Convective forcing of mercury and ozone in the Arctic boundary layer induced by leads in sea ice

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The ongoing regime shift of Arctic sea ice from perennial to seasonal ice is associated with more dynamic patterns of opening and closing sea-ice leads (large transient channels of open water in the ice)1–3, which may affect atmospheric and biogeochemical cycles in the Arctic4. Mercury and ozone are rapidly removed from the atmospheric boundary layer during depletion events in the Arctic4–7, caused by destruction of ozone along with oxidation of gaseous elemental mercury (Hg(0)) to oxidized mercury (Hg(II)) in the atmosphere and its subsequent deposition to snow and ice8. Ozone depletion events can change the oxidative capacity of the air by affecting atmospheric hydroxyl radical chemistry9, whereas atmospheric mercury depletion events can increase the deposition of mercury to the Arctic4,6,10, some of which can enter ecosystems during snowmelt11. Here we present near-surface measurements of atmospheric mercury and ozone from two Arctic field campaigns near Barrow, Alaska. We find that coastal depletion events are directly linked to sea-ice dynamics. A consolidated ice cover facilitates the depletion of Hg(0) and ozone, but these immediately recover to near-background concentrations in the upwind presence of open sea-ice leads. We attribute the rapid recoveries of Hg(0) and ozone to lead-initiated shallow convection in the stable Arctic boundary layer, which mixes Hg(0) and ozone from undepleted air masses aloft. This convective forcing provides additional Hg(0) to the surface at a time of active depletion chemistry, where it is subject to renewed oxidation. Future work will need to establish the degree to which large-scale changes in sea-ice dynamics across the Arctic alter ozone chemistry and mercury deposition in fragile Arctic ecosystems.

Profound changes that have occurred recently in the Arctic sea ice include historic minimum extents of perennial sea ice1 and a shift to thinner seasonal sea ice2, which experiences more dynamic patterns of opening and closing sea-ice leads3. These changes have consequences for the Arctic energy balance and the Earth’s radiation budget, with a positive feedback that can accelerate Arctic warming1. We here show that atmospheric mercury (Hg) and ozone (O3) depletion events near Barrow, Alaska, are directly linked to sea-ice dynamics in the Beaufort and Chukchi seas. We performed near-surface measurements of atmospheric Hg and O3 directly over the frozen Chukchi Sea during two field studies: the Bromine, Ozone, and Mercury Experiment (BROMEX)13 in March/April 2012, and the Ocean-Atmosphere-Sea Ice-Snowpack (OASIS) campaign14 in March 2009 (Fig. 1). We characterized the surrounding sea-ice conditions with daily Moderate Resolution Imaging Spectroradiometer (MODIS) satellite images and marked the location of open leads in the path of air masses during the previous 24 hours before the air masses arrived at the site. We consistently observed that periods of strong and concurrent atmospheric Hg depletion events (below 0.8 ng m⁻²) and O3 depletion events (below 5 p.p.b.v.) occurred when upwind areas consisted largely of consolidated sea-ice cover (completely frozen or containing fully refrozen leads). Periods when air masses travelled over open leads within about 150 km upwind of Barrow, however, were associated with higher, undepleted Hg(0) and O3 concentrations (Fig. 1).

Using high-temporal-resolution (every 4 hours) National Oceanic and Atmospheric Administration (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) back-trajectories, we show the effects of sea-ice leads on boundary layer Hg(0) and O3 for several periods associated with dramatic changes in Hg(0) and O3 concentrations (Figs 2 and 3 and Extended Data Figs 1 and 2). The first period in 2012 (Fig. 2) shows strong increases in Hg(0) and O3 concentrations when back-trajectories switched from areas dominated by consolidated sea ice to areas with open leads. Initially (cases 1 and 2), back-trajectories travelled entirely over consolidated sea ice; and although open leads occurred north of Barrow, back-trajectories did not intersect with these within the previous 24 hours. Therefore, the open leads were not affecting atmospheric Hg(0) and O3 concentrations for these two cases. During this period, Hg(0) and O3 concentrations in the Arctic atmospheric boundary layer were depleted (<0.6 ng m⁻³ and <15 p.p.b.v., respectively), indicating an ongoing atmospheric Hg and O3 depletion event. A new, 2-km-wide lead opened northeast of Barrow on 24 March 2012 (case 3). Although back-trajectories changed little since the two previous days (also supported by consistent wind velocities, see Extended Data Fig. 3), they now crossed this open lead and concentrations of Hg(0) and O3 dramatically increased to 1.2 ng m⁻³ and 33 p.p.b.v. within hours, approaching Northern Hemisphere background concentrations (roughly 1.5 ng m⁻³ for Hg(0) and 30 p.p.b.v. for O3).

In a second example period in 2009 (Fig. 3), open leads were present close to Barrow on 13 March, but air masses initially travelled over areas consisting of consolidated sea ice (case 4). This period was marked by decreasing O3 concentrations (starting at 40 p.p.b.v. and decreasing to <5 p.p.b.v.), showing an O3 depletion event. Early on 14 March, O3 concentrations increased threefold within 1–2 hours, and Hg(0) was correspondingly at near-background concentrations, exactly when the back-trajectories crossed a newly opened, 1-km-wide lead northeast of Barrow (cases 5 and 6). When this lead refroze on 14 March and the trajectories later moved south over consolidated sea ice during the next 2 days, concentrations of Hg(0) and O3 quickly depleted to near instrument detection limits (case 7). Again, on 16 March, concentrations of O3 quickly increased when air-mass trajectories crossed a newly developed lead 20 km from Barrow (case 8). As this lead continued to widen (case 9), both Hg(0) and O3 remained near background levels.

Two more time periods when highly dynamic patterns of Hg(0) and O3 were directly linked to sea-ice lead dynamics are shown in Extended Data Figs 1 and 2, demonstrating a total of 15 cases of this interaction. Patterns were consistent throughout all periods: when air masses travelled over consolidated sea ice or refrozen leads, Hg(0) and O3 were depleted or showed decreasing concentrations. When air masses crossed open leads within the 24 hours before measurements, Hg(0) and O3
concentrations were not, or were only slightly, depleted. One exception to this pattern occurred during a period after 18 March 2009 (Fig. 1), when the seasonal sea ice surrounding Barrow was characterized by large upwind leads (up to 30 km wide) and showed a complex mixture of frozen surface, open and refrozen leads. Both Hg(0) and O₃ showed dynamic fluctuations between depletion and background levels under these conditions, but the temporal and spatial resolution of the satellite imagery did not allow us to link open leads directly to Hg(0) and O₃ concentrations during that time. Mean concentrations of Hg(0) and O₃ during the eight distinct periods highlighted in Fig. 1 were statistically

Figure 1 | Time series of Hg(0) and O₃ concentrations. Concentrations of Hg(0) and O₃ for 2012 (a) and 2009 (b). Yellow boxes are periods when air masses crossed upwind areas of consolidated sea ice. Black boxes are periods when air-mass trajectories crossed open leads. The satellite images represent four typical sea-ice conditions that occurred during measurements. Original satellite images from Google Earth, Terrametrics.

Figure 2 | Impact of sea-ice leads on Hg(0) and O₃ in 2012. Hg(0) and O₃ concentrations between 21 March 2012 and 26 March 2012. Bold numbers correspond to time periods as numbered on the corresponding satellite images. Satellite images were taken at approximately 16:00 UTC (Coordinated Universal Time) each day. Colours represent 24-hour HYSPLIT back-trajectory arrival times near Barrow: orange, 04:00 UTC; blue, 08:00 UTC; red, 12:00 UTC; pink, 16:00 UTC; yellow, 20:00 UTC; black, 00:00 UTC (the next day); and purple, 04:00 UTC (the next day). Original satellite images from Google Earth, Terrametrics.
We considered both chemical and physical processes to explain observed linkages between lead dynamics and boundary-layer chemistry. Measurable levels of gaseous bromine species were reported during both years of measurements, consistent with active bromine chemistry, which is associated with O₃ and atmospheric Hg depletion events. Depletions of Hg(0) and O₃ occurring over consolidated sea ice were probably induced by active bromine chemistry in the area, although this process is not yet fully understood.

However, we cannot attribute the recoveries of concentrations within 1–2 hours of lead openings to chemical processes only. For example, bromine monoxide (BrO) concentrations, as identified from the Global Ozone Monitoring Experiment-2 (GOME-2) spectrometer, showed a consistent presence of large BrO clouds in the region. Therefore, the patterns of Hg(0) and O₃ were unrelated to coincidental patterns at the edges of BrO clouds (Extended Data Fig. 4). This was supported by direct measurements of BrO concentrations at the site that were not significantly correlated with Hg(0) or O₃. Even if atmospheric Hg- and O₃-depletion-event chemistry were to stop on contact with open leads, the Hg(0) and O₃ concentrations would remain depleted for some time and would not quickly recover within just a few hours. Sources of Hg leading to partial recovery of Hg(0) after atmospheric Hg depletion events could include the photochemical reduction of Hg(II) and re-emission from surfaces, but no such source exists for O₃ because O₃ is destroyed during oxidation. Another possible source for Hg(0) recovery involves emissions from Arctic Ocean water. However, this would not explain the simultaneous recovery of both Hg(0) and O₃ because O₃ is not typically emitted by the ocean. It is also striking that recoveries of both Hg(0) and O₃ consistently reached levels near Northern Hemisphere background concentrations, independently of the size of the leads. If there were an ocean source of Hg, this would not be expected.

We attribute the fast transitions from depleted to non-depleted Hg(0) and O₃ levels to changes in boundary-layer dynamics induced by sea-ice leads, which dominate the effects of underlying depletion chemistry. Lead openings generate large sensible and latent heat fluxes from the water surface to the atmosphere owing to strong temperature gradients (more than 20 K) between the warmer ocean water and cold polar atmosphere. This heat transfer causes significant convective mixing in the atmosphere directly above and downwind of leads (see video in ref. 13 of the lead cloud recorded during BROMEX). We propose that such convective mixing produces fast recoveries of surface Hg(0) and O₃ from air masses aloft. Vertical measurements of Hg(0) and O₃ in the stable polar boundary layer have shown that Hg(0) and O₃ depletions are limited to the surface layer, whereas air aloft is not depleted. We confirmed increased turbulent mixing using radiosonde data from Barrow during several periods when shallow boundary layers quickly grew in height in the presence of open sea water (for example, see Extended Data Fig. 3). It is also unlikely that increased wind speed alone—often associated with, and a cause of, opening sea-ice leads—would explain Hg(0) and O₃ recoveries through increased wind shear given the periods of Hg(0) and O₃ recoveries when wind speeds changed little and remained low (below 3 m s⁻¹; Extended Data Fig. 5).
The implications of the observed effects of the dynamics of sea-ice leads on atmospheric Hg and O_3 are large: the recovery of Hg(0) and O_3 via convective transport of Hg(0) and O_3 induced by open leads is probably a source of additional Hg(0) and O_3 to the atmospheric surface layer in the Arctic, all other factors remaining unchanged. Once in the surface layer, resupplied O_3 and Hg(0) from aloft can participate in renewed depletion chemistry, as the sea-ice leads occur at a time of active depletion chemistry, possibly increasing deposition loads attributed to Hg depletion events

and the total amount of O_3 destroyed in the atmosphere. It is probable that a warming environment and changes in sea-ice cover produce other changes in Arctic chemistry that may ultimately affect the quantity of Hg accumulating in biota (for example, shorter duration of sea-ice cover may cause increased photochemical reduction and photodegradation of methyl mercury). As seasonal sea increases at the expense of perennial sea ice and lead activity is expected to increase, large areas across the Arctic may experience increased convective replenishment of Hg(0) and O_3, affecting Hg oxidation and O_3 depletion chemistry, as observed in Barrow. Lead-induced shallow convective mixing of air in response to sea-ice—leads as shown for Hg(0) and O_3—could also affect the boundary-layer input of other pollutants—such as persistent organics, aerosols and other heavy metals.

**METHODS SUMMARY**

**Ground measurements.** Ground-based measurements in BROMEX chromatographed characterization of speciated atmospheric Hg (including Hg(0), Hg(II)gaseous, and Hg(II)particulate) and O_3 on the frozen Chukchi Sea (2 km off the coast) and on the frozen tundra (5 km inland), but only data from the sea ice were used for this study. During the OASIS campaign, atmospheric Hg speciation was measured at three different locations over the frozen Arctic Ocean. All the data presented passed strict quality assurance and control protocols, and the Hg(0) concentrations presented are hourly averages. To be consistent, all O_3 concentrations used for both satellite and ground measurements are hourly averages. To be consistent, all O_3 concentrations used for both years were from the NOAA-operated Barrow Observatory. Correlation analysis showed excellent agreement of data measured at all stations.

**Satellite images and HYSPLIT modelling.** Daily, densely gridded (250 m) MODIS images of ice conditions were composed of the 7-2-1 bands (2,105–2,155 nm, 841–876 nm and 620–670 nm wavelengths) from the NASA Terra satellite. These were combined with high-temporal-resolution NOAA HYSPLIT air-mass trajectories modelled 24-hour backtrajectories in time and generated every 4 hours based on meteorology data from the Global Data Assimilation System (GDAS) interpolated from a 1.0° by 1.0° grid in latitude and longitude. HYSPLIT model runs were performed at 2522 m and 4000 m. All back-trajectories presented are at 25 m and, owing to atmospheric stability, were verified at the heights of 225 m and 400 m. HYSPLIT trajectories generated with GDAS meteorological data were also checked with back-trajectories generated from Weather Research and Forecasting Model meteorological data to verify their paths. Daily-satellite images were used for both years to map sea-ice conditions precisely over several hundred kilometres around our measurement domain—including solid sea ice, open sea-ice leads and refreezing of previously open leads. The images were overlaid with the HYSPLIT back-trajectories to assess how O_3 depletion events and atmospheric Hg depletion events measured on the ground near Barrow related to the upwind footprint area of measured air masses.

**Online Content**

Any additional Methods, Extended Data display items and Source Data are available in the online version of the paper; references unique to these sections appear only in the online paper.

Received 24 June; accepted 29 November 2013.

**Published online 15 January 2014.**
METHODS

Ground measurements. In 2012, two measurement sites were operated simultaneously. The first site (71.32279° N, 156.74527° W), used for Hg(0) data analysis in this paper, was located 2 km off the coast on the frozen Chukchi Sea. A second site (71.27513° N, 156.64031° W) was located 5 km inland over snow/ice-covered tundra. Measurements at both sites included atmospheric Hg speciation and O3 concentrations along with wind speed and direction as well as air temperature. The ice site also included gradient micrometeorological flux measurements of Hg(0) and O3, net solar radiation, and relative humidity; fluxes are not presented in this paper. In 2009, measurements were made over the sea ice and are described in detail in ref. 14.

For Hg, all data shown in this manuscript are from sea-ice sites (one site in 2012; sites OOTI 1, OOTI 2 and OOTI 3 in 2009, as described in ref. 14). The measurement locations, however, did not have a strong effect on observed patterns and showed the same temporal dynamics at all sites. For example, in 2012, concentrations of Hg(0) and O3 measured over the sea ice and tundra were highly correlated ($r^2 = 0.83$ and 0.91, respectively). Therefore, measurements at both sites showed consistent temporal patterns and dynamics. O3 measured at the Barrow Observatory was also strongly correlated to that measured at the tundra site (in 2012, $r^2 = 0.95$) and the sea-ice site ($r^2 = 0.94$).

At all sites in 2009 and 2012, we used Model 2537B mercury vapour analysers (Tekran Inc.) to determine Hg(0) concentrations. Details of the ‘Out On The Ice’ (OOTI) system used to make measurements at the ice site in 2012 and at the OOTI sites in 2009 are also provided (with photographs) in ref. 14. The main difference in set-up between years was that the inlet height in 2009 was approximately 40 cm above the surface snow and in 2012 the height was approximately 90 cm. The Model 2537B preconcentrates (that is, amalgamates) Hg(0) from ambient air onto gold bead sample traps. The Hg(0) is thermally desorbed from the traps and measured with cold vapour atomic fluorescence spectrometry at a wavelength of 253.7 nm. Two other forms of atmospheric Hg were also measured, Hg(0)$_{\text{gaseous}}$ and Hg(0)$_{\text{particulate}}$ (see Extended Data Table 1). These species were measured with the Model 1130 and 1135 mercury speciation units, respectively (Tekran Inc.), which were connected upstream of the Model 2537B by a heated Teflon tube. Soda lime traps were attached just upstream of the Model 2537B. The Model 1130 contained a potassium-chloride-coated annular denuder for collection of Hg(0)$_{\text{gaseous}}$, whereas the Model 1135 contained a particulate filter and glass frit for collection of fine (<2.5 μm) particles. Air was drawn through the 1130 and 1135 models at ten litres per minute for one hour. At the end of the hour, the models were switched to analysis mode, which consisted of system flushing with Hg-free air for 10 min, then heating a pyrolyser in Model 1135 to ensure conversion of all forms of Hg to Hg(0).

HYSPLIT trajectories were also verified at 225 m and 400 m. HYSPLIT model runs were performed at 25 m, 225 m and 400 m. All trajectories presented are at the 25 m height, and owing to atmospheric stability, were also verified at 225 m and 400 m. HYSPLIT trajectories generated with NDAS meteorological data were also checked with trajectories generated from WRF meteorological data to verify paths. Daily satellite images were used for both years to map sea-ice conditions precisely over several hundred kilometres around our measurement domain—including solid sea ice, open sea-ice leads and refreezing of previously open leads. The images were overlaid with the HYSPLIT back-trajectories to assess bow O3 depletion events and atmospheric Hg depletion events measured on the ground near Barrow related to the upfront footprint of measured air masses.

12 sequential 5-minute Hg(0) measurements, during which time the Hg(0)$_{\text{gaseous}}$ and Hg(0)$_{\text{particulate}}$ samples were being collected; then Model 2537B would come offline for one hour to analyse the Hg(0)$_{\text{gaseous}}$ and Hg(0)$_{\text{particulate}}$ samples. Hg(0) data presented has been averaged to correspond to the hourly samples of the Hg(0)$_{\text{gaseous}}$ and Hg(0)$_{\text{particulate}}$.

Strict quality assurance and quality control procedures were applied to all Hg data, based on the standards discussed in ref. 31. In 2012 and by the Atmospheric Mercury Network (AMNet)$^{32}$. To assess system blanks, zero-air was flushed through the glassware without heat at the beginning and end of each desorption cycle. System blanks were calculated as three times the average Hg concentrations measured immediately before and after desorption cycles in zero-air. System blanks were subtracted from Hg(0)$_{\text{gaseous}}$ and Hg(0)$_{\text{particulate}}$. Concentrations to correct for these. Owing to the conditions, the Model 2537B analysers were calibrated only at the beginning and end of the studies, with an internal permeation source. The manufacturer’s reported detection limit for 5-minute samples measured with the Model 2537B analyser is $<0.02$ ng m$^{-3}$.

O3, wind speed and wind direction, we used data from the Barrow Observatory because it has the largest continuous set of O3, wind speed, and wind direction measurements. These data are publicly available from the National Oceanic and Atmospheric Administration Earth System Research Laboratory Global Monitoring Division FTP Data Finder site (http://www.esrl.noaa.gov/gmd/dv/data/index.php?site=brw).

Satellite images and HYSPLIT modelling. Daily, densely gridded (250 m) MODIS images of ice conditions were composed of the 7-2-1 bands (2,105–2,155 nm, 841–876 nm and 620–670 nm wavelengths) from NASA’s Terra satellite. All satellite images used in this analysis are available via the Internet (http://lANCE-modis.eosdis.nasa.gov/imagery/subsets?subset=BROMEX). These were combined with high-temporal-resolution (every 4 hours) 24-hour NOAA HYSPLIT back-trajectories based on GDAS meteorology and interpolated from a 1.0° by 1.0° grid in latitude and longitude. HYSPLIT model runs were performed at 25 m, 225 m and 400 m. All trajectories presented are at the 25 m height, and owing to atmospheric stability, were also verified at 225 m and 400 m. HYSPLIT trajectories generated with NDAS meteorological data were also checked with trajectories generated from WRF meteorological data to verify paths. Daily satellite images were used for both years to map sea-ice conditions precisely over several hundred kilometres around our measurement domain—including solid sea ice, open sea-ice leads and refreezing of previously open leads. The images were overlaid with the HYSPLIT back-trajectories to assess bow O3 depletion events and atmospheric Hg depletion events measured on the ground near Barrow related to the upfront footprint of measured air masses.

Extended Data Figure 1 | A second example of the impact of sea-ice leads on Hg(0) and O₃ in 2012. Time series of Hg(0) and O₃ concentrations near Barrow between 26 March 2012 and 29 March 2012. Bold numbers correspond to time periods, as numbered on the corresponding satellite images. 24-hour HYSPLIT back-trajectories were generated every four hours from 26 March to 29 March 2012. Satellite images were taken at approximately 16:00 UTC each day. Colours represent trajectory arrival times near Barrow: orange, 04:00 UTC; blue, 08:00 UTC; red, 12:00 UTC; pink, 16:00 UTC; yellow, 20:00 UTC; black, 00:00 UTC (the next day); and purple, 04:00 UTC (the next day). Original satellite images from Google Earth, Terrametrics.
Extended Data Figure 2 | A third example of the impact of sea-ice leads on Hg(0) and O₃ in 2012. Time series of Hg(0) and O₃ concentrations near Barrow between 29 March 2012 and 1 April 2012. Bold numbers correspond to time periods, as numbered on the corresponding satellite images. 24-hour HYSPLIT back-trajectories were generated every four hours from 29 March 2012 to 31 March 2012. Satellite images were taken at approximately 16:00 UTC each day. Colours represent trajectory arrival times near Barrow: orange, 04:00 UTC; blue, 08:00 UTC; red, 12:00 UTC; pink, 16:00 UTC; yellow, 20:00 UTC; black, 00:00 UTC (the next day); and purple, 04:00 UTC (the next day). Original satellite images from Google Earth, Terrametrics.
Extended Data Figure 3 | Meteorological and radiosonde data for 22–24 March 2012. The lower 1,000 m of a radiosonde launch from Barrow Airport at 05:30 UTC on 22 March 2012 (a), 23 March 2012 (b) and 24 March 2012 (c). The change in potential temperature with height (dΦ/dz) indicates a boundary layer height near 100 m for 22 March and 23 March (air mass over consolidated sea ice) and grows to 250 m on 24 March (the lead influence).

d. Wind rose for 22 March 2012 0:00 to 26 March 2012 0:00; wind directions are consistently from the east to the northeast, in support of the calculated air-mass trajectories in Fig. 2.
Extended Data Figure 4 | Atmospheric BrO around Barrow. BrO columns estimated from the Global Ozone Monitoring Experiment-2 (GOME-2) spectrometer on the Meteorological Operational satellite-Metop-A during the time period corresponding to patterns shown in Fig. 2. The image domain is 60°–80° N and 170°–270° E.
Extended Data Figure 5 | Low wind speed associated with an opening lead. O₃ and wind speed from 26 March 2012 to 30 March 2012. The bold number corresponds to case 13, when an opening lead influences concentrations of O₃ and Hg(0) that quickly recover to near-background levels. During this time period, wind speeds remained low (below 3 m s⁻¹), indicating that concentration recoveries were not linked to increased wind speed through increased wind shear.
Extended Data Table 1 | Key variable means during the two campaigns

<table>
<thead>
<tr>
<th></th>
<th>Mean Hg(0) (ng m⁻³)</th>
<th>O₃ (p.p.b.v)</th>
<th>Hg(II) gasous (pg m⁻³)</th>
<th>Hg(II) particulate (pg m⁻³)</th>
<th>Relative humidity (%)</th>
<th>Temperature (°C)</th>
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<tbody>
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<td>2012</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>No lead effect</td>
<td>0.33 ± 0.34</td>
<td>6.97 ± 7.48</td>
<td>59.7 ± 33.1</td>
<td>119.1 ± 64.2</td>
<td>57.9 ± 5.7</td>
<td>-30.6 ± 3.2</td>
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<tr>
<td>Lead effected</td>
<td>0.96 ± 0.48</td>
<td>28.56 ± 8.59</td>
<td>54.7 ± 45.9</td>
<td>53.0 ± 39.8</td>
<td>64.6 ± 3.3</td>
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<td></td>
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<tr>
<td>No lead effect</td>
<td>0.07 ± 0.04</td>
<td>7.32 ± 7.88</td>
<td>28.3 ± 8.1</td>
<td>785.0 ± 107.9</td>
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<tr>
<td>Lead effected</td>
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<td>9.7 ± 6.2</td>
<td>299.7 ± 79.4</td>
<td>83.0 ± 6.8</td>
<td>-27.9 ± 1.8</td>
</tr>
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</table>

Means (± one standard deviation) during periods when air-mass back-trajectories either did or did not cross an upwind lead (<20 km wide) for data from 2009 and 2012. All means are significantly different (P < 0.00001) except for 2012 Hg(II)gasous (P = 0.19).