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长三角背景地区秋冬季节大气气态总汞含量特征研究

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摘要:利用 Tekran 2537B 大气汞分析仪于 2009 年 9 月 15 日 ~ 2009 年 12 月 17 日对上海崇明岛大气气态总汞(TGM)进行了连续监测. 监测期间,崇明地区气态总汞的平均含量为 2.50 ng·m⁻³ ± 1.50 ng·m⁻³,高于北半球背景浓度 1.5 ~ 1.7 ng·m⁻³,月均浓度 9 月 < 10 月 < 11 月 < 12 月.9 ~ 12 月,崇明地区气态总汞含量的日变化规律为上午 08:00 ~ 10:00 出现峰值,白天气态总汞浓度水平与夜间基本一致. 崇明地区气态总汞含量和一氧化碳含量具有较好的相关性,表明崇明地区大气汞主要来自火电厂和工业锅炉的燃煤排放. 从风向和后向轨迹模型分析的结果来看,崇明岛的大气汞主要来自西部的大陆地区,尤其是位于崇明西北部的江苏、山东等地. 9 月和 10 月盛行来自海面的东风,因而气态总汞含量较低,11 月和 12 月盛行来自大陆工业区的西北风,导致崇明气态总汞含量较高.

关键词:气态总汞;长江三角洲;秋冬季;日变化;后向轨迹

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Characteristics of Total Gaseous Mercury Concentrations at a Rural Site of Yangtze Delta, China

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Abstract: The ambient concentrations of total gaseous mercury (TGM) in Chongming Island, Shanghai, were continuously observed using the Tekran 2537B mercury analyzer from 15th September to 17th December, 2009. The average concentration of TGM during our observation is (2.50 ± 1.50) ng·m⁻³, much higher than the background TGM of north hemisphere. The TGM concentration increased from September to December. During September to December, the concentration peaks appear during 08:00-10:00 am and the daytime TGM concentration approximately equaled that in the night. The TGM in Chongming significantly correlated with concentrations of CO, which indicates that TGM most likely comes from the coal combustion of power plants and industrial boilers. The back trajectory analysis demonstrates the atmospheric mercury in Chongming Island mainly comes from inland China, especially Jiangsu province and Shandong province which locate in the northwest of Chongming. The mercury concentrations are lower in September and October since the airflow is mainly from the eastern ocean. In November and December, the atmospheric mercury content is much higher because the northwest wind brings the pollutants from the western industrial areas.

Key words: total gaseous mercury; Yangtze River Delta; autumn and winter; daily variation; back trajectory analysis

大气中的汞有 3 种存在形式: 气态元素汞 (gaseous elemental mercury, GEM), 活性气态汞 (reactive gaseous mercury, RGM), 颗 粒 态 汞 (particulate mercury, HgP). 气态元素汞和活性气态汞总称为气态总汞(total gaseous mercury, TGM), 其中气态元素汞占气态总汞含量的 95% 以上,在大气中的停留时间为 0.5~2 a^[1]. 燃煤(包括燃煤电厂、工业锅炉和家庭用煤)和金属冶炼(包括锌、铅、铜、金等)是我国两个主要的人为汞排放源^[2]. 经济的高速增长使得对于能源和工业原料的需求大大增加,从而导致我国每年向大气中排放的汞也不断增加^[3]. 从 1995~2003 年,我国人为汞排放量以每年2.9%的幅度增加,并在 2003 年达到 696 t^[4].

20世纪80年代,由于在北欧和北美的偏远湖

泊中发现高甲基汞含量的鱼类, 汞被正式确定为一种可以通过大气进行长距离传输的全球性污染物^[5,6].由此, 大气汞的研究受到了国际上的广泛关注. Fitzgerald^[7]于 1995 年首先提出了建立全球范围的大气汞网络(atmospheric mercury network, AMNET)的设想.之后, 在北半球和南半球相继开展了大量的大气汞监测工作, 其中以欧洲^[8-10]和北美^[11-14]的大气汞监测网络最为完善, 站点最多. 我国日益严峻的汞污染形势也吸引了大批研究者的注意, 尽管我国大气汞的监测工作起步较晚, 但近年来

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也有一些城市和背景区域的大气汞监测结果相继被报道,如贵阳^[15]、北京^[16]、上海^[17],以及贡嘎山^[18]、长白山^[19]和雷公山^[20]等. 崇明岛位于我国东部沿海,地处长江入海口,是我国东部大气输出口,研究当地的大气汞含量对于分析我国的大气汞输出具有重要的意义.

1 研究区概况

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本研究中观测站点(图 1 中圆点所示)位于上海市崇明东滩湿地鸟类保护区管理处大楼楼顶(北纬31°31′20″,东经 121°54′30″,高度约 11 m),距离东滩湿地约 5 km. 崇明东滩鸟类保护区是国家级自然保护区,主要保护对象为迁徙鸟类及其栖息地.保护区位于崇明岛(中国第三大岛)的最东端,地处长江入海口,远离市区,受大陆风和海风影响. 崇明岛地处北亚热带南缘,属海洋性气候. 全年温和湿润,四季分明,冬季干冷,盛行偏北风,夏季湿热,盛行东南风,春秋季节是气候转换的季节,具有明显的季风气候的特点. 崇明东滩的年平均气温为15.3℃,7月和8月平均气温 27.5℃;1月最冷,月平均气温 2.9℃.

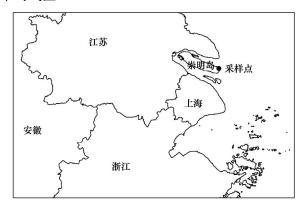


图 1 监测站点地理位置示意

Fig. 1 Location of sampling site in Chongming Island

2 研究方法

2.1 采样时间及方法

崇明大气气态总汞的监测工作于 2009 年 9 月 15 日~2009 年 12 月 17 日期间进行. 利用加拿大 Tekran 公司生产的 Tekran 2537B 大气汞分析仪对 大气气态总汞进行 24 h 的连续监测. Tekran 2537B 是目前国际通用的测定环境大气气态总汞含量的仪器. 该仪器的基本原理是以一定的流速(通常为1 L·min⁻¹)进行空气采样,当空气通过仪器内部的金管时,空气中的气态总汞被富集在金管中,采样时间一般为 5 min. 采样结束后进入分析阶段,通过加

热线圈对富集了汞的金管进行加热,使金管中富集的汞热解析出,再用冷原子荧光法测定汞含量. 该仪器的最低检出浓度为 0.10 ng·m⁻³,远低于大气中的气态总汞浓度.

监测站安装了戴维斯气象站对温度、风向、风速以及太阳辐射等常规气象参数进行监测,以提供和大气汞监测数据同步的气象数据. 另外还利用Thermo 公司生产的 Model 48i-TLE 一氧化碳分析仪进行 CO 的监测.

2.2 数据质量控制

仪器的数据质量通过仪器本身的自动校正和利用外置汞源的手动校正进行控制. Tekran2537B 内置标准汞蒸气发生源,每25 h进行一次自动校正,保证大气气态总汞的含量得到准确的测定. 手动校正利用 Tekran2505 产生标准汞蒸气,将零气(不含汞的空气)通入采样口,然后吸取已知含量的汞蒸气注射到 Tekran2537B 中进行测定,将测定的汞浓度和计算值进行比较,并根据测量结果作标准曲线.

3 结果与讨论

3.1 大气汞含量基本特征

从 2009 年 9 月 15 日 ~ 2009 年 12 月 17 日整个 采样期间,共获得大气气态总汞(TGM)小时平均数据2 111个(图 2). 采样期间气态总汞的平均浓度为 2. 50 $\text{ng·m}^{-3} \pm 1.50 \, \text{ng·m}^{-3}$,高于北半球大气气态总汞背景值 $1.5 \sim 1.7 \, \text{ng·m}^{-3}$ [21],这表明崇明地区受到了一定程度的大气汞污染. 较大的标准偏差表明当地气态总汞的浓度波动较大.

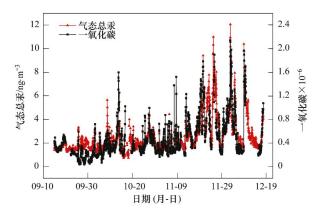


图 2 崇明地区大气气态总汞和一氧化碳的时间分布

Fig. 2 Time series for TGM concentration and CO concentration in Chongming Island

表 1 为崇明地区大气气态总汞月均含量的分布情况,从中可以看出从 9~12 月,气态总汞含量逐月增加,9 月和 10 月气态总汞含量较低,11 月和 12 月

气态总汞含量则较高. 另外从标准偏差可以看出,9 月和10 月崇明气态总汞含量相对稳定,波动较小 (标准偏差分别为0.34 和0.66),11 月和12 月气态总汞含量变化明显,波动较大(标准偏差分别为1.73 和1.98),最大小时平均浓度分别达到10.99 ng·m⁻³和12.05 ng·m⁻³,说明11 月和12 月崇明大气汞含量受到更多的人为活动影响.

表 1 崇明地区大气气态总汞含量统计/ng·m⁻³

Table 1 Statistical summary of TGM concentration

in Chong	ming	Island/	′ng•m	- 3
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-			0 0	U		
	月份	样品数	平均值	最小值	最大值	标准偏差
	9	383	1. 69	1. 23	3. 32	0. 34
	10	643	1. 99	0.70	5. 64	0.66
	11	688	2. 96	1. 19	10. 99	1. 73
	12	397	3. 31	1.43	12. 05	1. 98

3.2 大气气态总汞日变化规律

图 3 显示了崇明地区不同月份气态总汞的日变化规律. 为了避免重污染时段的影响,气态总汞的日变化规律采用中值进行分析. 崇明 9 月的各气象条件比较稳定,昼夜温差小,气态总汞的浓度在北半球背景浓度水平附近上下波动,日变化幅度非常小,10~12 月气态总汞浓度更高,日变化规律也更加显著. 9~12 月气态总汞均在上午 08:00~10:00 出现峰值,原因为昼夜大气边界层高度的变化,夜间温度和风速均低于白天,边界层较低,不利于大气污染物的扩散,边界层内的气态总汞不断累积,浓度从夜间开始不断升高,日出以后随着温度和风速的增加,边界层高度逐渐增加,大气中的气态总汞浓度开始扩散降低.

11 月除了在上午 10:00 出现了气态总汞的浓度峰值,在夜间 23:00 也出现了峰值,气态总汞的浓度从 21:00 开始升高,在 23:00 达到峰值,从 21:00

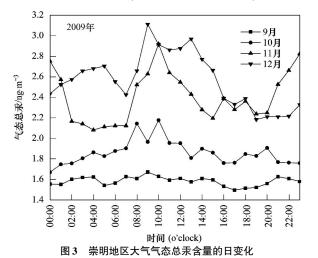


Fig. 3 Daily variation of TGM in Chongming Island

~次日 01:00 气态总汞均处于较高的浓度水平. Schmolke 等^[22]研究者在欧洲的一些监测站点也发现了类似的气态总汞日变化规律,研究认为夜间或清晨出现的气态总汞峰值与汞在夜间大气逆温层中的累积有关. 12 月高浓度气态总汞更多地出现在夜间(21:00~03:00),由于这一时段大气边界层高度低,从而更容易发生重污染过程.

监测期间崇明气态总汞白天和夜间的平均浓度没有明显差异,仅10月和11月白天略高于夜间.主要原因是边界层高度引起的扩散条件和源排放强度的影响相互抵消.白天工厂投入运行增加了人为源的汞排放,另外白天较高的温度和光照(图4)也促进土壤和水体中汞的释放^[23,24],增加了自然源的汞排放,但白天的气象条件更有利于污染物的扩散;夜间土壤和水体释放汞强度大大降低,电厂和锅炉燃煤排放的汞也大大减少,但处于不利的扩散条件,大气中的汞更容易累积.

3.3 污染来源分析

崇明岛受大陆风和海洋风影响,风向和风速是 影响大气汞含量分布的重要因素. 图 5 表示了监测 期间崇明地区风向相对频率分布和大气气态汞含量 的风向分布,可以看出,9月盛行东南风,10月风向 变换较为频繁,但主要为来自东北、正东和正南方 向的海洋风,11 月和 12 月则盛行来自西北方向的 大陆风. 从图 5(b) 可以看出,来自东部海面的海洋 风较为清洁,因而9月和10月气态总汞含量较低; 来自大陆的西北风则携带了大量大气汞,导致11月 和12月崇明气态总汞含量较高. 另外,从图5(b) 可以看出11月和12月均在西南风向上表现出较高 的气态总汞浓度,这是因为来自上海市区的西南风 也携带了大量气态汞,但由于西南风向并非主导风 向,因而上海市区并不是这段时间崇明地区大气汞 的主要来源. 需要指出的是,11 月在东南风向上也 显示了较高的气态总汞含量,而风向相对频率很低, 推测为来自东北的风在接近监测点时在局地发生偏 转,变为东南风,具体的论证将在3.4节中利用后向 轨迹模型进行.

崇明岛西北方向为境内拥有大量火电厂的江苏、山东、河北等省,燃煤污染是大气汞的主要来源,所以监测期间崇明地区的大气汞污染主要受崇明西北各省的燃煤电厂排放的大气汞影响. 监测期间崇明地区气态总汞和一氧化碳具有很好的相关性(图6),相关系数为 0.88,进一步表明燃煤排放为崇明地区大气汞的主要来源. 图 6 为采样期间崇明

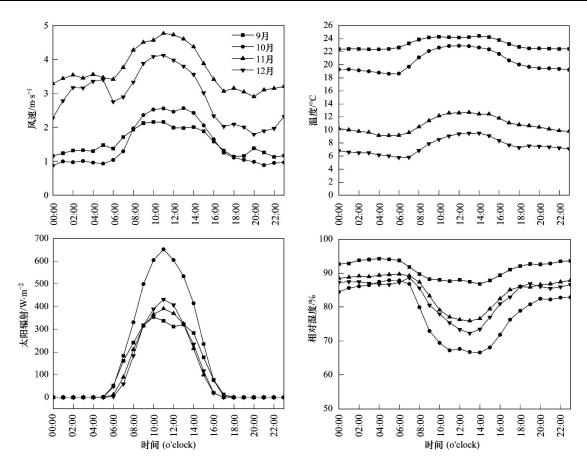


图 4 崇明地区气象条件(风速、温度、太阳辐射强度、湿度)的日变化

Fig. 4 Daily variation of meteorological conditions (wind speed, temperature, solar radiation, humidity) in Chongming Island

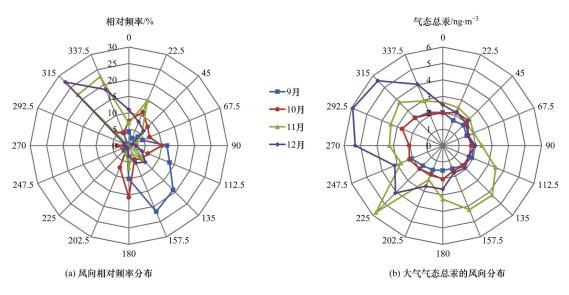


图 5 监测期间崇明地区风向相对频率分布和大气气态汞含量的风向分布

Fig. 5 Frequency distribution of wind directions and concentration distribution of TGM at wind directions in Chongming Island

地区各月份气态总汞和一氧化碳的相关关系,相关性为9月<10月<11月<12月,9月和10月气态总汞和一氧化碳的相关系数较小,相关性较弱,表明这两个月崇明地区大气汞人为源排放影响较小;11

月和12月气态总汞和一氧化碳高度相关,表明这段时间崇明地区大气汞主要受人为源排放影响.

3.4 典型污染过程的后向轨迹分析 从图 2 可以看出,从 11 月 19 日~12 月 12 日,

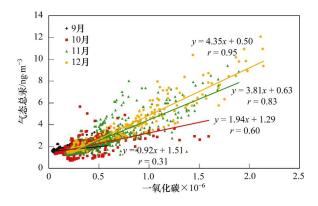


图 6 崇明地区气态总汞和一氧化碳的相关关系

Fig. 6 Relationship between TGM concentration and CO concentration

崇明地区连续出现了几个重污染过程,因此将这一段时间气态总汞的 5 min 监测数据列出,以便进行更深入的分析,如图 7 所示. 从中可以看出,11 月24~27 日,崇明地区经历了一个历时较长的重污染过程,这段时间崇明地区的风向变化较为频繁,且3.3 节中提及的东南风向上出现的高浓度现象也主要发生在这段时间(11 月 26 日 06:00~23:00),所以利用美国国家海洋和大气管理局(National Oceanic and Atmospheric Administration, NOAA)空气资源实验室(Air Resource Laboratory, ARL)的HYSPLIT 后向轨迹模型对这段时间进行了后向轨迹分析,分析结果如图 8 所示.

根据气态总汞的监测结果,11 月 24 日 21:00 之前气态总汞浓度处于较低水平(<3 ng·m⁻³), 从图 8(a)可以看出,这段时间,气流主要来自东海 海面;之后,来自海面的气流开始向陆地偏转,经 过浙江时气流中的汞含量增加,崇明地区的气态

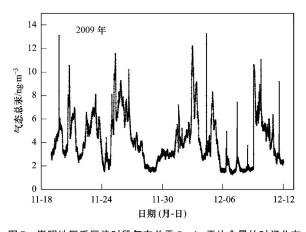


图 7 崇明地区重污染时段气态总汞 5 min 平均含量的时间分布 Fig. 7 Time series for 5 min average TGM concentration in Chongming Island

总汞含量也随之升到了 5 ng·m⁻³以上;至 11 月 25 日05:00,气流主要来自福建省,并途径浙江省 [图 8(a)中红线和蓝线所示],崇明地区的气态总汞含量进一步升至 9 ng·m⁻³以上.从 11 月 25 日 08:00 [图 8(b)],崇明地区的风向继续顺时针偏转,气流主要来自浙江、安徽和江苏;从 11 月 25 日 20:00 开始,气流主要来自山东,并在海面改变方向从东北方向进入崇明地区;期间崇明地区气态总汞一直保持较高浓度(~8 ng·m⁻³).从图 8 (c)可以看出,11 月 26 日 05:00 ~ 23:00 崇明地区持续东南风,气流主要来自江苏,在海面发生了180°偏转变成东南风将来自江苏的高浓度汞气流吹向崇明,造成该时段崇明地区气态总汞保持在

对其它重污染时段进行后向轨迹分析的结果表明,崇明大部分重污染过程中的大气汞来自位于崇

较高浓度(6.03 ng·m⁻³).

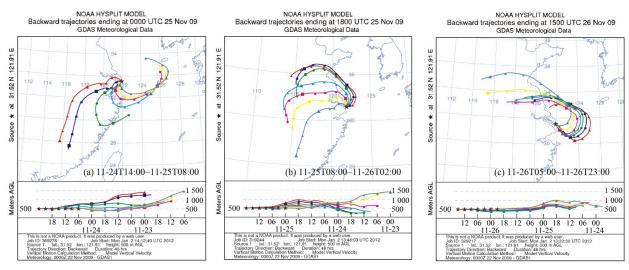


图 8 崇明地区后向轨迹分析结果

Fig. 8 HYSPLIT model results for Chongming Island

明西北部的江苏和山东. 12月3日出现的重污染过程主要受来自我国北方的内蒙、河北等地的影响. 12月9日的重污染过程,崇明的高浓度大气气态总汞主要来自江苏,此外,位于崇明西部的浙江、安徽和江西等地也有一定的贡献.

3.5 与其他监测站点的比较

表2列出了国内外一些城市点和背景点的大气

汞监测结果(TGM 和 GEM, GEM 占 TGM 95%以上). 监测期间,崇明站的 TGM 浓度远低于国内的城市 TGM 浓度,与国外一些城市相比,崇明 TGM 的含量也较低. 崇明的 TGM 浓度低于国内长白山、贡嘎山和雷公山等部分背景点的监测结果,但高于国外的一些背景点,如日本的冲绳以及美国的 Great Mountain Forest.

表 2 国内外城市和背景点 TGM 和 GEM 监测结果统计

Table 2 Statistical summary of TGM/GEM at urban and rural locations in China and abroad					
项目	地点	监测项目	浓度/ng·m ⁻³	采样时间	文献
	贵阳	TGM	8. 4	2001-11 ~ 2002-11	[15]
	北京	GEM	8.3 ± 3.6	2005-01	[16]
			6.5 ± 5.2	2005-04	
			4.9 ± 3.3	2005-07	
城市			6.7 ± 3.5	2005-10	
	广州	GEM	13. 5 ± 7.1	2005-01	[16]
	长三角地区	GEM	5.4 ± 4.1	2005-09	[16]
	上海	TGM	2.7 ± 1.7	2009-09	[17]
	韩国首尔	TGM	3.22 ± 2.10	2005-02 ~ 2006-02	[25]
	美国芝加哥	GEM	3.6 ± 2.9	1994-07 ~ 1995-10	[11]
	瓦里关	GEM	1.7 ± 1.1	2005-08	[16]
			0.6 ± 0.08	2005-12	
	贡嘎山	TGM	3. 98	2005-05 ~ 2006-06	[18]
背景	长自山	TGM	3.22 ± 1.78	2005-08 ~ 2006-07	[19]
	雷公山	GEM	2.80 ± 1.51	2008-05 ~ 2009-05	[20]
	日本冲绳	GEM	2.04 ± 0.38	2004-03 ~ 2004-05	[26]
	Great Mountain Forest,美国康涅狄格州	TGM	1.4 ~ 1.6	1999 ~ 2004	[12]

4 结论

- (1)监测期间崇明岛大气气态总汞平均含量2.50 ng·m⁻³ ± 1.50 ng·m⁻³,高于北半球背景值.月变化规律:9月<10月<11月<12月,9月和10月气态总汞浓度较低,受人为源影响较小;11月和12月浓度较高,受人为源影响较大.
- (2)崇明地区气态总汞含量表现出一定的日变 化规律,在上午 08:00~10:00 出现峰值,白天与夜 间气态总汞浓度无明显差异.
- (3)9月和10月崇明岛盛行来自海面的东风, 大气汞含量较低;11月和12月来自大陆的西北风 为主导风向,携带大量来自江苏、山东等地燃煤排 放的汞,导致这段时间崇明大气汞含量较高.

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