Arctic Sea Ice Reduction and Tropospheric Chemical Processes

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Abstract—**Arctic sea ice extent reached another historical record low in summer 2012. More importantly, perennial sea ice extent in 2012 set the new record low in the long period that extends back to the last half of the 20th century as observed by a combination of long-term measurements acquired by ocean buoys and decadal data acquired by satellite scatterometers. To investigate impacts of sea ice reduction on atmospheric chemical processes, we conducted the BRomine, Ozone, and Mercury EXperiment (BROMEX) in March-April 2012 around Barrow, Alaska. We present an overview of BROMEX and highlight results to document sea ice change and chemical processes. We found a large number of bromine explosion events occurred in the BROMEX area where seasonal sea ice dominated.**

Keywords-Arctic sea ice; bromine; ozone; mercury; BROMEX.

I. INTRODUCTION AND OBJECTIVE

In the context of Arctic change in the last half century, perennial sea ice, the class of thicker and older ice important to the stability of Arctic sea ice, has been declining most precipitously in the last decade. Nghiem et al. [1] reported the extent of perennial ice in the month of March (1950s to the 2000s), estimated by the buoy-based Drift-Age Model (DM) together with nearly a decade of QuikSCAT (QS) satellite observations within the domain of the DM (excluding peripheral seas such as Greenland Sea and the Canadian Arctic Archipelago). From the published data [1], perennial ice extent declined at rate of 0.5 million km^2 per decade in the 1970s-1990s while there was no discernable trend in the 1950s-1960s. Abruptly, the rate of decrease has tripled to 1.5 million km^2 per decade in the 2000s. Such rapid reduction has resulted in a regime shift of Arctic sea ice to being dominated by seasonal sea ice that grows and melts annually. The springtime distribution of these ice classes is critical in terms of preconditioning prior to the summer melt, and moreover it plays a major role in tropospheric chemical processes controlling bromine, ozone, and mercury photochemistry in the Arctic atmospheric boundary layer. To investigate impacts of sea ice reduction on atmospheric halogen and other chemical processes, we conducted the BRomine, Ozone, and Mercury EXperiment (BROMEX) in March-April 2012 around Barrow, Alaska, which was successfully carried out. In the sections below, we state the science issues (II), describe BROMEX (III), present the observations (IV), and provide the summary (V).

II. SCIENCE ISSUES

Observations of the state of Arctic sea ice using multiple satellites show drastic loss of perennial ice extent [1]. More importantly, an initial study has led to the first report of BrO in a clear vortex pattern, and a discovery of an apparent topographic control on the transport and distribution processes of bromine monoxide (BrO) [2]. The full field of the three-dimensional rising-air-parcel (RAP) model [2] forced by the NCEP/NCAR reanalysis [3] consistently reproduces the spatial pattern of BrO observed by the Global Ozone Monitoring 2 (GOME-2) satellite [4].

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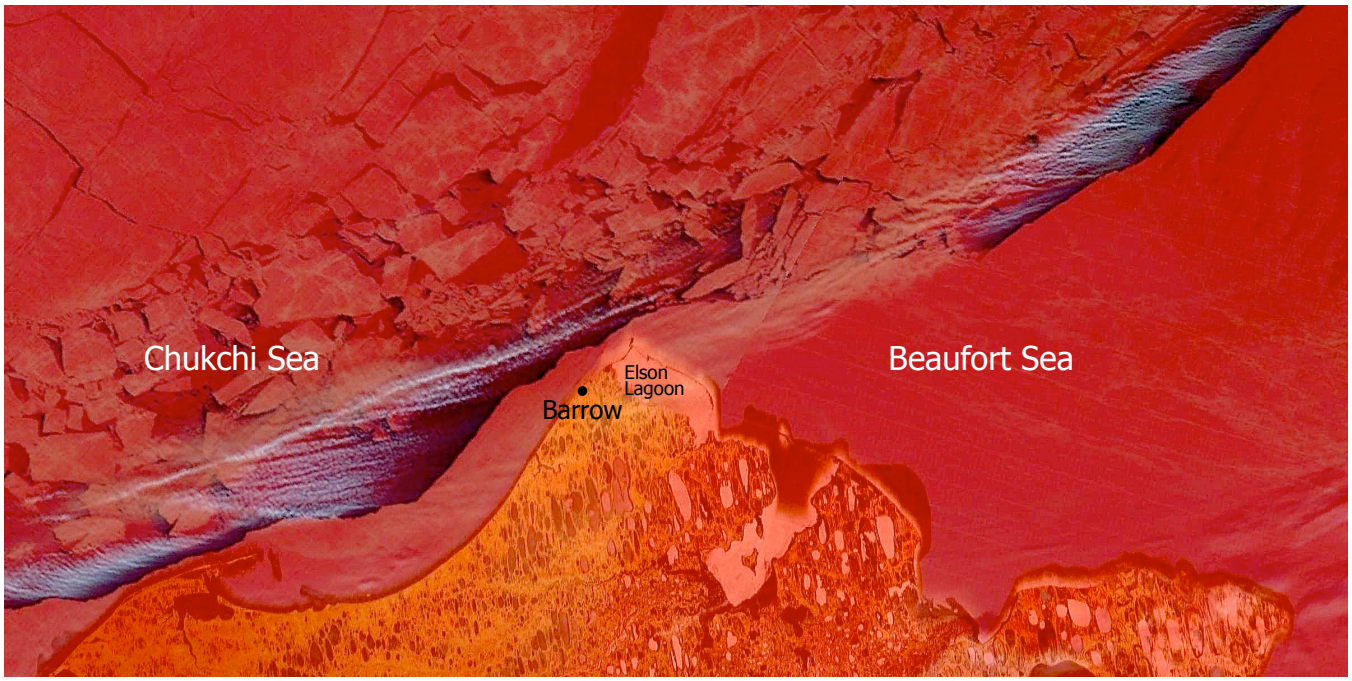

Figure 1. Image from a composition of bands 3, 6, and 7 acquired by the MODIS Terra satellite on 24 March 2012. Sea ice in the Chukchi Sea and the Beaufort Sea appears as red. Dark areas in the ocean are leads, formed by wind forcing that ruptured the sea ice cover. Grayish streaks are vapor plumes emanating from the leads. Landscape features are seen through the translucently overlain MODIS image with red-orange shades indicating snow cover.

The bromine explosion process is briefly summarized here. At the polar-sunrise time, gaseous bromine (Br_2) is photo-dissociated into Br atoms, which catalytically destroy ozone (O_3) and oxidize gaseous element mercury (GEM) with the Br atoms being regenerated in excess in this autocatalytic process. Hypobromous acid (HOBr) formed in a termination step deposits to saline surfaces, generating Br_2 that evolves to the gas phase, providing twice as many Br atoms back to the gas phase, and hence the term "bromine explosion" is used to describe this behavior [5].

Despite recent advances in understanding Arctic tropospheric halogen chemistry in the context of a changing state of Arctic sea ice, key science questions still remain to be answered in order to understand the impact of the recent drastic reduction of Arctic sea ice, such as how the bromine is initiated, recycled, transported, and terminated. Sea ice reduction has profoundly changed the Arctic environment and halogen chemical processes involved in the bromine explosion, ozone depletion, and mercury deposition events. This is a cascade of photochemical reactions that abruptly increase gaseous bromine such as bromine monoxide (BrO) in the atmospheric boundary layer, leading to the depletion of tropospheric ozone (O_3), and the oxidation of gaseous elementary mercury (GEM) to a reactive form that deposits onto land, ice, and ocean surfaces in the Arctic.

III. THE BROMEX FIELD CAMPAIGN

BROMEX was conducted in March and April 2012 around Barrow, Alaska (Figure 1). The field area extended from inland terrestrial sites to a large region of the sea ice offshore, where energetic dynamics created large leads, fortuitously providing a variety of sea ice and atmospheric conditions for this investigation. BROMEX included participation and contributions from more than thirty scientists, researchers, and support staff from multiple international institutions.

We coordinated and collected satellite data, including a number of near-real-time products, from multiple satellite instruments. These include the Moderate Resolution Imaging Spectroradiometer (MODIS), the Advanced Microwave Scanning Radiometer on the Earth Observing System (AMSR-E), the Special Sensor Microwave Imager/ Sounder (SSMIS), GOME-2, the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY), the Ozone Monitoring Instrument (OMI), the RADARSAT-2 Synthetic Aperture Radar (SAR), the Envisat Advanced SAR, the TerraSAR-X add-on for Digital Elevation Measurements (TanDEM-X) SAR, the Soil Moisture and Ocean Salinity (SMOS) radiometer, the CryoSat-2 altimeter, and the Oceansat-2 scatterometer. Multiple products derived from satellite data by the Jet Propulsion Laboratory (JPL), NASA Goddard Space Flight Center (GSFC), U.S. National Ice Center (NIC), University of Bremen, and University of Hamburg were used to plan for and to support the BROMEX campaign.

Three different aircraft were used for instrument deployments and data collections. We deployed atmospheric chemical and meteorological buoys in the Chukchi Sea and the Beaufort Sea. We set up instruments to measure multiple chemical species of mercury at sites on sea ice and on land. In addition to existing weather stations, we installed a meteorological tower and a snow measurement tower at a site on the tundra. At these sites, continuous atmospheric chemical and meteorological measurements were made for many weeks.

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We made measurements and collected samples of sea ice, snow, seawater, and air for physical, meteorological, chemical, biological, and acoustic studies. We obtained an enormous amount of satellite and field data. In the sections below, we present the BROMEX measurement components and discuss highlights of the results to document sea ice change and observed atmospheric and snow chemical processes.

IV. SATELLITE, ARICRAFT, AND FIELD OBSERVATIONS

A. Satellite Observations

Arctic sea ice extent reached another historical record low in summer 2012. More importantly, perennial sea ice extent in 2012 set the new record low in the extensive period from the last half of the 20th century up to date, as observed by a combination of long-term measurements acquired by ocean buoys and decadal data acquired by satellite scatterometers. The NASA SeaWinds scatterometer mission with the QuikSCAT (QS) satellite acquired the most stable and accurate Ku-band backscatter data [6] over a decade (1999-2009). The Oceansat-2 (OS2) scatterometer [7] launched by the Indian Space Research Organisation (ISRO) in September 2009 continues the scatterometer data collection to the present. The satellite data were used to identify, map, and monitor perennial sea ice versus seasonal sea ice that grows and melts annually [1]. To monitor winter perennial and seasonal sea ice, the DM [8] provides over a half-century estimate of Arctic sea ice age distribution to determine the long-term trend since 1955. From monthly gridded fields of ice motion based on buoy data, this model estimates sea ice age, and thereby identifies perennial and seasonal sea ice. In the DM spatial domain, model estimates and satellite scatterometers consistently determined the three-fold decrease rate of perennial sea ice extent in the 2000s.

Updated data from OS2 indicated that springtime perennial sea ice extent reached a new low record in 2012. Except for a narrow band of perennial sea ice in the north of the Alaskan coast, seasonal sea ice dominated the sea ice cover in the Chukchi Sea and the Beaufort Sea. The thinner and weaker seasonal ice, compared to perennial ice, facilitates the formation of large leads when strong winds force sea ice away from shore and creates long open water areas that quickly refreeze and support the growth of new (nilas) ice and frost flowers on the new sea ice surface. This new ice is highly saline and provides a potential source for lower atmospheric halogen chemical reactions. Figure 1, a MODIS image acquired on 24 Mach 2012 during BROMEX, shows a large lead in the Chukchi Sea to the west of Barrow. On the same day, both GOME-2 and OMI satellites detected extensive areas where a bromine explosion occurred above the ocean and across the terrestrial tundra land in the North Slope of Alaska.

B. Aircraft Observations

During BROMEX in March 2012, the Purdue Airborne Laboratory for Atmospheric Research (ALAR; see details at [9]) was used as a key platform to measure ozone (O_3, in-situ), BrO (produced entirely from Br atoms reacted with O_3, with Multi Axis – Differential Optical Absorption Spectroscopy or MAX-DOAS), and aerosol number and size distributions (in-situ). Eleven flights were conducted over continuous first-year sea ice in the Beaufort Sea, over new ice and leads with frost flower coverage in the Chukchi Sea, and over the snow-covered tundra between Barrow, Deadhorse, and the Brooks Range. All systems performed well during this campaign.

From initial analyses of the ALAR data, a number of interesting findings were apparent during the flights. For example, on 24 March 2012, we flew over the tundra, at a constant altitude, and observed the BrO distribution in the boundary layer. There was a relatively enhanced column of BrO over the tundra on this day. On other days, elevated aerosol layers were observed aloft, possibly suggesting long-range transport. Interestingly, on two flights, we observed deeper ozone depletions over the tundra, compared to the sea ice. Overall, the simultaneous O_3, BrO, and aerosol measurements will yield many insights into halogen cycling.

Through a coordination with the NASA IceBridge program, the NASA P3 aircraft was flown across the BROMEX field domain from the Beaufort Sea, across Elson Lagoon and the North Slope tundra, to the Chukchi Sea. Sensors on the IceBridge P3 aircraft measured multiple parameters including: sea ice thickness, snow thickness, surface height, and surface temperature. These parameters are useful for interpretation of sea ice and snow physical properties and for investigating their roles in halogen photochemical processes.

C. Field Observations

Field activities for BROMEX were focused on two types of locations: (1) buoy sites in the Arctic Ocean, and (2) snowmobile-accessible locations on land and shore-fast sea ice near Barrow, Alaska. A major goal of the near-shore sea ice work was to access a location where the landfast ice and pack ice meet so that when a major flaw lead opened we would have access to the open water. A safety plan was carefully developed and implemented for BROMEX since this area could be dynamic and frequented by polar bears.

A key component of BROMEX was the deployment of two chemistry buoys, fabricated by the University of Alaska Fairbanks (UAF): IceLander 1 (IL1) in the Beaufort Sea and IceLander 2 (IL2) in the Chukchi Sea. This deployment process had three steps: (1) defining the criteria for a suitable floe, (2) analyzing remote sensing imagery to select candidate floes, and (3) flying out to the candidate floes for an in-situ evaluation. The criteria were: first-year ice, center area of hundreds of meters without major ridges, snow covered, accessible by helicopter, and likely to survive into the melt season. With the high quality satellite imagery, the U.S. Army Cold Regions Research and Engineering Laboratory (CRREL) and UAF easily found the candidate floes and selected the best floe to deploy the IceLanders. Surveys of snow depths were performed, and results showed that average snow depths were comparable at the two sites.

IL1 was stationary until its retrieval in March 2012. IL2 drifted across sea ice under a variety of conditions from pack ice, to lead opening and closing, and unconsolidated sea ice in March to June 2012 when it was knocked over by

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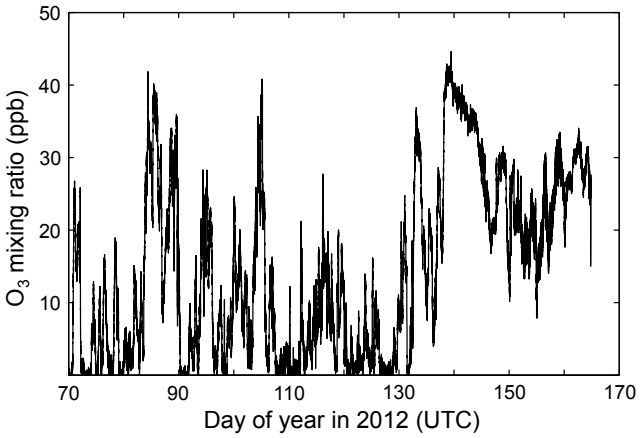

Figure 2. Ozone mixing ratio from IceLander 2 in the Chukchi Sea. Many ozone depletion events were observed in March to May 2012.

a polar bear. An identical DOAS system was also installed at a building in Barrow (BRWDOAS), which has been operating continuously to the present. An analysis has been completed for Level-2 (slant column density of gases versus elevation angle) data and also Level-3 (vertical profiles of BrO) data for BRWDOAS, IL1, and IL2. O_3 data were processed and finalized for IL1, IL2, and BRWDOAS. IL2 O_3 in Figure 2 revealed that ozone depletion events occurred frequently forcing O_3 level to near complete depletion in many instances in the Chukchi Sea region off the west coast of Barrow where it was dominated by seasonal sea ice. A preliminary analysis has been carried out for Lagrangian matching of data between IL1, IL2, and Barrow to look for changes in BrO and ozone related to sea ice. First examinations indicate that ozone and BrO changes between sites are small. An examination of the longer-term seasonal behavior of IL2 and BRWDOAS revealed a relationship between the BrO seasonal end date and snow properties. Additionally, snow microphysics is currently researched to obtain a solid interpretation of these observations.

To assess how changing sea ice conditions may affect the regional cycling of atmospheric mercury, Environment Canada's Out On the Ice (OOTI) system was deployed and operated at a site approximately 1 km from shore on the frozen Arctic Ocean during March-April 2012. This system measured speciated atmospheric mercury concentrations, including gaseous elemental mercury (GEM), reactive gaseous mercury (RGM), and fine particulate bound mercury (FPM). Environment Canada and the Desert Research Institute also collected data for surface-atmosphere fluxes of GEM and O_3, and meteorological variables, and measured speciated atmospheric mercury and O_3 concentrations at a second site 5 km inland in the Arctic tundra, co-located with in-situ halogen measurements by Purdue University. At each site, surface snow samples and a sample from a newly designed cold-plate sampler were collected daily for total mercury and major ion concentrations. Striking changes in GEM, RGM, and FPM concentrations occurred during the transition from a large open lead to newly frozen sea ice, furthering our understanding of the complexity of Arctic mercury depletion. The concentration of GEM was different at the inland tundra site and at the OOTI site over the sea ice reflecting the impact of sea ice on the mercury cycling.

At the inland tundra site, chemical ionization mass spectrometry was utilized to measure the concentrations of various bromine species, including Br_2, HOBr, and BrO, at a high temporal resolution. Together with co-located measurements of ozone and mercury, these ambient halogen measurements are being utilized to gain greater insights into the role of bromine in ozone and mercury depletion events. In addition, these BrO measurements on the tundra are being compared with other BrO measurements completed at other locations. Overall, a good agreement has been observed between the DOAS and chemical ionization mass spectrometry measurements of BrO. For the first time, collocated simultaneous mercury speciation and in situ halogen measurements were made to understand the link between these atmospheric components.

Accurate estimates of wind and temperature are required for understanding the myriad of processes involved in the transport and distribution of the chemicals. In-situ observations were provided by nearby National Oceanic and Atmospheric Administration (NOAA) weather stations, and to accurately document conditions at each field site, portable weather stations (PWS) were installed at each site. The Applied Physics Laboratory of the University of Washington (APL/UW) provided the PWS at the tundra site, and deployed buoys from the International Arctic Buoy Programme (IABP) offshore from the field camp to assure that there were adequate measurements for the analyzed weather fields produced by the National Weather Service (NWS), Alaska Weather Research Forecasts (WRF), and the National Centers for Environment Prediction (NCEP) reanalysis fields. Moreover, ten thermochrons provided by GSFC were deployed in different sea ice, snow, and built environments for comparison with MODIS satellite data collocated in time and in space with the surface measurements. Initial results revealed complexities that call for a rigorous protocol to be developed for accurate and consistent temperature measurements.

Understanding the role of snow in the chemical processes is crucial. During BROMEX, intensive fieldwork was carried out to document snow conditions around Barrow and under the flight line of the IceBridge P3 aircraft. The mean snow depth measured by the P3 aircraft was validated with thousands of snow depth measurements collected on sea ice and tundra. Overall, about 10,000 snow measurement samples were obtained across the BROMEX domain. The initial interpretation of the results suggests the P3 measurements appear to overestimate areas of low snow cover, and underestimate areas of high snow cover. To investigate the change in snow distribution over the tundra area after a strong wind event, the volume of snow that moved during this event was estimated. By examining the spatial lagged autocorrelation (i.e. how far did the snow dunes move), the distance that the snow transported during this event could be derived. Furthermore, photographic and videographic records were obtained to document snow and ice conditions, and BROMEX field activities.

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In an effort to trace the source and transport of bromide ions from the ice and snow reservoirs, two sea ice cores, 38 snow samples, and 20 blowing snow samples were collected at several locations in the vicinity of Barrow by Dartmouth College. The sea ice cores were each approximately 1.5 m long. Surface snow was collected at both sea ice core locations, at several locations in Elson Lagoon, at a tundra site, and along a 2-km East-West transect near the shoreline of the Chukchi Sea where samples were also collected for mercury analysis. Where possible, snow pits were dug to sample snow from different depths. Moreover, blowing snow samples were collected at four different heights (0.5 m, 1.5 m, 2.5 m, and 5.5 m) above the snow surface over a seven-day period. As a part of the ground-truthing efforts for the IceBridge aircraft measurement, surface temperature, surface height, snow depth, and ice thickness measurements were collected. In addition, in-situ measurements were taken to measure snow density and snow water equivalent.

The U.S. Naval Academy (USNA) also participated in BROMEX. CRREL and USNA personnel worked with members of the Barrow community to establish the opening of a 3 km of trail to the OOTI site and other locations in the Chukchi Sea. USNA successfully deployed IceGoat1 Buoy during BROMEX. This buoy reported near-real-time observations of air and sea temperatures, atmospheric pressure, position and images from two mounted webcams. Acoustic instrumentation systems were set up for under- and through-ice ambient noise and propagation loss measurements to characterize acoustic signatures of lead ruptures and closures. Sea ice, frost flowers, snow, brine icicle, and water samples were collected for chemical and biological analyses. Bulk snow and water samples were analyzed for ^{137}Cs and ^{134}Cs to look for the fingerprint of the 2011 Fukushima-Daiichi Nuclear Power Plant incident. Twenty bacterial cultures were made: 19 psychrophilic clones (obligate cold growth) isolated from ice core and seawater interface samples, and one psychrotolerant clone (in 4°C - 25°C) from a brine icicle sample. Also obtained was a small eukaryote to be studied for biofuels potential or for general characterization and potential toxicity.

V. SUMMARY

The BROMEX field campaign was successfully carried out across the sea, land, and air region around Barrow, Alaska. More than twenty institutions from U.S., Canada, U.K., and Germany participated in BROMEX. This is a major interdisciplinary science research to investigate impacts of Arctic sea ice reduction on atmospheric photochemical processes of bromine explosion, ozone and mercury depletion, and mercury deposition. Large datasets were obtained from the surface (sea ice and tundra snow), from aircraft (ALAR and NASA IceBridge P3), and from space (multiple international satellites). Valuable scientific findings have emerged from our initial analyses of BROMEX data, from which better measurement protocols and new research directions can be identified. While BROMEX helps improve our understanding of halogen processes in a changing Arctic, it has also initiated new science questions to be investigated in the future.

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