

Atmospheric gaseous elemental mercury in the Arctic: role of long-range transport and sea ice

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Abstract The potential sources of atmospheric mercury in the Arctic are still not well understood. Here, we analyzed the round-year observations of atmospheric gaseous elemental mercury (GEM) concentrations in 2010 at two sites in the Arctic: Zeppelin Observatory (78.90°N, 11.88°E) and Alert Observatory (82.47°N, 62.50°W). The results showed clearly different characteristics at these two sites. During the winter of 2010, the long-range transport of anthropogenic emissions was the dominant factor for elevated GEM at Zeppelin Observatory while which had little contribution to GEM at Alert Observatory. The change in GEM at Zeppelin Observatory during January, February and October was dominated by long-range transported anthropogenic emissions. The emission inventory combined with backward trajectory analysis suggested that the main sources were unintentional industrial sources, stationary combustion sources, and intentional use and product waste-associated sectors, which contributed 49%, 33%, and 18%, respectively. Potential source contribution function analysis was then conducted and found that Europe was the important source region. During the summer of 2010, sea ice concentration was an important factor affecting GEM at Alert Observatory while which had little effect on GEM at Zeppelin Observatory. These results further indicated the role of anthropogenic sources and climate warming on the spatial variation in GEM over the Arctic.

Keywords mercury, anthropogenic emissions, sea ice, Arctic

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1 Introduction

Gaseous elemental mercury (GEM) is the dominant form of atmospheric Hg and the main atmospheric Hg species transported to the Arctic (Lindberg et al., 2007; Schroeder and Munthe, 1998). After being transported to the Arctic, GEM can be chemically transformed into inorganic Hg²⁺ and then deposited in the Arctic environment, while the residual GEM can leave from the Arctic with the movement of air mass (UNEP, 2019). The models and field

studies showed that the long-range transport of atmospheric Hg emitted by anthropogenic emissions from source regions to the Arctic mainly takes place in winter and spring (Hirdman et al., 2009; Lamborg et al., 2002; Mason and Sheu, 2002; Seigneur et al., 2004; Stohl et al., 2007). Field studies suggested that atmospheric GEM can rapidly transform to Hg²⁺ (hours to days) and deposit to the surface during spring in the Arctic (Lindberg et al., 2001; Poissant and Pilote, 2003; Schroeder et al., 1998; Steffen et al., 2005). This phenomenon is known as atmospheric mercury depletion events (AMDEs) (Schroeder et al., 1998). The occurrence of AMDEs would make an increase in Hg deposition on the surface (e.g., snow, soil, vegetation) around the Arctic Ocean. A previous study surveyed the

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nationwide moss in Norway and found higher concentrations of mercury in moss growing at the Arctic coast (Berg et al., 2008). A model study suggested that the rate of mercury deposition in the area north of the Polar Circle was $89 \text{ t} \cdot \text{a}^{-1}$ and $208 \text{ t} \cdot \text{a}^{-1}$, respectively, through scenario calculations without and with an AMDE (Skov et al., 2004).

Previous studies suggested that halogen radicals play an important role in the occurrence of AMDEs in the Arctic, and the oxidation of Hg^0 by halogen radicals is an *in situ* process (Faïn et al., 2009a; Swartzendruber et al., 2006; Xie et al., 2008). Numerous studies have shown that Br atoms and BrO radicals are the main oxidants for Hg^0 during AMDEs in the Arctic (Balabanov and Peterson, 2003; Boudries and Bottenheim, 2000; Calvert and Lindberg, 2003; Goodsite et al., 2004; Khalizov et al., 2003; Lindberg et al., 2002; Skov et al., 2004; Tossell, 2003). Moreover, iodinated compounds can possibly influence the removal of GEM during AMDEs indirectly since I atoms and O_3 molecules can react rapidly and produce IO radicals, which can increase the concentrations of Br atoms and I atoms through the interaction with BrO radicals (Calvert and Lindberg, 2004).

Although there are no local major anthropogenic Hg sources in the Arctic, the effect of long-range transport of anthropogenic GEM emissions from other continents on atmospheric GEM in the Arctic is nonnegligible due to a relatively long atmospheric residence time of GEM (0.7–1.4 a). It has been reported that the transport process mentioned above mainly takes place during winter and spring (Raatz, 1984). Previous studies have reported the contributions of anthropogenic GEM emissions (including agricultural fires) from Europe and Asia to atmospheric GEM concentrations in the Arctic via long-range transport (Faïn et al., 2009b; Hirdman et al., 2009; Jaffe et al., 2005; Obrist et al., 2008; Stohl et al., 2007; Weiss-Penzias et al., 2007). These results indicated the importance of controlling anthropogenic emissions to decrease atmospheric GEM concentrations in the Arctic. Although these studies indicated the contributions of anthropogenic emissions to atmospheric GEM in the Arctic, the proportions of different sectors of emission sources in the long-range transported anthropogenic emissions and their corresponding source regions are still lacking.

The Zeppelin Observatory and Alert Observatory are both located in high Arctic regions; thus, they are considered to be the appropriate sites to study the characteristics of atmospheric Hg in the Arctic. The objective of this study is to investigate the potential influencing factors for atmospheric GEM concentrations at two sites (Zeppelin Observatory and Alert Observatory) in the Arctic in 2010 using backward trajectory, emission inventory. This study can help to improve the knowledge about the sources of atmospheric GEM in the Arctic and put forward more targeted strategies for controlling atmospheric GEM pollution in the Arctic.

2 Methods

2.1 Site locations

Zeppelin Observatory (78.90°N , 11.88°E) is located at Mount Zeppelin in Svalbard, Norway. It is 474 m above sea level and approximately 2 km from a small scientific community, with minimal effects from local anthropogenic sources. The Alert Observatory (82.47°N , 62.50°W) is a high Arctic site located at the northern tip of Ellesmere Island, Nunavut, Canada. The locations of these two Arctic stations are shown in Figure 1.

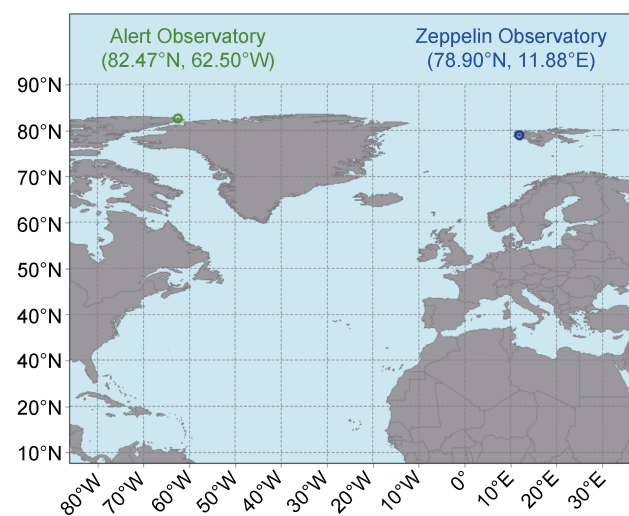


Figure 1 Schematic diagram of the location of the Zeppelin Observatory and Alert Observatory in the Arctic. The solid blue pentagram represents the location of the Zeppelin Observatory, and the solid green pentagram represents the location of the Alert Observatory.

2.2 Experimental methods

The 2010 gaseous elemental mercury concentrations data with an hourly resolution at Zeppelin Observatory and Alert Observatory were downloaded through the EBAS database website (<http://ebas-data.nilu.no/>).

TrajStat, a geographical information system-based software, and gridded meteorological data (Global Data Assimilation System, GDAS1) from the USA The National Oceanic and Atmospheric Administration was used to calculate 168-h backward trajectories of air masses arriving at a height of 200 m above ground level every 1 h from 2010-01-01 0:00 to 2010-12-31 23:00. The cumulative anthropogenic GEM emission (cumulative Hg^0 emission) was used to represent the contribution of long-range transport from anthropogenic emissions to GEM concentrations at the sites in the Arctic. In this study, the cumulative Hg^0 emissions at the Zeppelin Observatory and Alert Observatory with a daily resolution in 2010 were calculated by summing all the gridded emissions of the grids encountered by the air mass transported during the

past 168 hours based on the Arctic Monitoring and Assessment Programme (AMAP)/United Nations Environment Programme (UNEP) gridded GEM emission inventory for 2010 (Fu et al., 2019; UNEP, 2013). The fractional cumulative GEM emissions for three anthropogenic emission sectors (unintentional industrial sources sector, stationary combustion sources sector, and intentional use and product waste-associated sector) were respectively calculated by calculating the fraction of each sector's cumulative Hg^0 emissions to the sum of these three sectors' cumulative Hg^0 emissions in different months. More detailed methods can be found in Yue et al. (2021).

According to the gridded GEM emission inventory in 2010 published by AMAP/UNEP, the GEM emission sources were divided into stationary combustion sources, industrial sources, and intentional use and product waste associated sectors (UNEP, 2013). During the periods with a significant contribution of long-range transport from anthropogenic emissions to GEM concentrations at the two sites in the Arctic, the cumulative Hg^0 emission of these three different emission source sectors was calculated by summing up all the gridded emissions of the grids encountered by the air mass transported during the past 168 h, based on the gridded GEM emission inventory in 2010 published by UNEP (2013). The cumulative Hg^0 emissions of three different emission source sectors were used to indicate the contribution of long-range transport from anthropogenic emissions of different emission source sectors to GEM concentrations at Zeppelin Observatory during the periods with a significant contribution of long-range transport from anthropogenic emissions to GEM

concentrations at this site. The potential source contribution function (PSCF) analysis was conducted here to identify the potential source regions of different emission source sectors during these periods (the average value of atmospheric GEM concentrations during one month was chosen to be the pollution criterion in this study).

3 Results and discussion

3.1 The characteristics of atmospheric GEM concentrations at Zeppelin Observatory and Alert Observatory

Based on the atmospheric GEM concentrations data at Zeppelin Observatory and Alert Observatory in 2010 provided by the EBAS database website (<http://ebas-data.nilu.no/>), it was found that the atmospheric GEM concentrations ranged from 0.86 to 2.02 $\text{ng}\cdot\text{m}^{-3}$ and from 0.40 to 2.21 $\text{ng}\cdot\text{m}^{-3}$ at Zeppelin Observatory and Alert Observatory, respectively. The average values of atmospheric GEM concentrations in different seasons at these two sites are described in Table 1. The results showed that the atmospheric GEM concentrations at these two sites both reached the lowest value in spring, which indicated the effect of AMDEs on atmospheric GEM concentrations in the Arctic (Schroeder et al., 1998). The atmospheric GEM concentrations at two Arctic sites and other research areas are compared in Table 2. It was found that the average value of atmospheric GEM concentrations at two Arctic sites was generally lower than that in urban and rural areas worldwide.

Table 1 The average value of atmospheric GEM concentrations in different seasons at Zeppelin Observatory and Alert Observatory in 2010 (unit: $\text{ng}\cdot\text{m}^{-3}$)

	Spring		Summer		Autumn		Winter	
	mean	2SD	mean	2SD	mean	2SD	mean	2SD
Zeppelin Observatory	1.47	0.44	1.57	0.21	1.56	0.22	1.64	0.25
Alert Observatory	1.34	0.77	1.61	0.45	1.44	0.24	1.41	0.13

Table 2 Atmospheric GEM concentrations at two Arctic sites and other research areas

Location	Area type	Time	GEM (mean)/($\text{ng}\cdot\text{m}^{-3}$)	Reference
Hefei, China	Suburb	2013-07-2014-06	4.07	Hong et al., 2016
Beijing, China	Rural	2008-12-2009-11	3.22	Zhang et al., 2013
Guiyang, China	Urban	2009-08-2009-12	9.72	Fu et al., 2011
Detroit, USA	Urban	2004-01-2004-12	2.50	Liu et al., 2010
Dexter, USA	Rural	2004-01-2004-12	1.60	Liu et al., 2010
Houston, USA	Urban	2006-08-2006-10	1.66	Brooks et al., 2010
Florida, USA	Urban	2009-07-2010-07	1.30	Peterson et al., 2012
Maryland, USA	Suburb	2007-2015	1.41	Ren et al., 2016
Gothenburg, Sweden	Urban	2005-02-2005-03	1.96	Li et al., 2008
Nova Scotia, Canada	Urban	2010-01-2011-12	1.67	Cheng et al., 2014
Zeppelin Observatory, Arctic	Remote	2010-01-2010-12	1.56	This study
Alert Observatory, Arctic	Remote	2010-01-2010-12	1.45	This study

3.2 Seasonal variations at Zeppelin Observatory and the effect of emissions

The variation in atmospheric GEM concentrations at Zeppelin Observatory in 2010 and the variation in cumulative anthropogenic GEM emissions in 2010 are displayed in Figure 2. It was found that the variation trend of atmospheric GEM concentrations at this station was similar to the variation trend of cumulative anthropogenic GEM emission during three months (January, February and October). As shown in Figures 3a, 3b, and 3c, the correlations between atmospheric GEM concentrations and cumulative anthropogenic GEM emissions during these months were statistically significant positive ($r=0.51$ and $p<0.01$ in January, $r=0.41$ and $p<0.05$ in February, $r=0.55$ and $p<0.01$ in October). This suggested that the atmospheric GEM concentrations during these periods at Zeppelin Observatory were significantly contributed by the long-range transport of continental anthropogenic emissions. Moreover, the occurrence of the maximum value for atmospheric GEM concentrations in December, was accompanied by a sharp increase in cumulative anthropogenic GEM emissions, which may also indicate the contribution of the long-range transport of continental anthropogenic emissions.

For the periods with a significant contribution of long-range transport of continental anthropogenic emissions, based on the method that combined emission inventory and backward trajectory (mentioned in Section 2.2), our study investigated the fractional cumulative GEM emission for different anthropogenic emission sectors through the global GEM emission inventory of three different anthropogenic emission sectors (including unintentional industrial sources, stationary combustion sources, and intentional use and product waste associated sectors) (Figure 4). The results

showed that the average value of fractional cumulative GEM emissions for unintentional industrial sources was 49%. This suggests that unintentional industrial sources were responsible for nearly half of the anthropogenic GEM emissions, which had a significant contribution to the atmospheric GEM in 2010 at Zeppelin Observatory in the Arctic. Moreover, the average values of fractional cumulative GEM emissions for stationary combustion sources and intentional use and product waste-associated sectors were 33% and 18%, respectively, which indicates fewer contributions of these sectors to the anthropogenic GEM emissions mentioned above.

Based on the analysis above, PSCF analysis was conducted in this study to identify the potential source regions of anthropogenic GEM emissions that had a significant contribution to the atmospheric GEM in 2010 at the Zeppelin Observatory in the Arctic (Figure 5). The results showed that the potential source regions of the three anthropogenic emission sectors were generally similar during the same period. It was found that Europe was the most important source region of anthropogenic GEM emissions, which had a significant contribution to the atmospheric GEM in 2010 at Zeppelin Observatory in the Arctic. The results are in agreement with a previous report that Europe was an important anthropogenic source of Arctic mercury (Hirdman et al., 2009).

3.3 Seasonal variations at Alert Observatory and the effect of sea ice

The variation in atmospheric GEM concentrations at the Alert Observatory in 2010 and the variation in cumulative anthropogenic GEM emissions in 2010 are displayed in Figure 6. It was found that the variations in atmospheric GEM concentrations and cumulative anthropogenic GEM emissions did not show a similar trend

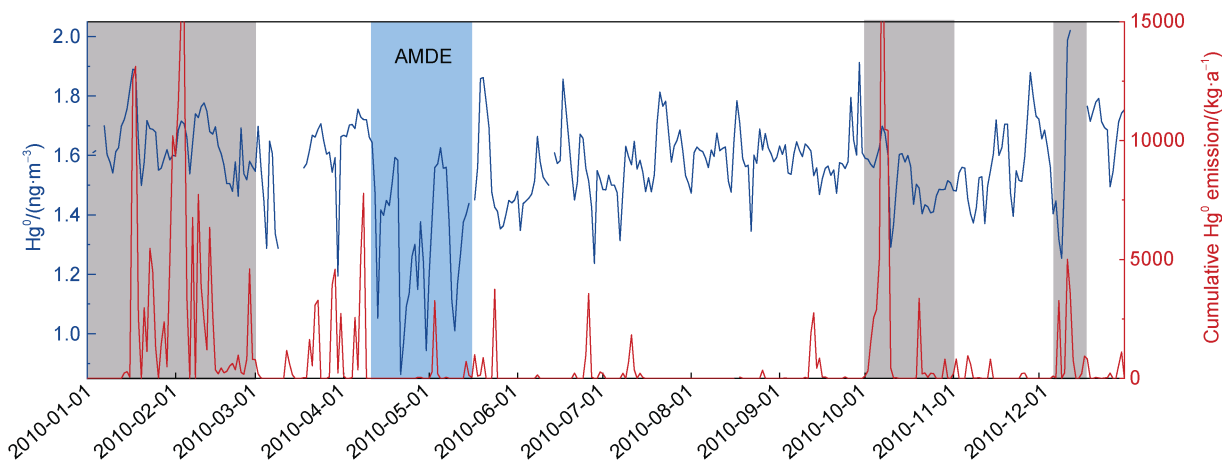


Figure 2 The variation in atmospheric GEM concentrations at Zeppelin Observatory (blue line) and the variation in cumulative anthropogenic GEM emissions (red line) in 2010. The gray shadow areas represent the periods with a significant contribution of long-range transport of continental anthropogenic emissions. The blue shadow areas represent the duration of AMDEs.

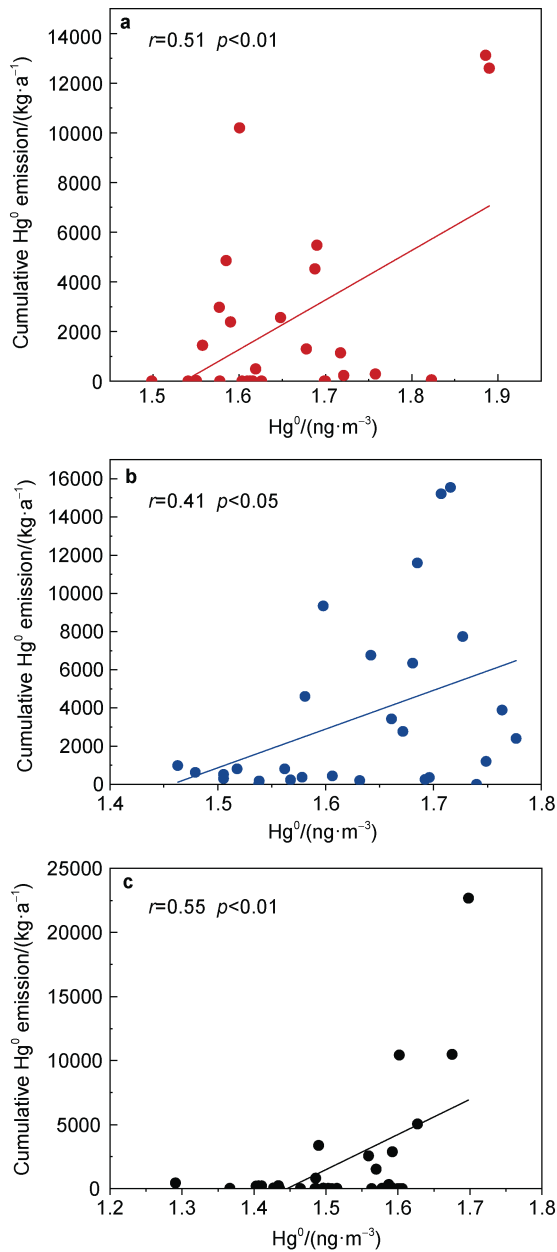


Figure 3 The correlations between atmospheric GEM concentrations at Zeppelin Observatory and cumulative anthropogenic GEM emissions in January 2010 (a), February 2010 (b) and October 2010 (c).

during any period in 2010. Moreover, the correlations between atmospheric GEM concentrations at the Alert Observatory and cumulative anthropogenic GEM emissions in 2010 showed that these two parameters had no correlations except during September (Figure 7). The cumulative anthropogenic GEM emissions during September 2010 were mostly not more than $2 \text{ kg}\cdot\text{a}^{-1}$, which was three orders of magnitude less than the average value of that for the Zeppelin Observatory during the periods with a significant contribution of long-range transport of continental anthropogenic emissions in 2010. These results

showed that the long-range transport of continental anthropogenic emissions may not be the dominant influencing factor for the atmospheric GEM concentrations in 2010 at the Alert Observatory in the Arctic.

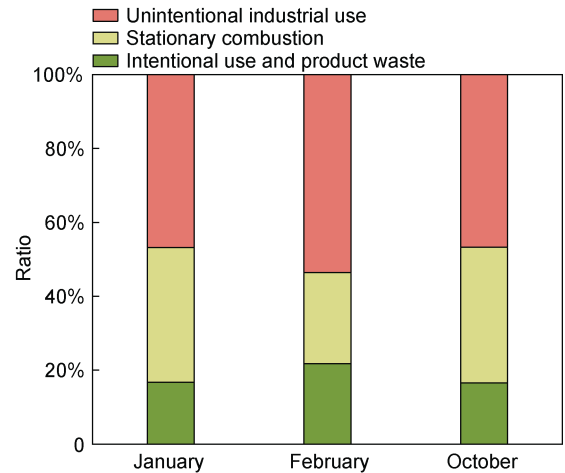


Figure 4 The fractional cumulative GEM emissions for three anthropogenic emission sectors (unintentional industrial sources, stationary combustion sources, and intentional use and product waste-associated sectors) during the periods with a significant contribution of long-range transport of continental anthropogenic emissions (January, February and October).

In this study, it was found that the atmospheric GEM concentrations at Alert Observatory showed an obvious increase after experiencing AMDEs, which was accompanied by a decrease in sea ice concentrations. The occurrence of the highest atmospheric GEM concentration was accompanied by the lowest sea ice concentration (Figure 8a). However, there was no correlation between the sea ice concentration and atmospheric GEM concentration at Alert Observatory during summer ($r=-0.04$), which may imply the non-linear relationship between oceanic Hg emission and sea ice melting, and/or influences of factors other than sea ice on GEM at Alert Observatory during this period. Moreover, it was found that the atmospheric GEM concentrations at Zeppelin Observatory varied slightly with an increase in sea ice concentrations during this period (Figure 8b), which may indicate that sea ice concentrations had little effect on GEM at Zeppelin Observatory during the summer of 2010.

On the one hand, previous studies have found that halogen radicals (especially Br radicals) are important oxidants for atmospheric GEM oxidation during spring in the Arctic (Fain et al., 2009a; Swartzendruber et al., 2006; Xie et al., 2008). During spring in the Arctic, the bromine-rich seawater in the open sea located in the sea ice areas or between the sea ice areas and the shore can refreeze due to the diurnal temperature difference (AMAP, 2011). During the refreezing of seawater, bromide will accumulate and concentrate on the surface of the sea ice and be released

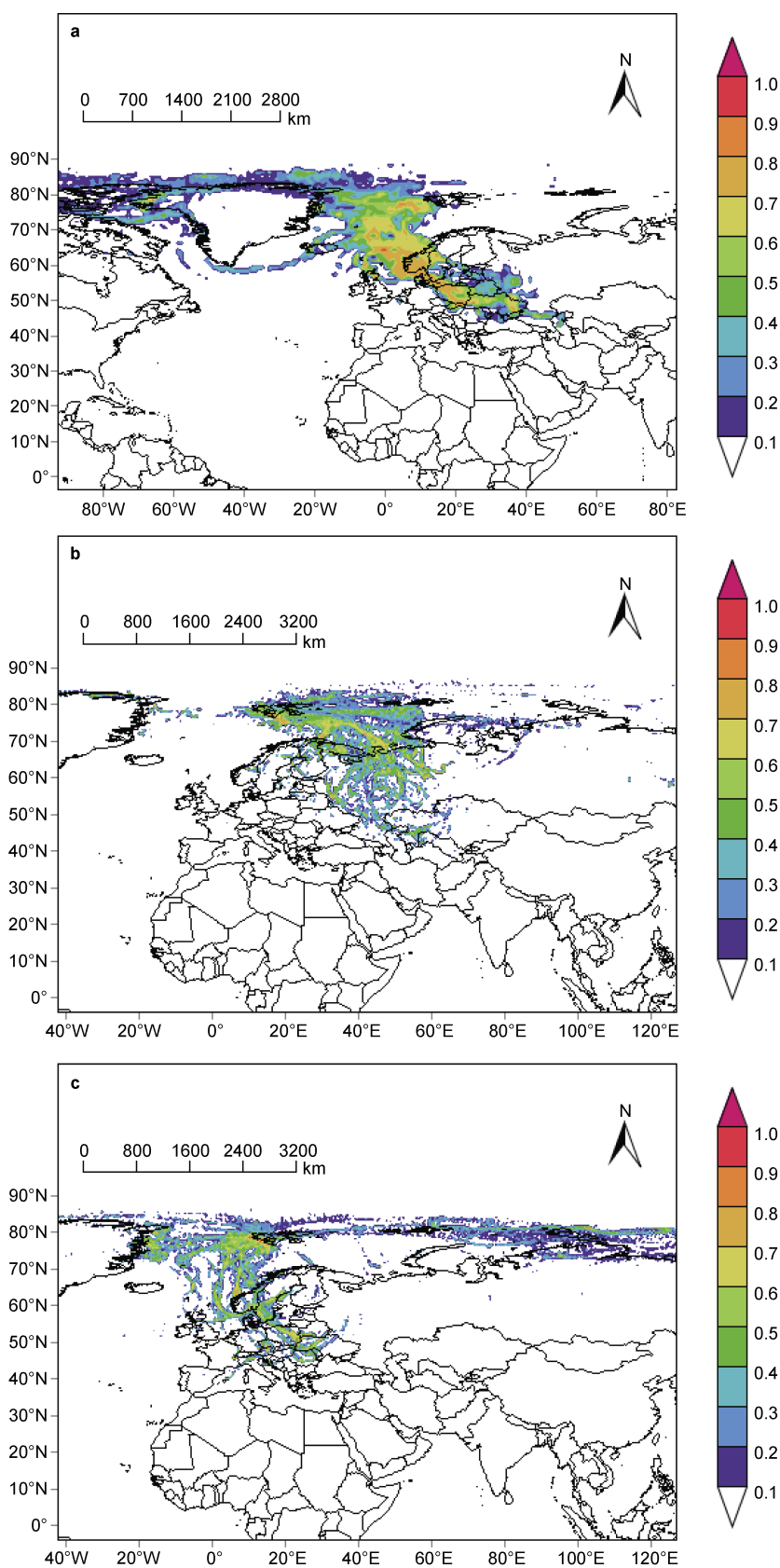


Figure 5 The potential source regions of anthropogenic GEM emissions that had a significant contribution to the atmospheric GEM in January 2010 (a), February 2010 (b) and October 2010 (c) at Zeppelin Observatory in the Arctic.

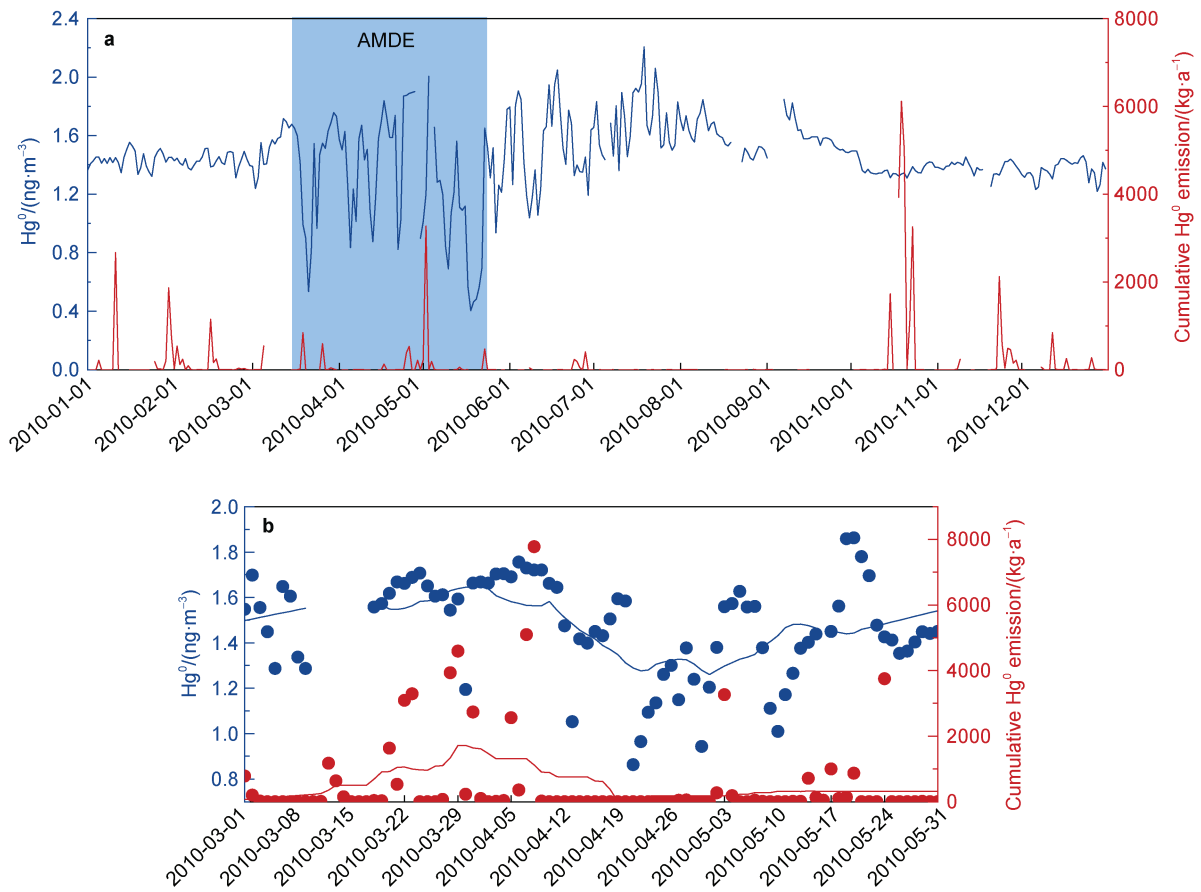


Figure 6 a, the variation in atmospheric GEM concentrations at the Alert Observatory (blue line) and the variation in cumulative anthropogenic GEM emissions (red line) in 2010. The blue shadow areas represent the duration of AMDEs. b, the trend lines of atmospheric GEM concentrations and cumulative anthropogenic GEM emissions during AMDEs in 2010 have been smoothed by Origin (the points of window are 20 and the polynomial order is 1).

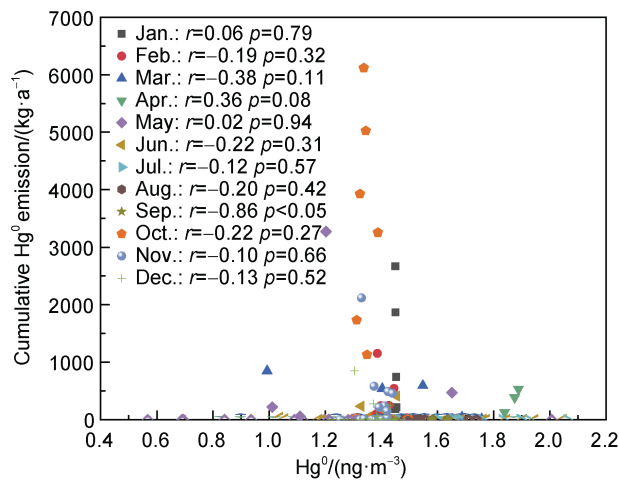


Figure 7 The correlations between atmospheric GEM concentrations at the Alert Observatory and cumulative anthropogenic GEM emissions in different months in 2010.

from the surface as Br_2 (AMAP, 2011). Then, the released Br_2 will react with ozone to generate BrO in the atmosphere, which will transform into Br radicals through a series of

reactions to make an important contribution to atmospheric GEM oxidation during spring in the Arctic (Simpson et al., 2007). Previous studies suggested that the formation of sea ice via the refreezing of seawater can provide halogen radicals as oxidants for atmospheric GEM in the Arctic (Kaleschke et al., 2004; Lindberg et al., 2002; Simpson et al., 2007), and the decrease in sea ice concentrations during summer (after AMDEs) will lead to a reduction in halogen radicals generated through the pathways above, thus weakening atmospheric GEM oxidation during this period. On the other hand, according to Henry's Law, the warming of seawater during summer can promote the volatilization of dissolved gaseous Hg (DGM) from surface seawater to the atmosphere, which may also be a reason for the elevated atmospheric GEM concentrations during summer (Dastoor and Durnford, 2014). Moreover, the photoreduction of mercury deposited during AMDEs was also considered to be an important reason for the summer peak of GEM in the Arctic (Dastoor and Durnford, 2014). This finding further confirms that sea ice concentration is an important factor affecting atmospheric GEM concentrations during summer in the Arctic (Aspmo et al., 2006; Hirdman et al., 2009).

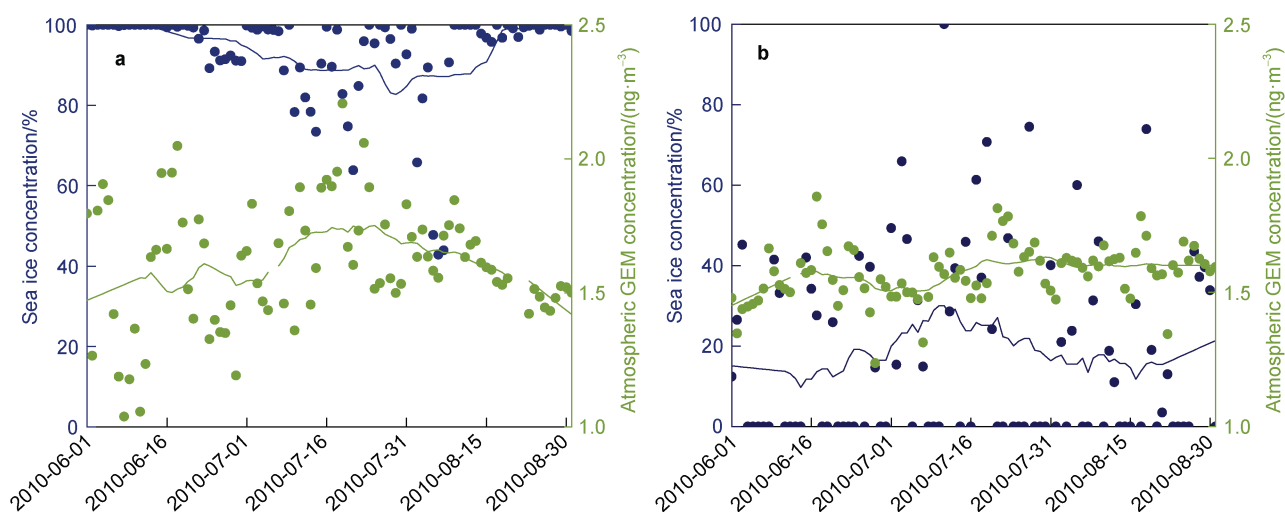


Figure 8 The sea ice concentrations and atmospheric GEM concentrations at Alert Observatory (a) and Zeppelin Observatory (b) during the summer of 2010. The trend lines of the two parameters during this period have been smoothed by Origin (the points of window are 20 and the polynomial order is 1). The sea ice concentration data at the two sites is a nine-grid mean based on the Sea Ice Remote Sensing dataset at the University of Bremen (<https://seaice.uni-bremen.de/databrowser/#p=sic>).

4 Conclusions

In this study, the round-year observations of atmospheric GEM concentrations in 2010 at two sites (Zeppelin Observatory and Alert Observatory) in the Arctic were analyzed. The change in GEM at Zeppelin Observatory was dominated by long-range transported anthropogenic emissions. The main sources were unintentional industrial sources (49%), stationary combustion sources (33%), and intentional use and product waste-associated sectors (18%). Further analysis suggested that Europe was the important source region. The change in GEM at Alert Observatory was mainly impacted by sea ice concentrations, especially in the summer season. This study also indicated that emission inventories combined with backward trajectories are a powerful method to identify anthropogenic contributions. With the elaboration of emission inventory in the future, the source compositions of atmospheric GEM that are long-range transported to Arctic regions might be identified more accurately through this method, thus providing theoretical support to control the contribution of anthropogenic emissions to atmospheric GEM in Arctic regions. Moreover, this study found that sea ice concentrations may affect atmospheric GEM concentrations in the summer of 2010 at Alert Observatory through various pathways. In the future, model simulations can be used to further investigate the effect of sea ice concentrations on atmospheric GEM concentrations in summer at Arctic sites in response to climate warming.

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