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Satellite observations of long range transport of a large BrO cloud in the Arctic

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Abstract

Ozone Depletion Events (ODE) during polar springtime are a well known phenomenon in the Arctic and Antarctic boundary layer. They are caused by the catalytic destruction of ozone by halogens producing reactive halogen oxides like bromine monoxide (BrO). The key halogen bromine can be rapidly transferred into the gas phase in an autocatalytic process – the so called "Bromine Explosion". However, the exact mechanism, which leads to an initial bromine release as well as the influence of transport and chemical processes on BrO, is still not clearly understood. In this study, BrO measurements from the satellite instrument GOME-2 are used together with model calculations with the dispersion model FLEXPART and Potential Frost Flowers (PFF) maps to study a special arctic BrO event in March/April 2007, which could be tracked over many days and large areas. Full BrO activation was observed within one day east of Siberia with subsequent transport to the Hudson Bay. The event was linked to a cyclone with very high surface wind speeds which could have been involved in the production and the sustaining of aerosols providing the surface for BrO recycling within the plume. The evolution of the BrO plume could be well reproduced by FLEXPART calculations for a passive tracer indicating that the activated air mass was transported all the way from Siberia to the Hudson Bay without further activation at the surface. No direct link could be made to frost flower occurrence and BrO activation but enhanced PFF were observed a few days before the event in the source regions.

1 Introduction

Strong depletion of tropospheric ozone in the polar boundary layer in spring was first reported in the 1980s for the Arctic at Barrow, Alaska (Oltmans, 1981) and Alert, Canada (Bottenheim et al., 1986). Later on measurements in the Antarctic at McMurdo (Sturges et al., 1993), Neumayer (Wessel et al., 1998), Syowa (Murayama et al., 1992) and Arrival Heights (Kreher et al., 1997) and at many other Arctic and Antarctic stations

ACPD

9, 20407–20428, 2009

followed, demonstrating that ODEs are a regular feature in the polar atmosphere. The importance of halogens for these events was suggested and validated by a clear correlation between low ozone and high values of filterable bromine (Barrie et al., 1989). Based on these data, a dominating bromine radical-catalysed cycle involving Br and BrO radicals was suggested to explain the destruction of ozone (Barrie et al., 1988). The occurrence of such ozone depletion events, lasting from a few hours to several days, is limited to the polar boundary layer in springtime and has been linked to low temperatures and the presence of sufficient sunlight. Under these conditions, bromine is supposed to be released from sea salt to the gas phase by different photochemical and heterogeneous reactions. The following autocatalytic sequence of reactions is the so called "Bromine Explosion" (Platt and Lehrer, 1996) which rapidly releases bromine from the liquid to the gas phase and explains the occurrence of the observed high BrO and low ozone values.

$$HOBr + H^+ + Br^- \longrightarrow H_2O + Br_2 \tag{R1}$$

$$Br_2 + h\nu \longrightarrow 2Br\cdot \tag{R2}$$

$$Br\cdot + O_3 \longrightarrow BrO\cdot + O_2 \tag{R3}$$

$$BrO\cdot + HO_2 \longrightarrow HOBr + O_2 \tag{R4}$$

$$H^+ + Br^- + HO_2 + O_3 \longrightarrow Br\cdot + H_2O + 2O_2 \tag{R5}$$

The first of the equations represents the heterogeneous reaction, which is required for initial release of gaseous bromine to the atmosphere. It can possibly proceed on surfaces with enriched concentrations of sea salt like fresh sea ice (Frieß et al., 2004; Simpson et al., 2007), sea salt aerosols (Vogt et al., 1996), frost flowers or aerosol from frost flowers (Kaleschke et al., 2004), sea salt deposited on ice or snow (McConnell et al., 1992) or blown snow which is enriched in sea salt (Yang et al., 2008). The production of reactive bromine species leads to an exponential growth of BrO radicals in the atmosphere, which can be measured in situ or by absorption spectroscopy from the

ACPD

9, 20407–20428, 2009

Satellite observations of long range transport of a large BrO cloud

M. Begoin et al.

ground and from space. BrO measurements from ground (Hausmann and Platt, 1994; Frieß et al., 2004) and satellite (Wagner et al., 1998; Richter et al., 1998) show a good correlation with depleted ozone concentrations (Hönninger and Platt, 2002; Lehrer et al., 1997) and are mainly observed above sea ice areas (Richter et al., 2002; Wagner et al., 2001). Due to their high reactivity, BrO radicals have a very short atmospheric lifetime and should vanish quickly from the atmosphere by consecutive reactions. Ground-based observations of ODEs are often limited to a few hours. However, BrO plumes can be observed in satellite data for many days and over large areas, indicating additional chemical mechanisms and the transport of the involved air masses. To date it is not clear whether these large-scale plumes indicate continuous generation of BrO from its surface sources even as an air mass moves around, or whether the BrO levels can be sustained in the air as it is transported away from surface sources of bromine. In this paper, we analyse an individual large BrO event observed in measurements of the GOME-2 instrument in March/April 2007. The sequence of satellite measurements is used to study the spatial and temporal evolution of the event and is compared to results from the FLEXPART dispersion model. In addition, maps of potential frost flowers coverage are used to investigate possible reasons for the initial bromine release.

2 Methods

2.1 GOME-2 BrO data

GOME-2 (Global Ozone Monitoring Experiment-2) is a UV/vis nadir-viewing grating spectrometer that measures the solar radiation scattered and reflected by the atmosphere (Callies et al., 2000). It was launched into a polar sun-synchronous orbit on the MetOp-A satellite (Meteorological Operational satellite-A) in October 2006 and has an equator crossing time of 09:30 h local time. With a spatial resolution of 40×80 km^2 and a scan width of 1920 km, a global coverage within one day can be achieved and several measurements per day are available at high latitudes under daylight conditions.

GOME-2 measures in the wavelength range between 240 and 790 nm with a spectral resolution between 0.2 and 0.4 nm. Retrieval of tropospheric BrO columns is performed in three steps: First, the BrO amount along the line of sight is determined using the well known Differential Optical Absorption Spectroscopy (DOAS) method (Platt, 1994). From this column, an estimate of the stratospheric contribution is subtracted and the resulting tropospheric column then converted into a tropospheric vertical column. For spectral analysis, a fitting window of 336 to 347 nm has been used as in the analysis of SCIAMACHY measurements (Afe et al., 2004). In the fit, the absorption cross-sections of O_3 (223 K and 273 K), NO_2 (223 K) and BrO are included as well as a Ring-pseudo-spectrum for correction of the effect of Rotational Raman scattering and a polynomial of order 4. The result of this analysis is the slant column SC which comprises both the tropospheric and stratospheric BrO. As here only the tropospheric component is of interest, a correction is performed subtracting a constant stratospheric vertical column of 4.5×10^{13} molec/cm^2 BrO based on results from SCIAMACHY limb measurements (Rozanov et al., 2005). While the assumption of a constant stratospheric field is an approximation, the uncertainty introduced for the discussion below is small. For the determination of vertical columns (VC) of BrO, a tropospheric Air Mass Factor (AMF) was calculated, assuming all BrO to be well mixed in a boundary layer of 400 m. A surface albedo of 0.9 was used as the measurements are performed over ice. Under these conditions, the exact height of the assumed boundary layer is not critical for the air mass factor. BrO retrievals using GOME-2 data have already been used in previous studies, e.g. investigating BrO in a volcanic plume (Theys et al., 2009).

2.2 FLEXPART model simulations

FLEXPART is a Lagrangian atmospheric particle dispersion model that simulates the long-range and mesoscale transport and diffusion of atmospheric tracers as well as loss processes such as dry and wet deposition or radioactive decay (Stohl et al., 2005). FLEXPART has been validated against large scale tracer experiments (Stohl et al., 1998). In this study, FLEXPART is used to simulate the transport of two BrO "clouds". The model was driven by operational meteorological analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) with a spatial resolution of 1 degree and a temporal resolution of 3 h. The model output had a resolution of 0.5 degree and a temporal resolution of 1 h. FLEXPART was initialized with the observed distribution of BrO at a particular time, and thereafter BrO was treated as a passive tracer that was tracked for about two weeks.

2.3 Potential frost flowers maps

Frost flowers have been discussed as a possible source of released bromine. They have a large surface area and a salinity up to three times higher than sea water, which can support the release of bromine to the atmosphere. Frost flowers grow by desublimation of humidity from the air on a thin brine layer on top of newly forming sea ice (Rankin et al., 2002) and are a source of sea salt aerosol. They have a lifetime of a few days and can be blown by strong winds or covered by snow (Perovich and Richter-Menge, 1994). Frost flowers can be a large source for sea salt aerosols. Sander et al. (2006) proposed a mechanism by which airborne sea-salt particles can be formed under PFF conditions. Here, GOME-2 BrO data will be compared with Potential Frost Flowers (PFF) maps describing the frost flowers area coverage from 0 to 100%. They are created using JRA-25 meteorological reanalysis data and SSM/I sea ice maps in combination with a thermodynamic model to simulate areas where frost flowers can potentially grow (Kaleschke et al., 2004).

3 Results

The BrO event investigated here started on 26 March in Laptev and East Siberian Sea with high tropospheric BrO values from 6.0×10^{13} molec/cm^2 up to maximum values of 1.9×10^{14} molec/cm^2 (Fig. 2). The days before, BrO values were significantly lower over the Arctic Ocean sea ice. The observations with the GOME-2 instrument indicate a fast transport of the fully developed BrO cloud on 26 March within the following 3 days towards Hudson Bay and the east coast of Greenland, where high BrO values could be detected until 5th April. At the same time, a few additional small BrO events can be observed from 31 March until 5 April in Kara, Barents and Greenland Sea. On 30 March the GOME-2 instrument was operated in narrow swath mode, which increases the spatial resolution but reduces coverage.

3.1 Meteorological situation

On 26 March 2007, a cyclone covered large parts of the Arctic Ocean, extending from the Laptev Sea to the North Pole. High surface wind speeds (Fig. 4) were associated with this cyclone. High BrO concentrations were first observed especially in the southern part of the cyclone north of the Laptev Sea where surface wind speeds were highest. This suggests that these high wind speeds were involved in the initial release of BrO. Over the next few days, the cyclone weakened while travelling eastwards. On 28 March, it was centered over the Beaufort Sea north of Alaska and Canada. Wind speeds were still high just north of Alaska/Canada but weakened substantially on 29 March. Since the BrO cloud first appeared on 25/26 March in the southern part of the cyclone where strong westerly winds prevailed and also the cyclone tracked eastward over the following days, the air mass containing the high BrO is expected to have travelled eastward towards Alaska/Canada, too. The comparison of surface wind fields from ECMWF model data (level 91) with observed BrO values gives a clear hint that transport processes must be involved in the evolution of the BrO field. Wind directions indicate a transport from the East Siberian Sea over the pole and from the Laptev Sea towards Hudson Bay.

3.2 FLEXPART model simulations

To localize the most likely initial BrO source in time and space, all single satellite orbits have been analyzed in detail. Owing to its large swath, GOME-2 provides several overpasses over each location at high latitudes at this season. The event appears to have started in two different, clearly separated regions. Three orbits were identified as covering the region of BrO initialization, one for the first event and two consecutive orbits which have to be combined to completely cover the second source region (Figure 1). To extract the most probable source region for the BrO events, a threshold was applied to the tropospheric BrO fields and only values above 6.0×10^{13} molec/cm^2 BrO VC were used. The exact time of BrO initialization cannot be determined from the satellite measurements. The process must have fully started some time between the measurement on 25 of March, when BrO was still low and the higher observations on 26 March. At that time, the BrO plume had possibly already expanded and moved away from the initial source region through transport. The fully evolved BrO distribution on 26 March derived in this way was then used as input for a FLEXPART simulation. The BrO distribution was remapped to a resolution of $0.5^{\circ}\times0.5^{\circ}$ and FLEXPART was initialized with the observed concentrations in these grid cells at the time of observation, assuming that all the BrO resided in the lowest 100 m of the atmosphere. Subsequently, the BrO was treated as a passive tracer that was transported by the ECMWF resolved winds as well as turbulence and convection parameterizations. To facilitate comparison with the satellite measurements, the 1 h FLEXPART model output had to be integrated vertically and sampled in a similar way as the daily averaged GOME-2 data. In a first step, each satellite orbit was matched with the FLEXPART model output closest in time and location. In the second step, each FLEXPART output chosen was masked spatially with its allocated GOME-2 orbit. Finally, the preprocessed FLEXPART data were combined in a single composite to get daily averaged data comparable to the satellite output. This was done for both FLEXPART model runs. Due to the mentioned clear separation in time and space of the two BrO events, the model results were added to create the final FLEXPART average to be compared to the GOME-2 satellite data. As the GOME-2 measurements, the daily averaged FLEXPART results show rapid transport of the passive BrO tracer towards Hudson Bay within five days. Further they show very high values in the Laptev Sea on 29 March before transport over the pole into the Barents Sea and towards Northern Greenland takes place (Fig. 3). Throughout the transport, the BrO tracer remains in the lowest atmospheric layers (mostly below 500 m) indicating transport close to the surface.

3.3 Potential Frost Flowers (PFF) maps

Values of the Potential Frost Flower coverage area until one week before the observed BrO event reach up to 50% but mainly lie below 1%. The Potential Frost Flower coverage area during and before the BrO event is shown in Fig. 5. From 20 to 25 March Potential Frost Flowers maps show increased values above 1% in the potential source regions identified by their large BrO columns on 26 March, but also afterwards from 27 to 30 March increased values can be identified. From 30 March until 3 April enhanced values can be also seen between the Canadian Arctic Archipelago, in the Hudson Bay and between Greenland and Spitsbergen and Franz Josef Land.

4 Discussion

4.1 Comparison of GOME-2 BrO with FLEXPART model data

The fast transport towards Hudson Bay seen in GOME-2 BrO data can be well described by modeled FLEXPART data (Fig. 3). The initialized BrO cloud is transported above the Beaufort Sea (27 March) towards the Canadian Arctic Archipelago (28/29 March) and finally towards Hudson Bay. On 31 March, the transported BrO in the FLEXPART model results is clearly isolated to a smaller region than observed in the GOME-2 BrO data, where BrO is distributed over a larger area with lower values. Nevertheless, the affected area showing enhanced BrO values stays the same, also in the following days until 5 April. In the model, the BrO cloud then moves towards Greenland, whereas an increase of BrO can be observed on 1/2 April North of the Hudson Bay in comparison to FLEXPART model results, indicating a fresh emission of BrO.

ACPD
9, 20407–20428, 2009

Title Page
Abstract Introduction
Conclusions References
Tables Figures
◀◀ ▶▶
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

From 27 till 29 March, the FLEXPART model data show very high values also in Laptev and East Siberian Sea, which can't be identified in the corresponding GOME-2 BrO maps. On 27 March BrO values in this region are much lower than predicted by the model, on 28 March high BrO values above 6.0×10^{13} molec/cm^2 can only be seen in a small part of the East Siberian Sea and on 29 March arctic BrO values above 6.0×10^{13} molec/cm^2 are no longer existent except for the Canadian Arctic Archipelago. These observations could be explained by the deactivation of BrO within the transported air masses, which is not taken into account in the model. Further the presence of clouds can't be excluded which could shield low BrO from the satellite's view. While the overall pattern agrees quite well, the absolute values differ: On the one hand the observed GOME-2 BrO is spread over a larger area with lower values than predicted by the model. On the other hand there are some regions, where BrO values show a strong increase, which indicates new BrO events and the additional emission of BrO.

4.2 Possible influence of frost flowers – Comparison of GOME-2 BrO with calculated Potential Frost Flowers (PFF)

Significant values of PFF>1% in the potential source region of the BrO event can be found in a large area on 20 and 21 March. Also afterwards, enhanced values can be found in northern Chukchi Sea until one day before the event starts. From this we conclude that for a direct initialization of the BrO event by frost flowers due to a wind induced release of sea salt aerosols to the gas phase a life time of frost flowers respectively their saline compounds of up to five days has to be assumed. Additional emissions of BrO during the time of the BrO event were observed from 31 March until 4 April in Kara Sea, on 1/2 April north of Hudson Bay and on 4/5 April near the north-eastern coast of Greenland, but in all cases a large life time of five days has to be assumed, if frost flowers should be a potential source for an additional release of bromine to the atmosphere. High PFF values from 1st to 3 April could explain a new bromine explosion on 4 April near the north-eastern coast of Greenland quite well, because the PFF affected area fits exactly to enhanced BrO values in the same region on 4 April. Further, enhanced BrO values in comparison to FLEXPART model results north of Hudson Bay (1 to 4 April) could be explained by additional release of BrO in the Canadian Arctic Archipelago, where high PFF values could be found from 30 March to 2 April. As alternative to frost flowers, first year sea ice was also discussed in the literature (e.g. Wagner et al., 2007; Frieß et al., 2004) as the relevant surface for bromine activation. Simpson et al. (2007) evaluated measurements in winter 2005 at Barrow (Alaska) and found better correlation of enhanced BrO columns with first-year sea ice contact than with potential frost flowers. No such statistical analysis was performed here, but it is interesting to note that due to the low altitude of transport in this event, sea-ice contact time was long independently of the mechanisms for initial activation.

4.3 Long range transport and life time of BrO

Due to its high reactivity, BrO radicals should vanish within hours from the atmosphere. In this special event, BrO has apparently been transported over thousands of kilometers from its probable source region in the East Siberian and Laptev Sea towards Hudson Bay and possibly also over the pole to the Barents and Kara Sea within more than one week and has not been produced in situ. A similar but smaller transport event was also described by Ridley et al. (2007). They explained an observed ODE over Hudson Bay during April 2000 by assuming a long-range transport of rapidly recycled BrO over 1000–1500 km using back trajectories and a polar regional model. A 3-D model study on BrO production and ozone depletion in the Arctic boundary layer for three different Arctic stations by Zhao et al. (2008) also indicated that local ozone destruction seems to be less important than transport of ozone depleted and BrO enriched air masses. This confirms the probable importance of transport processes for ozone depletion events and bromine compounds released in bromine explosions. In the potential BrO source region, high wind speeds dominated during the release process as well as during the complete observed transport event. High wind speeds with additional blowing snow being prevalent during a BrO event were also observed in the Antarctic at Halley station by Jones et al. (2009) using both, ground-based and satellite observations. These case studies suggest that high wind speeds and possibly also blowing snow (Yang et al., 2008) can play an important role in the release and recycling mechanisms of BrO. The movement of the BrO plume studied here agrees with the pattern predicted by FLEXPART model results for a passive tracer. For this reason, highly efficient chemical reactions have to be assumed to continually recycle BrO. For these reactions, three different possibilities exist:

1. Heterogeneous reactions on particles transported with the air mass, for example aerosols, blown snow or sea salt aerosols from frost flowers. Continuous presence of aerosol in the air mass could have been supported by the dominating high wind speeds during the whole BrO event.

2. Deposition and subsequent re-emission of bromine on the snow and ice surfaces. This grasshopping forward movement should result in a lagging of the observations behind the model predictions which only account for direct transport. This can't be validated by FLEXPART model results and GOME-2 BrO measurements and needs further systematic investigation of similar large BrO events.

3. Another possible explanation would be that the surfaces were already preconditioned for bromine release along the transport path and BrO within the apparent BrO plume served only to initiate the release of fresh bromine from the surfaces in the bromine explosion mechanism. This could also be an explanation for higher BrO values in comparison to FLEXPART model results observed north of Hudson Bay.

No firm conclusions can be drawn on the recycling mechanisms from the satellite data alone. However, the good agreement between transport calculation and observations as well as the relatively constant total BrO amount observed over several days and the high wind speeds involved suggest that at least for this event, recycling on aerosols within the air mass is more important than surface reactions.

5 Conclusions

In March/April 2007 a large BrO event could be observed in GOME-2 data over many days in the Arctic, reaching maximum tropospheric column values up to 1.9×10^{14} molec/cm^2. Full activation of the BrO took place between two GOME-2 overpasses which were 24 h apart. The BrO plume originated from two different source regions and over the next five days moved several thousand kilometers towards the Hudson Bay. The initialization and movement of the event was linked to a cyclone with very high surface wind speeds. Using the particle dispersion model FLEXPART and a passive tracer initialized in the boundary layer with the BrO observations from the first day, the satellite BrO values from subsequent days could be well explained by transport processes. Due to the short lifetime of BrO, highly efficient recycling processes have to be involved within the transported air masses to sustain the high BrO levels observed over this long period. The combination of high wind speeds and rapid movement of the BrO plume suggests that recycling of BrO took place on aerosols and/or snowflakes in the air mass rather than on the surface. The rapid activation of the BrO followed by several days of relatively constant total BrO amounts further supports the interpretation of the event as combination of an initial release with subsequent transport of BrO. To identify the recycling mechanism and probable source regions of BrO further statistical investigations of BrO events are needed. The appearance of additional BrO in the Kara and Greenland Sea several days after the initial event could be explained by reactivation of transported bromine compounds and the initialization of a new bromine explosion process. A direct correlation between high BrO values and enhanced values of PFF was not found, but high PFF values were observed a few days before high BrO values occurred. This could be an indication for initialization of bromine events by sea salt aerosols released from decaying frost flowers under the high wind speeds characteristic for this event or other possible halogen sources like first year sea ice or salt enriched snow. The observed long range transport of BrO supports earlier conclusions that low ozone events and BrO observed at coastal stations can originate from far away bromine sources in the Arctic Sea ice. The efficient recycling of BrO over several days indicates long-range transport of sea-salt aerosols in the cyclone from East Siberia to the Hudson Bay which is important for the analysis of in situ measurements and interpretation of ice cores.

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ACPD

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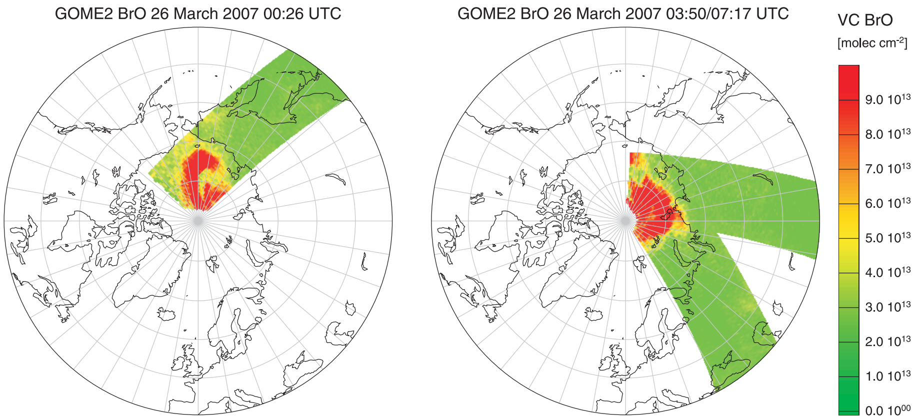

Fig. 1. The above figures show the three satellite orbits, which have been used as input to initialise the FLEXPART model calculations. For the second source region, two orbits have been combined to cover the complete BrO plume.

ACPD
9, 20407–20428, 2009

Satellite observations of long range transport of a large BrO cloud

M. Begoin et al.

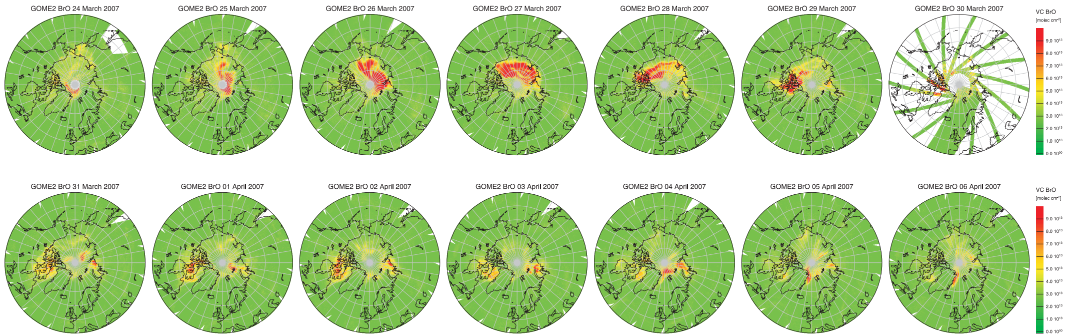

Fig. 2. Daily averaged vertical column of GOME-2 tropospheric BrO measurements. Enhanced values on 25 March in East Siberian Sea result from the last orbit at 22:44 UTC. The following orbit on 26 March at 00:26 UTC (Fig. 1) was used as one input for the FLEXPART model run. On 30 March GOME-2 was operated in narrow swath mode.

ACPD
9, 20407–20428, 2009

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◂◂ | ▸▸
◂ | ▸
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

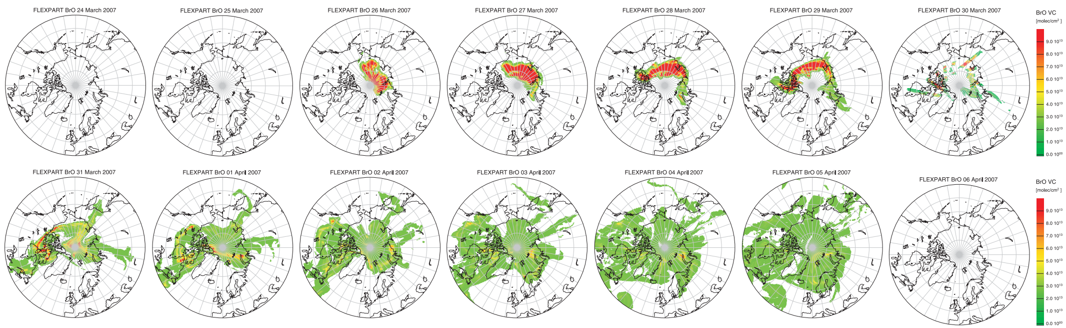

Fig. 3. Daily averaged FLEXPART model results adjusted to GOME-2 measurement data.

ACPD

9, 20407–20428, 2009

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
◀
▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

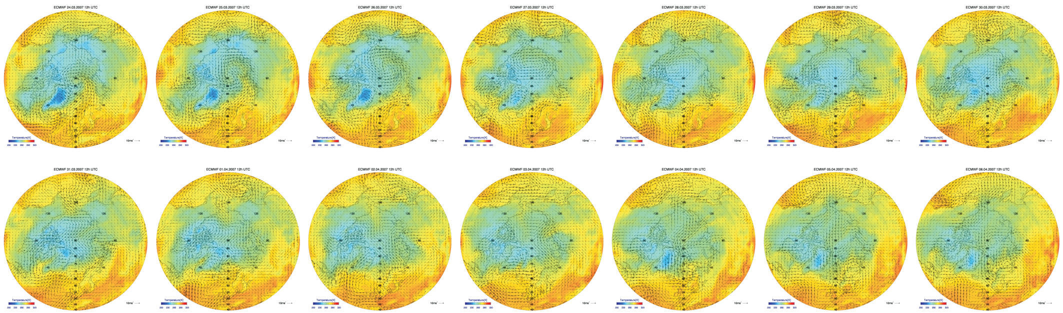

Fig. 4. ECMWF data, showing surface temperatures and wind fields during the BrO event (12:00 h UTC).

ACPD

9, 20407–20428, 2009

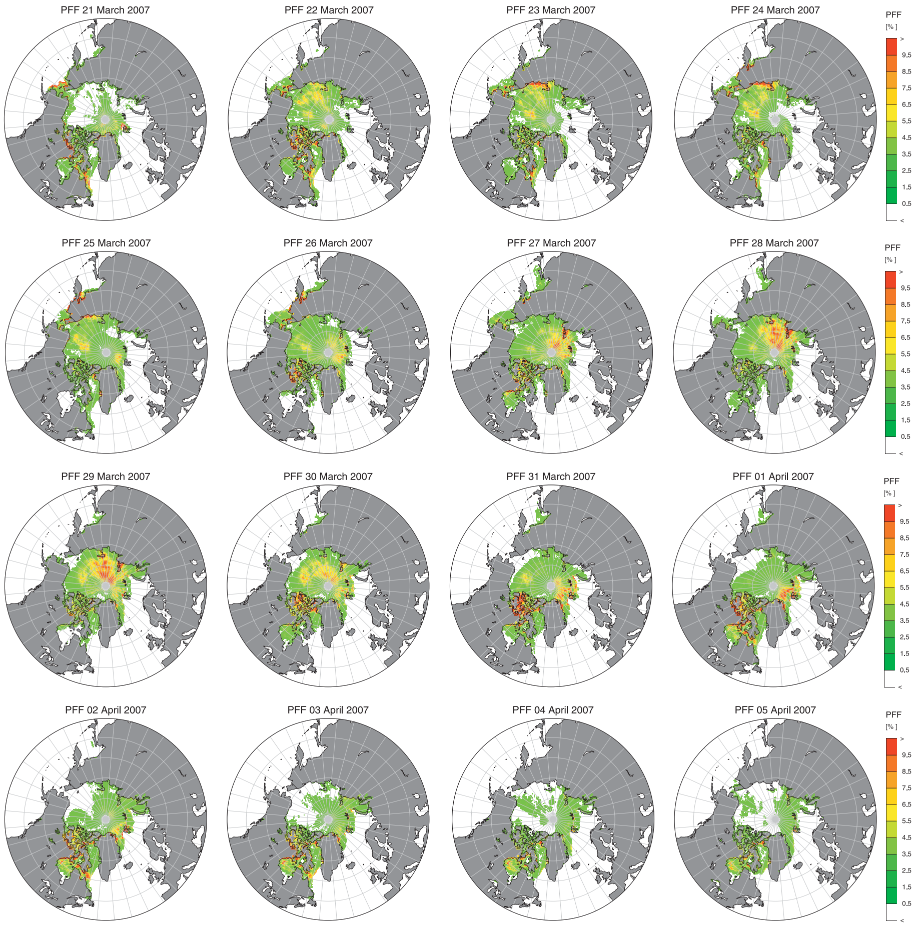

Fig. 5. Potential Frost Flowers maps showing PFF values before and during the BrO event.

ACPD
9, 20407–20428, 2009

Satellite observations of long range transport of a large BrO cloud

M. Begoin et al.