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Satellite observations of long range transport of a large BrO plume 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 to study an arctic BrO event in March 2007, which could be tracked over several days and a large area. Full BrO activation was observed within one day east of Siberia with subsequent transport to Hudson Bay. The event was linked to a cyclone with very high surface wind speeds, which could have been involved in the production and lifting of aerosols or blowing snow. Considering the short life time of BrO, transported aerosols or snow can also provide the surface for BrO recycling within the plume for several days. The evolution of the BrO plume could be reproduced by FLEXPART simulations of a passive tracer indicating that the activated air mass was transported all the way from Siberia to Hudson Bay. To localise the most probable transport height, model runs initialised in different heights have been performed showing similar transport patterns throughout the troposphere but best agreement with the measurements between the surface and 3 km. The influence of changes in tropopause height on measured BrO values has been considered, but cannot completely explain the observed high BrO values. Backward trajectories from the area of BrO initialisation show upward lifting from the surface up to 3 km and no indication for intrusion of stratospheric air. These observations are consistent with a scenario in which bromine in the air mass was activated on the surface within the cyclone, lifted upwards and transported over several thousand kilometres to Hudson Bay.
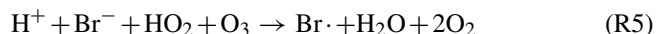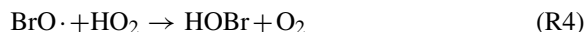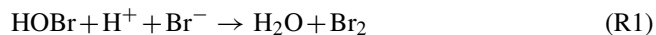
1 Introduction

Strong depletion of tropospheric ozone in the Arctic polar boundary layer in spring was first reported in the 1980s at Barrow, Alaska (Oltmans, 1981) and Alert, Canada (Bottenheim et al., 1986). Later, 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, demonstrated 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 believed 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.

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

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

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

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

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

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 young 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 ground (Hausmann and Platt, 1994; Frieß et al., 2004) and from space (Platt and Wagner, 1998; Richter et al., 1998). Ground based BrO measurements show good correlation with depleted ozone concentrations (Hönninger and Platt, 2002; Lehrer et al., 1997). In satellite measurements, enhanced BrO is mainly observed above areas covered by sea ice (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 and BrO events are often limited to a few hours, but also multi-day events can be observed. However, BrO plumes can be observed in satellite data for many days and over large areas, suggesting 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 the surface as an air mass traverses the source region, or whether the high BrO levels can be sustained in the air as it is transported away from surface sources of bromine. Large BrO columns in the satellite observations could also result from increases in stratospheric BrO column when the tropopause is unusually low. A clear anti-correlation between tropopause height and ozone columns was found in many studies (e.g. Hoinka (1996); Schubert et al. (1988)) and the same effect could also explain enhanced BrO columns in situations with a low tropopause.

In this paper, we analyse an individual large BrO event observed by the GOME-2 instrument in March 2007. The sequence of satellite measurements is used to study the spatial and temporal evolution of the event and is compared to transport simulations by the FLEXPART particle dispersion model (Stohl et al., 1998). To investigate the vertical distribution and origin of the BrO, several model runs initialised in different altitudes (boundary layer, lower troposphere, upper troposphere, lower stratosphere) were performed. A possible influence of stratospheric air masses on the high BrO values observed was investigated with data on tropopause heights and total ozone columns as well as backward trajectories.

2 Methods

2.1 GOME-2 BrO and ozone 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 9.30 h local time. With a spatial resolution of 40×80 km^2 and a scan width of 1920 km, global coverage is achieved within one day 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. BrO retrievals using GOME-2 data have already been used in previous studies, e.g. investigating BrO in a volcanic plume (Theys et al., 2009a).

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. To account for the influence of stratospheric BrO, a correction has been performed using data from the stratospheric BrO climatology developed by Theys et al. (2009b). This approach is based on a parameterisation using dynamical and chemical indicators together with output data from the 3-D chemistry transport model BASCOE (http://bascoe.oma.be). The BASCOE model includes all relevant chemical reactions in the stratosphere. Changes in BrO distribution due to stratospheric dynamics are taken

into account by a parameterisation based on measured ozone columns while NO_2 columns are used to deal with photochemical changes in BrO/Br_y ratios. The climatology has been optimised for bromine chemistry and budget and validated through comparisons using ground-based, balloon-borne and satellite limb (SCIAMACHY) stratospheric BrO observations. Compared to the changes in observed BrO values around 1.5×10^{14} molec/cm^2, stratospheric BrO climatology data only show small variations between 2.5–4.5×10^{13} molec/cm^2. The error budget of the stratospheric columns is estimated to be about 15%. As it is based on climatological relations, in cases of unusually low tropopause height climatology data could slightly underestimate the stratospheric column.

For the determination of BrO tropospheric vertical columns (VC), different Air Mass Factors (AMF) were calculated, assuming all BrO to be well mixed in a boundary layer of 400 m or located in a 1 km thick layer at 2/4/6/8/10 km, respectively. A surface albedo of 0.9 was used for the radiative transfer calculations as the measurements are performed over ice.

Clouds can interfere with the retrieval of BrO by shielding lower tropospheric BrO from view below a thick cloud and by enhancing the sensitivity to BrO above and within a cloud, in particular for optically thin clouds. The shielding effect is much less pronounced over snow and ice than over dark surfaces and up to cloud optical thickness of 20, the sensitivity of the measurements to boundary layer BrO might actually increase in the presence of clouds. In this study, GOME-2 BrO data have not been corrected for clouds as detailed cloud information over the polar region is not available at the time and location of measurements. This introduces some uncertainty and should be improved upon in future studies.

In this study we also use total ozone retrieved from GOME-2 data using the WFDOAS (Weighting Function DOAS) algorithm (Coldewey-Egbers et al., 2005; Weber et al., 2007) to investigate the possibility of stratospheric influences on the retrieved BrO columns (Section 4.2).

2.2 FLEXPART model simulations

FLEXPART is a Lagrangian 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 plumes. The model was driven by operational meteorological analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) with a horizontal resolution of 1 degree, a vertical resolution of 91 pressure levels and a temporal resolution of 3 h. The model output has a resolution of 0.5 degree and a temporal resolution of 1 hour. FLEXPART was initialised with the observed distribution of BrO at a particular time in different altitudes, and thereafter BrO was treated as a passive tracer that was tracked for about two weeks.

3 Results

The BrO event investigated here started on 25 March in Laptev and East Siberian Sea and reached its maximum on 26 March with highest tropospheric BrO values around 1.5×10^{14} molec/cm^2, if all BrO is assumed to be located and well mixed in the boundary layer below 400 m (Fig. 1). The days before, BrO values were significantly lower over the Arctic Ocean sea ice. Observations with the GOME-2 instrument indicate a fast transport of the fully developed BrO plume from 26 March over the following 3 days towards Hudson Bay. 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 and low temperatures (Fig. 2) were associated with this cyclone. High BrO concentrations were first observed especially in the southern part of the cyclone north of Laptev Sea where surface wind speeds were highest. This suggests that these high wind speeds could be involved in the initial release of BrO but higher solar insolation at lower latitudes could also contribute. 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 plume 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.

3.2 FLEXPART model simulations

To localise the most likely initial BrO source in time and space, all single satellite orbits have been analysed in detail. Owing to its large swath, GOME-2 provides several overpasses over each location at high latitudes per day 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 initialisation, one for the first event and two consecutive orbits which have to be combined to completely cover the second source region (Fig. 3). To extract the most probable source region for the BrO events, a threshold was applied to the tropospheric BrO fields and only values above 5.0×10^{13} molec/cm^2 BrO VC were used. The exact time of BrO initialisation cannot be determined
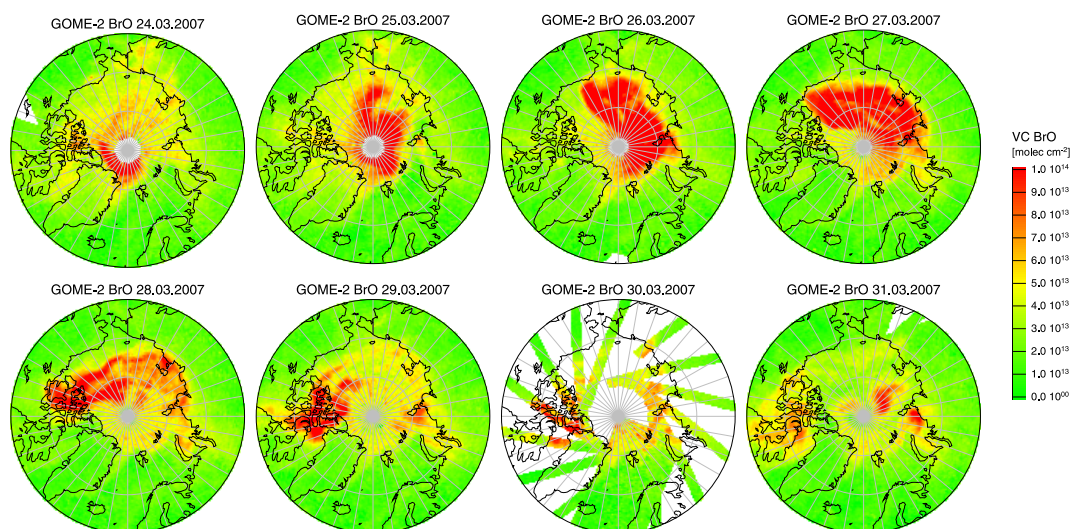
Fig. 1. Daily averaged vertical column of GOME-2 tropospheric BrO measurements corrected by stratospheric climatology data. For the radiative transfer calculations, BrO was assumed to reside in the lowest 400 m. Enhanced values on 25 March in East Siberian Sea result from the last orbit of that day at 22:44 UTC. The following orbit on 26 March at 00:26 UTC (Fig. 3) was used as one input for the FLEXPART model run. On 30 March GOME-2 was operated in narrow swath mode.
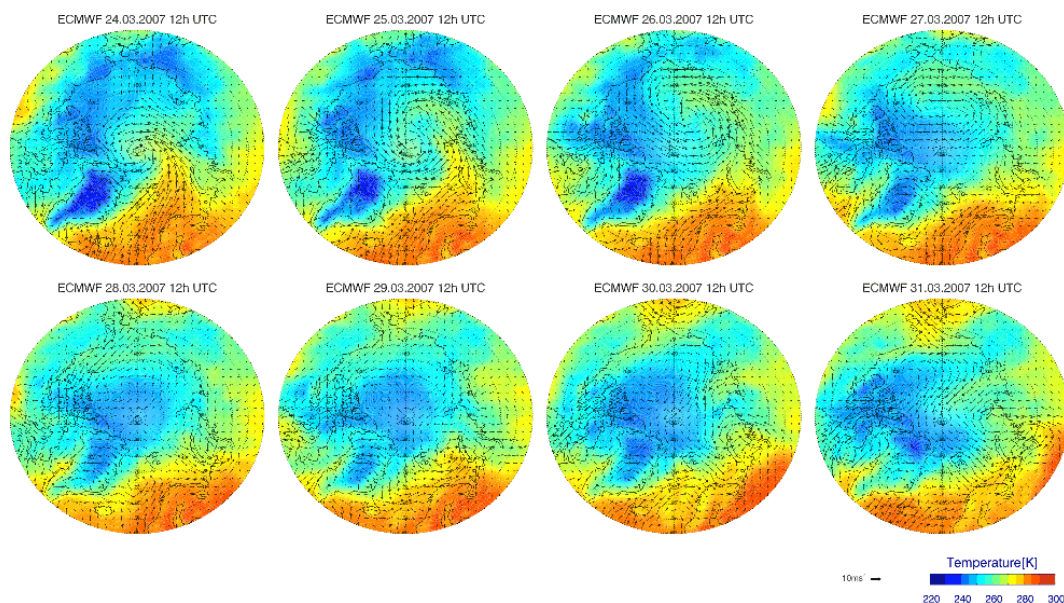

Fig. 2. ECMWF data, showing surface temperatures and wind fields during the BrO event (12:00 h UTC).

from the satellite measurements. The process must have started some time between the measurement on 25 of March, when BrO was still low and the highest observations on 26 March. At that time, the BrO plume had 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 $0.5° \times 0.5°$ and FLEXPART was initialised with the observed

concentrations in these grid cells at the time of observation. Because the exact distribution of BrO within the atmosphere is not known, several FLEXPART model runs have been initialised in different altitudes to get information on the potential origin of the observed BrO enhanced air masses. For these model runs all tropospheric BrO is assumed to be located within different layers of the atmosphere (0–1km/1–3km/3–5km/5–7km/7–9km/9–11km) and in the comparison to GOME-2 data, a corresponding AMF for the BrO amount
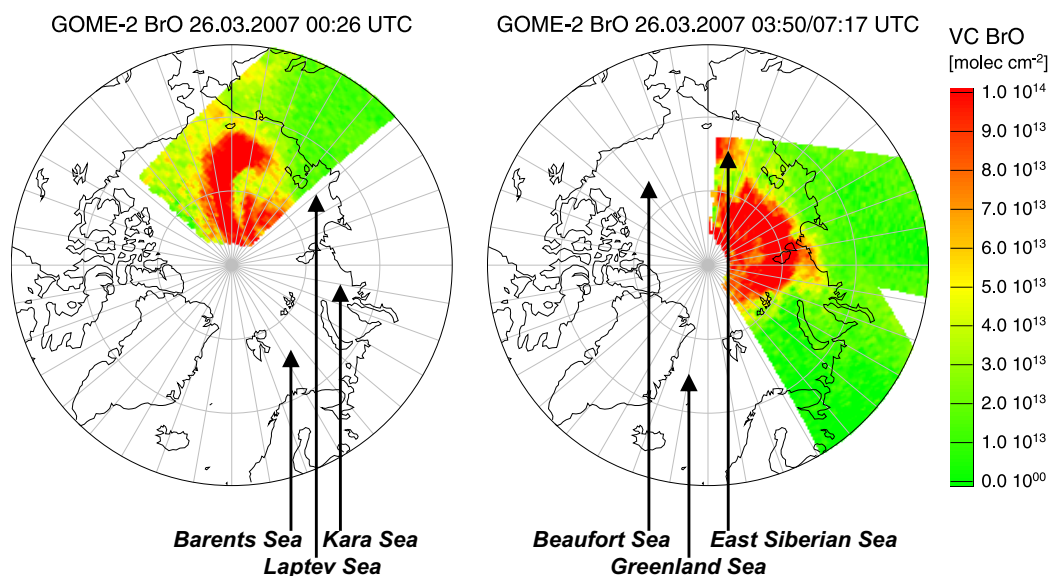
Fig. 3. Above figures show three satellite orbits, which have been used as input to initialise the FLEXPART model calculations. For the second source region, two orbits have been combined to cover the complete BrO plume.

was used. Subsequently, the BrO was treated as a passive tracer that was transported by the ECMWF resolved winds as well as turbulence and convection parameterisations.

To facilitate comparison with the satellite measurements, the 1h 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 corresponding GOME-2 orbit. Finally, the preprocessed FLEXPART data were combined in a single composite to get daily averaged data comparable to GOME-2 satellite measurements. This was done for all FLEXPART model runs. Due to the mentioned clear separation in time and space of the two observed BrO events, the model results were added to create the final FLEXPART average to be compared to GOME-2 satellite data.

As with the GOME-2 measurements, all daily averaged FLEXPART results below 9 km show rapid transport of the passive BrO tracer towards Hudson Bay within three days (Fig. 4). Above this altitude, the transport pattern shows substantial deviations from GOME-2 measurements and no transport towards Hudson Bay. Further, runs below 3 km show enhanced BrO values in the Laptev Sea on 29 March, which can't be seen in GOME-2 data. Above 3 km, model runs show export of BrO to low latitudes into Bering Sea on 26 March and transport of BrO from Hudson Bay over northern Greenland into the Barents Sea on 31 March 5. Both patterns are not observed in GOME-2 measurements. In all FLEXPART simulations from 26 until 31 March, the BrO tracer remains more or less at constant height throughout transport.

4 Discussion

4.1 Comparison of GOME-2 BrO with FLEXPART model data

The fast transport of BrO towards Hudson Bay observed in GOME-2 data can be seen as well in all FLEXPART model runs below 9 km altitude (Fig. 4). The transport patterns look quite similar but show differences in absolute values and some transport features are not seen in every model run (Transport into Bering Sea on 26 March/Higher BrO in Laptev Sea on 29 March/Transport over Northern Greenland into Barents Sea on 31 March (Fig. 5)). Nevertheless, the troposphere and lower stratosphere show similar dynamics during this special event. Above 9 km FLEXPART model runs show larger deviations from GOME-2 measurements, but below no specific altitude is clearly favoured by the model results within the first days of the event. Analysis of the daily linear correlation coefficient between measured and modelled BrO columns north of 60°N shows that the correlation between both remains high for several days and that best agreement is found for a FLEXPART initialisation between 1 and 3 km and 0–1 km (Fig. 6). This lower tropospheric location of the BrO is further confirmed by the rapid transport of BrO over Northern Greenland into the Barents Sea predicted by all FLEXPART runs initialised above 3 km altitude which is not seen in GOME-2 measurements.

4.2 Possible influence of stratospheric air

Strong changes in tropopause height can have an influence on the satellite measured BrO values as they impact on the stratospheric BrO column. As the tropopause descends and
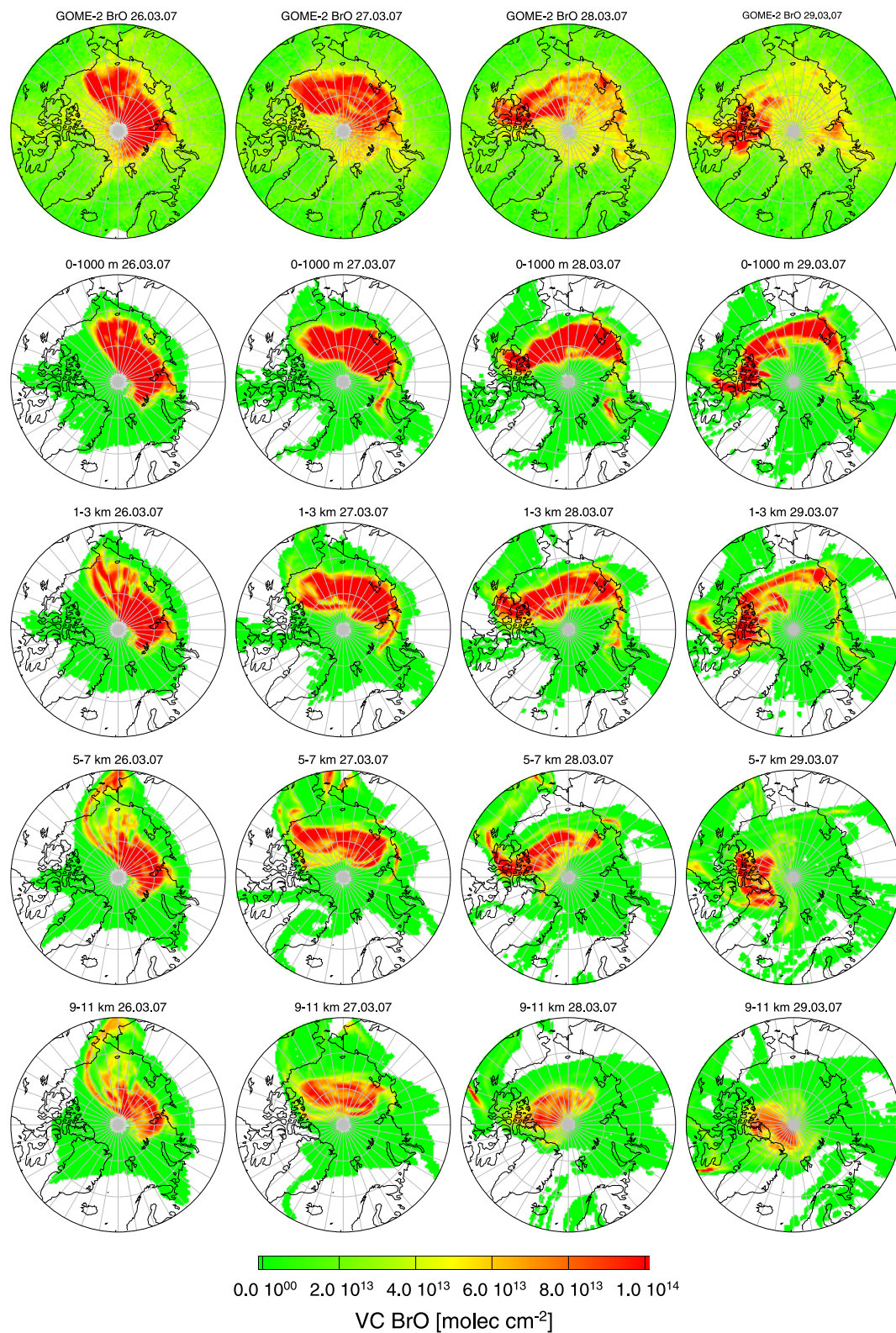
Fig. 4. Daily averaged FLEXPART model results adjusted to GOME-2 measurement data (26–29 March 2007). On top stratospheric corrected GOME-2 measurement data using boundary layer AMF (400 m) are shown.
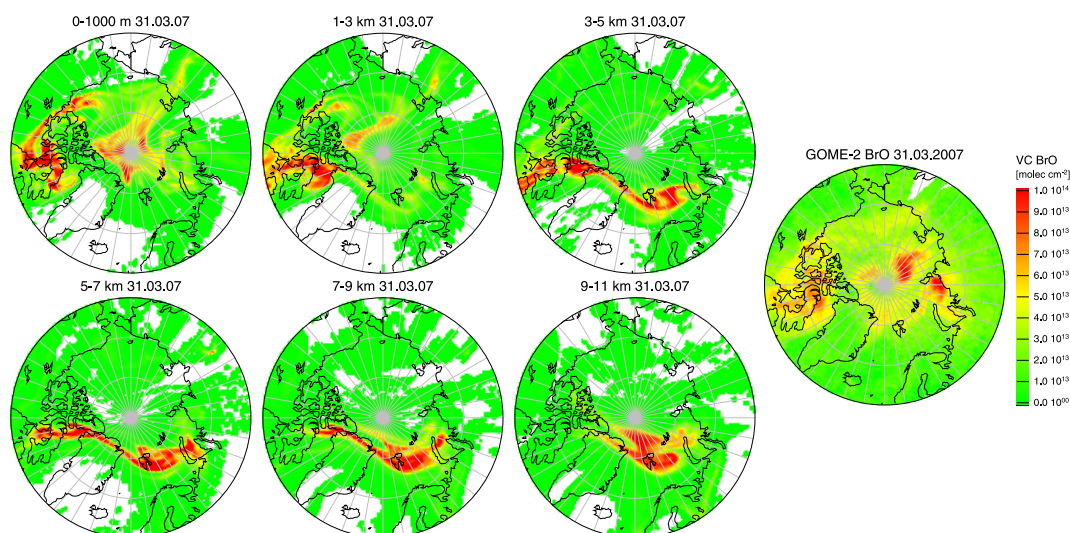
Fig. 5. FLEXPART model results for different initialisation heights on 31 March compared to GOME-2 measurments. Model runs above 3 km show transport over Greenland not seen in GOME-2 BrO
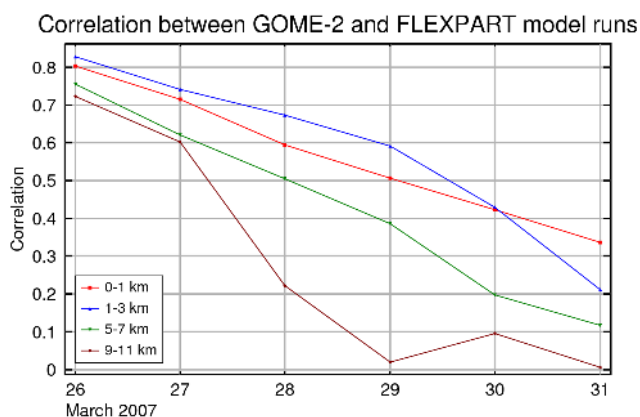

Fig. 6. Spatial correlation between GOME-2 BrO data and FLEX-PART model runs compared for different initialisation heights. All data North of 60° was used.

BrO rich air sinks to lower levels, the stratospheric BrO column increases. If not accounted for, this can lead to significant overestimation of tropospheric BrO values. As is illustrated in Fig. 7, the BrO event was at least partly linked to low tropopause heights so this effect needs to be considered. Assuming a simplified initial stratospheric BrO profile with a linear increase from 10 to 20 km (VMR: 0-15 ppt) and constant values above, a descent of stratospheric air by 5 km would lead to an increase of the BrO column by 85% or 3×10^{13} molec/cm^2 if this BrO is not converted into bromine reservoirs. While this is a significant effect, it could still not completely explain the observed high BrO columns around 1.5×10^{14} molec/cm^2. It should also be pointed out that the stratospheric BrO climatology applied will correct a large part of the tropopause change related to BrO increase through

the GOME-2 measured ozone column. However, as shown in Fig. 7, ozone columns were not particularly high during the event and therefore stratospheric BrO in the climatology does not show enhanced stratospheric values in this area.

The link between tropopause height, BrO, and ozone is further investigated by scatter plots of these variables for all data above 60°N during the first day of the BrO event (Fig. 8). Tropopause heights have been extracted from ECMWF data (horizontal resolution 0.5 degree/temporal resolution 6 h) using a potential vorticity surface of 2 PVU (courtesey of Andreas Dörnbrack, DLR) and have been allocated to corresponding GOME-2 orbits in the same way as FLEXPART model results. Ozone columns show the expected correlation with tropopause height, the largest values being observed at low tropopause height. The link is not as clear as in mid-latitude studies, perhaps a result of the complicating impact of chemical ozone depletion in the springtime ozone fields. Comparison of tropopause height and BrO columns shows, that enhanced BrO values above 1.0×10^{14} are always associated with lower tropopause heights between 5–9 km. This suggests an influence of tropopause height on BrO values. On the other hand, low tropopause is by no means always correlated with enhanced BrO values which can also be seen in Fig. 7 where many regions of low tropopause do not have corresponding high BrO columns. Therefore, low tropopause alone cannot be the explanation for the high BrO.

There are several possible explanations for the observed complex link between BrO and tropopause height:

1. The climatology data could possibly predict too low stratospheric BrO columns for these special weather conditions, leading to an overestimation of tropospheric
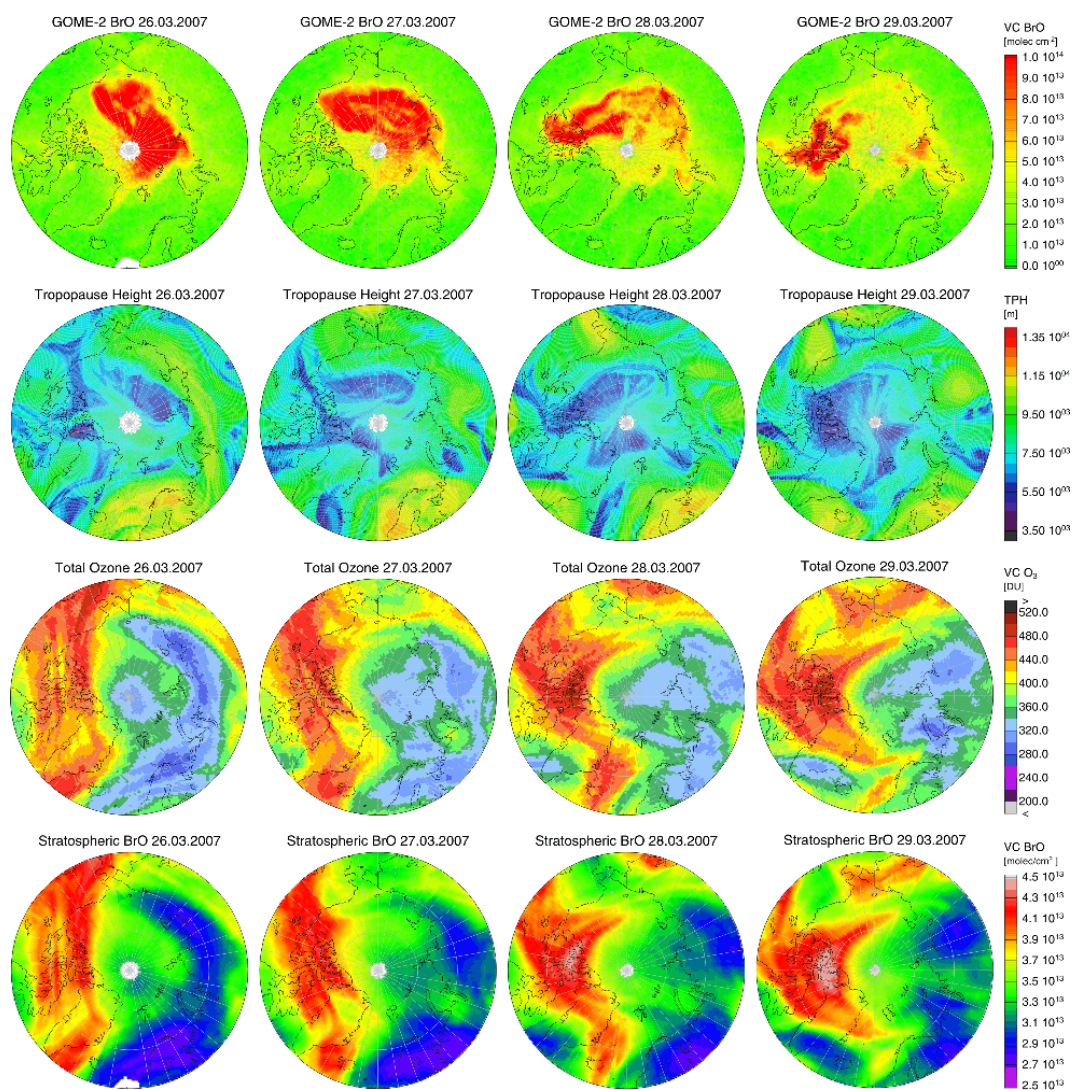
Fig. 7. Stratosphere corrected GOME-2 BrO (AMF 2000 m) in comparison to Total Ozone(GOME-2), Tropopause Height (ECMWF data adjusted to GOME-2 orbits-courtesey of Andreas Dörnbrack, DLR) and stratosperic BrO climatology data.
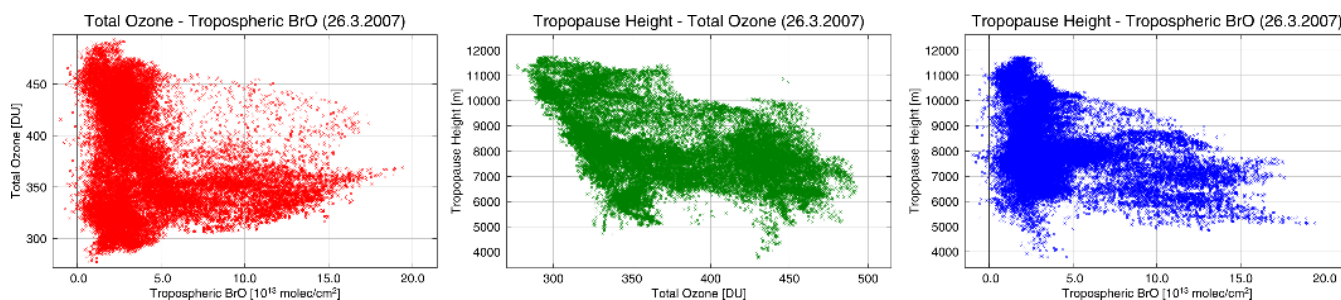

Fig. 8. Scatter plots of total ozone, tropopause height and tropospheric BrO from GOME-2
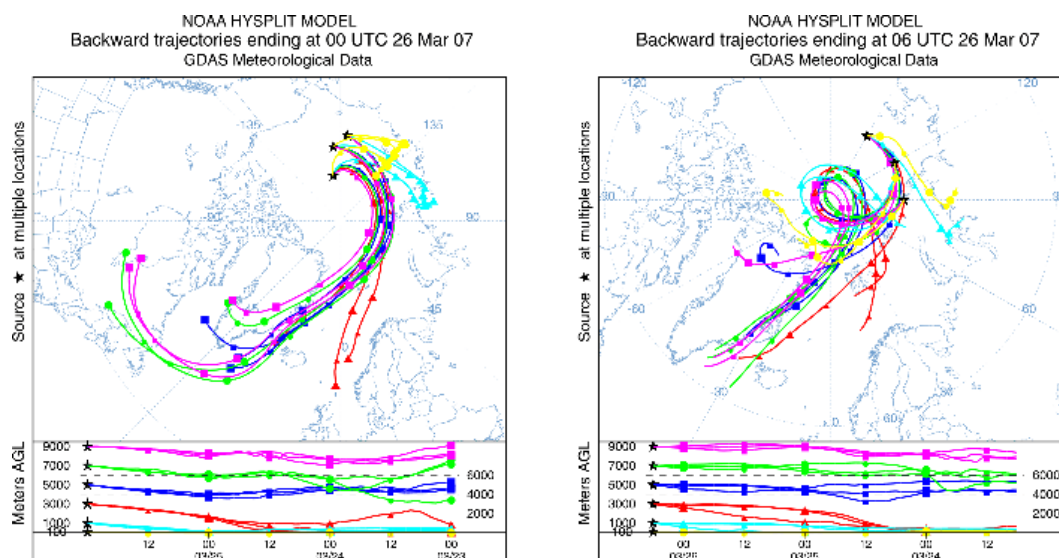
Fig. 9. HYSPLIT 72 h backward trajectories started within the FLEXPART initialisation plumes.

BrO in the presence of low tropopause heights. This effect should not be larger than 1.0×10^{13} molec/cm^2.

2. High BrO columns could depend on a different process which is linked to low tropopause heights. In the case discussed here, this could be the strong low pressure system which could have activated the BrO at the surface and lifted it up and at the same time have lead to a low tropopause height. This would imply that a low tropopause is not the reason for the BrO enhancement but just a by-product of the storm.

3. A low tropopause height in combination with the low pressure system could have lead to a stratospheric intrusion inserting BrO rich stratospheric air into the troposphere.

The third possibility is investigated by analysing backward trajectories from the region of BrO initialisation. If a stratospheric intrusion were responsible for the observed BrO enhancement, a strong downward motion should be apparent in the trajectories bringing stratospheric BrO into the lower troposphere. However, as shown in Fig. 9, backward trajectories calculated with the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph, 2005) initialised on 26 March within the two source regions (Fig. 3) do not indicate an intrusion. On the contrary, they show upward transport of surface air to altitudes of up to 3 km, in agreement with the results of the FLEXPART comparison which shows best agreement with GOME-2 data for BrO mixed up to this altitude.

Based on the results of the FLEXPART simulations, the correlations between BrO, ozone and tropopause height and the backward trajectories from the initialisation area we conclude that the most probable explanation of the observed BrO

plume is activation of BrO or precursors on the surface at very high wind speeds, upward transport to 3 km within the low pressure system and subsequent transport of the BrO rich air mass towards Hudson Bay in an altitude between the surface and 3 km.

4.3 Long range transport and life time of BrO

Due to their 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. According to the FLEXPART analysis, BrO has been transported in the lower troposphere at altitudes up to 3 km and therefore was only partly in contact with potential source regions at the surface after being initialised from sea salt on 25 March. 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 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 importance of transport processes for ozone depletion events and possibly also bromine compounds released in bromine explosions. Nevertheless ozone depleted air masses could also be transported over long distances after in situ ozone destruction, due to the lack of ozone production processes in the polar boundary layer.

In the potential BrO source region, high wind speeds dominated during the release process as well as during the

complete transport event. High wind speeds and blowing snow prevalent during an ODE were also observed in the Antarctic at Halley station by Jones et al. (2009) and could be linked to a BrO event observed by SCIAMACHY in the Weddell Sea. That case study suggests that high wind speeds and possibly also blowing snow (Yang et al., 2008) can provide a mechanism for the release and recycling of BrO in addition to the more classical ozone depletion events which are linked to low wind speed and a shallow boundary layer. Additionally high wind speeds are also accompanied by the formation of leads and polynyas on which young sea ice with a high salinity can form. The movement of the BrO plume studied here agrees with the pattern predicted by FLEXPART model results for a passive tracer in heights up to 3 km. For this reason, highly efficient chemical reactions have to be assumed to continually recycle BrO, independent from altitude. For these reactions, three possibilities exist:

1. Heterogeneous reactions on particles transported with the air mass, for example aerosols, blown snow or sea salt aerosols from frost flowers. The particles should be slowly removed from the atmosphere within a few days, but a 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 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 below 3 km 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 within the air mass is more important than surface reactions. This is also supported by backward trajectory calculations, which indicate an upward transport from the potential source region into higher altitudes up to 3 km at the beginning of the event.

In a recent study, Jones et al. (2010) investigated historical data sets of ozone profiles in the Antarctic and found

evidence of elevated layers of low ozone linked to low pressure systems. The Arctic BrO event discussed here appears to follow a similar pattern with initialisation by high winds in a low pressure system, upward movement and subsequent transport.

5 Summary and conclusions

In March 2007 a large BrO event was observed using GOME-2 data over many days in the Arctic, reaching maximum tropospheric column values around 1.5×10^{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 few days moved several thousand kilometers towards Hudson Bay. The initialisation 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 initialised in different altitudes with the BrO observations from the first day, the satellite BrO values from subsequent days could be explained by transport processes between the surface and 3 km. While initialisation in this altitude range provides the best agreement between model and measurements, vertical wind shear was small and the difference to runs initialised in the surface layer and between 5 and 7 km altitude is small.

Comparisons of stratospheric corrected BrO columns with total ozone and tropopause heights as well as backward trajectories from the area of initialisation indicate, that in this special event, BrO or precursors were probably initialised at the surface during high wind speeds, mixed upwards in the low pressure system and subsequently transported horizontally. In spite of the low tropopause heights present during the event, no indication was found for a stratospheric intrusion bringing BrO rich stratospheric air into the troposphere. However, backward trajectories indicate upward movement just before the start of the event. The effect of the increase in stratospheric BrO column through the low tropopause is thought to be corrected by the stratospheric BrO climatology used. Some remaining impact can not be excluded, but is believed to be small. The magnitude of the observed BrO column cannot be explained by tropopause height changes alone.

Due to the short lifetime of BrO, highly efficient recycling processes should be involved within the transported air masses to sustain the observed high BrO levels over this long period, no matter in which height interval the transport takes place. The combination of high wind speeds and rapid movement of the BrO plume suggests that recycling of BrO took place within the air mass, e.g. on aerosols and/or snowflakes 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 a combination of an initial release with subsequent transport

of BrO. To identify the recycling mechanism and probable source regions of BrO targeted in situ aircraft investigations of BrO events are needed.

The magnitude of the BrO event discussed here both in size and BrO columns and its link to a strong low pressure system underline the importance of dynamic BrO events. How relevant this type of BrO activation is in comparison to other BrO events at stable conditions needs to be investigated in more detail.

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. An efficient recycling of BrO over several days also indicates long-range transport of sea-salt aerosols in the cyclone from East Siberia to Hudson Bay. This transport is also important for the analysis of in situ measurements and the interpretation of ice cores taken far away from sea salt sources.

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