

Measurements of Atmospheric Mercury Species at a Coastal Site in the Antarctic and over the South Atlantic Ocean during Polar Summer

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Mercury and many of its compounds behave exceptionally in the environment because of their volatility, capability for methylation, and subsequent biomagnification in contrast with most of the other heavy metals. Long-range atmospheric transport of elemental mercury, its transformation to more toxic methylmercury compounds, the ability of some to undergo photochemical reactions, and their bioaccumulation in the aquatic food chain have made it a subject of global research activities, even in polar regions. The first continuous high-time-resolution measurements of total gaseous mercury in the Antarctic covering a 12-month period were carried out at the German Antarctic research station Neumayer (70°39' S, 8°15' W) between January 2000 and February 2001. We recently reported that mercury depletion events (MDEs) occur in the Antarctic after polar sunrise, as was previously shown for Arctic sites. These events (MDEs) end suddenly during Antarctic summer. A possible explanation of this phenomenon is presented in this paper, showing that air masses originating from the sea-ice surface were a necessary prerequisite for the observations of depletion of atmospheric mercury at polar spring. Our extensive measurements at Neumayer of atmospheric mercury species during December 2000–February 2001 show that fast oxidation of gaseous elemental mercury leads to variable Hg^0 concentrations during Antarctic summer, accompanied by elevated concentrations, up to more than 300 pg/m³, of reactive gaseous mercury. For the first time in the Southern Hemisphere, atmospheric mercury species measurements were also performed onboard of a research vessel, indicating the existence of homogeneous background concentrations over the south Atlantic Ocean. These new findings contain evidence for an enhanced oxidizing potential of the Antarctic atmosphere over the continent that needs to be considered

for the interpretation of dynamic transformations of mercury during summertime.

Introduction

Unlike other heavy metals that are almost exclusively associated with atmospheric aerosols, mercury exists in ambient air predominantly in the gaseous elemental form (Hg^0), which has an estimated mean global tropospheric residence time of about 1 year, making it subject to long-range atmospheric transport (1–3). The background concentration of gaseous elemental mercury (GEM) in the lower troposphere of the Northern Hemisphere and the Southern Hemisphere is generally around 1.7 and 1.3 ng/m³, respectively (4–6). Mercury is emitted into the atmosphere from a number of natural as well as anthropogenic sources. There is a growing body of evidence suggesting that current mercury emissions from anthropogenic sources are at least as great as those from natural sources (7, 8). At present, it is widely accepted that Hg^0 vapor constitutes by far the largest component of the total gaseous mercury (TGM) concentration in the troposphere (1). Other atmospheric mercury species such as total particulate-phase mercury (TPM) and reactive gaseous mercury (RGM) generally represent only a few percent of total airborne mercury in ambient air and were previously believed to be produced mostly by anthropogenic sources such as coal-fired power plants and waste incinerators (9), leading to much higher concentrations in urban air (10). Inorganic RGM species (e.g., HgCl_2) are water-soluble, and their dry deposition velocities and scavenging ratios are much higher than those for Hg^0 . Therefore, RGM species have a much shorter atmospheric lifetime than Hg^0 , and they can be expected to be removed closer to their sources (11–13).

However, American and Canadian researchers recently found very high levels of RGM and TPM species in the Arctic environment. Dramatically increased levels of TPM and RGM were measured during mercury depletion events (MDEs) in the time during and after polar sunrise at Alert, Nunavut Territory, Canada (14), and Point Barrow, AK (15, 16). Depletions of tropospheric mercury during and after polar sunrise, strongly correlated with ground-level ozone concentrations, were first observed in 1995 in the Canadian High Arctic (17). Later, MDEs have also been observed in the Antarctic (18) and, according to the proposed mechanism (19, 20), photochemically mediated reactions involving sea salt on snow/icepack or aerosols lead to chemical oxidation of elemental mercury in the troposphere. The frozen surfaces contain enriched concentrations of chloride and bromide ions which, through heterogeneous chemistry and photochemical processes, can form gaseous halogen atoms in the presence of sunlight (21). Laboratory experiments (22) have shown the existence of a dark heterogeneous reaction leading to the formation of Br_2 (Br activation) via the oxidation of Br^- , in Br^- -laden ice or snow, by O_3 . During and after polar sunrise, photochemical dissociation of the molecular forms of the halogens (Br_2 and/or Cl_2) results in the production of the corresponding atomic species. The halogen (preferentially bromine) atoms react with ozone, forming halogen oxide radicals (BrO^\bullet and/or ClO^\bullet), which in turn can oxidize elemental mercury vapor to Hg(II) species. The primary halogen atoms (Br and/or Cl) may also directly oxidize elemental mercury to Hg(II) . RGM is formed, which according to Lindberg et al. (16) could be HgO , HgBr_2 , and/or HgCl_2 . However, HgO (i.e., mercuric oxide) is incorrectly classified

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as a reactive gaseous mercury species; its vapor pressure is too low for it to be in the gaseous phase even in temperate locations let alone the polar regions, nor is it particularly reactive chemically.

These studies have revealed clear evidence that long-lived GEM is transformed to TPM and/or RGM during polar springtime but have also raised new questions, such as the following:

- (i) Why do MDEs abruptly end at polar summer?
- (ii) Is RGM produced in an on-site reaction?
- (iii) Can RGM only be produced in the marine boundary layer?
- (iv) Do high levels of RGM in polar regions only occur simultaneously with depleted concentrations of tropospheric ozone?

Sudden increases in the tropospheric BrO^* mixing ratio during spring have been found in the Antarctic using ground-based differential optical absorption spectrometry (DOAS) measurements (23) and observations from satellite-borne instruments (24, 25). Friess (26) documented, from trajectory calculations for Neumayer Station, that air masses coming from the sea-ice surface accompanied by BrO^* enhancements are found to be a necessary prerequisite for the observations of surface ozone-depletion events. A similar link to MDEs has not yet been established, and an open question is: why do MDEs stop abruptly during polar summer, even though solar radiation intensity is still high or even increasing as compared with spring and sea salt continuous to be in plentiful supply.

Up to the present, RGM measurements in the Antarctic were performed at only one other location—Terra Nova Bay from November 2000 to January 2001 (27). RGM concentrations ranged from 10 to 330 pg/m^3 and revealed the high oxidation potential in the atmosphere during Antarctic summer. The measurements reported here comprise the first data set of different atmospheric mercury species in Antarctica and over the south Atlantic Ocean. Indications for a variety of dynamic species transformations of atmospheric mercury during Antarctic summer are presented, and we show that, besides solar radiation, the origin of the air parcels and their contact with sea ice are important and could be a major requirement for the observation of MDEs at Neumayer Station. It is also explained how a different oxidation mechanism might be leading to high RGM levels during Antarctic summer, accompanied by surface-level ozone concentrations that are negatively correlated with Hg^0 concentrations.

Experimental Section

Sampling Location. In the period December 23, 2000–February 5, 2001, highly time-resolved measurements of atmospheric mercury species were carried out at the German Antarctic Research Station Neumayer, operated throughout the year by the Alfred Wegener Institute for Polar and Marine Research, Bremerhaven. The site is located at 70°39' S, 8°15' W on the Ekströmsen, about 8 km from Atka Bay. At Neumayer, the maximum solar incidence angle is 42.8°. On average, the sun stays permanently above the horizon from November 19 to January 24 and is permanently below the horizon from May 19 to July 27. The analyzers were installed at the Neumayer Air Chemistry Observatory (ACO), which is approximately 1.6 km south of the main site. The new observatory (since January 1995) was designed as a container building placed on a platform some meters above the snow. Prevailing wind directions are from the east, and northern wind directions are very rare; therefore, the risk of a potential contamination from the base is minimized. Nevertheless, supplementary control of potential contamination situations was carried out by measurements of wind direction, wind

speed, condensation nuclei concentrations, snow drift, and PAH concentrations.

After completing the measurement program at Neumayer Station in February 2001, the instruments for the determination of TGM, GEM, and RGM were operated onboard the German research vessel (R/V) *Polarstern* on the cruise from Neumayer to Punta Arenas, Chile, during February 2001 (for the cruise plot see Figure 5). The Tekran analyzers were placed inside an air chemistry research container on the upper deck about 12 m above sea level.

Instruments. Two Tekran gas-phase mercury vapor analyzers (model 2537A) were installed at Neumayer Station in January 2000 for the determination of an annual time-series of atmospheric mercury concentrations in the Antarctic (18). Until December 2000, the two Tekran analyzers were operated with a 15-min integrated sampling frequency at a flow rate of 1.0 L/min. This measurement program was enhanced throughout the ANT-XVIII (18th Antarctic program of the German Alfred Wegener Institute for Polar and Marine Research) summer campaign (December 2000–February 2001) and for the ship cruise from Neumayer to Punta Arenas (February 2001) by extensive measurements of atmospheric mercury species. RGM was measured with a Tekran 1130 mercury speciation unit, which gave one Tekran 2537A mercury vapor analyzer the ability to concurrently monitor both elemental (GEM) and reactive gaseous mercury (RGM) species in ambient air at the picograms per cubic meter level. A KCl-coated quartz annular denuder captured reactive gaseous mercury while allowing elemental mercury to pass through. At Neumayer, the heated denuder module with the sample inlet was located 2 m above the snow surface, mounted on an aluminum mast about 5 m beside the ACO. A Standard Operating Procedure for the determination of RGM using a KCl-coated annular denuder has been recently published by Landis et al. (28).

During the 60-min sampling (adsorption) phase for RGM, the model 2537A provided 5-min integrated samples for GEM concentrations at a flow rate of 1.5 L/min. An additional pump module integrated in the 1130 speciation unit pulled 6 L/min ambient air and increased the total flow rate for the denuder to 7.5 L/min. During the 40-min analysis (desorption) phase (including 8 steps for 5 min: 3× cleaning, 3× desorption, 2× cooling), the denuder was flushed with zero air and heated to 500 °C to thermally decompose and reduce RGM to the elemental form. Altogether 12 GEM values and 1 RGM value were achieved from one measurement cycle of 100 min. Downstream of the denuder, a 0.45- μm PTFE filter in front of the sample inlet of the Tekran 2537A prevented the fine particulate phase mercury (particle size < 2.5 μm) from penetrating to the gold traps. The second Tekran 2537A continued to measure TGM and was operated with a 5-min integrated sampling frequency. The sample inlet with a drift protection (no filter) was located 6 m above the snow surface on top of the ACO. The air was sampled at a flow rate of 1.5 L/min through a heated sample line about 10 m in length ending up with a 0.45- μm PTFE filter in front of the inlet of the analyzer. The accuracy and precision of the Tekran 2537A have been assessed during field intercomparisons at an urban/industrial site (29) and at a remote marine background location (30). The analyzers were calibrated every 25 h with an internal automatically triggered permeation source injection (see Quality Control). The detection limit for Hg^0 in this operation mode is 1.1 pg absolute.

Total particulate-phase mercury (TPM) was measured using an AESminiSamplR developed by the Meteorological Service of Canada (31). This miniaturized device, a movable quartz tube with a 6-mm quartz fiber filter disk, served as both particulate trap and pyrolyzer for airborne particulate mercury species. The airborne particulate matter was collected at 2 m height on two AESminiSamplR devices sampling

in parallel. The total sample volume ranged between 4 and 6 m³ of ambient air with an integration time of 48 h. The collection device was used in combination with a system of amalgamation/thermal desorption of Hg⁰ onto a gold trap followed by a cold vapor atomic fluorescence spectrometer. The procedural detection limit of this method for a typical 24-h sample is ≈ 2 pg/m³.

Ozone mixing ratios were measured continuously by means of an Ansyco ozone analyzer (model O341M) on the basis of UV absorption with a detection limit of ≈ 2 μ g/m³.

Quality Control. The Tekran 2537A performed automated recalibrations for Hg⁰ using an internal permeation source, which provides approximately 1 pg/s of Hg⁰ at 50 °C into a zero airstream. The periodicity of the calibrations for the Tekran 2537A in connection with the mercury speciation unit 1130 was 2500 min (25 RGM cycles) and for the second Tekran 2537A measuring TGM was 1500 min (25 h). In addition, automated standard additions were carried out for the Tekran 2537A (without speciation unit) every 355 min over the entire measurement period to check the sensitivity of the signal and, therefore, the stability of the detector response and the internal permeation source. The recovery rates for the standard additions ranged from 90 to 116% with a mean recovery of $101 \pm 3\%$. The accuracy of the measurements was verified by manual injections of a known volume of air from a saturated mercury vapor atmosphere at a known temperature. This procedure was applied before, during, and after the Tekran 2537A analyzers were running in the chemical observatory at Neumayer and onboard the R/V *Polarstern* between January 2000 and February 2001 (18). Over this long time period (more than 1 year), maximum deviations of $<5\%$ between the theoretical and the measured values were found; therefore, adjusting of the perm source was not necessary. To demonstrate the trueness of the results, further comparative measurements of TGM with two different methods were carried out before the instruments had been set up at Neumayer (18).

A direct primary calibration method for RGM measurements with the Tekran speciation unit 1130 does not yet exist. However, Landis et al. (28) have suggested a secondary calibration protocol used to calibrate the TEKRA 1130 through the use of a HgCl₂ permeation device. Presently it is not possible to find a representative certified reference material for a reproducible calibration of gaseous Hg(II) species on a KCl-coated denuder. Field intercomparison studies of different methods for the determination of RGM were performed at Mace Head, Ireland, and Sassetta, Italy (30, 32). They revealed a good precision for the denuder method and no significant breakthrough. It was also reported that the thermal desorption and reduction of RGM to Hg⁰ has an efficiency of nearly 100% (10, 33, 34). Comparable results were also obtained for different methods such as multistage filter packs, refluxing mist chambers, and KCl-coated denuders (35). These major findings of different field intercomparison exercises have been recently reconfirmed by a detailed laboratory study published by Landis et al. (28). Although the paper published by Landis et al. (28) is a significant advancement in the harmonization and standardization for the determination of RGM in ambient air, an international debate is still ongoing about the validation and measurement uncertainty for these operationally defined methods in polar regions (Arctic Atmospheric Mercury Research Workshop in Toronto, Canada, August 26–28, 2002). The analytical gold trap in the detection system for TPM analysis was calibrated with daily manual injections of a known volume of air from a mercury vapor saturated atmosphere at a known temperature.

Definition and Chemical Composition of the Parameter TGM. In recent years, controversial discussions about the chemical composition of TGM at high levels of RGM and/or

TPM have led to different interpretations of the parameter TGM measured with a Tekran model 2537A analyzer. Because RGM has been shown to adsorb on many materials, the question was whether, at high levels of RGM, the gaseous divalent inorganic mercury species (commonly assumed to be HgCl₂) could pass through the sample line and the particulate filter in front of the sample inlet of a Tekran 2537A to sum up, with the Hg⁰, to TGM? We used both a Tekran speciation unit 1130 connected to a Tekran 2537A to provide GEM and RGM concentrations and a traditional Tekran 2537A with a heated sample line and a 0.45- μ m PTFE particulate filter in front of the analyzer to measure TGM concentrations. All 2-h averaged atmospheric mercury species concentrations during the complete summer period (December 2000–February 2001) containing RGM concentrations >0.15 ng/m³ are presented in Figure 1. The lower graph represents the absolute values of the residuals $|TGM - (GEM + RGM)|$ and the respective standard deviation for the 2-h averaged TGM concentration. Most of the single residuals are lower than or within the variation of the standard deviation of the 2-h averaged TGM levels, even at levels of RGM up to 0.4 ng/m³, showing that the residuals are in the range of the white noise process and the uncertainty of the TGM measurements. A single large residual, with more than 0.8 ng/m³ difference between TGM and GEM + RGM, is due to some outliers in the highly time-resolved data set and is also reflected in the large standard deviation during that 2-h period. As well as the residual analysis in Figure 1, Figure 6 shows exemplarily (during a 10-day period in January 2001) that TGM concentrations are higher at midday (January 6–9), simultaneously with high RGM concentrations, and can be roughly estimated as the sum of GEM + RGM.

These figures suggest that, at least under Antarctic conditions with low air humidity, high levels of reactive gaseous mercury can pass through a heated Teflon sampling line and a PTFE particulate filter to be amalgamated on gold cartridges and subsequently detected as Hg⁰ in a Tekran 2537A.

In addition, Table 1 shows the arithmetic mean of the 2-h averaged TGM and GEM and the calculated sum of GEM + RGM concentrations for the complete measurement period during Antarctic summer. The three data sets are normally distributed (Kolmogorov–Smirnov test), and their standard deviations are based on the same variance of the entire population (*F*-test for $P=0.95$). The mean TGM concentration during this summer period differs significantly from the mean GEM concentration (result of the *t* test: $t = 5.37 > t[P=0.95, f=1041] = 2.60$). Moreover, a comparison of the mean values of the TGM concentrations and the calculated sum of GEM + RGM concentrations reveals no significant difference between these two aggregate concentrations for this summer period (*t* test: $t = 0.75 < t[P=0.95, f=1017] = 2.60$). These statistical results indicate for these particular Antarctic conditions that it is possible to measure TGM with a heated sample line and a Tekran 2537A as the commonly defined “sum parameter” of both gaseous inorganic mercury species (GEM + RGM) in ambient air, even at periods with relatively high RGM concentrations.

Results and Discussion

End of Mercury Depletion Events during Antarctic Springtime. Ebinghaus et al. (18) have shown that Antarctic MDEs coincided with ozone-depletion events in the lower troposphere and supported the hypothesis that BrO[•] free radicals are involved. It was hypothesized that Br atoms and/or BrO[•] radicals are involved in a chemical reaction that destroys ozone and that the subsequent depletion of atmospheric boundary layer mercury (at times up to an altitude of ≈ 1 km) was due to a reaction between gaseous elemental mercury and BrO[•] free radicals and/or halogen atoms. The MDEs

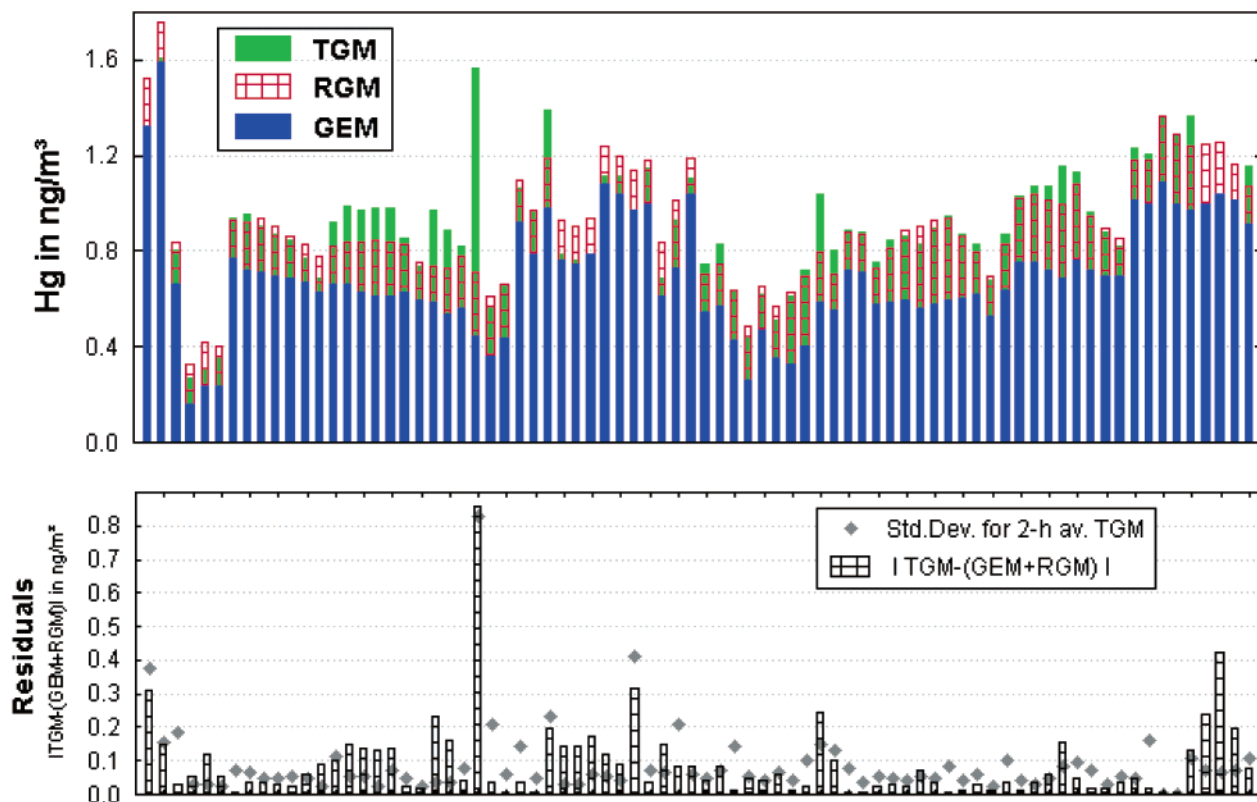


FIGURE 1. Selected 2-h averaged mercury species concentrations during the summer period (December 2000–February 2001) containing RGM levels >0.15 ng/m^3 and the respective absolute values of the residuals $|\text{TGM} - (\text{GEM} + \text{RGM})|$ as compared with the standard deviations for the 2-h averaged TGM concentrations.

TABLE 1. Arithmetic Mean of 2-h Averaged TGM and GEM and Calculated Sum of GEM + RGM Concentrations during Antarctic Summer (December 2000–February 2001) at Neumayer Station

parameter	instrument	<i>n</i>	arithmetic mean and SD (ng/m^3)	max (ng/m^3)	min (ng/m^3)
TGM	Tekran 2537A	515	1.08 ± 0.29	2.34	0.27
GEM	Tekran 2537A + speciation unit 1130	528	0.99 ± 0.27	1.89	0.16
GEM + RGM	Tekran 2537A + speciation unit 1130	504	1.07 ± 0.23	1.95	0.33

ended at the beginning of November, but a possible explanation for their sudden termination was left open. The results of the TGM measurements and ground-level ozone concentrations for the time period from January 2000 to January 2001 are presented in Figure 2. The bars in the bottom graph show the weekly correlation coefficients (r [$P = 0.99$]) for surface-level ozone and TGM concentrations. The correlation analysis for the fall and dark winter months (March–July) yields very variable correlation coefficients; hence, considering the complete winter time, no significant linear relationship among these two variables is evident. The TGM concentration pattern between August and October 2000 shows a strong positive correlation with ozone (max correlation coefficient $r > 0.9$) covering that time of the year when depletion events are caused by photochemically mediated destruction of ozone and species transformation of mercury (during MDEs) (18). The MDEs suddenly end at the beginning of November when TGM and ozone concentrations start being negatively correlated (max $r \approx -0.8$) until the beginning of February 2001 (end of the mercury measurements at Neumayer Station). This is a first indication that the atmospheric processes for the oxidation of mercury during springtime MDEs should be based on a different mechanism as compared with the Antarctic summer period from November to February.

But why do MDEs suddenly end at the beginning of November 2000 during Antarctic springtime? Figure 3 shows the smoothed trend of the 12-h averaged TGM concentrations since May 2000. The gray bars represent the number of coordinates from each of the five-day (120-h) backward trajectories that were located over sea ice with more than 40% ice coverage. All backward trajectories were based on the output of the global model for Antarctic trajectory calculations (36) and were obtained for Neumayer Station every 12 h by the German Weather Service. The trajectories were calculated for surface pressure and consist of 21 coordinates (every 6 h) representing the backward transport of the surface air masses (37). The weekly sea-ice analysis charts were obtained from the web page of the National Ice Center from the National Oceanic and Atmospheric Administration (NOAA) in Washington. Our category with sea-ice coverage $>40\%$ is based on the classification of the National Ice Center. A number of 21 would be related to an air mass that originated from the sea ice having a maximum contact time with sea ice over the south Atlantic Ocean (coverage $>40\%$) before reaching Neumayer, whereas no contact with sea ice with more than 40% ice coverage would result in a number of 0. The dashed line represents the astronomical sunshine duration (ASSD) for 24 h at Neumayer Station. During the year 2000/2001, the sun stayed permanently above

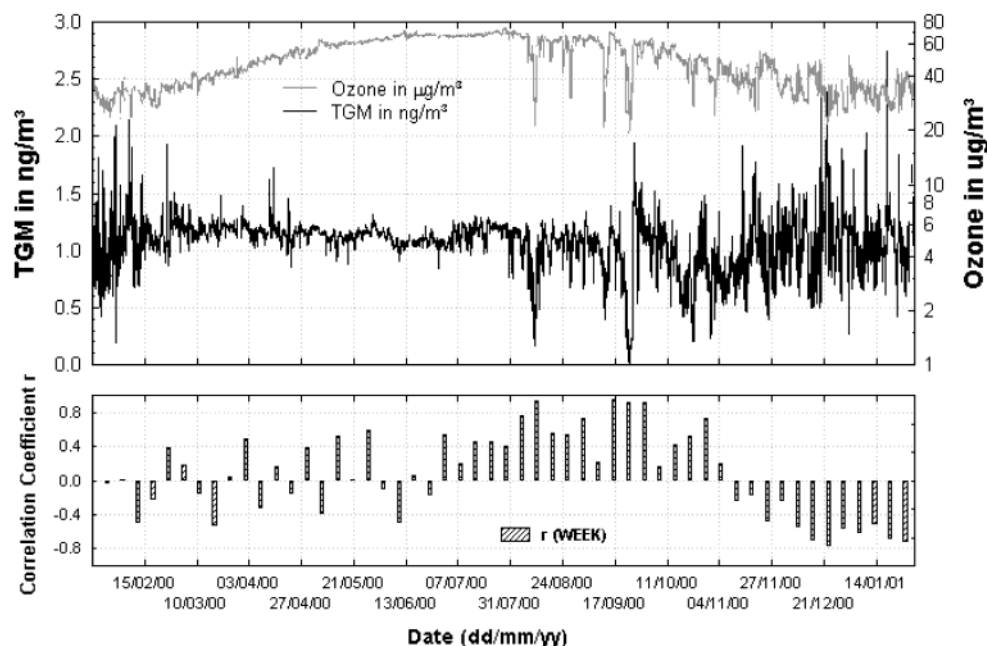


FIGURE 2. Annual time series of 1-h averaged TGM and surface-level ozone concentrations at Neumayer, January 2000–February 2001 and their weekly mean correlation coefficients (r). The TGM concentrations were obtained from two separate Tekran analyzers, and mean values were used if both analyzers were operating in parallel.

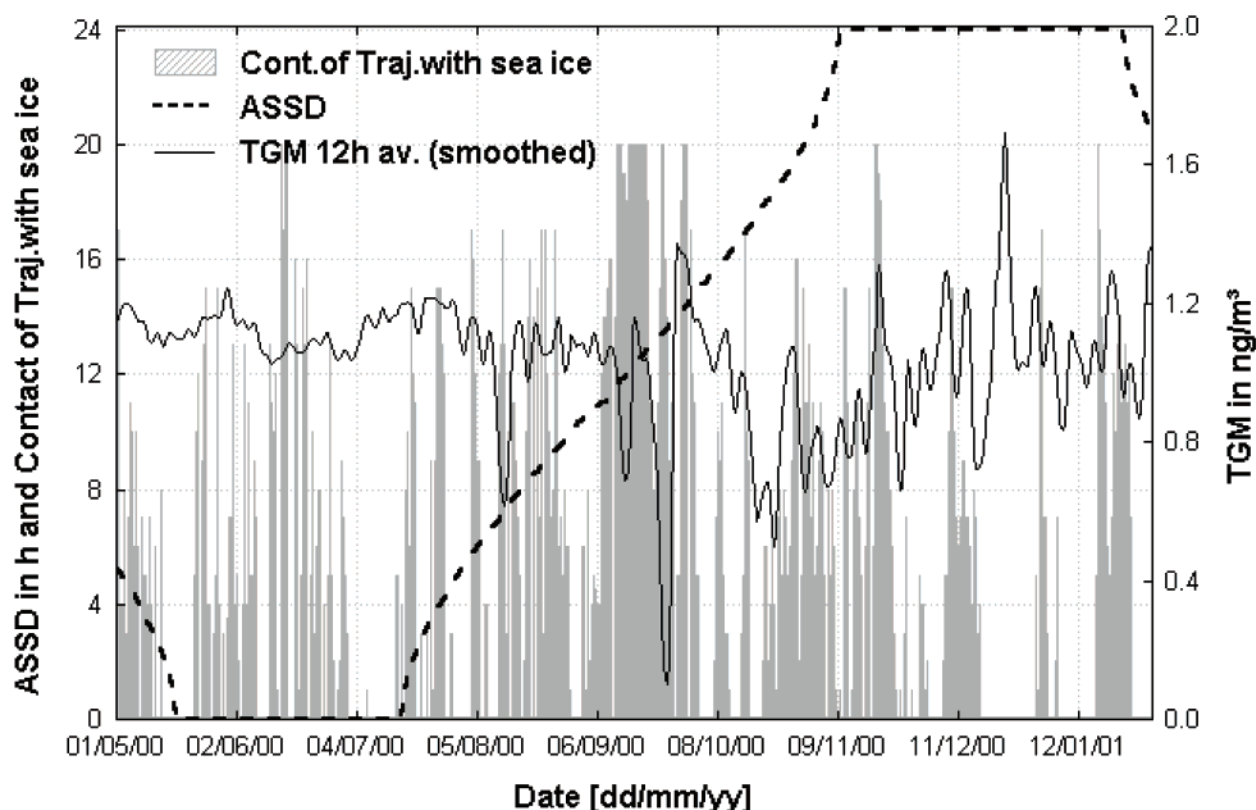


FIGURE 3. Comparison of the 12-h averaged TGM concentrations (smoothed) and the duration of sea-ice contact of the surface air parcels obtained from trajectory calculations ending at Neumayer at 12:00 and 24:00 UT. The dashed line represents the astronomical sunshine duration (ASDD) for each day.

the horizon from November 10 to January 25 (ASDD = 24 h) and permanently below the horizon from May 17 to July 17 (ASDD = 0 h).

Figure 3 gives clear evidence that shortly after polar sunrise (polar sunrise = July 17, 2000), at the beginning of August, the first strong MDEs occurred at Neumayer. This coincides

with the arrival of air masses that resided most of the time over the ocean, covered with more than 40% sea ice, before reaching Neumayer Station. This good agreement between the TGM measurements during MDEs, and the results from trajectory calculations in combination with sea-ice charts support the theory that reactive bromine, which destroys

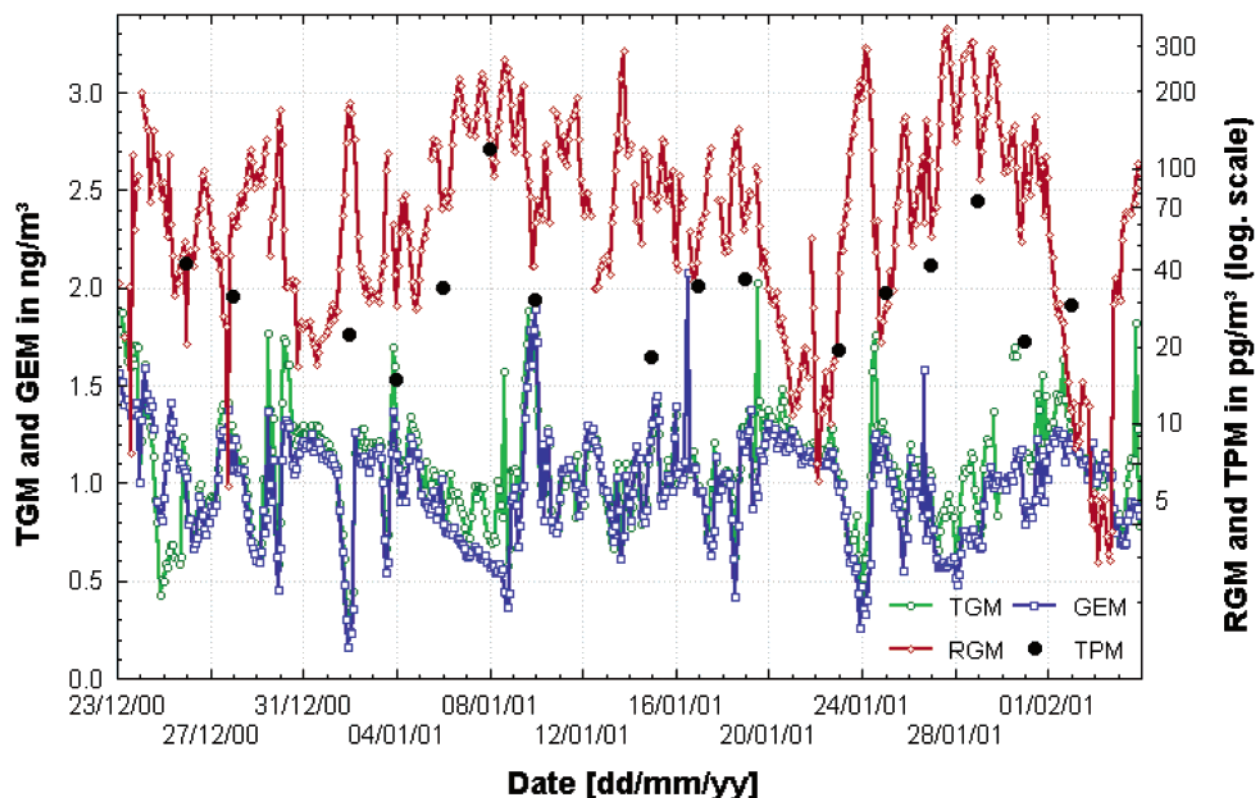


FIGURE 4. Atmospheric mercury species concentrations at Neumayer Station between December 23, 2000, and February 5, 2001. TGM, RGM, and GEM levels are 2-h mean concentrations. TPM concentrations were obtained from 48-h integrated samples. Missing data in the RGM concentrations were due to the automatic calibration cycles for the Tekran analyzer.

ozone and can oxidize elemental mercury in a subsequent reaction, is released from sea salt, which is associated either with sea-ice surfaces or sea-salt aerosols.

These reactions must end when the sea ice in the Weddell Sea around Neumayer starts to vanish at the end of November, thus reducing, within a short time, the size of the surrounding sea ice that in winter covers an area of around 1.5 times the area of the continent (around 20 million km in October, 3 million km in February). The consequences are shown in Figure 3. Although photochemical reactions liberating reactive halogen species from sea salt would be supported by strong solar radiation in the time after November 1, the occurrence of strong MDEs decreased and the duration of air mass contact with sea ice decreased as well. Release of reactive bromine species from sea-salt surfaces, as a precursor for surface ozone destruction, and the oxidation of atmospheric mercury (Hg^0) after polar sunrise can only take place within a short time period, ending with the disappearance of the sea ice around the Antarctic continent.

Exact determinations of the source region of air masses during enhanced BrO^* events at Neumayer in August and September 2000 including detailed trajectory analysis underlain by sea-ice maps have been carried out by Friess (26). TGM concentrations during Antarctic summer were still highly variable; however, these variations are considered to be due to other atmospheric reactions, as explained in the following section.

Mercury Species Concentrations during Antarctic Summer. The results of mercury species measurements for the time period December 2000–February 2001 are presented in Figure 4. The arithmetic mean of all TGM measurements between December 23, 2000, and February 5, 2001, was $(1.080 \pm 0.293) \text{ ng/m}^3$ and is in the range of typical background concentrations of TGM for the Southern Hemisphere as

reported in refs 18 and 27. Mercury levels in the remote Southern Hemisphere are generally lower than in the Northern Hemisphere and in the Arctic. Several studies (4, 5, 18, 38) have shown that total gaseous mercury background concentrations over the Atlantic Ocean in the Southern Hemisphere are very homogeneous and are about 25% lower than the Northern Hemisphere TGM background concentration. Nevertheless TGM and GEM concentrations during the sunlit summer period varied more than during the dark winter months, and depletions events with concentrations $<0.3 \text{ ng/m}^3$ were observed.

RGM concentrations during this summer period varied between 5 pg/m^3 and maximum levels of more than 300 pg/m^3 . These peak concentrations are comparable to those measured in the vicinity of strong anthropogenic sources such as coal-fired power plants and waste incinerators (35). Comparable values have been reported for the springtime depletion period in the Arctic at Point Barrow, AK (16), and for the nondepletion period (November–December 2000) in the Antarctic at Terra Nova Bay (27) with maximum values of 900 and 300 pg/m^3 , respectively.

Previous RGM measurements at a rural European site over a 2-month time period showed average values of $5\text{--}90 \text{ pg/m}^3$ (39). This is in good agreement with very homogeneous RGM levels with a median of 5.5 pg/m^3 ($n = 75$; lower quartile = 3.8 pg/m^3 , upper quartile = 16.3 pg/m^3) obtained onboard the R/V *Polarstern* in February 2001 over the south Atlantic Ocean (see Figure 5).

As shown in Figure 4, TPM concentrations at Neumayer were in the range of $15\text{--}120 \text{ pg/m}^3$, showing roughly the time trend of the RGM concentrations only at a lower level. It was not possible to compare the RGM and TPM time trends statistically because of the much lower time resolution for TPM, caused by the long sampling time of 48 h. However, our data clearly indicate an important change in the

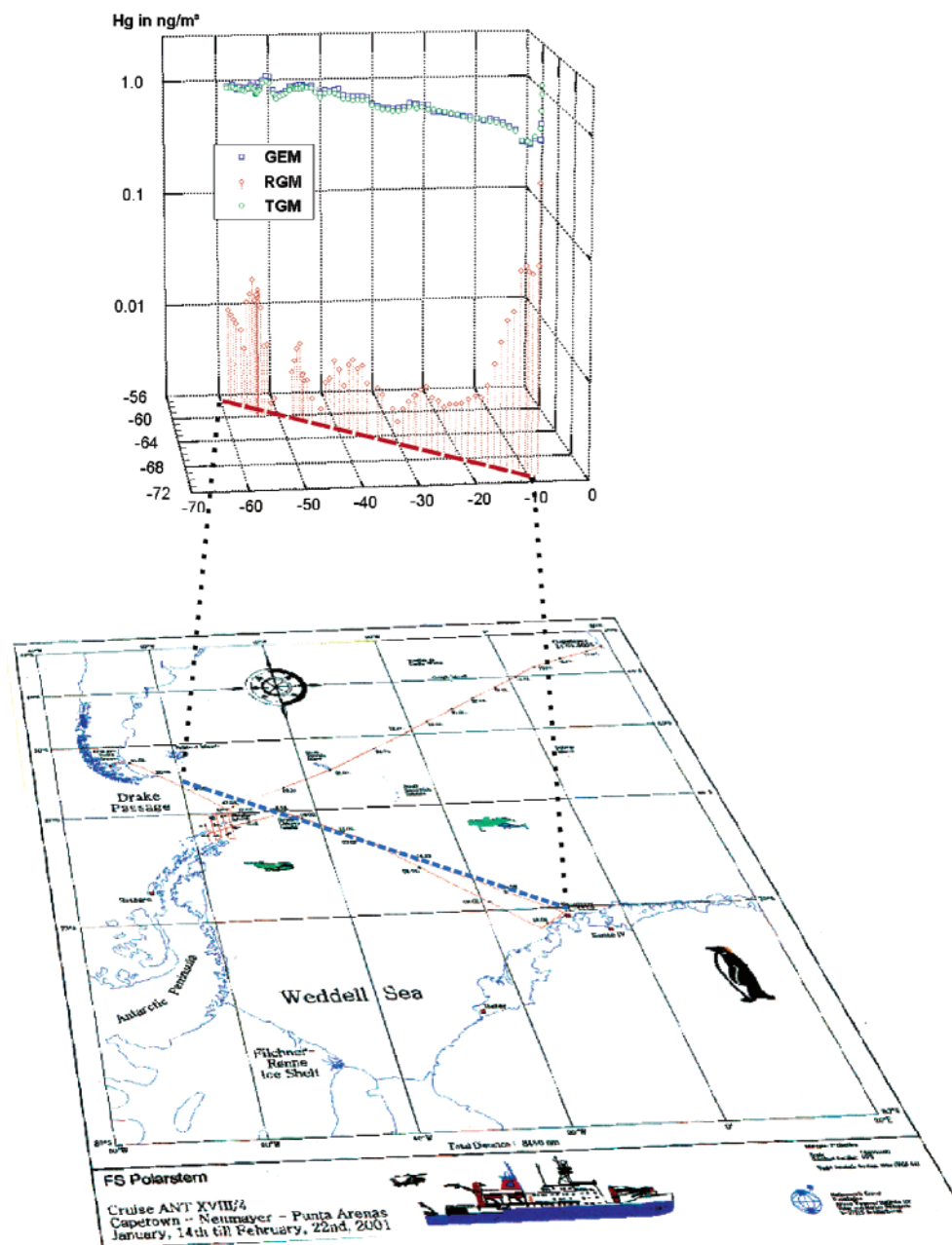


FIGURE 5. Two-hour averaged atmospheric mercury species concentrations over the south Atlantic Ocean measured onboard the R/V *Polarstern* during cruise ANT XVIII/4 (Neumayer–Punta Arenas, February 2001).

speciation of atmospheric mercury during Antarctic summer at Neumayer Station, producing elevated levels of RGM and/or TPM at this remote site.

Mercury Species Concentrations over the South Atlantic Ocean. To reconfirm the homogeneous distribution of atmospheric mercury in the remote Southern Hemisphere and to point out the exceptionally high RGM levels found at Neumayer Station during Antarctic summer, additional mercury species measurements were carried out onboard the R/V *Polarstern* after finishing the measurement campaign at Neumayer. The results of the ship cruise from Neumayer to Punta Arenas are presented in Figure 5. TGM, GEM, and RGM remained very stable at low levels without high variations over the entire measurement period. The mean TGM concentration was $(1.1 \pm 0.2) \text{ ng/m}^3$ and is in good agreement with average Southern Hemisphere background TGM concentrations from previous studies (4, 5, 18, 38). The first RGM measurements obtained onboard a ship in the

Southern Hemisphere yielded very low concentrations in the range of $1\text{--}30 \text{ pg/m}^3$; thus, revealing no significant differences between GEM and TGM during the whole cruise. The few outliers at the beginning of the cruise were due to sample contamination caused by the ship stopping several times because of heavy sea ice. No obvious trend over the south Atlantic Ocean or influence from the South American continent can be seen. These supplementary measurements confirm the hypothesis that the very high RGM concentrations at Neumayer Station measured in the time before (December 2000–February 2001) should be influenced by local production of oxidized gaseous mercury species over the Antarctic continent or shelf ice during polar summer.

Dynamics and Chemistry of Atmospheric Mercury Transformations during Antarctic Summer. As we have shown in the previous sections, the summertime oxidation of elemental mercury and transformation to more reactive gaseous species at Neumayer is based on a different reaction

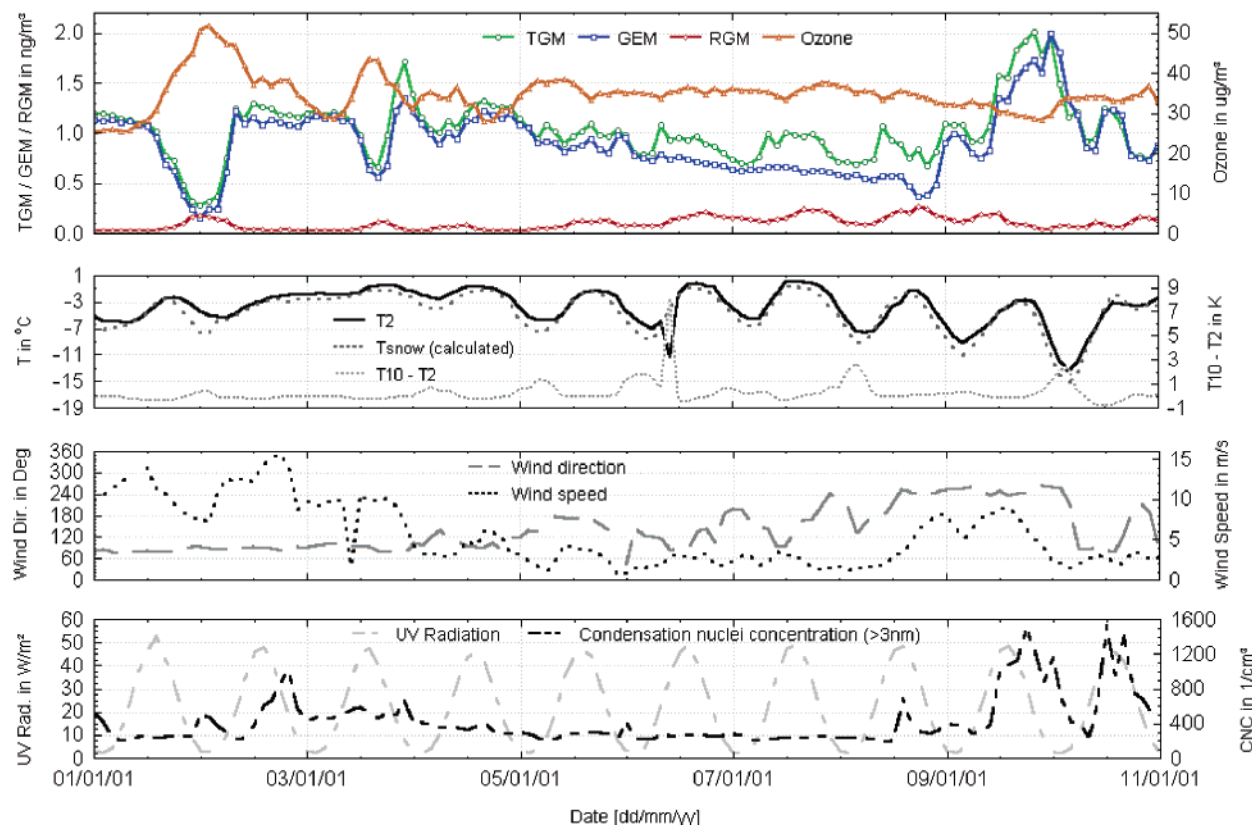


FIGURE 6. Two-hour averaged atmospheric mercury species (TGM, GEM, RGM) and ozone concentrations together with selected meteorological and chemical parameters for a 10-day period during Antarctic summer. T2 and T10 represent the air temperatures at 2 and 10 m height.

mechanism as compared with the springtime gas-phase oxidation of Hg^0 by BrO^\bullet and/or bromine (or chlorine) atoms and the concurrent destruction of surface-level ozone during springtime depletions (16, 18). To examine potential influences of meteorological factors, anthropogenic emissions, or solar radiation, a representative short time period of 2-h mean atmospheric mercury species concentrations and surface-level ozone concentrations together with selected additional meteorological and chemical parameters is presented in Figure 6. During this Antarctic summertime 10-day period, highly variable mercury concentrations were observed, with significant negatively correlated surface-level ozone and TGM ($r = -0.61$ [$P = 0.99$; $n = 120$]) or GEM concentrations ($r = -0.58$ [$P = 0.99$; $n = 120$]). At the same time, RGM concentrations are positively correlated with ground-level ozone concentrations ($r = 0.30$ [$P = 0.99$; $n = 117$]).

During this time period (January 1–11, 2001), TGM and GEM concentrations decreased temporarily from normal background levels of 1.1–1.2 to $<0.4 \text{ ng/m}^3$, whereas RGM levels increased simultaneously from 30–40 to more than 200 pg/m^3 , and surface-level ozone concentrations exceeded normal background concentrations ($30 \text{ } \mu\text{g/m}^3$ during Antarctic summer, representing the minimum of the annual ozone concentrations caused by photolysis of ozone from higher UV-B radiation [see Figure 2]) rising to more than $50 \text{ } \mu\text{g/m}^3$. T2 and T10 represent the temperatures measured at 2 m and 10 m height. The strength of the inversion, in terms of the stability of the lower troposphere, can be estimated by the temperature difference $T_{10} - T_2$.

In particular, during January 1 and 2, a strong decrease of TGM and GEM was observed. For this depletion and all following events, no significant correlation to any of the given additional parameters could be found. We hypothesize that the oxidation of elemental mercury to divalent atmospheric

mercury species and a concurrent production of ozone has already occurred before the air parcels were advected to the measurement location. On the basis of several recent studies providing evidence that ozone is photochemically produced at the South Pole (SP) surface during Antarctic summer, together with a high atmospheric oxidizing power on the Antarctic plateau (40, 41), we propose a gas-phase oxidation of elemental mercury by potential oxidants such as OH^\bullet , HO_2^\bullet , $\text{O}(^1\text{D})$ and ^3P , NO_3^\bullet , etc. associated with high levels of NO resulting from photodenitrification processes in the snowpack that serve as an ozone precursor (conversion of NO to NO_2 , followed by photolysis of NO_2). This hypothesis is confirmed by the latest results from the ISCAT program (Investigation of Sulfur Chemistry in the Antarctic Troposphere) revealing high concentrations of TPM in aerosol samples together with a simultaneous production of ozone at SP in November 2000–January 2001 (42). Furthermore, Sommar et al. (43) recently investigated the reactivity and kinetics of hydroxyl radicals toward Hg^0 in the gas phase, finding that the reaction is slightly exothermic with a reaction rate constant of

$$k_{\text{OH}} = (8.7 \pm 2.8) \times 10^{-14} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$$

If this reaction is included in global models, a natural lifetime of Hg^0 of 4–7 months can be derived (43). However, with the rate coefficient estimated by Sommer et al. (43), our measurements revealing fast oxidation of Hg^0 during Antarctic summer cannot be completely explained.

Zhou et al. (44) showed that the snowpack photochemical production of HONO could affect the concentrations of hydroxyl radicals in the Arctic boundary layer. They found sustained concentrations of HONO during the Polar Sunrise Experiment 2000 high enough that upon photolysis HONO became the dominant hydroxyl radical source.

Additional cross-correlation analysis of TGM and ozone concentrations at the highest time resolution (5 min) for different events where Hg^0 concentrations decreased and surface-level ozone increased always lead to a maximum negative correlation in the cross-correlation function at a lag of zero. These results confirm that the suggested reaction in the atmosphere, producing ozone and oxidizing elemental mercury, must be faster than 5 min or was already completed before the air parcel sampled arrived at the sampling site. The second explanation is probably more plausible because Hg^0 minima are infrequent during this time and very short-lived, and no significant correlation with other parameters such as UV radiation and wind speed measured on-site at Neumayer Station were detected. However, the actual place of the oxidation reaction of elemental mercury in the atmosphere and the origin of the depleted air masses should be relatively close to Neumayer Station. High RGM levels were measured at this site, and RGM species are not expected to undergo long-range transport because of their relatively short atmospheric lifetime (11, 13). Additional atmospheric measurement data of potential precursor compounds (NO_x , CO, and OH radicals) and isentropic trajectory calculations are necessary to explain the reaction mechanism and the origin of the air masses reaching Neumayer during Antarctic summer with concurrent high concentrations of RGM and surface-level ozone.

High CNC levels (condensation nuclei concentration) measured on January 9 and 10, 2001, at the end of this event were associated with TGM and GEM concentrations exceeding normal background concentrations by more than 50% ($>1.5 \text{ ng/m}^3$). Higher concentrations of elemental mercury during Antarctic summer can be due to anthropogenic emissions. From late-December to mid-February, increased numbers of visiting scientists are operating in the vicinity of the main station, and it is well-known that human activities produce large concentrations of condensation nuclei (45). They cannot be totally excluded at Neumayer although the ACO is located 1.6 km south of the main station and vehicles are not allowed to approach the ACO.

Re-emission of Hg^0 from Arctic snowpack following the springtime depletion period has been reported recently (16, 46). However, Hg^0 peaks at Neumayer are infrequent and very short-lived, and no significant correlation with UV radiation or temperature can be seen. Therefore, we would exclude re-emission of Hg^0 from snow and/or water surfaces by photoreduction and volatilization to the atmosphere as an explanation for the results we observed.

What Can We Learn from the Behavior of Atmospheric Mercury during Antarctic Summer? Tropospheric mercury concentrations in polar regions are very sensitive to species transformations caused by photochemically driven oxidation processes during the sunlit time of the year. Gaseous elemental mercury is a globally distributed air pollutant with a long atmospheric lifetime and can be transported to remote areas such as the Arctic or Antarctic (47). In the past few years, intensive research has been initiated and carried out in the polar regions of the globe after the discovery of the so-called MDEs in the Arctic (17) and later in the Antarctic (18). Since the initial discovery at Alert, ON, Canada, in 1995 (17), MDEs have been observed at several circumpolar sites [Alaska (15, 16), Spitsbergen (48), Quebec (49), and Greenland (50)], and recent environmental process studies (14, 16, 51, 52) now provide a better understanding of this phenomenon, which is expected to lead to a significantly increased input of atmospheric mercury into polar ecosystems (53, 54). It is assumed that atmospheric oxidation of elemental mercury by enhanced levels of BrO^* or halogen atoms is followed by adsorption on aerosol surfaces and/or direct deposition of reactive gaseous mercury and particulate-phase mercury, both leading to an effective removal of mercury from the

lower polar troposphere (14, 16). It is noteworthy that MDEs coincide with polar sunrise ozone depletion events in the lower polar troposphere where ozone is destroyed by a photochemically initiated reaction between ozone and reactive gaseous halogen species (especially bromine).

We have shown that these phenomena in the Antarctic are limited to the time after polar sunrise and could be related to the spatial coverage of annual sea ice, decreasing dramatically during Antarctic summer. After the end of MDEs in November 2000, we found a different behavior of TGM/GEM and surface-level ozone during Antarctic summer. Ozone was not destroyed but could rather have been produced via a photochemical reaction of ozone precursors such as NO and NO_2 over the Antarctic continent. This production (source process) could be associated with an enhanced oxidation potential affecting the transformation of various chemicals (including Hg^0) leading to different exchange processes between the atmosphere, snow, and ice (51).

The dynamic species transformations of atmospheric mercury during Antarctic summer that are presented here illustrate the complexity of photochemical reactions in polar regions and have revealed the limitations in our understanding of the chemical cycling of mercury and other atmospheric constituents/contaminants in remote regions with seasonally variable sea-ice coverage. Further studies on possible reaction mechanisms and chemical kinetics of these phenomena are necessary to assess the resulting net input into the polar biosphere.

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Note Added after ASAP Posting

This paper was released ASAP on 11/21/02 with errors in the units used (m^2 instead of m^3). The correct version was posted on 11/27/02.

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