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# 冰冻圈甲基汞研究进展

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**摘 要:** 作为气候系统五大圈层之一,冰冻圈在全球汞的生物地球化学循环中扮演着重要角色。人类活动排放的汞污染物通过大气环流迁移到偏远的冰冻圈地区,并在环境中转化为更具毒性的甲基汞,经食物链传递、富集和放大,会对冰冻圈生态环境构成重大威胁。为了深化理解冰冻圈甲基汞研究现状,本文总结了典型冰冻圈介质甲基汞的浓度水平和分析方法,重点梳理了甲基汞在冰川、冻土、积雪和海冰中的迁移、转化和归趋等生物地球化学过程,特别是对冰冻圈微生物汞甲基化进行了详细的归纳和总结。同时关注气候变化背景下冰冻圈甲基汞分布、行为和环境影响,这对于评估人类和野生生物在冰冻圈中的甲基汞暴露风险至关重要。鉴于目前对低温环境中甲基汞变化过程和机理的认识尚处于起步阶段,文章对未来冰冻圈甲基汞研究方向进行了展望。

**关键词:** 甲基汞; 冰冻圈; 冰川; 冻土; 积雪; 海冰

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## 0 引言

汞是一种全球性污染物,由于元素汞具有较高蒸汽压,在大气中的停留时间长(0. 5~2年),是唯一能以气态形式进行远距离传输的重金属元素[1]。人类活动排放的汞污染物可以通过大气环流迁移到地球偏远地区,并通过干、湿沉降进入陆地和水生生态系统,最终富集于冰川、植被、水体和土壤等地表环境之中[2-3]。当今人为活动使大气中的总汞浓度已超出自然水平的4. 5倍[4],大气沉降到地表环境之中的无机汞可以通过生物和非生物甲基化作用转化为毒性更大的甲基汞[5-7]。甲基汞具有很强的神经毒性,它在生态系统食物链中具有显著的生物放大和生物积累特性[1,8]。2013年,联合国环境规划署通过了旨在减排汞污染物的国际公约—《关于汞的水俣公约》,以遏制人为汞污染向环境排放输入,协议的主要目标之一是最大程度上降低与甲基汞接触相关的健康风险。

冰冻圈在地球上覆盖范围超过9. 6×10$^7$ km$^2$,包括冰盖、山地冰川、多年冻土、积雪、海冰、冰架等,它们主要分布在南北极和青藏高原("第三极")[9-10]。在人类活动深刻影响自然环境的背景下,远离人类活动区的冰冻圈亦受到可全球传输的污染物影响。人类活动释放的汞经长距离传输进入偏远地区,封存于冰冻圈,使冰川、冻土等冰冻圈要素成为汞的临时储库[11-17]。Schuster等[15]估算了整个北半球冻土区的汞储量约为(1 656±962) Gg, Mu等[17]估算出青藏高原活动层储存汞约16. 6 Gg。沉积于北极冰川中的汞总量约为(2 415±22) Gg, Huang等[18-19]估算出中国西部冰川区每年冰尘中汞总量为34. 3 kg。然而,随着全球气候持续变暖,冰冻圈要素快速退缩变化,这将导致储存于冰冻圈中的污染物发生"二次释放",将对生态环境和人体健

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康产生不利影响[4,9]。据估计，格陵兰冰盖西南部融水径流中汞通量约为 3~42.4 kg·a$^{-1}$[20]。Schaefer 等[21]模拟了 RCP8.5 排放情景下冻土退化导致 Yukon 流域汞输出将增加一倍（约为 11 Mg·a$^{-1}$）。Dastoord 等[18]估算出亚北极地区（60° N 以北）河流向北冰洋输出溶解汞和颗粒汞总量约为（41±4）Mg·a$^{-1}$，主要来源于季节性积雪和多年冻土区。

不仅如此，气候变化会刺激冻土、融雪和湿地环境微生物汞甲基化，同时冻土融化的增强、降水的改变以及融雪导致地表水流量的增加，促使甲基汞向下游生态系统输送[22-25]。冰川、冰盖和海冰的减少也会对冰冻圈汞生物地球化学循环产生重大影响[26-27]。以上事实都表明，冰川、冻土、积雪和海冰等冰冻圈介质融化将成为陆地和海洋生态系统汞的一个重要来源，越来越多的证据亦指出冰冻圈消融向下游生态系统输送的汞总量巨大，这无疑会加剧甲基汞暴露风险[24-25,28-31]。甲基汞可以在生物体内积累，进而对高营养级生物甚至人类产生潜在危害[1,8]。已有研究报道北极生物体内有高水平的汞含量（如鸟类和鱼类），Franklin 海湾冰下鳕鱼的总汞浓度为 0.38 μg·g$^{-1}$，其中 80% 为甲基汞[32]。有研究报道北极地区居民每周从海洋食品摄入汞约 846 μg，远超过世界卫生组织限定的阈值[33]。

冰冻圈具有地球上独特的极端环境特征，甲基汞在冰冻圈的积累、迁移和转化等行为是全球汞生物地球化学循环过程中非常重要的环节（图 1）[34-36]。然而控制冰冻圈甲基汞浓度的因素复杂多变，以及缺乏足够灵敏的超痕量形态分析方法使得甲基汞浓度难以量化，迄今全球对冰冻圈甲基汞的认识仍处于初级阶段，存在很大的不确定性。冰冻圈汞甲基化在生物和非生物方面都具有其独特性，目前对冰冻圈各要素中甲基汞浓度的测定和微生物多样性的认识仍匮乏，这对于研究探讨寒冷环境条件下微生物与汞甲基化的关系成为挑战。总而言之，我们对冰冻圈甲基汞的来源、产生/降解机制以及对控制甲基汞迁移和转化的生物地球化学过程的因素仍然知之甚少。

基于梳理冰冻圈甲基汞研究进展和未来研究方向，本文全面综述了冰冻圈典型介质（冰川、冻

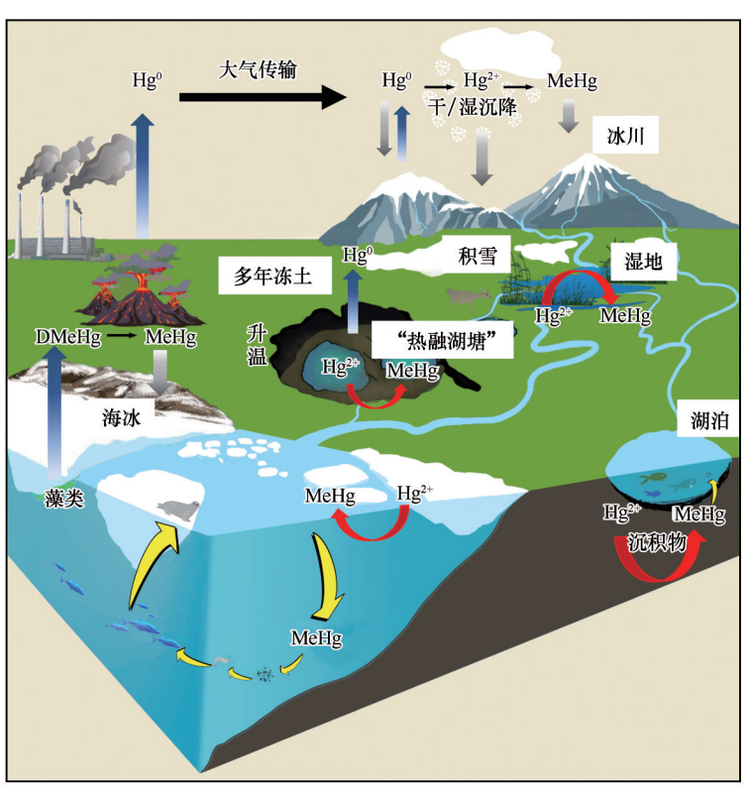


图 1　冰冻圈甲基汞迁移和转化等过程示意图（黄色箭头为甲基汞在冰冻圈中迁移和富集等过程；红色箭头为甲基汞在冰冻圈中转化过程；蓝色箭头和灰色箭头为汞在地表-大气界面交换过程）

Fig. 1　Schematic diagram of biogeochemical processes such as the migration and transformation of MeHg in the cryospheric environment [The illustration describes the migration and enrichment of methylmercury (yellow arrow), methylation (red arrow), and the surface-atmosphere exchange of mercury (blue arrow and gray arrow)]

土、积雪和海冰)中甲基汞的研究现状,归纳总结了冰冻圈各要素甲基汞的浓度水平和生物地球化学过程,包括汞甲基化机制、转化、迁移、归趋及甲基汞暴露风险等,并重点关注汞甲基化微生物对冰冻圈甲基汞产生机制和变化过程的影响,对未来冰冻圈甲基汞研究提出了新的展望。

## 1 甲基汞产生机制

汞的甲基化包括生物和非生物两种途径[5,37-40]。在自然条件下汞的非生物甲基化效率非常低,通常认为环境中的甲基汞是由微生物主导产生[38-40]。Jensen等[40]首次通过实验验证了沉积物中的甲基汞主要是由微生物甲基化产生的,主要的汞甲基化微生物包括硫酸盐还原菌(Sulphate reduction bacteria, SRB)、铁还原菌(Iron-reducing bacteria, FeRB)和产甲烷菌(Methanogenus)等。在过去很长一段时间内,对汞甲基化的遗传机制并不清楚。直到2013年美国橡树岭国家实验室研究人员报道了*hgcA*和*hgcB*两个基因簇,可以直接调控微生物汞甲基化,从而揭示了汞的微生物甲基化遗传机制[41]。其中*hgcA*编码类咕啉蛋白,*hgcB*编码铁还原蛋白,这两种蛋白分别在甲基运输以及电子提供中起着重要的作用[41-42]。研究表明,在所有已被证实的汞甲基化细菌中都存在这两个基因簇,*hgcAB*基因的发现成为可靠的甲基化微生物的分子标记物,可以预测汞甲基化过程的存在[41-43]。目前使用这一分子探针已识别到30个门类的汞甲基化细菌,Gilmour等[43-44]还在许多新环境鉴定出了汞甲基化细菌以及除硫酸盐还原菌等常见菌种之外的许多细菌。随着对微生物甲基化分子机制理解的深入,可以根据环境中*hgcAB*基因的含量进行甲基汞风险预测,已有证据表明土壤中*hgcA*丰度与甲基汞浓度呈显著正相关[45]。

汞的生物甲基化是甲基汞的重要来源,它受到多种环境因素的控制,例如刺激无机汞的可用性、微生物群落以及基因调控的变化等都可以影响到汞生物甲基化[46-47]。除了直接控制汞甲基化的基因*hgcA*和*hgcB*,还有一些通过控制环境中汞化合物形态和迁移转化进而间接影响甲基汞分布的基因[48]。*merA*基因编码汞还原酶,介导的细菌能够将氧化态汞($Hg^{2+}$)还原为气态汞($Hg^0$),由于$Hg^{2+}$是甲基化反应的底物,抗汞细菌亦可能在降低甲基化过程的底物供应方面发挥作用[48-49]。*merB*基因编码烷基汞裂解酶,控制着去甲基化过程的发生,从而降低环境中甲基汞的总水平[48]。鉴于影响甲基汞的环境因素众多且转化机理繁杂,本文仅重点关注冰冻圈汞的微生物甲基化,而对去甲基化等过程不予开展过多讨论。

## 2 分析方法

### 2.1 甲基汞浓度

冰冻圈中甲基汞含量极低(pg级),因此要特别注意在采样和分析过程中潜在的汞污染问题,需严格遵循“clean hands-dirty hands”的操作规范[50]。甲基汞浓度测定通常采用EPA推荐的方法(Method 1631)进行测试,即“蒸馏-乙基化-吹洗和捕获-冷原子荧光光谱法(CVAFS)”相结合的方法[51]。样品在进行预处理后,还需要对汞形态进行分离,最常用的汞形态分离技术为气相色谱(GC)或高效液相色谱技术(HPLC)。国内对甲基汞检测方法的研究起步较晚,早期一直采用气相色谱法测定甲基汞,但方法灵敏度较低[52]。近些年来,气相色谱与冷原子荧光光谱法联用技术被广泛应用于各种环境样品甲基汞的测定[17,19]。之后,电感耦合等离子体质谱法(ICP-MS)、同位素稀释-电感耦合等离子体质谱法(ID-CV-ICP-MS)以及同位素稀释固相微萃取-气相色谱-电感耦合等离子体质谱法(SPME-GC-ICP-MS)等手段也陆续被应用到甲基汞的检测分析[53-55]。这些方法灵敏度高,检出限低,能很好地满足冰冻圈样品中低水平甲基汞的测定,目前常用的甲基汞测试方法列于表1。

### 2.2 汞甲基化微生物

早期对汞甲基化微生物的鉴定依赖于实验室

表1 不同类型环境样品甲基汞测试方法

Table 1 Methods for the determination of MeHg in various environmental samples

| 样品类型 | 测定方法 | 检出限 | 文献来源 |
|---|---|---|---|
| 雪冰 | GC-CVAFS | 0.02 ng·$L^{-1}$ | [56] |
| 雪冰 | GC-AFS | 0.002 ng·$L^{-1}$ | [57] |
| 冻土 | HPIC-CVAFS | 0.018 ng·$g^{-1}$ | [58] |
| 冻土 | CVAFS | 0.006 ng·$g^{-1}$ | [17] |
| 海冰 | GC-ICP-MS | 0.02 ng·$L^{-1}$ | [59] |
| 冰尘 | GC-CVAFS | 0.02 ng·$g^{-1}$ | [19] |
| 融水 | GC-AFS | 0.02 ng·$L^{-1}$ | [60] |
| 降水 | GC-CVAFS | 0.002 ng·$L^{-1}$ | [61] |
| 鱼类 | GC-CVAFS | 0.12 ng·$g^{-1}$ | [62] |
| 水稻 | GC-CVAFS | 0.003 ng·$g^{-1}$ | [63] |

培养，测试从厌氧沉积物中培养的分离物在添加无机汞后产生甲基汞的能力[64-65]。然而，不同汞甲基化细菌的甲基化能力差异悬殊，如果忽视它是一个物种或菌种的特异性功能，使用这种方法预估环境中汞的甲基化潜力是欠准确的[43]。目前主要通过16S rRNA基因的分析，评估微生物群落结构与环境中甲基汞和生物地球化学特性的关系[45,66-67]。高通量测序作为一种新兴的免培养分子生物学技术，检测快速、准确、信息全面丰富，随着高通量测序技术的不断升级换代，测序通量、读长和准确度也有了很大提升，为认识各种环境中汞甲基化微生物多样性、群落结构组成及其生态功能提供了有利手段[68]。Illumina公司的Solexa和Hiseq是目前全球使用量最大的第二代测序机器。现代分子生物学技术将从环境样品中提取的核酸（DNA或RNA）为分析对象，以进行目标片段的聚合酶链式反应（PCR）扩增，得到的PCR产物通过测序技术来鉴定微生物组成及其多样性。Mpbio公司的FastDNA Spin Kit for Soil试剂盒目前已广泛应用于DNA的提取，它是获得雪冰微生物高分子量、高纯度的基因组DNA非常重要的前提。

对*hgcAB*基因簇、*mer*基因簇等分子层面的研究极大地拓宽了我们对汞甲基化机制的认识[41,48]。Gionfriddo等[69]开发出了一套全新的引物，并将汞甲基化细菌基因序列参考库扩展到了283个，从而可以更准确地获取汞甲基化微生物多样性和环境中甲基汞产生的潜力。当今采用高通量测序获取基因和微生物多样性的生物学信息，开辟了冰冻圈汞甲基化研究的新领域。冰冻圈汞甲基化微生物分析方法概括归纳如图2所示。分析冰冻圈中*hgcAB*基因的方法一种是基于聚合酶链式反应技术，利用*hgcAB*特异引物获得样品中的*hgcAB*基因序列和丰度；另一种是通过BLAST方法从样本宏基因组数据中筛选出*hgcAB*同源序列[34]。

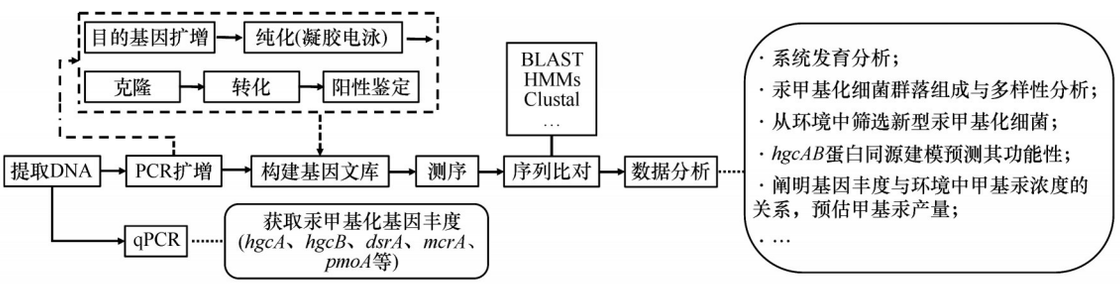


图2　冰冻圈汞甲基化微生物分析方法流程图

Fig. 2　Flow chart showing the analysis of microbial mercury methylation in the cryospheric environments

## 3　冰冻圈典型介质甲基汞研究现状

### 3.1　冰川

冰川作为冰冻圈重要组成因素，全球山地冰川（不包含格陵兰冰盖和南极冰盖）覆盖面积约为$17\times10^4$ km$^2$，共约$11\times10^4$条冰川，主要分布在亚北极和青藏高原等地[10]。早期Jitaru等[70]报道了欧洲阿尔卑斯山雪冰中溶解性甲基汞的含量（0.3~4.0 ng·L$^{-1}$）。近年来科研人员对我国青藏高原冰川也开展了研究工作，Huang等[19]对中国西部冰川上冰尘甲基汞含量进行测试分析，测得甲基汞平均含量为（1.0±0.4）ng·g$^{-1}$，并初步估算出每年冰尘中的甲基汞总量约为（0.7±0.3）kg。微生物群落在汞的生物地球化学转化过程中非常活跃，冰川微生物活动可能发生汞微生物甲基化[71-74]。大气汞沉降到青藏高原冰川之后在冰尘中积累，由于冰尘洞会发生季节性融化，这里便成为各种微生物群落的栖息地[75]。前人研究指出我国西部冰川冰尘甲基汞与总汞的比值较高，说明冰尘表面可能是冰川汞甲基化的潜在位点[19]。Zhang等[56]对青藏高原冰川开展了汞微生物甲基化初步研究，结合老虎沟12号冰川末端6个不同类型样品的汞形态和宏基因组学分析，发现在含大量碎屑和粉尘的尘埃层中汞浓度和汞甲基化基因（*hgcA*）丰度最高。不过该研究中汞甲基化细菌的未分类序列占比很高，微生物群落组成仍不明晰，尚未能全面地阐明汞甲基化的微生物学机制，需要进一步深入研究。

冰川对气候变化响应非常敏感，由于气候变暖，冰川正在以前所未有的速度退缩[9]。冰川消融不仅给下游生态系统带来水资源，而且还可能释放出在冰川中存储了几个世纪的汞[20]。在消融初期，雪冰中的汞以脉冲式释放到冰川径流，近90%的汞

会被释放进入融水[76]。Sun等[77-78]估算出喜马拉雅地区融水径流汞输出通量为337 kg·a$^{-1}$,青藏高原冬克玛底流域中汞输出总量约为747.4 g·a$^{-1}$,亚北极Yukon流域汞输出总量约为4 400 kg·a$^{-1}$[79-80]。Zolkos等[81]对六条北极主要河流进行全年采样调查,估算出总汞输出通量为20 Mg·a$^{-1}$,Dastoor等[18]估算了北极冰川补给流域总汞年产率为(0.03~3.9 g·km$^2$·a$^{-1}$)。尽管青藏高原河流输出总量较低,但山地冰川(如扎当冰川等)的汞输出通量要高于大部分河流,表明青藏高原山地冰川补给流域具有更高的汞释放效率[30-31,78]。在未来全球升温1.5 ℃情景下,到21世纪末青藏高原及周边地区冰川储量将减少2 169~3 217 km$^3$,预估计在未来约11~16吨汞污染物将从冰川释放到下游地区(冰川中汞平均浓度为5.6 ng·L$^{-1}$)[82-84]。富含湿地的冰川流域通常有着更高的总汞和甲基汞含量,生物群落中的汞含量亦较高[85]。阿拉斯加东南部冰河湾下游集水区,存在湿地的溪流溶解汞的比例大得多(0.1~3.4 ng·L$^{-1}$),而裸露的冰川径流溶解汞含量仅为0.1~0.9 ng·L$^{-1}$[85]。湿地可以提供丰富的溶解性有机物,同时这种厌氧环境还有利于使汞甲基化的硫酸盐还原菌生存,可以增强微生物的活性和汞甲基化[85-87]。汞污染物进入湿地之后被汞甲基化微生物转化为毒性更强的甲基汞再次进入径流,对下游地区人类健康和生态系统存在潜在威胁[88-91]。因此,冰川流域生物群落中的甲基汞浓度与流域湿地覆盖率之间存在密切关系,冰川流域湿地可能成为冰川径流中甲基汞净来源,并将其输送到下游生态系统[85,88,92-95]。

大量研究表明偏远冰冻圈下游地区水生生态系统甲基汞的富集效应是非常显著的[85,96-97]。格陵兰冰盖下游湾峡水生生态系统中,融水径流汞浓度(44~348 ng·L$^{-1}$)比北极其他河流(0.3~2.8 ng·L$^{-1}$)高出1~2个数量级,甲基汞浓度(1.0~3.5 ng·L$^{-1}$)甚至高于大多数淡水,与湿地水平相当[20]。我国学者亦研究发现青藏高原河流和湖泊中鱼类甲基汞含量(24.9~1 196 ng·g$^{-1}$)为中国野生鱼类汞浓度的最高水平[96]。此外,前人在冰川补给湖观测到甲基汞的峰值出现在化学跃层,甲基汞占总汞的比例高达57%,同时16S rRNA数据显示检测出硫酸盐还原菌,因此受冰川融水补给的冰川湖化学跃层存在着较强的汞甲基化潜力[97-98]。

### 3.2 多年冻土

多年冻土蕴藏着大量的汞存储量,有研究估算了北极苔原土壤储存汞约184 Gg;青藏高原活动层储存汞约16.6 Gg[16-17]。冻土甲基汞的分布和转化等过程更为复杂,受各种物理、化学和生物因素控制,包括温度、溶解性有机质(DOC)、碱度、pH、总汞浓度、微生物以及其他金属元素或离子(如铁、锰)等[22,46,99-101]。MacMillan等[22]发现冻土环境中甲基汞与溶解有机碳等营养物质,以及甲基汞与微生物之间有很强的相关性。当温度较高(8 ℃)时,在富含有机物和矿物质的土壤层中,甲基汞净产量比温度较低(−2 ℃)时增加了10倍以上[23]。可溶性土壤有机碳类型及其生物可利用性,对促进甲基汞的初始合成特别重要[99]。当冻土活动层融化时,低分子量溶解有机质增加促使形成大量生物可利用的Hg-DOM复合物,从而使无机汞的可利用性增加,更有利于冻土汞甲基化[23-24,100]。

由于气候变暖,多年冻土快速融化、塌陷,形成热融喀斯特地貌,其中最显著的是形成热融湖塘,此过程更有利于微生物将无机汞转化为甲基汞[22-24,102-104]。Tarbier等[24]在亚北极多年冻土区测量了178个土壤样品,甲基汞的浓度范围为0.01~28 ng·g$^{-1}$,与其他地区土壤甲基汞相比要高得多(例如阿拉斯加北部<0.1~3.4 ng·g$^{-1}$),同时发现热融池水和热岩孔隙水中甲基汞含量亦较高(0.01~3.1 ng·L$^{-1}$)[22,24,58,100]。在融化阶段,塌陷的冻土沼泽地比完整冻土带活动层的净甲基化程度还要高一个数量级(甲基汞占总汞的比例高出13倍),表明当热融湖塘形成时,冻土区的汞甲基化率将会显著提高[24]。这主要是由于冻融塌陷使得冻土泥炭表面被淹没在水位以下,从而形成缺氧环境,这种条件非常有利于具有汞甲基化能力的厌氧微生物生存(包括硫酸盐还原菌、铁还原菌及产甲烷菌等)[58,91,100,104-105]。

Lehnherr等[91,104]在加拿大高纬度地区的湿地湖塘中观测到汞甲基化率很高,来自原位甲基化产生(1.8~40 ng·m$^{-2}$·d$^{-1}$)的甲基汞远高于大气沉降(0.03~0.05 ng·m$^{-2}$·d$^{-1}$)。Fahnestock等[100]发现在亚北极多年冻土消融区汞甲基化微生物有着更高的丰度和多样性。通过对全球多种环境样品的微生物基因组序列比对发现,融化的冻土中汞甲基化基因(*hgcA*、*hgcB*)丰度较高,而且这些基因主要存在于产甲烷菌和韧皮菌中[106]。北极快速变暖和降水增多正在加速多年冻土的退化,并促进了历史遗留汞释放到现代生物地球化学循环中,多年冻土大幅度退化已经被认为是北极流域汞通量快速增加

的主要控制因素[29]。前人已在高原观测到大量汞从融化的多年冻土活动层中释放到热融湖塘和其他水体之中[107]，然而青藏高原冻土甲基汞浓度的测定以及汞甲基化过程研究目前还处于空白阶段，这些过程极有可能会对高原汞循环产生直接和重大的影响。

### 3.3　积雪

降雪是冰冻圈大气降水最重要的表现形式，是大气汞进入冰冻圈的重要环节[108]。积雪亦是一种活跃的媒介，消融后将成为环境汞的重要来源[60,109-110]。在南北极地区，南极（0.1~1 ng·$L^{-1}$）与北极（1~5 ng·$L^{-1}$）表层雪冰中的总汞浓度在冬季均很低；然而在春季，受大气汞亏损事件（Atmospheric Mercury Depletion Events，AMDEs）的影响，南北极雪冰中汞的浓度均显著增加[111-119]。早期研究表明积雪中甲基汞与大气沉降密切相关，但在AMDEs期间甲基汞与无机汞表现出完全不同的行为模式[108,111]。Constant等[120]调查了加拿大亚北极地区AMDEs时期雪冰中的甲基汞变化规律，结果表明甲基汞的来源与AMDEs无关。雪冰甲基汞与氯化物之间存在较强的相关性，表明沿海地区积雪甲基汞可能来自海盐气溶胶[108]。海洋中浮游植物可以产生二甲基汞（DMeHg），由于二甲基汞具有较高的挥发性，它可以从海洋释放到大气中，被自由基氧化成单甲基汞，然后随海盐沉降到附近的雪面之上[108,121-123]。值得注意的是新沉降的甲基汞在积雪中并不稳定，15%~56%的甲基汞会在夜间通过去甲基化或以其他方式损失[124]。

积雪中甲基汞的来源在过去被归因于大气沉降，然而随着研究的不断深入，新的证据表明在低纬度苔原的积雪中可能存在着较强的汞甲基化活动[108,111,123-127]。在融雪期间，雪冰融水中甲基汞占总汞的比例增大（从2.7%增加到7.6%）[126]。导致这一现象的原因一方面由于甲基汞从无机/有机物中浸出，在雪冰融化时趋于集中；另一方面是由于雪冰内部可能发生甲基化作用，微生物活动和反应可能在积雪中产生了甲基汞[108,120,123-127]。极地积雪的pH值通常为酸性，有机物以及微生物等存在于雪层中，因此在消融的积雪中存在微生物汞甲基化的可能[120]。随着气候变暖，温度升高会促使积雪中微生物种群数量增长，从而导致微生物影响汞甲基化的强度大幅增加[25]。Loseto等[60]估算出北极地区积雪中甲基汞产量约为1.5 mg·$km^{-2}$·$d^{-1}$，并指出北极康沃利斯岛陆地积雪融化是地表径流中最重要的甲基汞来源。

### 3.4　海冰

海冰中的甲基汞来源主要包括大气沉降和原位汞甲基化。海洋中的浮游植物生产的二甲基汞从海面释放，在大气中降解为甲基汞，最后又沉降到海冰或海水中，这是海冰甲基汞重要来源之一[121]。Baya等[128]研究指出海冰覆盖和初级生产是促进海洋浮游植物生产二甲基汞的主要驱动因素。海冰甲基汞来源除了大气和海洋之外，海冰原位产甲基汞亦非常重要。大量研究已表明海冰中下部和冰水界面是已知的可能产生甲基汞的区位[28,129]。因此，海冰中汞的原位甲基化使得海冰融化极有可能成为海洋生态系统新的甲基汞来源。

Cossa等[129]在南极地区开展了“积雪-海冰-卤水-海水”连续系统中的汞循环研究。结果表明，尽管海冰与海水的甲基汞浓度范围大致相同，但在整个系统中分布较为复杂，海冰底部的甲基汞浓度达到峰值。Beattie等[28]在北极海冰中亦观测到了类似的分布模式，海冰中下部甲基汞与总汞比值通常非常高。相比底层水体，海冰具有更高的微生物活性、丰度和多样性[130-133]。大气汞通过干湿沉降的方式积累在海冰和海水中为微生物甲基化提供了底物[134-135]，并且海冰内部存在局部厌氧条件，成为硫酸盐还原菌和铁还原菌繁衍的氧化还原区，这种现象非常有利于海冰汞甲基化的发生[131]。此外海冰-海水界面通常具有丰富的藻类、叶绿素和有机质[132-133]。在春季和夏季，由于冰雪融化和浮游植物生长，无机汞被浮游植物吸收利用，这些过程极有可能在海冰-海水界面发生甲基化[133-134]。已有研究表明，在南极和北极海冰底部都观测到较高水平的甲基汞含量，这与原位产生甲基汞密切相关，因为海冰底部也有较高的细菌丰度和有机质含量，而且厌氧环境亦有利于汞甲基化细菌的生长繁殖[28,129]。

最新的研究不断揭示出潜在的新型海洋汞甲基化微生物，它们具有更强的耐氧性和更广泛的生境范围，这些都是有利于海冰汞甲基化的理想条件[59]。Gionfriddo等[59]通过对南极海冰汞甲基化研究表明，南极海冰中存在甲基汞的富集现象，主要是由微嗜氧细菌-硝化刺菌属（*Nitrospina*）主导产生的，参与调控微生物汞甲基化的基因为*hgcA-like*。这与之前的认知有所不同，过去认为只有厌氧条件支持显著的汞甲基化过程，且汞甲基化微生物都属

于厌氧细菌。因此,海冰汞的研究不仅揭示了更多潜在的新型海洋汞甲基化微生物,而且极大地拓宽了我们对汞甲基化微生物耐氧性的认识。气候变暖背景下,海冰变薄、季节性海冰区扩大、表层海洋分层加剧和浮游生物生态动力学的变化,这些过程都会造成海洋甲基汞产量增加[26,136]。从海冰融化释放出来的大量甲基汞进入海洋生态系统之后会在食物链中富集和放大,将会对南北极水生食物链中高营养级的动物构成很大威胁[26,28,59,134]。通过对北极多年海冰的融化情况的估算,每年输入北冰洋的总汞和甲基汞的通量估计分别为420 kg·a$^{-1}$和42 kg·a$^{-1}$[28]。南极海冰融化亦会给海洋分别带来超过9 600 kg·a$^{-1}$总汞和18 kg·a$^{-1}$甲基汞,总汞的估算值要比北冰洋高出一个数量级[59,134]。为方便后续研究,将冰冻圈总汞和甲基汞浓度水平相关信息汇总于表2。

表2 冰冻圈总汞和甲基汞浓度水平

Table 2 Concentrations of MeHg in the cryospheric environment

| 地点 | 样品类型 | 甲基汞 | 总汞 | 文献来源 |
|---|---|---|---|---|
| Devon Island, Canada | 多年冻土 | 0.3~3.1 ng·g$^{-1}$ | — | [58] |
| Barrow, Alaska | 热融湖塘 | 0.3~2.3 ng·L$^{-1}$ | 25.3~141.2 ng·L$^{-1}$ | [23] |
| Tibetan Plateau, China | 多年冻土 | — | 5.6~39.3 ng·g$^{-1}$ | [17] |
| Fennoscandia | 多年冻土 | 0.004~28 ng·g$^{-1}$ | 1.1~210 ng·g$^{-1}$ | [24] |
| Tussock tundra, Alaska | 多年冻土 | — | 56~226 ng·g$^{-1}$ | [16] |
| Laohugou No. 12, Tibetan Plateau, China | 雪冰 | 0.005~0.5 ng·L$^{-1}$ | 24.4~172.1 ng·L$^{-1}$ | [56] |
| Tibetan Plateau, China | 冰尘 | 0.4~1.7 ng·g$^{-1}$ | 17.9~114.5 ng·g$^{-1}$ | [19] |
| French Alps | 表层雪 | 0.002~0.3 ng·L$^{-1}$ | 1.8~169.5 ng·L$^{-1}$ | [57] |
| Spitsbergen Island, Norway | 表层雪 | 0.004~0.5 ng·L$^{-1}$ | 1~90 ng·L$^{-1}$ | [123] |
| Resolute Bay, High Arctic, Canada | 表层雪 | <0.015~0.2 ng·L$^{-1}$ | 2.6~150 ng·L$^{-1}$ | [125] |
| Hudson Bay, Canada | 表层雪 | 0.04~0.14 ng·L$^{-1}$ | 1.6~5.1 ng·L$^{-1}$ | [120] |
| Alberta, Canada | 表层雪 | 0.02~0.3 ng·L$^{-1}$ | 0.8~9.0 ng·L$^{-1}$ | [137] |
| Antarctic | 积雪 | <0.02~0.3 ng·L$^{-1}$ | 1.0~30.6 ng·L$^{-1}$ | [59] |
| McMurdo/Ross Island, Antarctic | 积雪 | — | 40~430 ng·L$^{-1}$ | [114] |
| Casey Station, Antarctic | 海冰 | <0.009~0.09 ng·L$^{-1}$ | 0.2~5.7 ng·L$^{-1}$ | [129] |
| Antarctic | 海冰 | <0.02~0.2 ng·L$^{-1}$ | 1.4~179 ng·L$^{-1}$ | [59] |
| Beaufort Sea and McClure Strait, Arctic | 海冰 | <0.02~0.5 ng·L$^{-1}$ | 0.1~12.2 ng·L$^{-1}$ | [28] |
| Southern Ocean | 海水 | <0.02~0.1 ng·L$^{-1}$ | 0.2~23 ng·L$^{-1}$ | [59] |
| Arctic Ocean | 海水 | 0.006~0.07 ng·L$^{-1}$ | 0.09~1.4 ng·L$^{-1}$ | [135] |
| Resolute Bay, High Arctic, Canada | 海水 | 0.06~0.1 ng·L$^{-1}$ | 0.1~0.2 ng·L$^{-1}$ | [125] |

注:表格中海水均为海冰覆盖水域。

Note: the seawater refers to the area covered by sea ice in the table.

## 4 冰冻圈甲基汞暴露风险

自工业革命以来,人类活动已深刻改变了全球汞循环过程,造成大量人为汞污染物进入冰冻圈。近期研究发现,嗜冷菌微生物已经进化出了特殊机制来克服低温环境,并积极参与到冰冻圈汞甲基化过程,将冰冻圈自然环境中无机汞转化为甲基汞[34,138-139]。由于生物富集和累积作用,海豹、鲸鱼、海鸟及北极熊体内的汞含量非常高,这些处于食物链高层的捕食者体内的汞几乎完全是甲基汞形式[140-152]。在北极生物群落中海鸟作为顶级捕食者,寿命长、分布广泛而且对汞吸收很敏感,因此海鸟经常被用作海洋生态系统中汞富集研究的生物指示剂[145]。研究发现海鸟羽毛中的汞浓度已超过5 μg·g$^{-1}$(dw)的毒性阈值,而这种高水平浓度通常被认为有害健康和影响生殖[144-145,153-154]。南极磷虾是污染物向上层营养级生物转移的重要环节,磷虾体内总汞浓度范围为4.04~19.4 ng·g$^{-1}$,甲基汞浓度范围为0.26~2.94 ng·g$^{-1}$,依赖磷虾的消费者(如幼鱼)通过食物链大量富集和积累甲基汞[146-148]。已有研究报道南极海洋食物链顶端的物种(如企鹅和哺乳动物)体内甲基汞富集系数高达多个数量级,在北极地区捕食性鱼类中甲基汞含量甚至比海水高出约一百万倍[149-150]。

冰冻圈甲基汞富集作用同样对生活在该区域的人类健康构成重大威胁。北极地区居住着土著居民(如因纽特人),他们依赖于捕获鱼类和海洋哺乳动物为食,通过饮食摄入是当地居民暴露甲基汞的主要途径[3,155-157]。北极因纽特人原住民由于常年捕食海洋生物,人体内的甲基汞含量都远超过了毒理学阈值[104,158]。由于甲基汞对成人和儿童的毒性效应有所不同,处于发育中的儿童中枢神经系统比成年人对甲基汞更为敏感,而孕妇长期处于低剂量甲基汞暴露将会对婴儿的智力发育产生严重影响[33]。研究表明,北极一些地区50%以上的母亲和育龄妇女的血液中的汞含量已超过美国环境保护署建议的标准浓度(5.8 μg·$L^{-1}$),且在格陵兰岛部分地区近乎90%的育龄妇女血液中的汞含量都超标,这些都表明我们迫切需要重视冰冻圈甲基汞暴露风险[33,157-158]。

## 5　结论与展望

在气候变暖背景下,冰冻圈温度升高能够促进微生物活动,积极参与汞甲基化过程,提高了甲基汞的净生产率。不仅如此,储存于冰冻圈的汞污染物重新释放到环境中参与汞的地球化学循环并输送到下游水生生态系统,当重新释放的汞进入湿地、湖泊和湾峡等环境,这里将成为高效的甲基汞生产场所。一旦甲基汞从冰冻圈环境进入水体,在水生食物链中富集并放大,会对人类和野生生物构成潜在威胁。目前有关温带环境陆地和海洋上甲基汞生产过程已经有了大量的认识,然而人们对冰冻圈系统中甲基汞的来源和生产/降解等研究得较少。

本文综述了冰冻圈甲基汞研究进展,总结了甲基汞在冰冻圈(冰川、冻土、积雪和海冰等)中的迁移、转化和归宿等生物地球化学过程,阐明了汞甲基化微生物对冰冻圈甲基汞产生机制和变化过程的影响,并指出了甲基汞在冰冻圈存在较大的生态暴露风险。冰冻圈甲基汞是近年来新兴的研究领域,迄今对其认识仍非常有限。基于目前研究现状及存在的不足,提出以下几点研究展望:

(1)加强冰冻圈汞甲基化过程与机理研究。冰冻圈在全球汞的生物地球化学循环中起着非常重要的作用,其中汞甲基化是特别关键的过程,然而目前对冰冻圈中甲基汞的认识仍处于初级阶段,存在高度不确定性。基于温带或者热带的甲基化机理认识并不完全适用于冰冻圈,并且可能会限制对于这一特殊环境中的汞甲基化过程的认识。因此,迫切需要加强冰冻圈特殊环境中甲基汞过程与机理研究。

(2)学科交叉引入新研究手段和方法。环境中微生物甲基化是主导甲基汞生产最重要的因素,然而目前对全球冰冻圈中具有产甲基汞能力的微生物多样性的认识仍很匮乏。需要引入新的手段和方法,结合环境影响要素,共同揭示冰冻圈甲基汞现代过程和演化规律。通过利用宏基因组学(元基因组学和元转录组学)来解析汞甲基化微生物群落组成,扩大汞甲基化细菌基因序列参考库,全面解译甲基汞微生物生产机制,这对于深化理解冰冻圈甲基汞的认识至关重要。

(3)关注气候变化对冰冻圈甲基汞循环的影响及环境效应。冰冻圈是对全球变暖响应最为敏感的地球圈层,作为全球重要的汞储库,随着全球气候变暖加剧,封存于冰冻圈中汞的行为和命运将很大程度上影响和改变全球汞生物地球化学循环模态。未来阐明气候变化对汞甲基化的影响对于冰冻圈生态系统非常重要,需要进一步深化开展气候快速变化情景下冰冻圈甲基汞分布、行为和环境影响的研究。在冰冻圈建立起对不同介质(冰川、冰盖、多年冻土、积雪和海冰等)汞消融释放及生物区系汞污染物的动态长期监测,科学评估气候变化下冰冻圈甲基汞未来变化趋势及冰冻圈汞污染物二次释放等环境过程构成的生态环境安全威胁。

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# Research progress on methylmercury in the cryosphere

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**Abstract**：The cryosphere is one of the climate system component which plays an important role in the global biogeochemical mercury cycling. Anthropogenic mercury has been transported to remote cold and high regions worldwide through atmospheric circulation，and then transformed into methylmercury in the cryosphere. As one of the highly toxic environmental pollutants，methylmercury can be greatly bioaccumulated and biomagnified through the food web，which potentially poses threats to human and wildlife as well as the global cryospheric environments. In order to gain a full picture of research progress on methylmercury in the cryosphere，our study comprehensively summarized the concentration levels and analytical methods of methylmercury，and biogeochemical processes such as the migration，transformation and fate of methylmercury in various cryospheric environments including glacier，permafrost，snow ice and sea ice. We particularly made the literature review of microbial mercury methylation and evaluated the risk of methylmercury exposure to human and wildlife in the cryosphere. Meanwhile，we focus on distribution，behavior，and environmental effects of methylmercury in the cryosphere against the backdrop of climate change，which is essential for assessing the exposure risk of methylmercury to humans and wildlife. Perspectives of methylmercury researches in the cryosphere have also been highlighted in this review，though there is existing a knowledge gap of biogeochemical methylmercury cycling in the low temperature environments which merits further study.

**Key words**：methylmercury；cryosphere；glacier；permafrost；snow ice；sea ice

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