Reviewer 1

The authors present an analysis of satellite BrO data, time series of surface ozone and air temperature measured at three Arctic locations (Barrow, Alert, and Ny-Alesund), and aircraft BrO and ozone sonde measurements during the ARCTAS and ARCPAC campaigns. These measurements are very heterogeneous regarding their temporal and spatial coverage. Nevertheless, the authors use correlation between the different measurements in an attempt to characterize conditions and properties of springtime ozone depletion events in the Arctic. Any analysis of correlations between different observed species must be done with great care. The authors present some new ways of performing such correlation analysis, which may be helpful to better understand some of the processes relating the activation of reactive bromine and the depletion of ozone in the springtime boundary layer of the Arctic. Therefore, this subject is well suited for publication in ACP. However, several major issues remain before the manuscript will be in a publishable form. One major issue concerns the determination of tropospheric BrO columns, which recently became the issue of a scientific debate. Not being an expert in the field, I feel quite uncomfortable with the way this is handled in the manuscript. In fact, the manuscript makes this issue even more confusing instead of clarifying it. In addition, the authors draw several conclusions from their analysis, which in my opinion are not warranted and which also neglect results from previous publications. In contrast, I believe that some issues related to the processes during the depletion of ozone may be better addressed with the available data set. Further details and some suggestions are given in the major comments below.

We thank the reviewer for the detailed comments. We will provide quick responses to facilitate further discussion in the review process. The time and effort spent by the reviewer on this paper are greatly appreciated.

A major issue raised by the reviewer is how we presented the satellite BrO products. Ideally we would want to have one "standard" tropospheric BrO productions from OMI or GOME2 measurements and carry out our analysis related to ODEs. However, there is not such a product. In the paper, we discussed the reasons in sections 2.2.1 and 4. The next question is if the satellite measurements can be applied judiciously to understand ODE process. We stated in section 4 "Considering the observations available from the ARCTAS and ARCPAC experiments, the current quantitative constraints on the magnitudes of tropospheric (or stratospheric) column BrO are poor. However, judicious use of correlation analysis provides useful scientific insights into the processes of bromine-related ODEs as we have shown in this study."

We feel that it is important to acknowledge the various problems related to the tropospheric column BrO products, even though it makes our analysis more difficult to carry out (and adds a large supplement section in this paper). In describing tropospheric BrO vertical columns in section 2.2.1, we discussed the rationale and approach in this study to use the satellite products. We agree that many readers may not necessarily want all the information we provided in the paper. Therefore, we included most figures related to the different satellite BrO products in the supplement. A general reader can easily skip the supplement (and section 4) if he/she does not need that information.

This is a paper of data analysis. There are many aspects of the measurement data that we will not be able to explore in one paper. The main goal of this paper is to understand better the characteristics of ODEs. We did not attempt to write a paper on how to best derive tropospheric column BrO from OMI or GOME2. Therefore, the problems of the satellite data are acknowledged and discussed, but we did not have all the solutions.

Major comments:
The authors calculated six different versions of the tropospheric BrO columns using two different satellite products (OMI, GOME2) and three different stratospheric BrO columns (called 20th, SCIA2ND, RAQMS). I assume that these multiple calculations were caused by the ongoing scientific debate of how to extract tropospheric BrO columns from the satellite products that deliver only total BrO columns. I further guess that these multiple calculations were performed to avoid getting dragged into this debate and to demonstrate that the determination of the tropospheric BrO column is not a critical issue. However, the presentation of the results is rather confusing to me. I admit that I am neither an expert in the field of satellite data retrieval nor in stratospheric BrO. The brief presentation of the RAQMS approach (stratospheric columns are too low and are scaled using zonal mean values from the 20th method) and the neglect of the RAQMS products in the further data analysis lead me to the conclusion that the RAQMS stratospheric BrO columns are almost useless. If that is the case, why are they presented at all? A quick search on publications regarding the comparison of BrO columns from OMI and GOME2 delivered no real result, but rather some statements in the internet that they agree quite well. Accordingly, the authors present in Fig.2 the monthly average for April 2008 of the tropospheric BrO distribution from GOME2-SCIA2ND, which in fact compares well with the OMI-SCIA2ND shown in Fig. S5 as also stated by the authors. If however the OMI and GOME2 BrO columns agree well, what is the use in using these two different satellite products? Of course, if the total BrO columns are similar and the same stratospheric BrO columns are subtracted, they should give the same tropospheric BrO distributions and the same results in the further correlations. Interestingly, the correlation coefficients between surface ozone at Alert and Zeppelin and the tropospheric BrO shown in Fig. 3 are markedly different for GOME2-SCIA2ND and OMI-SCIA2ND. There are also almost always large differences between GOME2-20th vs. OMI-20th and GOME2-RAQMS vs. OMI-RAQMS as shown in Figs. S1 to S4. What is the explanation for this difference? Do only the monthly averages agree but not the daily maps, which are used in the correlation analysis? If the two satellite products are similar and one of the methods to obtain the stratospheric BrO columns is useless, the six possible products melt down to only two independent tropospheric BrO distributions. If the two satellite products are not similar this number increases to four, but it would require a thorough discussion how and why the OMI and GOME2 BrO columns differ.

Response
We thank the reviewer for carefully reading the paper. Comparing only monthly distributions hides the difference (sometimes very large difference) in spatial and temporal variations. We stated in section 2.2.1 “Retrievals of tropospheric BrO columns from satellite measurements are quite uncertain, particularly in the estimate of stratospheric BrO columns (e.g., Choi et al., 2012). During our analysis period, in situ BrO observations are too limited and they do not provide enough quantitative constraints

to validate satellite tropospheric BrO column products (the details will be discussed in section 4)." Without adequate validation measurement constraints, we stated "In order to take into account of the uncertainties in the estimates of stratospheric BrO vertical columns, we take the approach of using three different estimate methods." We then stated "These methods give different estimates of latitudinal/longitudinal variations in stratospheric column BrO and consequently in tropospheric column BrO. Most importantly, the estimated stratospheric BrO columns using these methods do not introduce in the resulting tropospheric BrO columns an unphysical correlation with tropospheric ozone." The OMI/GOME2-RAQMS results are shown in the Supplement to support the above statements.

The reasons for product difference could be the instrument sensitivity, retrieval algorithm, cloud interference, and the estimates of the stratospheric BrO columns. Resolving the quantitative contribution from each to the differences among different OMI and GOME2 tropospheric BrO columns will take a different paper and more validation measurements than we have. What we did in the paper is to figure out how to use the satellite data despite of the difference in the products.

In the first paragraph of chapter 3.1.2 and in Fig. 2 the authors present the monthly average for the tropospheric column of BrO for April 2008. In Fig. 2 they also show a map of first- and multi-year sea ice suggesting a relationship between the ice properties and the BrO columns. Such a relationship could be tested since satellite data of BrO are available since the 1990s (By the way, references to publications on this subject are completely missing!) and could be used to detect if trends are in line with the modified sea ice properties of the Arctic Ocean. However, such an analysis is missing and likely beyond the scope of the manuscript. Since the authors do not refer further to the relationship of sea ice properties and BrO and since it is not used for further analysis I recommend to delete this part because it remains to superficial.

Response
We agree. The FYI discussion is not essential for the analysis in this paper and we will remove the relevant discussion in the revision.

In several cases the authors discuss correlations between in situ measurements (surface ozone, ozone sondes, BrO, Br2+HOBr, soluble bromide) with tropospheric BrO columns. The in situ aircraft measurements are averaged into altitude bins. Nevertheless, the in situ observations and the columns are completely different quantities and can only be compared under certain assumptions. In the case of BrO, in situ concentrations and tropospheric column should only correlate if changes in the column are related to changes in the specific altitude bin. On the other hand, high concentrations in a certain altitude range could show up in the in situ measurements, but could be masked in the tropospheric column if at the same time the BrO in a different altitude range decreases by the same (or larger) amount. Therefore, calculating correlations between in situ BrO and tropospheric BrO columns implies strong hypotheses about the tropospheric BrO distribution with high concentration close to the ground and low and constant BrO concentrations throughout the troposphere. In fact this assumption

is probably well justified for Arctic springtime conditions, but they should be discussed in detail in the manuscript before presenting the correlations.

Response
The good correlations would indicate that the variations are driven primarily by lower-tropospheric BrO. It doesn't necessarily indicate that BrO in the free tropospheric is insignificant.

The correlations are empirical evidence based on measurements. They are not based on any a priori assumption we placed. In writing the paper, we consciously avoided discussing the implication of the correlations on BrO distributions since we cannot quantify its distributions with correlation analysis alone.

In the case of Br2+HOBr and soluble bromide the authors claim to test if the observed quantities "capture to some extent the distribution of lower tropospheric BrO" (page 8). But why should these compounds correlate (besides the implied assumption about the vertical distribution like in the case of BrO)? According to the chemical mechanism the BrO maximum does not occur simultaneously with concentrations of HOBr, Br2, or HBr (included in soluble bromide) (e.g. Lehrer et al., 2004). So, the absence of a significant correlation makes the result ambiguous: the mechanism and the BrO columns are both correct or they are both incorrect. I recommend to delete this part.

Response
We stated in section 4 "…Without additional BrO measurements, a true validation study based on in situ BrO measurements is therefore infeasible." and in the next paragraph "One approach is to focus on correlation analysis between tropospheric column BrO and other related in situ observations.". More details are given in that section. In short, there was a good correlation between satellite-derived BrO products and DC-8 data (two flights only) and there was no correlation between the satellite-derived BrO products and NOAA P3-B data (5 flights). In contrast, "The measurements of Br_2+HOBr were reported for 7 ARCTAS fights and 5 ARCPAC flights (Neuman et al., 2010; Liao et al., 2012) and soluble bromide measurements were also available in the ARCTAS flights (Liao et al., 2012)."

These correlations were only included in the Supplement figures. We stated in section 2 "While not quantifying the uncertainties in the derived tropospheric BrO columns, the large separation of correlation coefficients does indicate that the products have different characteristics." The more subtle point is "In fact, even the values of (anti)correlations between ozone and BrO are not that important. It is the change of the (anti)correlation between ozone and BrO with time or altitude that provides useful information on the importance of in situ chemistry relative to transport and on the vertical extent of bromine-driven ozone loss." The last statement is for BrO-ozone correlation but it applies to satellite BrO correlations with in situ measurements of other bromine species. A large portion of the correlation is driven by the absence and presence of bromine species along the flight tracks (not the absolute values of bromine species). In the Supplement, we used these data together with DC-8 in situ BrO correlations.

The most complex case is probably the comparison of in situ ozone with the BrO columns. Again, in my opinion all comparisons are based on the assumption that the relationship between ozone and BrO is restricted to the boundary layer and that other layers remain constant in ozone and BrO.

Response
Yes, we assumed that the BrO variation occurred in the same layer of ozone variation since neither satellite measurements nor our various estimates of stratospheric BrO columns are correlated with tropospheric ozone.

On page 11 and in Fig. 3 the authors describe a stronger anti-correlation for Alert and Zeppelin without a time lag or with a delay of only 1 day. For Barrow, the anti-correlations are strongest with time delays of 1 to 3 days. The authors conclude that at Alert and Zeppelin local processes are responsible, while at Barrow transport is more important for low ozone values. I believe that this interpretation is an oversimplification. I agree with the conclusion that if along the back trajectories in the last 0 to 3 days BrO columns are high, surface ozone concentrations will probably be low (or lower than background). By the way, can these results be exploited to determine how fast ozone is removed? However, there is also the possibility that air masses with low ozone concentrations persisted for longer periods (i.e. longer than the backward trajectories), which were than transported to the observational sites. In fact, Bottenheim et al. (2010) and Jacobi et al. (2010) presented surface ozone time series form the Arctic Ocean indicating periods of longer than 5 days with low ozone concentrations. Thus, it could well be possible that air masses depleted in ozone were transported to the coastal station without enhanced BrO concentration along 5-day back trajectories. Such events have the potential to mask correlations between ozone and BrO. I assume that with the available data set these processes can probably be tested in more detail.

Response
If the transport took longer than 5 days, we would not capture it in the analysis. However, the selected cases by Jacobi et al. (2010) are ODEs that lasted several days. One possibility is that we did not observe ODEs that lasted over an extended period of time (see Figure 1) unlike the TARA observations shown by Bottenheim et al. (2009) and the 1994 and 2003 cases selected by Jacobi et al. (2010). If > 5 day of low O_3 transport was involved in the presence of a larger area of stable boundary layer, one would expect to find ODEs lasting several days. Like the surface observations, the ODEs in ozonesonde and aircraft observations in this study are not near-0 ozone cases either. The fact that we did not find this type of long-range transport cases does not rule out its existence. We will modify the conclusion section to include the discussion of how our results relate to these two previous studies. Since these two previous studies did not have a proxy for BrO concentrations, the method we developed in this study could be used to quantify the time lag between the occurrence of ODE and the time of the ODE observation.

Third paragraph on page 11: In characterizing the different source regions of ozonepoor air the authors refer to Fig. 2 and describe the Chukchi Sea, "where tropospheric BrO columns are high", the Beaufort Sea, "where we did not find the enhancement of

*tropospheric BrO", and the area northwest of the Zeppelin Station, "where there is moderate increase of tropospheric BrO". By looking at Fig. 2 I would say that average concentrations over the Chukchi Sea, the Beaufort Sea, and in the area northwest of the Zeppelin Station were approximately 6*10^13, 5*10^13, and 5*10^13 molecules/cm2. Therefore, the Beaufort Sea can not be considered as a region with low BrO columns. This needs to be corrected affecting also some of the conclusions.*

Response
We agree. We will modify the paragraph to only compare the BrO columns over the two back trajectory source regions.

Chapter 3.1.3: The entire discussion of the relationship between the temperature and the occurrence of low ozone is not at the current state of knowledge. Several previous publications have shown that low ozone has been observed at temperatures above -20 _C. Some authors also have suggested that the relationship observed between temperature and ozone in many cases is not related to the chemical mechanisms, but rather to the meteorological conditions (e.g. Jacobi et al., 2010). Moreover, if the authors conclude that for Barrow low ozone is rather related to transport processes than to local chemistry, what does the temperature measured at Barrow as used in Fig. S9 tell about the conditions during the ozone depletion? I still believe that lower temperature favor the halogen activation making the chemical depletion of ozone more efficient. However, data to prove this effect is difficult to obtain. Maybe the authors can again find information on this using their aircraft data?

Response
The temperature-ozone correlation analysis is based on data only. We had no a priori assumption on the mechanisms. It is true that the Barrow site could be strongly affected by short-range transport. But low temperature and low ozone in the air mass can both be transported to this site, especially since the boundary layer is stable during ODEs at Barrow (please see Figure 7). We don't feel that it is a problem.

If we inadvertently misreported previous research results in section 3.1.3, we would be very happy to make the corrections. In the last paragraph of the section, we discussed some possible mechanisms. We will add the analysis results from Bottenheim et al. (2009) and Jacobi et al. (2010) that there was not a temperature threshold for ODEs, which are consistent with our results. In our analysis, we cannot tell if the process is driven by chemistry or transport and did not attempt to extrapolate our results. We stated in the conclusions "There was a significant correlation between ozone and temperature during ODEs, although we did not find evidence for a threshold temperature value, which implies that temperature variation is a stronger factor for ODE formation." The temperature variation could be a reflection of the pressure change discussed by Jacobi et al. (2010), which we will add in section 3.1.3. However, it is not obvious from the data presented in this study that mesoscale transport is the sole driving force for ODEs we analyzed, given the obvious anticorrleation between BrO column and ozone in a time delay of < 3 days. We will add this discussion in section 3.1.3.

We will address the minor problems in the revision of the paper. We hope to hear your thoughts on our responses so that we may resolve the major issues before the end of the discussion period. It is a complex paper and would take more time than usual to review. We thank you again for your time and effort in reviewing this paper.