

水/气界面间汞交换通量的研究进展*

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摘要 本文综述了近年来国内外大量文献, 对有关水体和大气中汞的存在形态、特性及水/气间汞交换通量的影响因素进行了总结, 描述了目前国内外相关领域的研究现状, 并对该领域下一步的研究方向进行了探讨.

关键词 水/气界面, 汞, 通量.

汞是惟一可以在常温下以液态存在并具有挥发性的金属, 它以多种化学形态存在于环境中, 并在水体、大气、土壤和生物体间不断迁移转化. 其中, 水体与大气互为汞的“源”或“汇”, 一方面, 汞从水体向大气的释放减轻了水中汞的负荷, 另一方面, 也增加了大气中汞的含量, 反之亦然. 水体和大气界面间汞的交换通量决定着汞传输方式——进入大气向周边环境扩散, 还是留在水中进一步转化为甲基汞并随水生食物链逐级富集到大型食肉性鱼类体内, 最终对人类健康构成威胁. 因此, 水体和大气界面间汞交换通量的研究受到国内外研究者的广泛关注.

1 水体和大气中汞的存在形态与特性

1.1 汞在大气中的存在形态与性质

单质汞在常温下具有很高的饱和蒸汽压, 且多数汞化合物也具有较强的挥发性. 大气中, 95%以上的汞是气态单质汞 (gaseous elemental mercury, GEM 即 Hg^0)^[1-3], 其余不足 1%—5% 是活性气态汞 (reactive gaseous mercury, RGM) 和颗粒态汞 (particulate gaseous mercury, PGM)^[4-5]. GEM 挥发性高和水溶性低且在大气中相对惰性^[6-10], 可以长时间 (约 0.5—2 年) 滞留在大气中^[11-12], 并随全球大气循环发生大范围长距离的迁移. RGM 主要是以 $HgCl_2$ 为主的汞的卤化物^[5, 13-14], 具有较高的表面活性和水溶性^[15], 很容易通过降雨再次进入陆地生态系统, 对局地或区域性环境具有重要影响^[5-16]. PGM 根据其颗粒粒度的大小不同, 主要沉降在污染源附近, 并随污染源距离的增加含量逐渐减少.

1.2 汞在水环境中的存在形态和性质

水环境中, 汞的主要存在形态为溶解气态汞 (dissolved gaseous mercury, DGM, 主要是 Hg^0)、二价无机汞 (Hg^{2+})、单甲基汞 (methylmercury, MMe $_2$) 和少量二甲基汞 (dimethylmercury, DMMe $_2$). DGM 在全球海水中占总汞的 10%—30%^[17-18]. DGM 主要存在于浅表水层, Lindberg 等^[19]曾搜集了 11 组水体表层 1 cm、100 cm 和沉积物上部 5 cm 的水样进行研究, 结果发现有 9 组 DGM 含量是随着水深增加而减少的, 这支持了 DGM 源于表层水的观点. 水中 DGM 含量的高低受几个竞争性因素控制: 可利用溶解态活性 Hg^{II} (DRM), DRM 光致还原率或其它产生过程, 新生 DGM 的水平对流, 包括氧化和释放在内的 DGM 的损失^[19].

由于 Hg^0 在水中的溶解度很低, 通常它在天然水体, 特别是表层水体中处于超饱和状态, 导致大量 Hg^0 会从水体向大气释放^[5]. 研究表明, 水体是大气汞的重要自然释放源之一^[4, 20-21], 而水体向大气的排汞过程成为汞从水体移除的一个主要途径^[22]. 目前对水中 DGM 形成的机理还不清楚, 但现有的研究表明水中 DGM 形成的机理可能很多, 其中最重要的是水中微生物将二价汞还原为单质汞^[23], 或者是非生物在腐殖质存在的情况下将二价汞还原为单质汞^[24], 或者有机汞化合物的降解产生单质汞^[18, 25]. 最近研究也显示, 二价汞的光致还原是 DGM 产生的另一个重要的机制^[26-27]. 水中 DGM 的损失主要是向大

2010 年 3 月 12 日收稿.

* 国家自然科学基金资助项目 (No. 40803036, No. 40973083).

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气的释放和被氧化成二价汞(如在氯离子存在的情况下)^[28].

2 水/气界面间的汞交换及其影响因素

水/气间汞交换过程是汞在大气、陆地和水体之间生物地球化学循环的一个非常重要的环节^[21]。一方面,水体向大气释放单质汞,减轻了水体汞的负荷,减少了水中汞的甲基化几率,从而减少了鱼体内甲基汞富集的可能。另一方面,大气中单质气态汞可参与全球大气循环传播和扩散,随大气干、湿沉降进入更广阔的环境系统,因此水体又是大气汞的重要来源。

大量的研究显示,水体每年向大气释放的汞约占大气汞天然来源的32%—77%^[4, 20, 29-30]。王定勇^[31]对重庆土壤和水体汞释放通量的研究结果显示,水体的汞释放通量明显高于土壤。由此可见,水体对大气汞的贡献是非常可观的。水/气间汞交换通量方面的研究,一直受到研究者的广泛关注。目前,这方面的研究主要集中在水/气间Hg⁰的交换通量上^[31-35],也有少量研究对水/气间甲基汞的释放通量进行了初步探讨^[36]。

热力学上,水/气界面的汞交换受亨利法则控制($[Hg^0]_{air}/[Hg^0]_{water} = H$), $[Hg^0]_{water}$ 即水中DGM的含量, $[Hg^0]_{air}$ 即大气中气态单质汞的含量。通常,水气间交换通量采用大气中TGM与水中DGM的浓度,通过平衡方程计算得到,公式如下: Flux = $K(C_aH^{-1} - C_w)$,这里 C_a 代表大气中气态单质汞的含量, C_w 代表水中溶解气态汞的含量, K 表示单质汞在水体和大气间的质量传输系数, H 即亨利系数。

水/气间汞交换通量的研究显示,汞交换通量存在如下三个规律:昼夜变化规律、季节变化规律和晴雨天变化规律。(1)昼夜变化规律。水体向大气汞的释放通量极大地取决于光照强度,并与光照强度具有显著的相关性,这种现象主要表现为水体向大气汞的释放通量在白天和光照较强的时间段出现峰值,夜晚和光照较弱的时间段出现谷值^[37-41]。Gårdfeldt^[42]等的研究认为这主要是因为白天比夜晚有更强的紫外光照射,特别是UVA,促进了汞的光致还原作用,从而使表层水中产生更多的DGM,他的研究结果也证明了这点,即DGM的释放通量与光照呈现显著相关性,相关系数 $R^2 = 0.99$ 在此基础上,部分环境参数的改变如水深、DOC含量,将改变水体垂直方向上的受光照程度,从而产生一般水体的DGM随水深减少的趋势,以及水气界面汞通量晴天大于阴天,白天大于夜晚的现象。另外两个点出现底部水中DGM含量也较高,这意味着除了光照之外,还有别的条件会影响到DGM的产生和逸出。(2)季节变化规律。春、夏季节(或暖季节)释放通量高于秋、冬季节(或冷季节)^[31, 35, 40-41]。如Zhang和Dill^[43]对一个水库进行为期一年的研究显示,水中DGM含量在夏季的六到八月最高,之后逐渐降低,12月达到最低值,春夏比秋冬季节有更高的DGM平均产率,且季节变化趋势也显示出似乎接近日光辐射变化规律。其次,光照对温度的影响会影响到水温,温度的变化会影响到DGM在水中的饱和度,暖季节水温上升,DGM在水中的溶解度下降,趋向于向大气的释放,因此导致水体向大气汞释放通量增加。(3)晴雨天的变化规律。晴天表现为水体向大气汞的释放,阴天和降雨期间表现为大气向水体的汞沉降^[35, 44]。对雨天汞出现负通量的原因和机理还不清楚,目前认为可能的原因是Hg被通量箱壁或水表本身吸收,一旦太阳出来,蒸发开始,通量再次变成正值^[34]。

就目前的认识来看,水体中的DGM主要来自于表层水中二价汞的还原,水中DGM的产生量与驱动DGM释放的因素是直接影响水体向大气汞释放通量的主要因素。对于DGM从水中向大气扩散的驱动力方面,则普遍认为主要是受到水气界面间DGM浓度梯度及热力学方面的影响。另外,许多物理化学参数可以加速这种转化增加水体向大气汞的释放量,包括光照强度、水温、pH、和DOC浓度^[26, 28, 45-47]等。这些参数中,光照强度是影响DGM产生的主要因素,光照的增加会促进DGM的产生,同时,光照增加使水体温度增加,降低了DGM在水中的溶解度,从而表现为水体向大气释放汞的通量增加^[39, 48-49],而其它参数对天然水体中DGM的产生所起的作用仍然存在争议。比如:对贵州省境内的红枫湖水体与大气间交换通量的最新研究显示,在天气光照不足 $140W \cdot m^{-2}$ 的阴雨天气,水体与大气间汞交换通量并没有明显的昼夜变化规律,且与光照没有显著正相关关系^[33, 50]。此外,水体汞释放还受盐度、风速、浪花等影响^[44],其相互的关系也不是简单的正负相关关系,而是比较复杂的函数关系。而紫外光的类型(UV-A或UV-B)对DGM也有影响,春夏比秋冬季节有更高的DGM平均产率,且季节变化趋势也显示出似乎接近日光辐射变化规律。

为了查明各项参数(特别是光照强度及其与之共同作用的其它参数)对水体与大气间 DGM 交换量的主要影响机制,除了野外的实测,也有人进行了部分室内模拟研究。研究发现,日光辐射通过一系列的途径产生活性氧化形态,且常常只有不到 1 s 的环境半衰期。水中 DGM 被羟基^[7,42] O、O₃^[6,51],有机过氧功能团^[52],和氯化物及有机化合物所氧化^[26,28],导致含量降低。然而,也有研究发现 DGM 在黑暗中也会减少^[22,26,49,53]。这意味着水中存在受长效光介质控制的氧化作用或者是存在非光致氧化作用。法国的一项研究显示^[54],颗粒相(微生物、氧化物等)与还原过程有关。该研究对过滤和不过滤的天然水用 300—450 nm 的光在有氧和无氧状况下照射 4 d 发现 DGM 在黑暗期间也能观察到,且在过滤的水中 DGM 似乎形成的量很低,而在未过滤水样中观察到明显的还原过程,且只有在通氮气时该过程变的较为显著。

3 问题与展望

据 Park 等^[55]对韩国南部 Juam 水库的研究和近年来美国部分湖泊、中国贵州省的百花湖夏季 DGM 浓度的比较结果显示,中国的淡水湖水体 DGM 远高于韩国,而韩国又高于美国。他认为韩国被研究区域水体没有直接的人为汞污染源,其 DGM 偏高的原因除了源于本地少数上风向的工厂之外,很有可能是来自中国这样的高汞释放源区。本文作者对贵州省百花湖水体向大气释汞通量的现场测定也显示较高的结果:其中平均值为 4.0—9.7 ng m⁻² h⁻¹,最大值高达 51 ng m⁻² h⁻¹^[35],显著高于意大利的西西里海峡(0.1—0.3 ng m⁻² h⁻¹^[40])、美国北明尼苏达湖(0.04—0.05 ng m⁻² h⁻¹^[56])、美国阿拉斯加湖(均值为 1.2±0.4 范围为 0.5—1.7 ng m⁻² h⁻¹^[57])、美国威斯康辛湖(0.4—2.3 ng m⁻² h⁻¹^[58])、加拿大安大略湖(0.04—1.3 ng m⁻² h⁻¹^[59]; 中值为 0.8—1.9 ng m⁻² h⁻¹^[33]) 和密西根湖(1.0±0.6 ng m⁻² h⁻¹^[60])。由于我国是目前公认的大气汞排放国家,以上两个事例暗示我国较丰富的湖泊/水库系统可能是我国及周边地区大气汞的一个重要的天然来源。由于我国相关研究十分有限,因此对不同类型水域的深入研究十分必要。第二,尽管向大气汞的贡献主要来自于海洋,但对于淡水系统向大气释放汞的研究可作为海洋相关研究的重要补充。第三,根据目前国内外研究现状可知,水/气界面间甲基汞通量测定方面,无论是实验方法还是实验结果都很少。第四,尽管环境样品中甲基汞的含量极低,但因其毒性很强,仍是应当尝试研究的领域。在 DGM 的形成机制和 DGM 向水体释放的驱动机制方面,需要更详细的室内模拟实验和野外测试结果来验证。

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RESEARCH DEVELOPMENT ON WATER/AIR EXCHANGE FLUX OF MERCURY

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ABSTRACT

Recent work reported in a large number of references on the water/air exchange flux of mercury from both China and abroad is reviewed in this paper. Mercury species and characteristics in the water and air environment as well as the main factors affecting the mercury exchange flux between water and air are summarized. Based on the summary, the current research status and future research direction are discussed.

Keywords water/air interface, mercury, exchange flux