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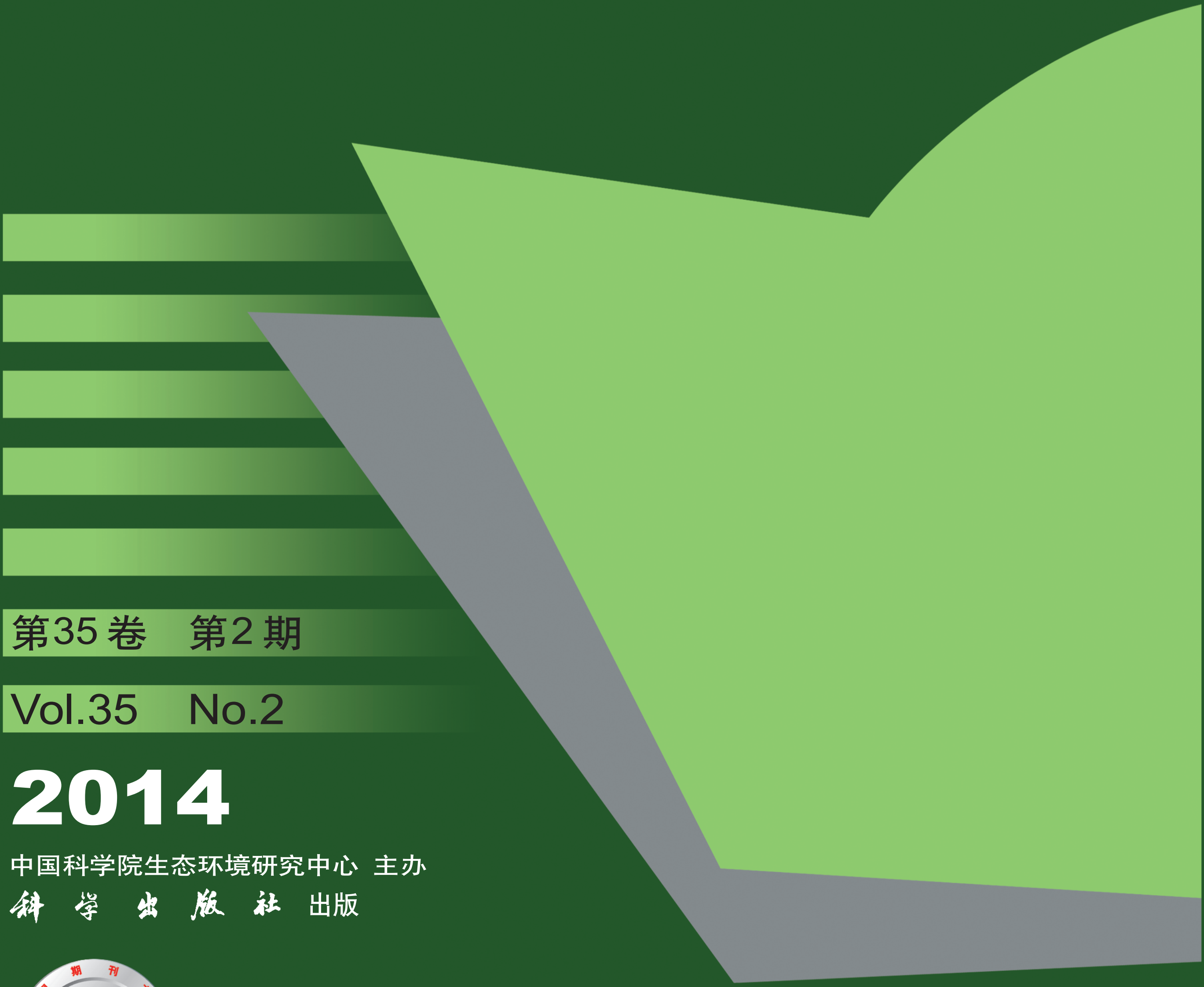
环境科学

(HUANJING KEXUE)

ENVIRONMENTAL SCIENCE



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北京城区PM$_{2.5}$中致癌重金属季节变化特征及其来源分析

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摘要:于2009年4、7、10月和2010年1月在北京城区采集了PM$_{2.5}$样品,采用电感耦合等离子体质谱仪分析得到29种金属元素,对7种致癌重金属浓度、富集程度及其可能的来源进行了分析.结果表明,7种致癌重金属As、Cd、Co、Cr、Ni、Pb和Se年均值浓度分别为(11.6±14.0)、(2.6±2.4)、(1.0±0.7)、(11.3±9.4)、(4.0±2.4)、(142.5±98.9)、(3.3±2.2) ng·m^{-3},其中仅As年均值浓度超过WHO参考限值的0.8倍.7种致癌重金属仅As、Cd、Pb和Se等4种重金属有明显富集现象,富集因子均超过500,其中夏季富集因子明显高于其它季节.春、秋和冬季4种高富集致癌重金属主要来源于北京周边的燃煤和城区机动车排放,夏季则主要来源于区域性污染源的输送.

关键词:PM$_{2.5}$;砷;健康风险;富集;来源

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Seasonal Variation of Carcinogenic Heavy Metals in PM$_{2.5}$ and Source Analysis in Beijing

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Abstract: During April, July, October 2009 and January 2010, daily (24-h average) PM$_{2.5}$ samples were collected at urban sites in Beijing and 29 metal elements were analyzed by the ICP-MS. The characteristics of 7 carcinogenic heavy metal mass concentrations, enrichment, and possible sources were discussed. The annual average concentrations of As, Cd, Co, Cr, Ni, Pb and Se were (11.6±14.0), (2.6±2.4), (1.0±0.7), (11.3±9.4), (4.0±2.4), (142.5±98.9) and (3.3±2.2) ng·m^{-3}, respectively. Only annual average concentration of As exceeded WHO standard by a factor of 0.8. Higher enrichment factors of As, Cd, Pb and Se were found and their enrichment factors exceeded 500. Their enrichment factors in summer were much higher than those in other seasons. The local coal combustion and vehicle exhaust should be the dominant sources for the above four carcinogenic heavy metals in spring, autumn and winter, while regional transportation contributed more in summer.

Key words: PM$_{2.5}$; arsenic; health risk; enrichment; source

城市高浓度的细颗粒物(PM$_{2.5}$)污染不仅会导致能见度下降影响人们的视觉感受还会影响人体健康[1~3].尽管富集在PM$_{2.5}$中重金属占其质量浓度不足5%,但其对人体健康具有重要影响[4].美国1990年空气清洁修正法案列出了188种有害空气污染物(HAPs),其中含锑(Sb)、铅(Pb)、砷(As)、锰(Mn)、汞(Hg)、镉(Cd)、镍(Ni)、铬(Cr)、硒(Se)和钴(Co)等10种重金属[5].美国毒物与疾病登记署(ATSDR)将As、Cd、Co、Cr、Hg、Ni、Pb和Se等重金属列入致癌物质[6].可见,了解城市大气环境中重金属的含量及其可能的来源,对于保护公众健康以及制定切实可行的控措施具有重要意义.

国外对PM$_{2.5}$中重金属的观测主要用于PM$_{2.5}$来源解析以及对人体健康影响研究[7~9],而我国则主要用于PM$_{2.5}$来源解析研究[10~12],很少关注其健康影响.此外,我国新颁布的《环境空气质量标准》(GB 3095-2012)仅将Pb列入常规监测指标,而将Cd、Hg、As以及Cr(Ⅵ)列入附录A的参考指标,

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且其它致癌重金属尚未涉及. 由此可见,我国对大气环境中致癌重金属关注还不够.

北京作为我国的首都,同时也是我国政治、经济和文化中心,人口超过2 000万. $PM_{2.5}$浓度一直处于较高的水平,对公众的健康影响不容忽视. 过去十几年,北京开展了大量关于$PM_{2.5}$的污染特征及其来源解析的研究[13~16],而对$PM_{2.5}$中重金属的研究相对较少[17~21],尤其对$PM_{2.5}$中致癌重金属污染特征的长时间序列的观测研究更少[22]. 本研究收集了2009~2010年度北京城区$PM_{2.5}$中主要重金属的较长时间序列的浓度资料,与世界卫生组织(WHO)相关标准进行对比,同时分析了致癌重金属的季节变化特征及其可能的来源,以期为环保部门制定相关控制措施和政策提供科学依据.

1 材料与方法

1.1 采集地点

采样地点设在北京大学理科楼顶(39°59′N, 116°18′E),距离地面约25 m,采样点周围是校园和商住区, 5 km范围内无明显大气污染源,周围无建筑物遮挡,视野比较开阔.

1.2 采样方案及采样方法

于2009年4月1 ~ 30日(代表春季)、7月1 ~ 31日(代表夏季)、10月1 ~ 31日(代表秋季)和2010年1月1 ~ 31日(代表冬季)利用一台frmOMNITM(BGI Co., USA) 颗粒物采样器($PM_{2.5}$切割头)每天采集一张Teflon膜(Whatman PTFE, England) 样品. 采样时间为当日的10:00到次日的10:00,采样器流量为5.0 $L\cdot min^{-1}$,Teflon膜采样前后放在恒温恒湿箱(温度20 ~ 23℃,相对湿度35% ~ 45%) 平衡24 h,再用精度为1 μg电子天平(ME36, Sartorius, Göttingen, Germany) 进行称量,采样完成后样品放入冰箱冷冻柜中 -18℃保存待进行化学分析.

1.3 其它资料

收集2009 ~ 2010年北京市气象台(编号54511, 39°48′N, 116°28′E) 风速风向资料. 收集2009 ~ 2010年海淀区万柳环境监测站SO_2和NO_2日均值资料.

1.4 重金属分析方法

将Teflon膜用陶瓷剪刀切割成两份,其中1/2张Teflon膜用混酸(5 mL HNO_3 + 2 mL HF, Merck, Germany) 溶解,放入高通量密闭微波消解系统(MARSXpress, CEM, Matthews, NC) 进行消解,具体消解程序见文献[23]. 消解溶液利用电感耦合等离子体质谱(ICP-MS)测量Al、Fe、Na、Mg、K、Ca、Ba、Ti、Mn、Co、Ni、Cu、Zn、Mo、Cd、Sn、Sr、Sb、Pb、Tl、Se、Ge、Cs、Ga、V、Cr、As、Se和Rb等合计29种元素[24]. ICP-MS质控程序参照美国NIST SRM-1648(城市颗粒物),其中Se, As, Cs, Sb和Rb回收率 < ±15%,其余元素回收率 < ±10%.

2 结果与讨论

2.1 致癌重金属时间变化特征

尽管本研究获取$PM_{2.5}$中29种金属元素浓度,但本研究仅重点讨论As、Cd、Co、Cr、Ni、Pb和Se等7种致癌重金属(其中颗粒态的Hg未能用ICP-MS测量). 整个观测期间,北京城区$PM_{2.5}$年均值为(135 ± 63) $\mu g\cdot m^{-3}$,约是世界卫生组织(WHO)过渡时期目标$PM_{2.5}$年均值(35 $\mu g\cdot m^{-3}$)的4倍. As、Cd、Co、Cr、Ni、Pb和Se年均值浓度分别为(11.6 ± 14.0)、(2.6 ± 2.4)、(1.0 ± 0.7)、(11.3 ± 9.4)、(4.0 ± 2.4)、(142.5 ± 98.9)、(3.3 ± 2.2) $ng\cdot m^{-3}$,合计占$PM_{2.5}$质量浓度的(0.13 ± 0.05)%,其中As、Cd、Ni和Pb年均值是WHO参考限值的1.8、0.5、0.2和0.3倍,仅As超标.

本研究收集了采用ICP-AES或ICP-MS分析方法获取的国内主要城市致癌重金属的浓度资料(表1). 由表1可知,尽管北京As年均值超过WHO标准6.6 $ng\cdot m^{-3}$,但是从2006年以来呈现逐年下降的趋势,Cd和Pb(2008年除外)浓度比较稳定,无显著变化. 与上海比较,北京As和Cr年均值浓度较低,Pb年均值则略高,其它致癌重金属年均值浓度基本相当. 与广州相同季节比较,广州As、Cd、Cr、Pb和Se浓度显著地高于北京. 而与香港相同季节比较,北京As、Cr和Pb浓度明显高于香港,但Ni浓度则比较接近. 综合来看,北京$PM_{2.5}$中7种致癌重金属浓度水平略高于香港,但低于上海和广州.

就季节变化而言,Cd,Pb和Se浓度季节变化不明显,Co和Ni具有相同的季节变化特征:冬 > 春 ≥ 秋 > 夏. As和Cr季节变化特征则相对比较明显,分别为:夏 > 春 > 秋 > 冬和秋 > 冬 > 春 > 夏. 结合图1可知,除了个别样品,Cd、Pb和Se与$PM_{2.5}$的变化趋势基本一致,且可能与$PM_{2.5}$主要贡献源燃煤密切相关. Co和Ni二者变化趋势基本一致,但与$PM_{2.5}$的变化趋势差异明显,可判断二者可能共源,但来源于燃煤的可能性较小. 通常情况下,大气中Ni主要

来源于燃煤,然而其浓度变化与典型燃煤元素 Cd 等元素差异明显,因此可以判断其可能主要来源于机动车或非移动式柴油车[28]. 值得注意的是,As 和 Cr 浓度相对较高,尤其是 As 年均值浓度超过 WHO 标准,然而 As 和 Cr 分别在夏季和秋季出现明显较多的峰值,且与 $PM_{2.5}$峰值出现的时间明显不同. 由此可推断,夏季和秋季某种特殊污染源对 As 和 Cr 浓度贡献显著.

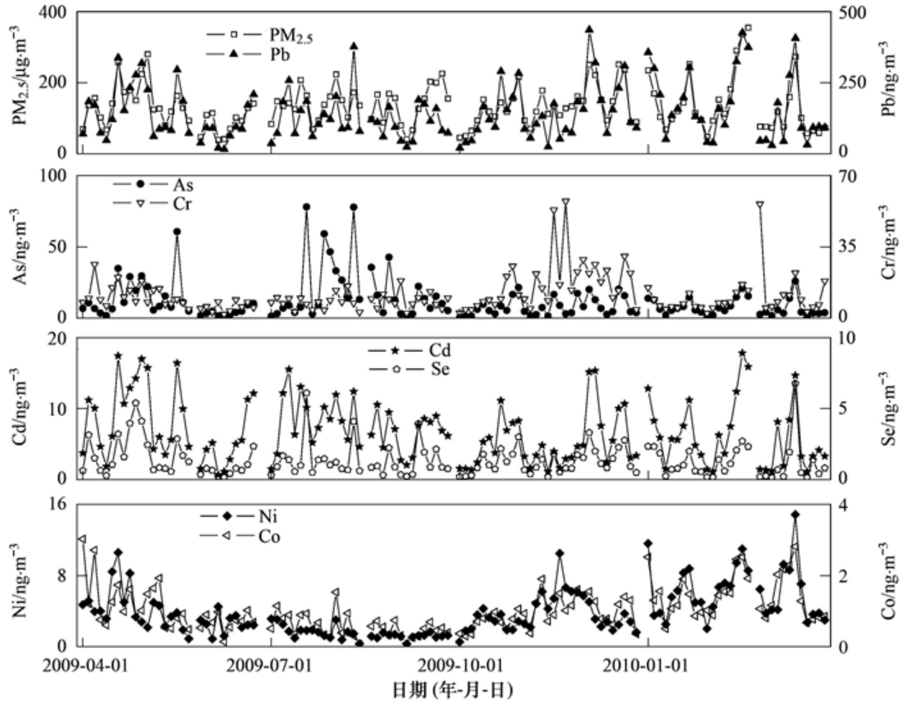

图 1　北京城区 $PM_{2.5}$中致癌重金属质量浓度逐日变化特征

Fig. 1　Daily variation of carcinogenic heavy metals in $PM_{2.5}$ in Beijing

表 1　我国主要城市 $PM_{2.5}$中致癌重金属浓度/$ng\cdot m^{-3}$

Table 1　Concentrations of carcinogenic heavy metals in $PM_{2.5}$ in China/$ng\cdot m^{-3}$

城市	时间(年-月)	样本量(个)	As	Cd	Co	Cr	Ni	Pb	Se	文献
北京	2009-04 ~ 2010-01	120	11.6 ± 14.0	2.6 ± 2.4	1.0 ± 0.7	11.3 ± 9.4	4.0 ± 2.4	142.5 ± 98.9	3.3 ± 2.2	本研究
	2009-04	29	11.4 ± 12.9	2.9 ± 2.7	0.9 ± 0.5	9.1 ± 5.5	4.2 ± 2.6	132.9 ± 92.2	3.8 ± 2.6	
	2009-07	30	19.5 ± 21.7	2.7 ± 2.6	0.4 ± 0.2	8.3 ± 3.7	2.3 ± 1.3	122.5 ± 74.2	3.8 ± 1.7	
	2009-10	31	7.9 ± 6.4	2.5 ± 1.7	0.9 ± 0.5	17.1 ± 13.1	3.8 ± 1.7	146.7 ± 99.1	2.7 ± 1.9	
	2010-01	30	7.6 ± 6.1	2.4 ± 2.6	1.6 ± 0.8	10.4 ± 9.8	5.8 ± 2.6	168.0 ± 122.8	3.1 ± 2.4	
北京	2008	233	10	2	—	–	—	91	—	[22]
	2007		19	3	—	–	—	169	—	
	2006		22	4	—	–	—	191	—	
	2005		12	2	—	–	—	139	—	
上海(宝山)	2004-03 ~ 2005-03	52	32 ± 27	4.6 ± 3.2	1.1 ± 0.5	58 ± 39	20 ± 14	149 ± 122	4.5 ± 2.8	[25]
上海(普陀)			28 ± 19	3.7 ± 1.9	0.8 ± 0.5	15 ± 10	8 ± 3	143 ± 117	3.1 ± 1.9	
上海(黄埔)			36 ± 28	1.7 ± 1.6	0.4 ± 0.2	19 ± 17	6 ± 4	75 ± 74	2.5 ± 2.2	
上海(嘉定)			27 ± 24	1.6 ± 1.0	0.5 ± 0.3	17 ± 15	6 ± 5	67 ± 51	1.6 ± 1.3	
广州(白云)	2004-08 ~ 2004-09	8	41 ± 12	9 ± 3	—	38 ± 11	—	261 ± 76	18 ± 5	[26]
广州(荔湾)		8	81 ± 17	14 ± 3	—	51 ± 11	—	373 ± 80	34 ± 7	
广州(天河)		8	46 ± 10	11 ± 3	—	68 ± 16	—	304 ± 69	25 ± 6	
广州(海珠)		8	64 ± 28	17 ± 8	—	64 ± 28	—	403 ± 178	27 ± 12	
香港(红磡)	2000-11 ~ 2001-02	—	3.93 ± 3.44	—	–	2.43 ± 1.65	5.34 ± 2.74	76.86 ± 33.90	—	[27]
香港(观塘)			5.07 ± 3.99	—	—	4.51 ± 3.67	6.00 ± 5.14	91.62 ± 50.11	—	

2.2 致癌重金属富集程度

富集因子(EFs)用于表征气溶胶中元素相对于地壳表层元素的富集程度[29]. 该方法通常被用于识别气溶胶的元素来源于自然源还是人为源,其计算公式如下:

$$\mathrm{EFs} = (c_{\mathrm{element}}/c_{\mathrm{reference}})_{气溶胶}/(c_{\mathrm{element}}/c_{\mathrm{reference}})_{地壳}$$

式中,c_{element}为元素质量浓度; $c_{\mathrm{reference}}$为参比元素质量浓度.

Al和Fe是常用的参比元素[30],考虑到北京地区Al大气排放源相对于Fe少,因此本研究选取Al作为参比元素. 地壳上层中参比元素浓度资料见文献[31].

由图2可知,As、Cd、Co、Cr、Ni、Pb和Se的EFs年均值分别为508、1 860、4、20、12、599和3 145. 当EFs < 10时,通常认为该元素没有富集[32]. 按照该判别标准,As、Cd、Pb和Se富集程度明显,Cr和Ni富集程度较低,而Co几乎没有富集. 由此可见,As、Cd、Pb和Se富集程度高与人为源排放密切相关. 从各个致癌重金属EFs季均值来看,As、Cd、Pb和Se夏季EFs显著高于其它季节. 除As外,其它6种致癌重金属的季节均值浓度最大值均未出现在夏季,但其EFs最大值却出现在夏季,其主要原因是夏季参比元素Al浓度最低,为0.69 $\mu g \cdot m^{-3}$,约是其它季节的0.3倍,从而使得夏季气溶胶中致癌重金属元素显著富集.

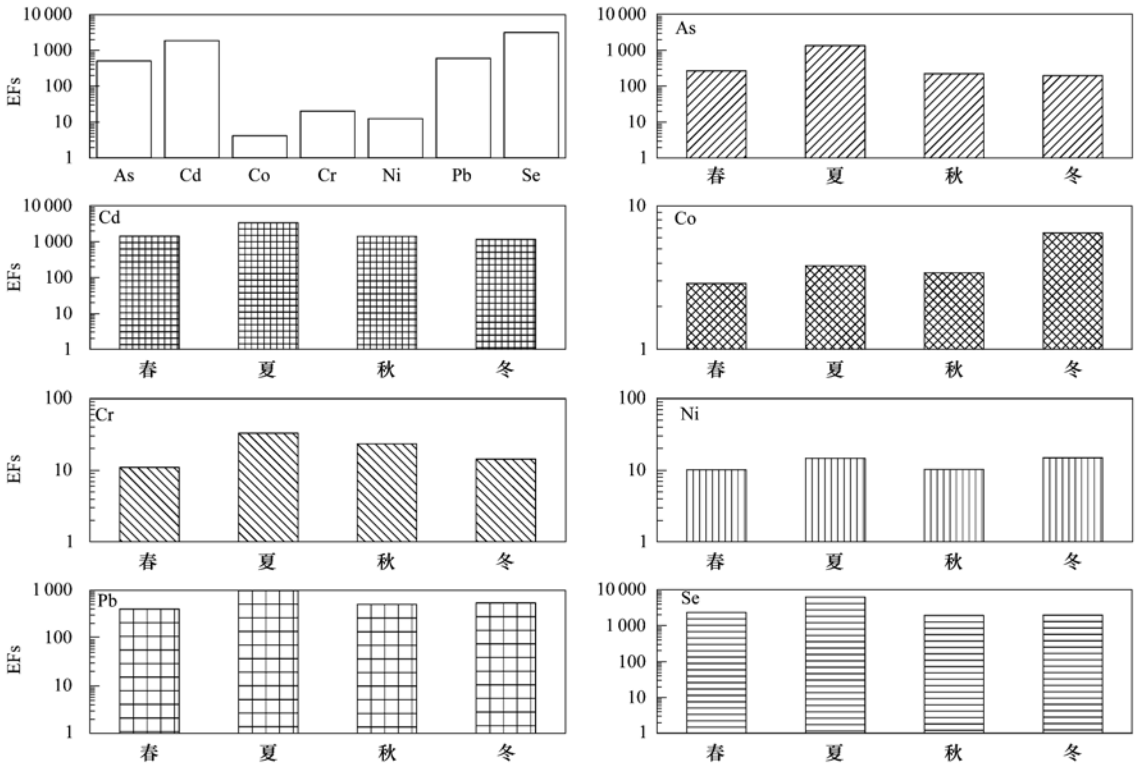

图2 北京城区致癌重金属的富集因子年均值和季节变化

Fig. 2 Annual average enrichment factors and their seasonal variations in Beijing

2.3 致癌重金属来源分析

为了解4种高富集致癌重金属可能的来源,对As、Cd、Pb、Se、Al、SO_2和NO_2等7种污染物进行相关性分析(表2). 由表2可知,地壳元素Al与上述致癌重金属无相关性,表明上述致癌重金属主要来源于人为排放. 春季4种致癌重金属与SO_2之间的相关性较好,表明4种致癌重金属与北京周边燃煤源密切相关. Pb和Se与NO_2也有较好的相关性,表明机动车排放对Pb和Se也有一定的贡献. 秋冬季4种致癌重金属与SO_2和NO_2相关性相对较好,表明上述致癌重金属与燃煤和机动车排放关系密切. 值得注意的是,夏季4种致癌重金属与SO_2之间几乎不相关或相关性较差,与NO_2不相关,表明夏季上述致癌重金属元素主要来源于区域性输送.

本研究采用PSCF (potential source contribution function) 模型来进一步了解4种高富集致癌重金属可能的来源以及输送路径. 由图3可知,As的PSCF高值区主要分布在石家庄至天津污染带上,甚至山

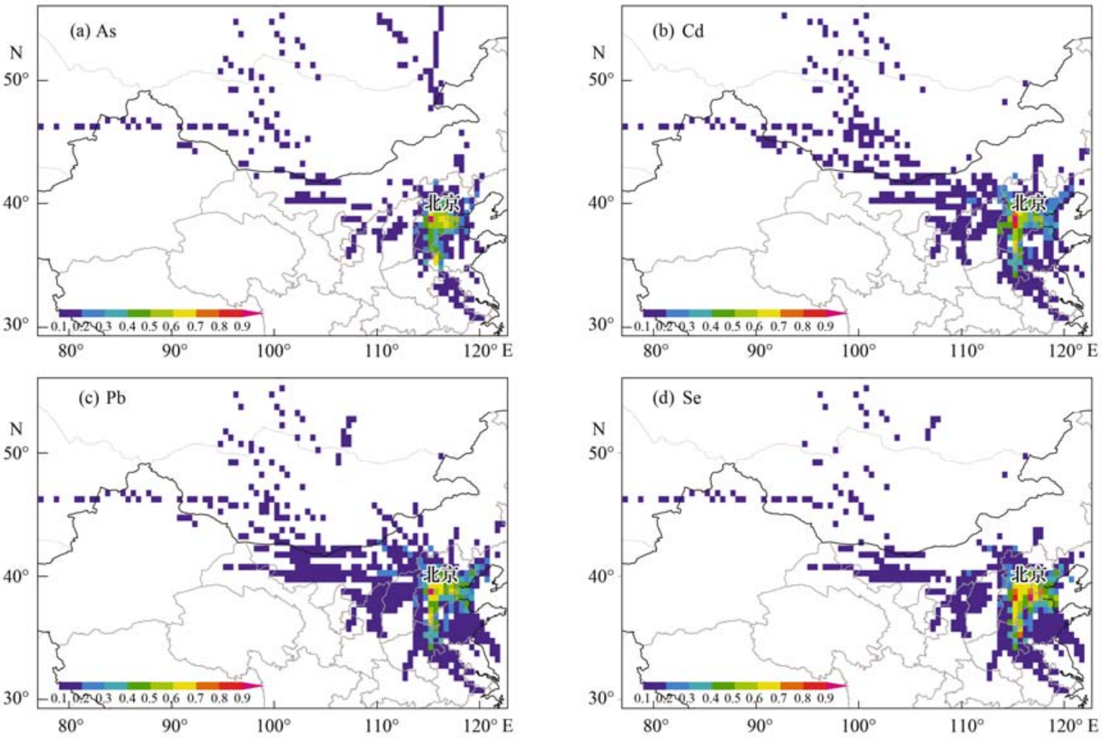

图 3　北京城区高富集致癌重金属 PSCF 空间分布

Fig. 3　PSCF spatial patterns of higher enrichment carcinogenic heavy metals in Beijing

表 2　北京不同季节高富集重金属之间的相关性(1)

Table 2　Correlation between higher enrichment carcinogenic heavy metals in different seasons in Beijing

季节	污染物	As	Cd	Pb	Se	Al	SO_2	NO_2
春季	As	1.00						
	Cd	0.69**	1.00					
	Pb	0.82**	0.90**	1.00				
	Se	0.79**	0.86**	0.98**	1.00			
	Al	0.21	0.21	0.19	0.15	1.00		
	SO_2	0.74**	0.81**	0.87**	0.88**	0.43*	1.00	
	NO_2	0.41*	0.42*	0.65**	0.66**	0.09	0.68**	1.00
夏季	As	1.00						
	Cd	0.73**	1.00					
	Pb	0.64**	0.70**	1.00				
	Se	0.47**	0.47**	0.82**	1.00			
	Al	-0.09	0.06	0.03	-0.03	1.00		
	SO_2	-0.02	0.15	0.43*	0.52**	0.44*	1.00	
	NO_2	-0.07	-0.06	0.00	-0.02	-0.09	-0.10	1.00
秋季	As	1.00						
	Cd	0.89**	1.00					
	Pb	0.81**	0.95**	1.00				
	Se	0.70**	0.85**	0.95**	1.00			
	Al	0.05	-0.11	-0.19	-0.27	1.00		
	SO_2	0.50**	0.69**	0.80**	0.80**	0.02	1.00	
	NO_2	0.68**	0.83**	0.86**	0.84**	-0.17	0.87**	1.00
冬季	As	1.00						
	Cd	0.91**	1.00					
	Pb	0.96**	0.83**	1.00				
	Se	0.94**	0.78**	0.98**	1.00			
	Al	0.10	0.18	-0.03	-0.04	1.00		
	SO_2	0.82**	0.69**	0.86**	0.89**	-0.13	1.00	
	NO_2	0.74**	0.61**	0.76**	0.83**	-0.16	0.83**	1.00

1) ** 表示 $\alpha = 0.01$ 水平上，相关性极显著（双侧检验）；* 表示 $\alpha = 0.05$ 水平上，相关性显著（双侧检验）

东菏泽地区对北京 As 也有一定的贡献. Cd 和 Pb 的 PSCF 高值区主要分布在石家庄地区,但中值区则主要分布在天津至唐山污染带上. Se 的 PSCF 高值区分布范围较大,包含了整个河北省南部地区、天津和山东菏泽地区.

综合来看,北京夏季 $PM_{2.5}$中高富集重金属主要来源于区域性输送,而其它季节则主要来源于北京周边燃煤源和城区机动车排放.

3 结论

(1)北京城区 7 种致癌重金属中仅 As 年均值超标,其夏季浓度显著高于其它季节.

(2)北京城区 7 种致癌重金属仅 As、Cd、Pb 和 Se 等 4 种重金属有明显富集,夏季富集因子显著高于其它季节.

(3)北京城区 4 种高富集致癌重金属主要来源于周边燃煤源、城区机动车排放以及区域性污染源的输送.

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