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目 次

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中国沉积物多环芳烃的时空分布及驱动因子

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摘要: 根据 2000 ~ 2020 年中国湖泊、河流和河口沉积物中多环芳烃(PAHs)的时空分布,并通过构建结构方程模型和重力模型分析了驱动 PAHs 时空分布的社会经济因素. 结果表明,不同地区沉积物中 PAHs 含量由高到低依次为:北部沿海 > 东北 > 东部沿海 > 南部沿海 > 黄河中游 > 长江中游 > 西南 > 西北. 南部沿海、长江中游和东部沿海地区高分子量 PAHs 的比例相对较高,东北、西北、北部沿海和黄河中游地区低环 PAHs 的比例相对较高. 沉积物中 PAHs 的含量从 2000 年起逐渐增加,2006 年以后逐渐减少,且不同地区沉积物中 PAHs 含量到达峰值的年份有显著差异. 经济发达地区沉积物中 PAHs 的含量在达到峰值后逐渐下降; 发展中地区由于经济发展较快导致污染物积累较快. 偏远或欠发达地区 PAHs 含量逐渐增加,但累积率低于发达地区. 城市化和工业化对沉积物中 PAHs 的影响显著,其中对沉积物中 PAHs 分布影响最大的因素为经济发展.

关键词: 多环芳烃(PAHs); 内陆沉积物; 社会经济因素; 贝叶斯模型; 重力模型

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Spatiotemporal Distribution and Driving Factors of Polycyclic Aromatic Hydrocarbons (PAHs) in Inland Sediments of China

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Abstract: This study reviewed the spatial and temporal distributions of polycyclic aromatic hydrocarbons (PAHs) during 2000-2010 in inland sediments of China and quantified the underlying socioeconomic determinants based on the structural equation model and gravity model. We found that PAHs concentrations in the sediments of eight different regions followed the order of Northern coast > Northeast > Eastern coast > Southern coast > Middle Yellow River > Middle Yangtze River > Southwest > Northwest. The Southern coast, Middle Yangtze River, and Southern coast regions showed large contributions to the high-molecular weight PAHs, whereas the Northeast, Northwest, and Middle Yellow River regions showed high contributions to the low-molecular weight PAHs. PAHs concentrations continuously increased from the year 2000 followed by a gradual decline after 2006, with significant differences in the year when PAHs levels reached their peak. PAHs concentrations of sediment in developed regions declined in recent years following a continuous increase in the 2000s; however, they still increased rapidly in developing regions owing to fast economic development. In addition, the increment rate of PAHs concentrations in sediment at the remote or less-developed regions was slower than that at the developed regions. Urbanization and industrialization had an important effect on PAHs in the sediments, and the largest influencing factor was the economic development.

Key words: polycyclic aromatic hydrocarbons (PAHs); inland sediment; socio-economic factors; Bayesian estimation; gravity model

多环芳烃(polycyclic aromatic hydrocarbons, PAHs)是含有 2 个以上苯环的芳香烃类化合物以及由它们所衍生出的各种化合物的总称, 其主要来源是以垃圾焚烧和燃煤为代表的能源和化工生产过程中有机物质不完全燃烧[1,2]. PAHs 因其广泛的分布、持久性、毒性和生物积累能力而受到全世界的关注,它们在环境介质中总致突变活性占很大比例(在空气悬浮颗粒中占 35% ~ 82%,在沉积物中占 10% ~ 240%)[3,4]. 根据现有的排放清单[5],1995 ~ 2008 年,虽然全球 PAHs 排放量从 59.2 万 t 大幅下降至 49.9 万 t,但在许多原始地区(如深海表层沉积物,北极和南极洲以及森林土壤),PAHs 的含量没有显示显著下降的趋势. 全球 PAHs 排放量下降速度放缓,主要是由于快速工业化的发展中国家大量排放 PAHs. 中国人为排放的 PAHs 占东亚地区总排放量的四分之三,超过了全球总排放量的四分之一[6]. 沉积物中 PAHs 的时空分布与经济活动和环境过程(如大气输送和沉降)密切相关,深入理解 PAHs 的生物地球化学循环链过程有助于更科学地评估和防治沉积物的污染[7],但目前的报道较少涉及社会经济因素与其的定量关系. 本文基于 2000 ~ 2020 年已公布的国家沉积物类型 PAHs 数据,分析影响 PAHs 时空分布的潜在人为和自然因素,并通过构建结构方程模型和重力模型解析其与社会经济因素之间的关联.

1 材料与方法

1.1 数据排序

本文为采样时间在 2000 ~ 2020 年的中国沉积物(中国香港、澳门和台湾资料暂缺)中 PAHs 的浓度和组成数据(表 1),文献检索工具为 Web of

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表 1 中国沉积物中 PAHs 的采集数据1)

Table 1 Acquisition data of PAHs in inland sediments in China

位置	采样时间（年-月）	样本数（n）	最小值 /ng·g^{-1}	最大值 /ng·g^{-1}	平均值 /ng·g^{-1}	主要成分	文献	位置	采样时间（年-月）	样本数（n）	最小值 /ng·g^{-1}	最大值 /ng·g^{-1}	平均值 /ng·g^{-1}	主要成分	文献
百花湖		16	715	1 167	863	2 环和 4 环	[8]	天津海河	2004	16	775	255 372	27 074		[67]
白洋淀		16	101	1 495	353	高环	[9]	厦门湾	2005	14	98	325	173	2 环和 3 环	[55]
白洋淀	2009-01	16	97	2 402			[10]	海南岛	2017	16	67	990	697		[68]
白洋淀	2016-03	16	163	861			[11]	汉丰湖		15	22	692	204	2 环和 3 环	[69]
白洋淀	2007-01	6	467	1 366	756	2 环和 3 环	[12]	杭州	2002	10	140	2 342	596	3 环	[70]
北部湾	2017	16	2	359	110		[13]	杭州湾	2015	16	7	335			[71]
清远北江	2016	15	54	819	55		[14]	杭州湾	2011	16	10	507			[71]
扬州京杭运河	2009	16	505	4 532	2 359		[15]	杭州湾	2012	16	21	457			[71]
渤海湾	2012-09	16	52	163	108		[16]	杭州湾	2013	16	32	384			[71]
渤海湾	2007-08	16	141	301	188		[17]	杭州湾	2014	16	34	242			[71]
渤海湾	2012-08	16	275	586			[18]	红枫湖		9	82	310	185	菲和芴	[72]
博斯腾湖	2014-07	16	25	491	145		[19]	红枫湖	2010	16	848	3 725			[73]
博斯腾湖		16	38	185		2 环和 3 环	[20]	洪湖	2014	16	93	431	244	菲和芴	[74]
博斯腾湖	2016-12	16	57	360			[21]	淮河中游		16	83	2 599	525		[75]
博斯腾湖	2007	16	335	1 394	821 ±236		[22]	黄浦江	2016-05	16	25	9 910			[76]
查干湖	2016	16	306	1 214	586		[23]	黄浦江	2006-01	18	107	1 707			[77]
长荡湖	2013	14	181	441			[24]	黄浦江	2010-07	16	132	6 800			[78]
昌江	2011-05	16	1 113	44 311	12 456		[25]	黄浦江	2012-12	16	140	5 966	1110		[79]
巢湖	2012	16	11	33 090			[26]	黄浦江	2013-07	16	253	1 025			[80]
巢湖南淝河	2016	16	252	498			[27]	黄浦江	2012-11	16	723	38 541			[81]
巢湖	2009	16	61	10 200	1 230		[28]	黄浦江	2003-04	16	244	2 805			[82]
巢湖	2011	15	81	30 365	5 255		[29]	呼伦湖	2016	16	283	1 057	597		[23]
巢湖	2010	16	110	6 246	908 ±1 878		[30]	胶州湾	1997	23	82	4 567			[83]
巢湖	2010-08	14	116	2 832	2 899		[31]	锦州湾	2009-08	16	133	594			[84]
巢湖		18	785	6 993	2 873		[26]	厦门西海	1999-06	9	59	1 177	334 ±337	3 环和 5 环	[85]
巢湖	2009	16	909	1 878			[28]	九龙江河口	1999	16	248	675			[85]
十五里河	2017	16	175	340			[27]	克钦湖	2007	16	140	363	223		[86]
巢湖河口	2012	28	136	70 500			[26]	莱州湾	2012-08	16	161	448			[18]
三峡水库	2015	16	202	2 291	990		[32]	莱州湾		16	239	697			[87]
胶州湾	2014	16	22	635	239		[33]	莱州湾	2016-12	16	269	895	613		[88]
大伙房水库	2010	16	323	912	592 ±139	2 环和 4 环	[34]	青藏高原	2006	16	98	595			[89]
大九湖		16	49	985	303	4、5 和 6 环	[35]	雷州湾		16	22	320			[90]
大辽河河口	2007-08	35	276	1 607	743	2 环和 3 环	[36]	连环湖	2007	16	198	1 056	446		[91]
大辽河河口		16	374	11 588			[37]	连环湖		16	293	2 247	767	4 环和 6 环	[86]
大辽河流域	2005-08	12	62	841	287	4 环和 6 环	[38]	连环湖	2007	16	297	1 327	672		[92]
达里诺尔湖	2014	16	100	1 036	480	4 环和 6 环	[39]	连环湖	2007	16	477	2 247	1 019		[93]

续表 1

位置	采样时间（年-月）	样本数（n）	最小值 /ng·g^{-1}	最大值 /ng·g^{-1}	平均值 /ng·g^{-1}	主要成分	文献
大通湖	2008	16	82	310	185		[40]
大亚湾	2003-12	16	43	158	126		[41]
大亚湾	1999-08	14	115	1 134	481	3 环和 4 环	[42]
大冶湖	2015	16	36	2 033	941	4 环和 5 环	[43]
后海湾	2004-02	16	184	582	354	3 环和 4 环	[44]
滇池	2012	14	210	11 070			[45]
滇池	2006	16	479	4 561			[46]
滇池		16	558	6 418	1 324 ±136	4 环和 6 环	[47]
滇池	2012	16	590	23 250			[48]
滇池	2006	16			2 249 ±1 497		[22]
滇池河口	2012	16	230	12 271			[45]
滇池流域北	2013	16	142	2 131			[49]
淀山湖	2015	16	55	1 331	373		[50]
滴水湖		16	74	579	273	二氢苊和芴	[51]
武汉东湖	2012	16	11	2 478	686		[52]
东平湖		13	49	134	83	2 环和 3 环	[53]
洞庭湖		15	78	566	232	苊烯	[54]
廉江-汕头	2005	14	153	401	272	2 环和 3 环	[55]
中国南海	2007	15	8	180			[56]
中国南海	2015-03	16	61	355	146		[57]
鄂陵湖	2016	16	11	55	29		[58]
洱海湖		16	32	559	257	5 环	[59]
巢湖丰乐河	2014	16	71	3 372			[60]
抚河	2011	16	568	7 335	3 633		[25]
抚仙湖	2015	16	83	262	143	4 环	[61]
抚仙湖	2011-09	12			1 550 ±234		[62]
关河河口		16	90	218			[63]
南岭山地湖泊		19	86	778		4 环和 6 环	[64]
汕头	2005	16	102	395	228	2 环和 3 环	[55]
海河	2010	16	259	11 297			[65]
海河河口		16	92	15 886			[66]
海河，天津		16	23	10 209			[67]
周溪湾	2011-11	12	41	384	89		[123]
普莫雍错湖	2014-08	15	30	229		5 环和 6 环	[124]
钱塘江	2005	15	91	614	313	2 环和 3 环	[125]
青岛湖	2007	16	256	1 884	685	4 环	[126]
青岛海	2006-12	16	221	2 151			[127]
青海湖		16	334	3 903	805		[128]
青海湖	2006	16	495	1 173	657	芴和菲	[129]
青海湖	2006	16	11	279			[130]

位置	采样时间（年-月）	样本数（n）	最小值 /ng·g^{-1}	最大值 /ng·g^{-1}	平均值 /ng·g^{-1}	主要成分	文献
辽东湾	2012	16	22	274			[18]
辽东湾	2007-08	16	272	1 606			[36]
辽河	2010-05	15	38	9 014	1 612		[94]
辽河	2012-02	16	46	1 167			[95]
辽河	2014-06	16	83	25 374	3 149	2 环和 3 环	[96]
辽河	2010	15	124	21 233	3 208		[94]
辽河	2012-05	16			2 394		[97]
辽河，吉林	2014	16	160	461	303		[98]
滦河河口		16	5	545			[99]
滦河	2006	16	7	1 586			[100]
滦河	2014	16	21	287	115		[101]
滦河		16	40	328			[102]
滦河	2015	16		478			[103]
骆马湖	2009	16	189	726	443		[104]
澳门海岸	1999	16	294	12 741			[105]
黄河中下游	2004-06	12	17	903	170	3 环	[106]
闽江河口	1996	16	315	1 260	437		[107]
闽江河口	1999-11	9	112	877	433	5 环和 6 环	[108]
密云水库		16	619	1 088	842	菲和芴	[109]
磨盘山水库		16	89	749		2 环和 3 环	[110]
墨水河	2015-01	16	197	8 549	3 320		[111]
南湖		13	284	7 935	2 199	4 环	[112]
南沙红树林湿地	2011	28	332	1 652			[113]
南四湖		16	20	929		高环	[114]
南四湖	2001	16	160	32 600			[115]
珠江	2012-12	16	126	3 829	564		[116]
珠江	1997	16	1 434	10 811	4 892		[117]
珠江	2009	16	4 788	8 665	7 078		[118]
珠江三角洲	2002-01	25	294	1 100	749		[119]
珠江三角洲	1997-03	16	323	14 812			[120]
珠江口		16	138	1 100			[121]
鄱阳湖	2011-05	16	33	369	157 ±63		[122]
鄱阳湖	2012-08	16	73	367	193		[166]
太湖梅梁湾	2011-11	16	782	1 105	944		[167]
太湖梅梁湾	2007	16	180	1 600			[151]
太湖梅梁湾	2003-07	16	1 207	4 754	2 563		[168]
太湖梅梁湾	2005-11	16	1 690	5 033	3 498		[169]
太湖梅梁湾	2003	16	2 092	4 094			[170]
太湖五里湖湾	2000	16	13	963			[171]
谭江	2010-11	16	57	1 564	551		[172]

续表 1

位置	采样时间（年-月）	样本数（n）	最小值 /$ng \cdot g^{-1}$	最大值 /$ng \cdot g^{-1}$	平均值 /$ng \cdot g^{-1}$	主要成分	文献	位置	采样时间（年-月）	样本数（n）	最小值 /$ng \cdot g^{-1}$	最大值 /$ng \cdot g^{-1}$	平均值 /$ng \cdot g^{-1}$	主要成分	文献
青海湖	2012	16	366	966			[131]	三峡水库	2016	16	186	2 056	726		[173]
琼州海峡	2015	17	21	990			[132]	天津河		16	370	5 607	2 041		[174]
泉州湾	2009-03	16	183	721			[133]	天目湖		16	288	714	459	高环	[169]
三沙湾	2013-05	16	8	353			[134]	北京通惠河	2002-04	16	127	928	540	2 环和 3 环	[175]
赛里木湖	2013-01	16	129	643		2 环和 3 环	[135]	望花湖	2007	16	215	1 308	694	2 环和 3 环	[86]
第二松花江	2006-01	5	447	20 331	3789		[136]	渭河	2016	16	113	1 703			[176]
上海城市湖		16	56	4 928	1 131	4 环和 6 环	[137]	渭河	2015-08	16	459	7 576			[177]
珊珀湖		16	91	901	365	萘和芴	[138]	微山湖	2009-07	16	325	1 577	699	高环	[178]
嵊泗海，黄海	2017-06	16	46	196	109		[139]	卧龙湖		12	1 413	3 948		茚苯[1,2,3-cd]芘	[179]
深圳湾	2014	16	225	2 205			[140]	乌梁素海	2014-08	16	129	2 162	405		[39]
深圳西海	2014	15	102	2 336			[140]	乌梁素海	2011-08	16	293	2 372	915		[39]
石梁河水库	2014		17	840	214		[141]	乌梁素海	2008	15	119	568			[180]
玛珥湖	2009	16	474	2 289			[142]	厦门港	1998-07	16	247	480	367	3 环和 4 环	[181]
松花湖	2006	16	2 948	17 180	7 119		[143]	厦门港和筼筜湖	2002	16	98	1 377			[182]
松花江	2008-01	16	68	654	234		[144]	西江	2014	16	53	141	92		[183]
南海	1993	16	25	275	146		[145]	新丰江水库	2016	16	56	381			[184]
南海北	2015	15	11	66			[146]	杏林湾	2012-01	16	413	2 749	950		[185]
武汉南湖		16	336	5 358			[147]	鸭绿江口		16	68	1 500			[186]
苏北浅滩	2017	16	nd	25	6		[148]	羊卓雍错	2012-08	16	7	58	19	菲和䓛	[187]
苏州河		16			1 269		[149]	长江口和沿海	2006	18	235	1 516	605	2 环和 3 环	[188]
太湖	2015	11	nd	280			[150]	长江口		16	737	1 812	1 154		[188]
太湖	1997	16	877	5 730			[151]	长江口		16	38	308			[189]
太湖	2014	16	12	2 281			[152]	长江口		16	65	955			[190]
太湖	2007-07	16	91	1 040	63		[153]	长江口		16	90	502			[191]
太湖	2017-01	12	164	520	276		[154]	长江口	2005-12	17	107	633	309		[77]
太湖	2010-05	15	209	3 843	584		[155]	长江武汉段	2005	16	31	4 813	1 315	2 环和 4 环	[192]
太湖	2010	15	255	1 059	580 ± 236		[156]	长江宜宾至泸州段	2015-08	16	5 071	9 324	7 000		[193]
太湖		15	257	1 709	829	高环	[157]	阳宗海		14	201	1 914		2、4 和 6 环	[128]
太湖	2011-05	16	262	1 087	491		[158]	澄海湖	2013	16			659		[194]
太湖		16	280	840	460		[159]	黄河三角洲		16	11	252			[195]
太湖		16	379	1 047			[160]	渤海口		16	100	537			[196]
太湖	2000-02	16	410	5 260	1437		[161]	黄河，河南	2004-06	13	31	133	77		[106]
太湖	2014	16	430	831	572		[162]	黄河，河南	2011	15	36	45 862			[197]
太湖	2009	15	437	1 335	957 ± 67		[163]	黄河，兰州	2003-11	16	7	98			[198]
太湖水体		16	11	210			[152]	黄河，兰州	2003-07	7	778	12 357	4 680	4 环	[106]
太湖竺山湾	2012-11	16	1 099	1 167	1133		[164]	扎龙湿地	2007-01	15	31	290	130		[199]
太湖梅梁湾	2009-12	9	612	1 703	1257		[165]	漳卫新河		16	23	10 209			[102]

1）空白处为文献中没有对应的参数

Science 和 CNKI. 通过数据筛选,识别并丢弃噪声数据(即样品来自点源污染附近区域和远离人类活动的背景区域). 本文所采集的 PAHs 数据90% 以上为 16 种 PAHs,故统称为 16 种 PAHs. 所有 PAHs 分析的样品回收率在 47.2%~119.4% 之间,平行样品的相对标准偏差小于 15%; 本研究采用柯尔莫哥洛夫-斯米尔诺夫检验法检验正态分布. 当数据不符合正态性假设时,使用曼-惠特尼和克鲁斯卡尔-沃利斯检验; 当数据不符合正态分布,则进行斯皮尔曼检验. 统计学显著性设定为 $P<0.05$ 水平.

1.2 贝叶斯结构方程模型

为了避免高度相关变量之间的干扰,首先采用主成分分析进行降维分析. 从 29 个人为参数中提取 6 个主成分(表 2),分别归为 6 个因素,即能源消耗、经济发展、城市化水平、植被、工业化和森林火灾. 从 6 个自然参数(平均气压、平均气温、极端最低气温、降水量、日照时数和相对湿度)中提取出 1 个成分,归为气候因素.

表 2 人为参数的旋转分量矩阵1)

Table 2 Rotation component matrix of man-made and natural parameters

项目	组分					
	1	2	3	4	5	6
人口	0.49	0.78	−0.09	−0.12	0.07	0.14
人口密度	−0.07	0.06	0.85	−0.16	−0.17	−0.09
国内生产总值/区域	−0.12	0.00	0.87	−0.10	−0.20	−0.11
城市化	0.02	0.09	0.90	−0.16	−0.09	0.12
国内生产总值	0.41	0.83	0.28	−0.07	0.02	0.04
第一产业占国内生产总值的比例	−0.04	−0.13	−0.81	0.04	−0.20	0.17
第二产业占国内生产总值的比例	0.29	0.13	−0.18	0.02	0.86	0.04
第三产业占国内生产总值的比例	−0.23	−0.04	0.64	−0.04	−0.65	−0.14
公路交通客运	0.11	0.82	−0.14	−0.20	0.10	0.11
水路交通客运	−0.06	0.72	−0.03	−0.09	−0.08	0.28
公路交通货运	0.54	0.64	−0.07	−0.05	0.29	0.19
水路交通运输	−0.12	0.74	0.30	−0.11	0.30	−0.06
森林火灾面积	−0.00	0.07	−0.10	0.39	0.13	0.73
草原火灾区域	0.24	−0.20	0.17	0.78	0.29	0.12
烟尘总排放量	0.92	−0.03	−0.15	−0.03	0.13	0.01
氮氧化物总排放量	0.89	0.35	−0.04	0.09	0.16	−0.05
二氧化硫总排放量	0.90	0.19	−0.16	0.10	0.14	0.03
城市污水	0.34	0.89	0.17	−0.08	0.06	0.10
总用水量	0.20	0.70	−0.21	0.18	0.01	−0.12
耗煤量	0.92	0.16	−0.00	0.15	0.14	−0.07
焦炭消耗量	0.79	0.09	−0.04	−0.18	0.12	−0.06
石油消耗量	0.54	0.54	0.27	−0.12	−0.20	0.05
城市天然气供应	0.04	0.75	0.41	0.02	−0.15	0.01
总热功率	0.75	0.44	0.16	0.14	0.17	−0.19
森林面积	−0.02	0.04	−0.39	0.78	−0.04	0.38
森林保护区	−0.17	−0.02	−0.39	0.69	−0.24	0.15
森林覆盖率	−0.25	0.29	−0.18	−0.12	−0.01	0.83
草原地区	−0.06	−0.23	−0.23	0.82	0.01	−0.34
总能量输出	0.87	0.05	0.04	−0.09	−0.16	−0.07
方差/%	23.22	21.43	15.17	9.80	6.44	6.36

1)提取方法:主成分分析、旋转法和 Kaiser 标准化最大方差法

为构建非信息先验贝叶斯结构方程模型,假设: ① 所有参数均为截面数据,利用气候因子与各种人为因素之间的潜在因果关系表征其空间和年际变化,并综合考虑了不同地区气温和降水的差异; ② 对于中间组分,减小环境投资可能直接影响大气污染物排放. 同时,环境投资也会受到经济发展和城市化水平的影响,而森林火灾也可能影响植被. 其中,经济发展用 GDP、交通和用水量来衡量; ③ 气候因素对沉积物中 PAHs 的影响途径分为直接(大气沉降)和间接影响(改变人为因素的参数); ④ 人为因素会直接或间接影响 PAHs 的分布. 贝叶斯估计依赖于 Gelman-Rubin 收敛. 为了避免局部最优解,将迭代次数设置为100 000次,并丢弃前5 000次数据. 潜在的规模折减系数小于 1.1,表明模型达到

了收敛性[200]. 采用后验预测 P 值和卡方差值 95% 置信区间对模型拟合进行判断. 当后验预测 P 值在 0.5 左右[201]，卡方差异值的 95% 置信区间以零为中心且基本对称，说明模型拟合良好. 若 95% 置信区间路径系数为零，说明研究假设只有部分得到支持，否则说明研究假设均可得到支持.

1.3　重力模型

重力模型计算公式如下：

$$I_{ij} = GP_iP_j/d_{ij}^r$$

式中，I_{ij}为地区 i 和 j 之间的重力，P_i 和 P_j 分别为地区 i 和 j 的质量，d_{ij}为两个地区之间的距离，G 为重力系数，r 为重力衰减系数($r=2$).

沉积物 PAHs 表征网络计算公式如下：

$$T_{ij} = C_iC_j/d_{ij}^2$$

式中，C 为沉积物中 16 种 PAHs 总含量的平均值.

人类活动关系(国内生产总值、人口、用水量、道路客运和货物运输)的计算公式如下[202]：

$$H_{ij} = \sqrt[5]{E_iN_iW_iT_iF_i} \cdot \sqrt[5]{E_jN_jW_jT_jF_j} \;/\; d_{ij}^2$$

式中，E 为国内生产总值，N 为总人口，W 为用水量，T 为公路客运量，F 为公路货运量，H 为社会经济发展指数，d 为两个地区重力点之间的距离[203].

2　结果与讨论

2.1　沉积物中 PAHs 的时空分布

中国沉积物中 16 种 PAHs 总量的地理分布见图 1. 污染水平最高的是 2003 年天津市河流(775 ~ 255 372 ng·g^{-1}，平均值：27 074 ng·g^{-1})，3 环和 4 环约占总含量的比例分别为 44% 和 41%，主要来源于附近的矿物和煤炭燃烧[68]. 污染水平最低的是 2017 年江苏省苏北浅滩(0 ~ 25.2 ng·g^{-1}，平均值：5.88 ng·g^{-1})，4 环 PAHs 占 42%，其次为 5 环 PAHs(占 35%)，主要来自高温燃烧[148]. 根据国务院发展研究中心《地区协调发展的战略和政策报告》将中国大陆划分的 8 个经济区，不同地区沉积物中 PAHs 含量分布由高到低依次为：北部沿海 > 东北 > 东部沿海 > 南部沿海 > 黄河中游 > 长江中游 > 西南 > 西北. 总体而言，东部高于西部；河口低于河流；北方高于南方.

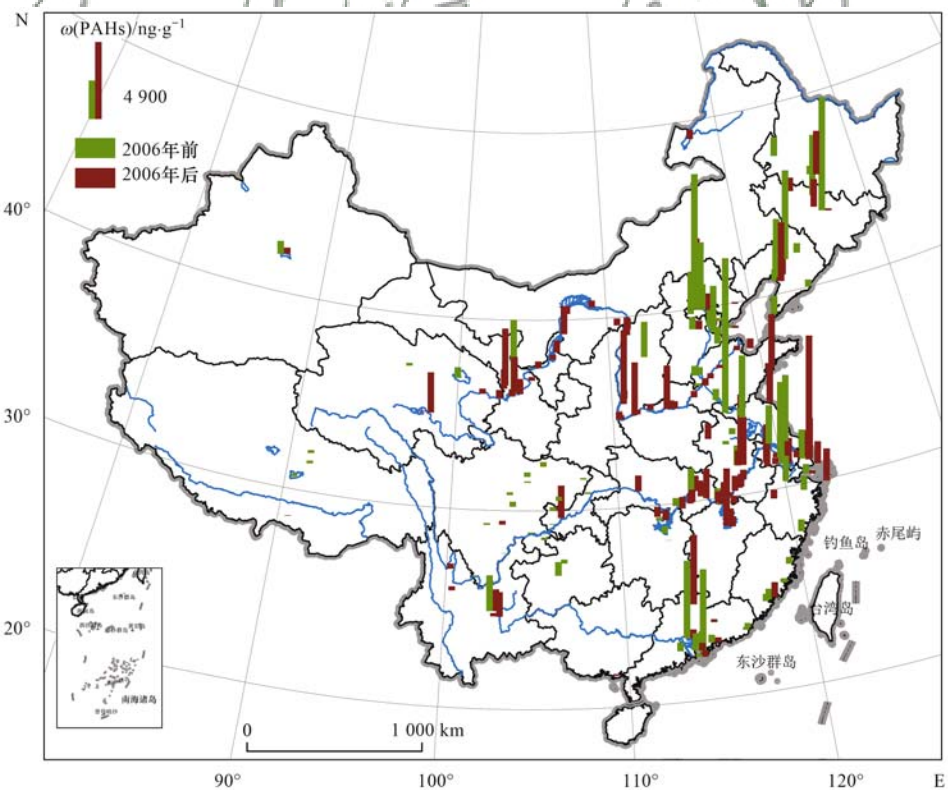


图 1　沉积物中 PAHs 含量地理分布

Fig. 1　Geographic distribution map of PAHs concentration in sediments

PAHs 含量的时空变化趋势表明，沉积物中 PAHs 的含量从 2000 年起逐渐增加，2006 年以后逐渐减少(图 2). 2006 年以前，PAHs 高污染区域主要集中在沿海地区，如东北、京津冀、长三角和珠三角地区，这些地区的城市化水平很高，人口和工业集中. 东北地区污染网络的中心位于辽河流域，该地区的经济发展以重工业为主. 北部沿海地区污染最严重的地方是海河下游. 长江中游污染网中心点位于京杭大运河的安徽段[15]，表明航运对内河运河中沉积物的污染较大. 而东部沿海地区和南部沿海地区

的污染网络中心是黄浦江和珠江,分别位于上海和广州,这些地区作为经济发达地区,机动车排放源的污染贡献率相对较大[77,120]. 2006 年以后,长江中游和黄河中游污染网络密度增加,黄河中游的污染网络节点位于三门峡段(渭河)[176]. 上游地区宝鸡、西安等城市沉积物中 PAHs 含量也随之增加,可能与当地化学工业排放和煤炭燃烧有关[177].

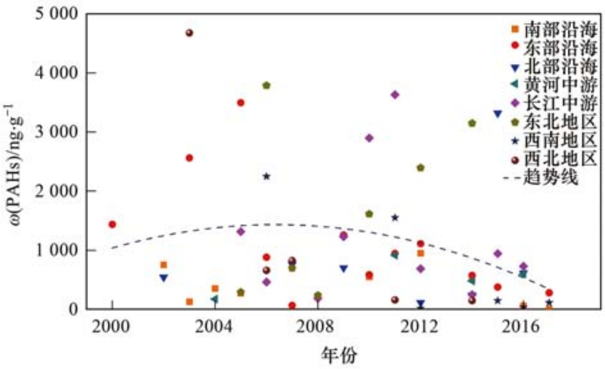


图 2　沉积物中 PAHs 含量的历史趋势

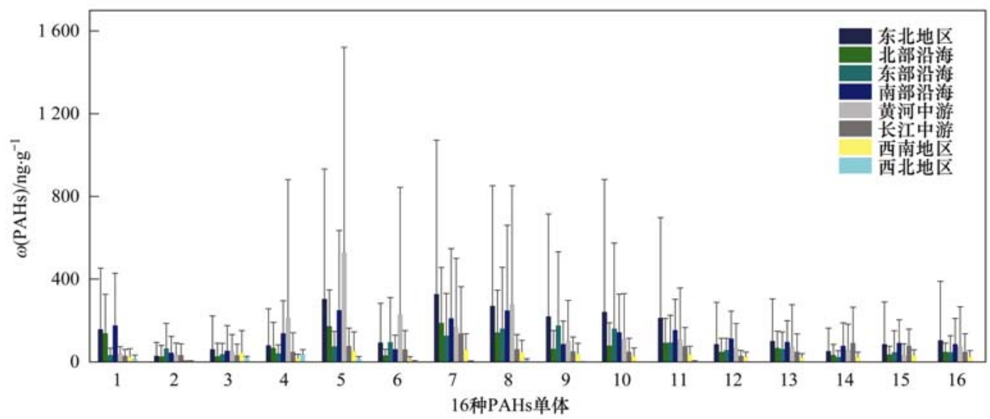
Fig. 2　Historical trends of the PAHs concentrations in sediments

不同地区沉积物中 PAHs 含量到达峰值的年份有显著差异. 北部沿海地区如白洋淀的 ω(PAHs)在 2009 年时出现最大值为2 402 $ng \cdot g^{-1}$,在 2016 年最大值为 861 $ng \cdot g^{-1}$[10,11]. 东北松花江 ω(PAHs)在 2005 年的最大值为15 310 $ng \cdot g^{-1}$,2010 年后,最大值为3 877 $ng \cdot g^{-1}$[204,205]. 东北辽河 ω(PAHs)的中位数由 2009 年的3 281 $ng \cdot g^{-1}$下降至 2014 年的 178 $ng \cdot g^{-1}$[157,206]. 长江中游地区如长江武汉段沉积物中 ω(PAHs)在 2005 年范围为 26 ~ 7 136 $ng \cdot g^{-1}$,在 2016 年范围为 46 ~ 424 $ng \cdot g^{-1}$[192,207]. 东部地区巢湖沉积物中 ω(PAHs)在 2009 年最大值为10 200 $ng \cdot g^{-1}$,但 2016 年时最大值降至1 032 $ng \cdot g^{-1}$[27,28]. 2003、2005 和 2011 年太湖梅梁湾沉积物中 ω(PAHs)分别为2 563、3 498和 944 $ng \cdot g^{-1}$[168~170],说明该地区湖泊污染的治理效果显著. 青海湖、博斯腾湖等偏远地区沉积物中的 PAHs 含量总体较低,但沉积物中的 PAHs 含量呈逐年增加的趋势,且高环 PAHs 的比例逐年上升[208].

2.2　沉积物中 PAHs 的来源解析

在大多数地区,沉积物中 4 环及以下 PAHs 的比例(20.5% ~ 45.1%)高于 5 环和 6 环 PAHs(图 3),这可能与我国以煤炭为主要能源有关[6]. 南部沿海、长江中游和东部沿海地区由于经济发达和交通繁忙,导致高分子量 PAHs 的组成比例高于其他地区. 西北地区低分子量 PAHs 的组成比例显著高于其他地区,可能是由于该地区经济发展水平较低和人口数量较小. 北部(东北、北部沿海和黄河中游)低环 PAHs 含量相对较高,且主要为源自生物质和煤炭燃烧源的菲、芴、芘和萘,这与北方地区的生产和生活能耗一致[5,6]. 南部和东部的 PAHs 含量差异不明显,但高环 PAHs 的比例普遍高于北部,南方的强降雨、高温和长时间的日照是造成这种分布现象的重要因素[209].

为进一步解析不同地区沉积物中 PAHs 的来源,采用特征比值法来确定 PAHs 的可能排放源. 可以看出 2006 年前与 2006 年后 8 个区域的 PAHs 比值有显著变化,不同地区的比值也不相同(图 4),比如 2006 年前 8 个区域 Ant/(Ant + Phe)的比值普遍比 2006 年后的大,表明化石能源消费的增加加剧我国内陆沉积物中 PAHs 污染[210~212]. 黄河中游地区作为我国最大的煤炭开采和深加工基地,沉积物中 Ant/(Ant + Phe)比值最大且 2006 年后明显减少,可能是由于 2006 年后我国对煤炭开采进行严格管理



1. 萘(Nap),2. 苊烯(Acy),3. 二氢苊(Ace),4. 芴(Flu),5. 菲(Phe),6. 蒽(Ant),7. 荧蒽(Fla),8. 芘(Pyr),9. 苯并[a]蒽(BaA),10. 䓛(Chry),11. 苯并[b]荧蒽(BbF),12. 苯并[k]荧蒽(BkF),13. 苯并[a]芘(BaP),14. 二苯并[a,h]蒽(DahA),15. 茚并[1,2,3-cd]芘(InP),16. 苯并[ghi]苝(BghiP)

图 3　中国 8 个地区的 PAHs 单体含量

Fig. 3　PAH concentrations in eight Chinese regions

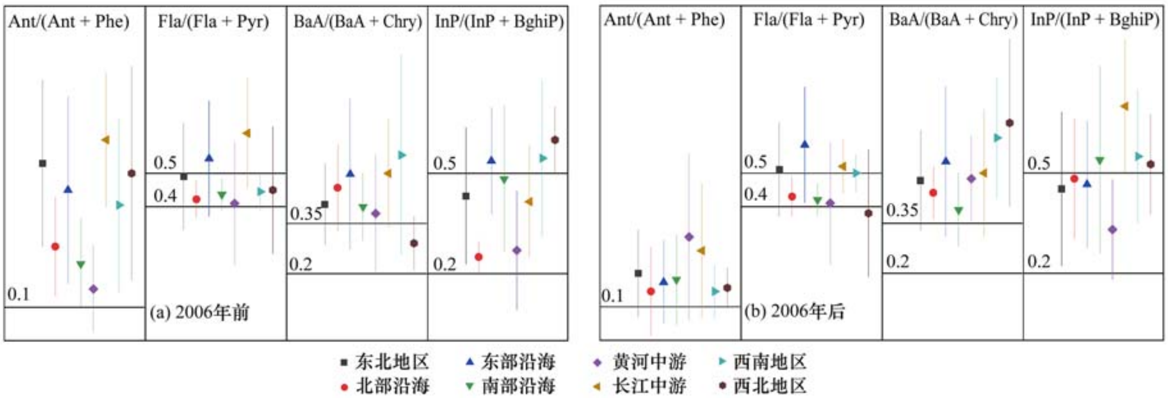


图 4　中国 8 个区域的 PAHs 比值

Fig. 4　Ratio of PAHs in eight Chinese regions

并修订《中华人民共和国煤炭法》，此外，2006 年后西北地区 BaA/(BaA + Chry)比值显著比 2006 年前大，可能是由于 2006 年以后西部地区经济快速发展引起机动车数量剧增. 西北地区和西南地区的 InP/(InP + BghiP)和 BaA/(BaA + Chry)的值较高，可能是由于其经济发展水平较低，大量燃烧草、木和煤作为供热和发电[213]. 而经济较为活跃的东部沿海地区的 Fla/(Fla + Pyr)比值一直较高，反映了其地区人口数量多和交通发达[214].

2.3　人为和自然因素与沉积物中 PAHs 分布的关系

利用贝叶斯结构方程模型计算得到各社会经济因素对 PAHs 含量影响的路径系数，从而构建了人类活动强度和全国沉积物中 PAHs 污染分布网络. 根据标准化路径系数(λ)从大至小，影响沉积物中 PAHs 的分布的因素分别为：经济发展($\lambda = 0.33 \pm 0.18$)、能源消耗($\lambda = 0.20 \pm 0.10$)、城市化水平($\lambda = 0.02 \pm 0.01$)、植被($\lambda = -0.25 \pm 0.12$)、森林火灾($\lambda = 0.13 \pm 0.17$)和工业化($\lambda = 0.06 \pm 0.03$). 从环境投资和能源消费的角度看，经济发展对 PAHs 的分布直接影响和间接影响均最大(图 5). 此外，气候因素和植被对沉积物中 PAHs 的分布显现负相关，因为高温和降水较多的环境能够有效溶解和稀释 PAHs[215]. 植被则具有吸附和降解功能，同时还可以通过减少水土流失，防止 PAHs 的环境迁移. 本研究结果与中国潮间带 PAHs 分布模式影响的最新研究结果不同[209]，可能是不同的研究区域，不同流域和省份的参数带来的差异.

2.4　经济活动关联与 PAHs 分布的关系

中国不同省、市之间存在普遍的空间溢出效应. 从 2000 ~ 2019 年，点向和点出入度发生了显著变化，区域经济活动由少中心向多中心转变(图 6). 经济效益关联区逐年增加，但增长的速度和方式各不相同. 在北部沿海、东部沿海和南部沿海比较发达的地区，增长速度明显加快，而黄河中游和长江中游增长缓慢，西南地区增长最慢.

沉积物中 PAHs 含量的时空变化也显示出类似

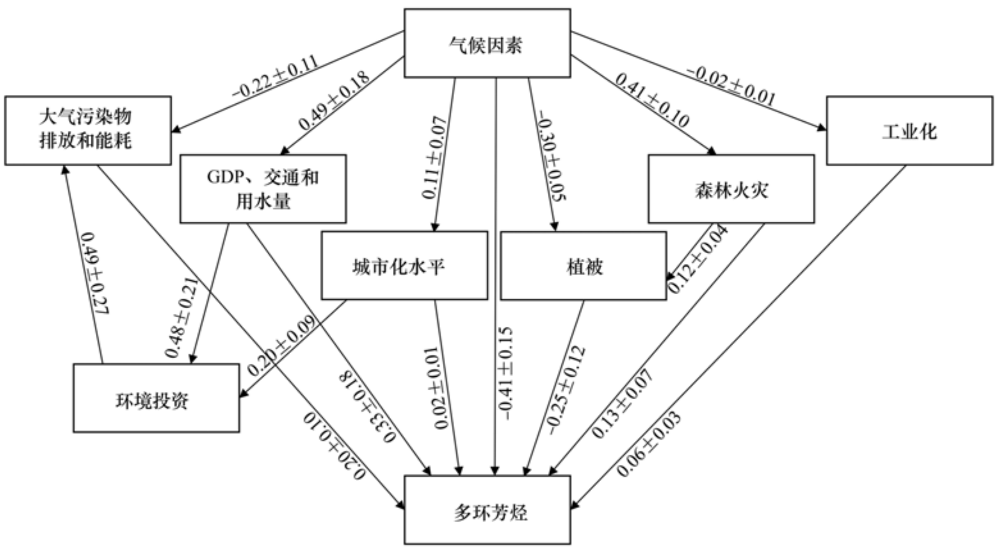


图 5　贝叶斯结构方程模型的结果

Fig. 5　Results of Bayesian structural equation model

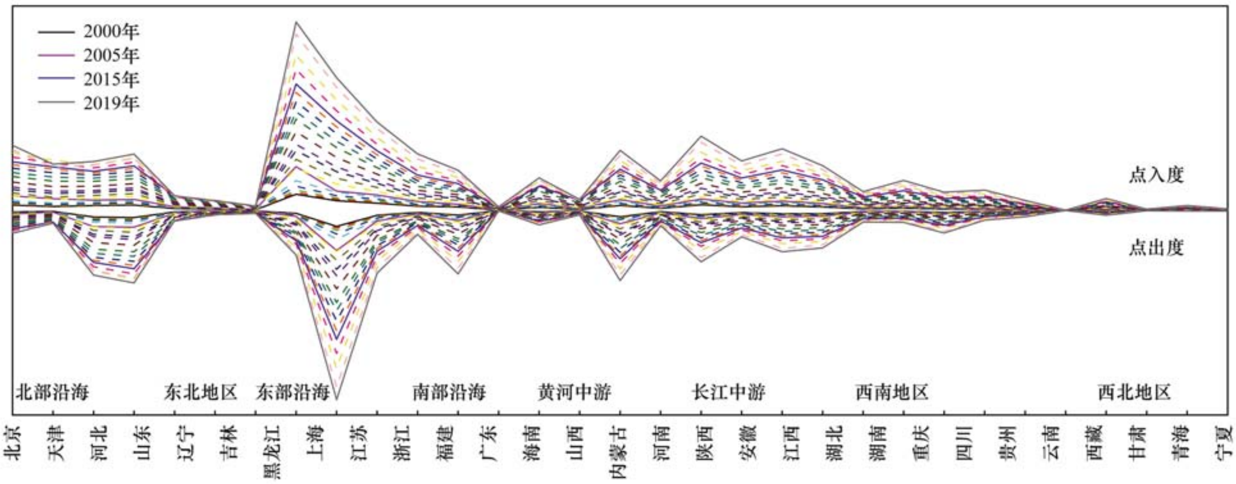


图 6　2000 ~ 2019 年中国 31 个地区的投入产出分析结果

Fig. 6　Input-output analysis results of 31 regions in China from 2000 to 2019

的规律. 2005 ~ 2015 年,人为因素的强相关网络整体密度显著增加,关联区域从 2005 年的沿海地区转移到 2015 年的内陆地区(图 7). 与此同时,沉积物中 PAHs 的时空分布发生了变化,高污染区域也发生了转移. 2005 年前后,高污染区出现在东北辽河,天津海河和广州珠江[38,68,119]. 2015 年前后,黄河中游的高污染区分别为河南黑石关、甘肃兰州、云南滇池、江苏太湖、漳渭新河和上海黄浦江[216]. 随着第二产业比重呈先上升后下降的趋势,并在 2010 年前后出现拐点,PAHs 的污染组分发生了变化,高环 PAHs 的比例逐渐增加,如东北辽河和巢湖[30,94]. 在青海湖和滇池等也出现了类似的变化[45,131]. 由此可见,城市化和工业化对沉积物中 PAHs 的影响显著. 此外,人类活动与污染的相关网络图发生变化,节点数量增加,密集的强相关网络由沿海地区扩展至中西部地区. 随着全国地区经济快速发展,城市化水平高的地区有了足够的财政用于排放控制、清洁生产和专项污染控制等环境治理,从而有效降低了污染

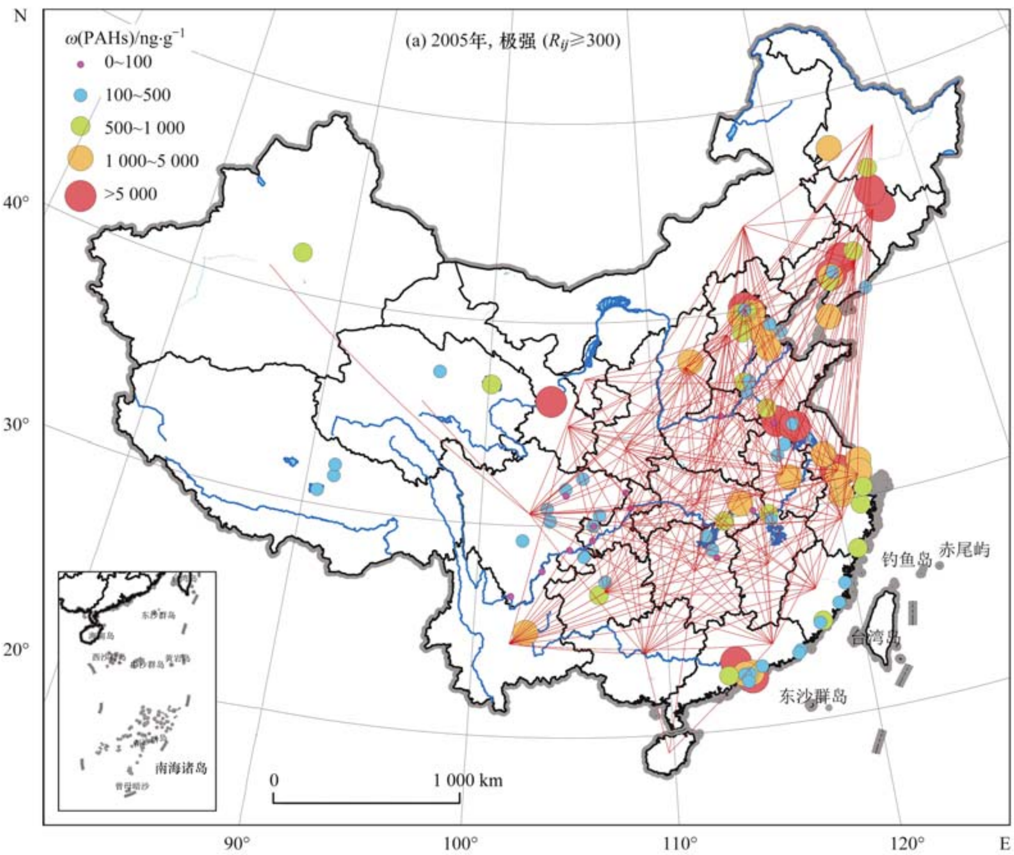
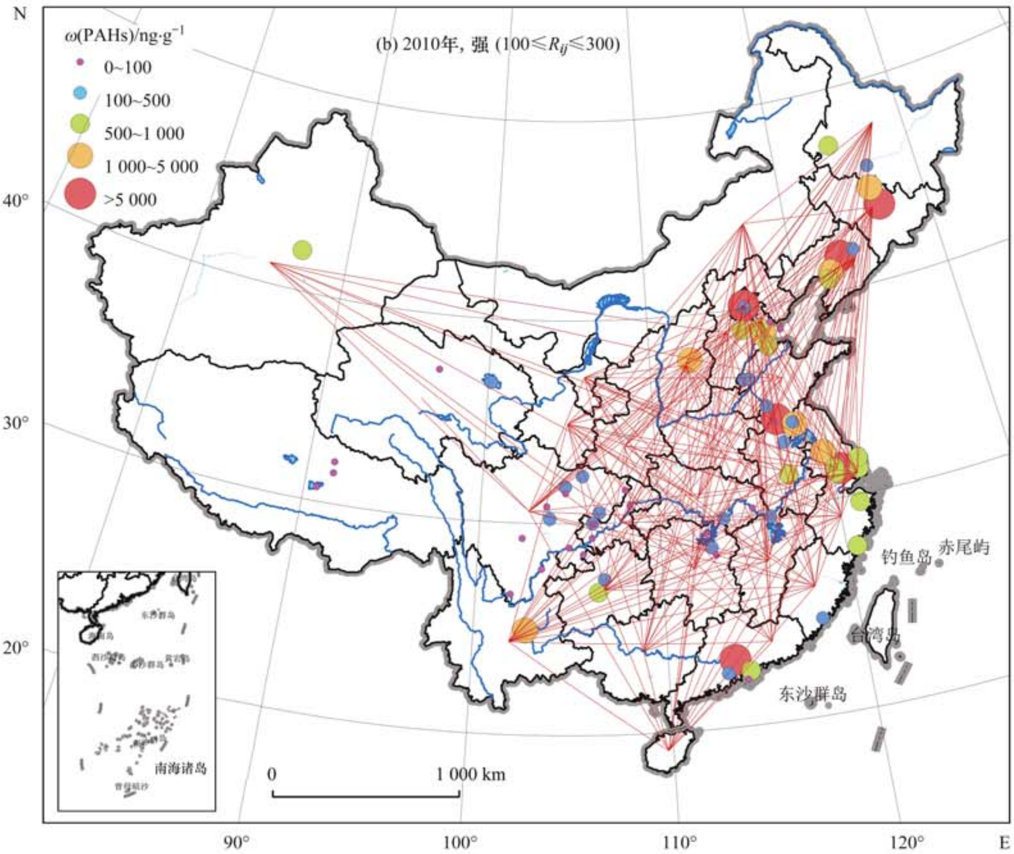
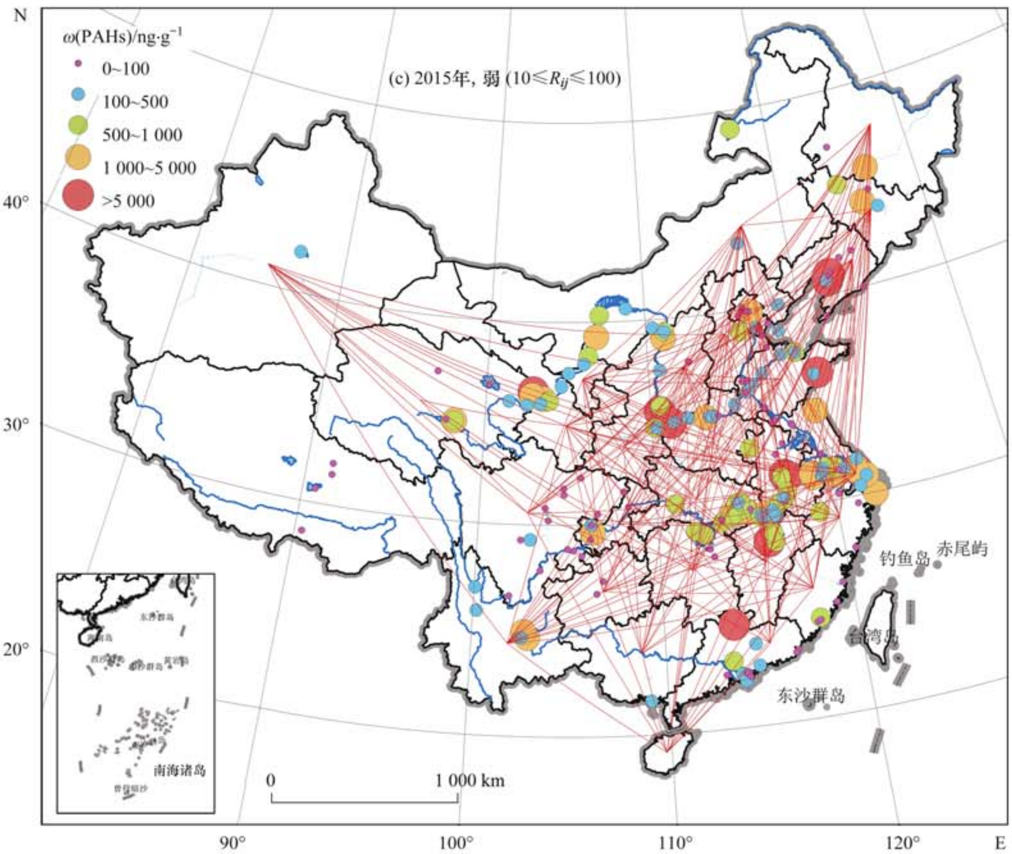


图 7　改进的重力模型结果

Fig. 7　Improved gravity model results

续图7





水平.

总的来说,各地区沉积物中 PAHs 污染程度的减少和增长各不相同,表现出复杂的关联机制. 随着区域经济的发展和关联性的增强,经济发达地区加大了环境管控和治理措施,并促进周边地区的经济活动. 经济发达地区沉积物中 PAHs 的含量在达到峰值后逐渐下降. 发展中地区的经济发展较快,污染物积累较快. 偏远或经济欠发达的地区,污染物含量

逐渐增加,但累积率慢于快速发展地区.

3 结论

(1)高污染区域主要集中在北部和东部沿海地区,沉积物中4环PAHs占比较高,燃煤和生物质燃烧是沉积物中PAHs的主要来源.

(2)通过贝叶斯结构方程模型,得出对沉积物中PAHs分布直接影响最大的因素是经济发展.

(3)沉积物中PAHs的时空分布与经济发展有关.强关联区域的污染网络由沿海地区向内陆地区扩展,内陆地区PAHs含量呈上升趋势,而这与沿海地区情况相反,这表明当区域社会经济发展到一定水平后,由于环境投资的增加和能源结构的调整,沉积物中PAHs含量变化趋势逐渐平稳.

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HUANJING KEXUE

Environmental Science (monthly)

Vol. 44 No. 1

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CONTENTS