,土壤样品中的回收率在60.48%~141.12%,平行样相对标准偏差小于5%,符合分析要求。

2 结果和讨论

淮南潘一矿于1983年建成投产^[26],其境内潘一杨庄塌陷水域(研究区域)地理坐标为E116.817°~E116.819°,N32.801°~N32.816°。塌陷水域面积约为4 km²,平均水深3.6 m,20 a左右沉陷覆水时间,整个水域为失地农户渔网分割养鱼,但无饵料添加,为自然散养型^[27]。

采样点TR01,TR02,TR03,TR04及TR05为农田土壤样;TR06采样点位于当地堆积已久的覆土煤矸石山腰上,并进行了柱状采样,只采集了0~15 cm的样品,表层为土壤样,余下为碎屑状煤样,另外又在附近采集了一个煤矸石样;TR07和TR08为塌陷水域岸边样,被杂草覆盖,TR07距岸边20 cm左右,由于采样前两天,采样区域处于阴雨天气,TR08被雨水淹没5 cm左右,但并未与水体直接相连;TR09为槐树林土壤样,TR10为泥河入口岸边土壤样;TR11为公路附近采样点(不慎遗失15~20 cm土样),由于煤炭运输和附近煤矸石的堆积,土壤中夹杂煤屑和细碎煤矸石,土壤整体呈现煤黑色。DN01, DN02, DN08, DN09, DN10及DN11水深在3.5 m左右,其余底泥采样点水深在4~6 m,深度较大。

由于TR06, TR11, DN03, DN06和DN08采的样品土壤和沉积物层数不全,并未将这5个点土壤和沉积物中的PCBs含量纳入统计范围。为了便于比较,底泥样、土壤样以及煤和煤矸石中的PCBs含量分别统计。

2.1 PCBs的含量特征

PCB18, PCB28, PCB31, PCB44, PCB52和PCB118检出率达100%,呈现出面污染,底泥和土壤样品中均未检测出PCB138,高氯代联苯PCB101, 105, 149, 153, 174, 180和194呈点状污染。

由图2可以看出,各采样点主要以三氯联苯和五氯联苯同系物为主,这与我国主要生产和使用1号PCBs与2号PCBs是相符合的。土壤和底泥中PCBs含量残留范围为1 581~5 510 ng/kg和3 278~5 973 ng/kg,平均值为2 447 ng/kg和4 339 ng/kg(干重),低于淮河淮南段6 340 ng/kg^[28]。7种指示性PCBs^[29-31]常被用来研究PCBs的污染状况,土壤和底泥中7种指示性PCBs的平均含量为800 ng/kg和1 516 ng/kg,污染比较严重,而且底泥的污染程度要大于土壤。为了进一步研究PCBs污染状况,将其与国内外其他水体沉积物及土壤中PCBs的含量做比较,结果见表1、2。与表1其他地区相比较知,塌陷区水体底泥沉积物多氯联苯污染处于较轻污染水平,与表2其他地区相比较知,塌陷区土壤中PCBs含量水平处于世界土壤背景范围内,与其他研究相比处于较低水平。加拿大环境委员会制定了沉积物环境质量标准(CA-SQG)^[42],规定的沉积物环境质量标准参考值ISQG(interim sediment quality guideline)为34 100 ng/kg,研究区域底泥沉积物中PCBs含量未超过ISQG值,对暴露的生物体威胁尚可接受,极少产生生物负面效应。荷兰规定7种指示性PCBs的土壤修复目标值和干预值分别为20 000 ng/kg和10⁶ ng/kg^[43],而研究区域土壤中7种指示性PCBs平均含量

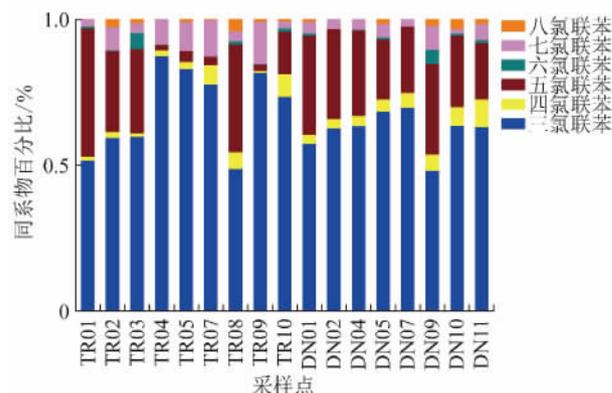


图2 各采样点同系物组成

Fig. 2 Composition of PCBs homologs in every sampling station

为 800 ng/kg, 低于土壤修复目标值, 更远低于干预值, 因此该采煤塌陷区土壤污染水平较低, 不需要进行土壤修复。

表 1 国内外其他水体沉积物中 PCBs 的质量含量

Table 1 Comparison of PCBs concentrations in different areas

地点	多氯联苯种类	多氯联苯含量/ (ng · kg ⁻¹)	参考文献
Busan Bay 韩国	22	5 710 ~ 199 000	[23]
Lerma River 墨西哥	7	54 000 ~ 123 000	[32]
Marseille Bay 法国	7	14 200 ~ 88 100	[33]
白洋淀	41	5 960 ~ 29 610	[34]
太湖	56	1 350 ~ 13 800	[35]
汾河	123	ud ~ 126 490	[36]
本文	14	3 278 ~ 5 973	

注: ud 表示未检出

表 2 其他国家和地区土壤中 PCBs 的质量含量水平

Table 2 Levels of soil PCBs in other countries

地点	多氯联苯种类	多氯联苯含量/ (ng · kg ⁻¹)	参考文献
世界土壤背景值	9	26 ~ 96 900	[37]
巴基斯坦奇纳布河土壤	33	700 ~ 30 500	[38]
波兰城市固废垃圾填埋场	7	2 471 ~ 12 062	[39]
上海城区土壤	74	232 ~ 11 325	[40]
山东省农田土壤	—	1 460 ~ 19 200	[41]
本文	14	1 581 ~ 5 510	

2.2 PCBs 的垂向分布特征

潘一采煤塌陷水域, 形成年限相对较短, 水体底泥沉积物是农田土壤转化而来, 有别于沉积年代久远的河流、湖泊、海湾和近海等地区的沉积物。图 3 表明各采样点总 PCBs 含量随深度的变化规律不明显, 总体上讲 0 ~ 20 cm 范围内, 自下而上, PCBs 含量有逐渐减少的趋势。当以所检出的 13 种 PCB 做统计, 分析其在深度上的变化规律时, 由图 4 得出, 13 种 PCB 含量在不同深度上处于同一污染水平。可见当以总 PCBs 平均含量做统计时, 并不能很好的得出 PCBs 含量随深度的变化规律, 若是单独计算每种 PCB 平均含量, 可以较好的得出 PCBs 含量随深度的变化规律, 这种差异的原因可能是高氯联苯迁移性差, 易于富集在污染源附近导致的, 这也与实际检测结果相符。13 种 PCB 含量在不同深度上处于同一污染水平, 这是因为塌陷水域在沉陷积水以前作为农用耕地, 0 ~ 20 cm 的土壤被不断的翻耕, 使不同种类的 PCBs 混合均匀。马召辉等^[44]对太湖沉积物中 PCBs 研究结果显示, 在 0 ~ 15 cm, PCB118 含量随深度增

加而明显降低, 但 15 cm 以下则无明显变化。而本研究中 PCB118 并不呈现出上述变化规律, 可见土地的原始利用类型对采煤塌陷水体中 PCBs 的分布有重要影响。采煤塌陷水体底泥沉积物中 PCBs 的分布有其独特的规律。

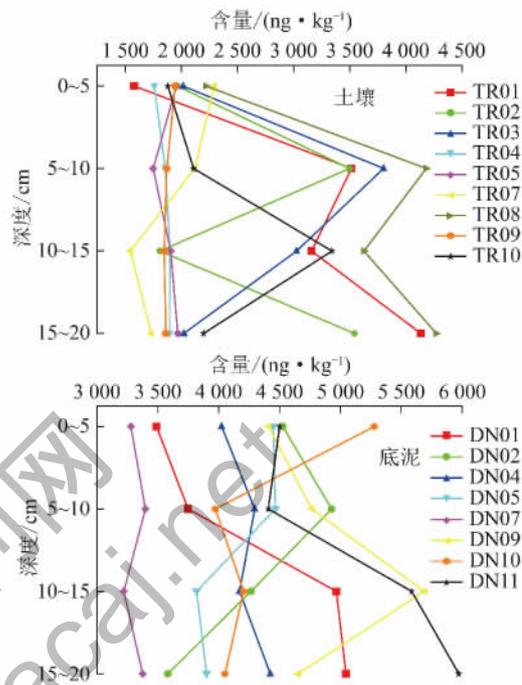


图 3 土壤和底泥中 13 种 PCB 总含量随深度的变化
Fig. 3 Total PCBs concentrations in soils and sediments along with depth

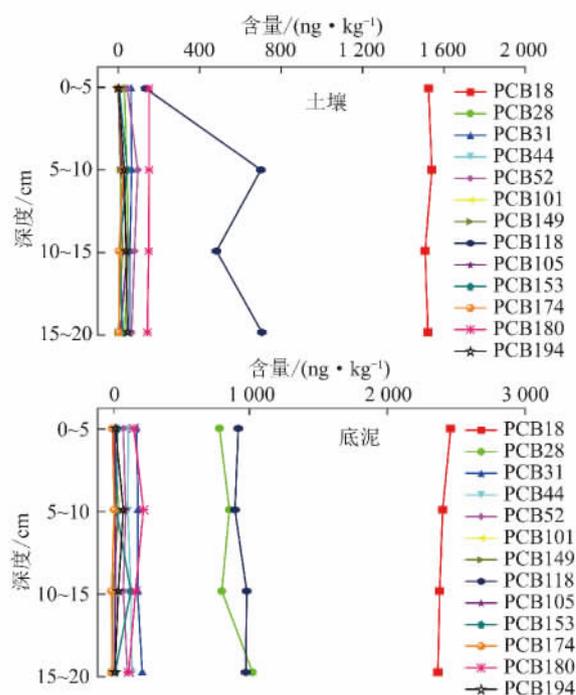


图 4 土壤和底泥中每种 PCB 含量随深度的变化
Fig. 4 Every PCB concentration in soils and sediments along with depth

2.3 PCBs的横向分布特征

如图1所示,线路I上分布着TR01,TR02和TR03采样点,线路II上分布着TR04,TR05,TR06,TR07和TR09采样点。由图5可以看出,线路I上的采样点(TR01,TR02和TR03)13PCBs平均总含量高于线路II(TR04,TR05,TR07和TR09)上的采样点。线路I和线路II上的采样点均为农田土壤样,所处环境基本相同,差异较大的是线路I两侧遍布煤矸石山,因此可以推测,线路I上的采样点13PCBs平均总含量高于线路II上的采样点,其最可能的原因是由于煤矸石堆置引起的。

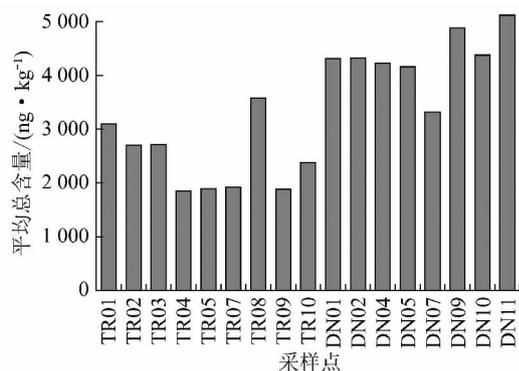


图5 各采样点13种PCB的平均总含量

Fig. 5 Average total 13PCBs concentrations in every station

对工业经济发达地区土壤及沉积物中PCBs含量^[25,45-49],以及世界背景值^[37]进行分析研究,经过总结,PCB138和PCB153在工业经济发达地区,均有较高的残留水平和检出率,且总PCBs含量处于高水平,而研究区域并未检出PCB138,作为PCB138同系物的PCB149和PCB153含量很低,且塌陷区土壤和底泥中总PCBs含量处于较低水平,在检测样本中高氯联苯PCB101,PCB149,PCB105,PCB153,PCB174和PCB194呈点状污染,再加上研究区域相对封闭^[26],因此研究区域中土壤和底泥沉积物中PCBs的主要来源不是工业污水及生活污水的排放,泥河河水的补给对采煤塌陷水域中PCBs的含量影响不大。

研究区域地表,因采煤挖空逐渐沉降,形如漏斗,成为雨水的汇集之地。多氯联苯能随着水土流失进行迁移^[50],采煤塌陷塘的底泥由农田土壤直接演化而成,同时还有部分农田土壤流失沉积而成,根据张瑞等^[51]研究结果,农业土壤中多氯联苯的输入是底泥沉积物中多氯联苯的重要来源,图5的结果表明底泥中多氯联苯的含量普遍高于周围土壤样中的含量,与上述研究相符合,农田土壤的水土流失,会增加底泥沉积物中PCBs的含量。从图5中可以发现,土壤中多氯联苯含量的峰值出现在TR08,土壤采样点

TR08地势低洼,雨水在此处汇集,属于即将淹没区,从图5可以发现采样点TR08处土壤中的PCBs含量高于其他土壤采样点,这恰能有力的说明农田土壤的流失,最终会导致底泥中PCBs含量的增加。底泥中多氯联苯含量的峰值出现在DN11采样点,沿着河流流向(图1)观察,再结合图5可以发现,DN09,DN10和DN11的13PCBs平均总含量高于其他底泥采样点。在实际采样过程中,DN08,DN09,DN10和DN11河水深度较浅,河床位置较高,由于河床的阻碍作用,河水停留时间较长再加上河水流速减缓,水中的悬浮物和颗粒物等在此处的沉降量增加,有文献研究^[52-54]指出颗粒物和悬浮物中含有大量的PCBs,因此悬浮物和颗粒物沉降量的增加可能是导致DN09, DN10和DN11的13PCBs平均总含量较高的原因。

2.4 煤矸石中PCBs的赋存

图6的结果表明,煤和煤矸石中含有大量的PCBs,是土壤平均含量的3倍多,矸石山表层土壤(TR06)中PCBs的含量亦高于土壤平均含量,煤和煤矸石中7种指示性PCBs的含量为5872 ng/kg和7728 ng/kg,远低于荷兰规定的7种指示性PCBs干预值,不需要采取干预措施。TR11采样点,0~5,5~10,10~15 cm的含量分别为5510,4944,4577 ng/kg,峰值出现在0~5 cm表层土壤,也是所检测土壤样中PCBs含量的最大值,TR11平均PCBs含量将近为土壤平均含量的2倍,再结合TR11的采样点信息可以判断,TR11采样点土壤PCBs含量较高,应当是煤灰和煤矸石污染引起的。非邻位的PCBs(PCB77,81,126,169)是在燃煤、有色金属冶炼及再生、工业与市政废弃物燃烧等高温过程中产生的特征产物^[55-56],研究区域遍布煤矸石山,且农村有焚烧生活垃圾的习惯,再加上有用煤作为燃料的历史,应加强对PCB77,81,126,169的检测,以便评估煤炭开采对塌陷区环境的影响。

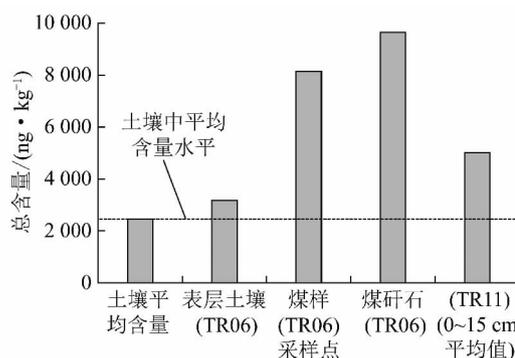


图6 煤和煤矸石中13PCBs的总含量

Fig. 6 Total 13PCBs concentrations in coal and gangue

3 结论与展望

(1) 淮南潘一采煤塌陷水域周围农田土壤和底泥中除 PCB138 未检出,其他 13 种多氯联苯总量变化范围为 1 581 ~ 5 510 ng/kg 和 3 278 ~ 5 973 ng/kg, 平均值为 2 447 ng/kg 和 4 339 ng/kg(干重),三氯联苯和五氯联苯为其主要污染特征,与我国多氯联苯生产和使用历史相符合。

(2) 与国内外研究相比,该塌陷水域土壤和底泥沉积物中多氯联苯污染处于轻度水平,塌陷水域土壤中 7 种指示性 PCBs 含量较低,不需要进行土壤修复;当探讨采煤塌陷水域周围农田土壤和底泥中 PCBs 随深度变化规律时,对每种 PCB 进行分析,能更好的总结出 PCBs 的垂向分布规律;采煤塌陷塘的底泥由农田土壤直接演化而成,区别于河流、湖泊、海湾和近海等地区的沉积物,在 0 ~ 20 cm 范围内,潘一采煤塌陷水域中土壤和底泥中 14 种 PCB 含量处于相同水平,尤其是呈面状检出且含量较高的 PCB18。

(3) 采煤塌陷水域周围农田土壤中 PCBs 的输入,是淮南潘一采煤塌陷水域底泥 PCBs 含量增加的重要原因,塌陷水域是其周围农田土壤中 PCBs 等污染物的汇。研究区域煤矸石中 PCBs 含量较高,但煤矸石中 7 种指示性 PCBs 含量远低于荷兰规定的 7 种指示性 PCBs 干预值,不需要采取干预措施,煤矸石山的堆置,对周围土壤中 PCBs 含量有重要影响,会使土壤中 PCBs 含量增加。

(4) 此次研究,仅仅从含量方面,研究了潘一采煤塌陷水域土壤和底泥沉积物中 PCBs 分布特征,而温度、pH、有机质、水体悬浮物和氧化还原条件的改变等对 PCBs 在环境中行为的影响尚未涉及,尤其是采煤塌陷区中 PCBs 在含煤有机质介导下 PCBs 迁移转化特征和降解环境条件等尚不清楚,有待进一步加强这些方面的研究。鉴于 PCB77,81,126,169 是燃煤和工业活动等过程中产生的,应当加强对 PCB77,81,126,169 的检测,以便确定煤炭开采活动和煤矸石堆置,对塌陷区 PCBs 含量的影响和贡献大小,还应当扩大检测范围和多氯联苯检测种类,为多氯联苯污染源解析做好准备。

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