Interactive comment on "Seasonal variation of tropospheric bromine monoxide over the Rann of Kutch salt marsh seen from space" by C. Hörmann et al.

Anonymous Referee #1
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We would like to thank Referee #1 for the detailed and helpful comments and suggestions he/she made to improve the quality and clarity of our manuscript.
For reference, the original comments (**black**) are always included below, followed by our response (**blue**). Modifications of the original manuscript (**green**) are indicated in **red**.

Hörmann et al. report seasonal variations of tropospheric BrO abundances over the Rann of Kutch (salt desert/seasonal lake at the border of India and Pakistan) using BrO tropospheric column retrieved using OMI UV measurement during 2005-2014. This is a first attempt to quantify tropospheric BrO over salt lakes using satellite measurements. This study agrees well with the scope of Atmospheric Chemistry and Physics. I recommend this article to be published in ACP given that the following major and minor concerns are addressed.

Major comments:

1) Total column BrO retrieval using DOAS (Differential Optical Absorption Spectroscopy) has significant uncertainty depending on selection of fitting window, up to ≈50% or higher, which may affect the magnitude of tropospheric column BrO and thus the BrO mass abundance quantified using that column. The tropospheric column BrO and BrO mass abundance is as correct as the magnitude of total column BrO retrieved in the given fitting window, and it should be mentioned in the manuscript (in Sect. 3). In addition, comparing the BrO mass abundances from authors' retrieval and those from the OMBRO operational product (using the same approach) will give a good, solid example to show possible uncertainties in mass abundances calculated from satellite BrO measurements.

   We agree with the referee that the uncertainty of the retrieved slant column density depends on the selection of the fit window as thoroughly discussed by Vogel et al. (2015). In addition, the final total (and especially the tropospheric) column density/BrO mass depends on the a-priori assumptions used for the radiative transfer calculations.
   We compared the results of our own OMI BrO retrieval at MPIC with those from the NASA OMBRO operational product in order to illustrate the differences as suggested by the referee. The calculation of tropospheric BrO VCDs was therefore applied to the BrO slant columns from the OMBRO product in the very same way as for the SCDs retrieved at MPIC. Mean BrO VCDs were calculated for April (the month with the highest BrO VCDs) and October (close to zero BrO VCDs directly after the monsoon season) for the whole time period 2005-2014. Finally, the resulting BrO VCDs over the Rann area (22.5-25.5°N, 67.5-72.5°E) were integrated along the longitudinal direction (yielding so called *line densities*) to allow an easy comparison of the results (Figure R1).

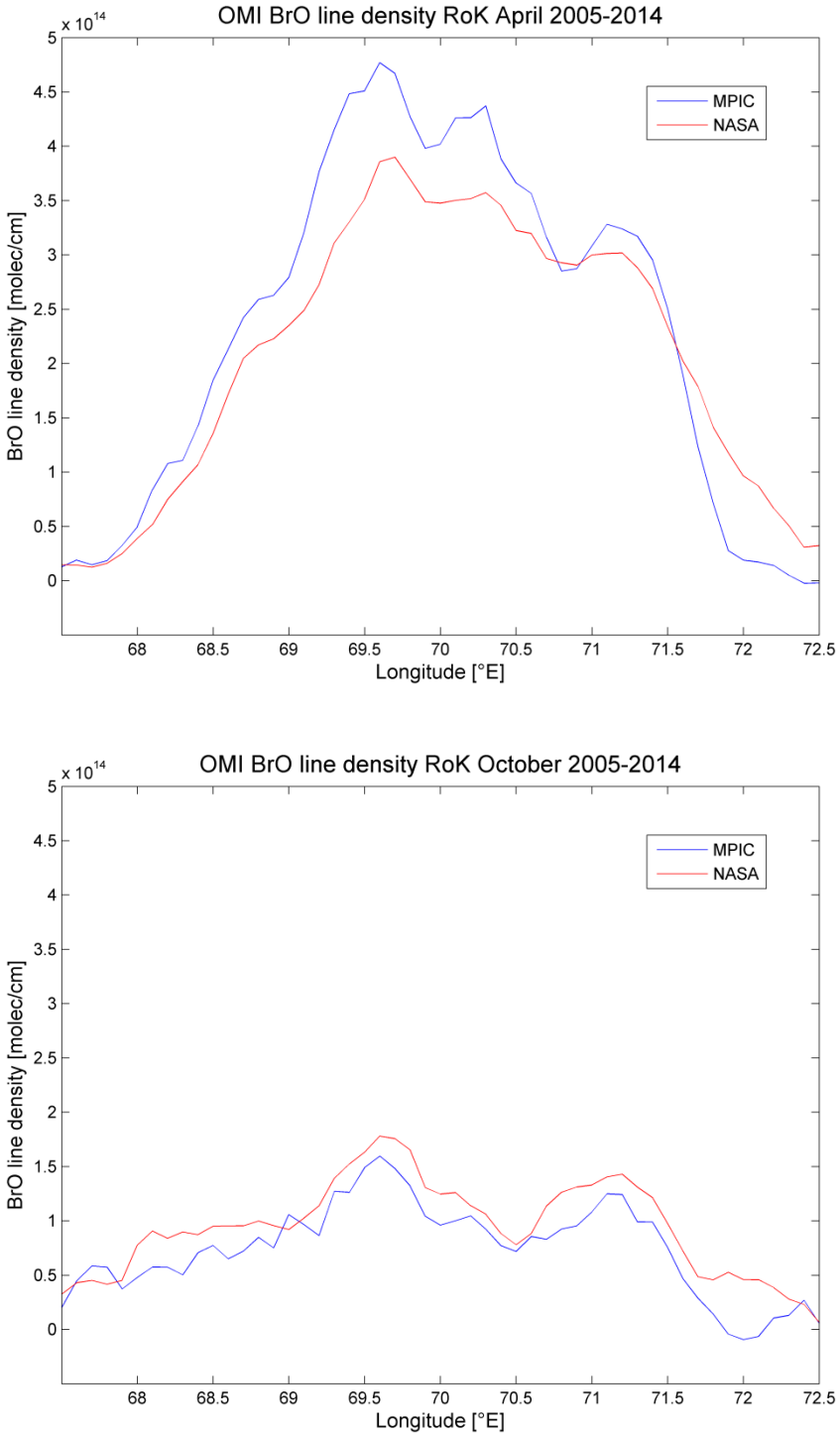

Fig. R1: Comparison of the BrO line densities from the NASA OMBRO product (red) with our (MPIC) evaluation (blue) for April (top panel) and October (bottom panel) of the 2005-2014 period.

As can be seen from Figure R1, the resulting BrO line densities for the MPIC and NASA data are very similar, showing essentially the same features and differences of typically 10%.
We added this information to the manuscript at the end of the first paragraph of the "*Results*" section:

"A detailed comparison of the BrO VCDs with those from the operational NASA OMBRO product showed only small differences of typically 10%. For this purpose, the calculation of troposphere BrO VCDs was applied to the BrO slant columns from the OMBRO product in the very same way as for the SCDs retrieved at MPIC."

Additional reference:

Vogel, L., Sihler, H., Lampel, J., Wagner, T., and Platt, U.: *Retrieval interval mapping: a tool to visualize the impact of the spectral retrieval range on differential optical absorption spectroscopy evaluations*, Atmos. Meas. Tech., 6, 275-299, doi:10.5194/amt-6-275-2013, 2013.

2) Although stratospheric column BrO has little variations at the region of study, the "magnitude" of assumed stratospheric column BrO can affect the magnitude of tropospheric column BrO and thus the magnitude of mass abundances of tropospheric BrO. Such uncertainties in the magnitude are particularly important in this study, since this study aims "quantification" of BrO mass abundances instead of merely tracking the seasonal and temporal variations. How does the estimate of stratospheric column BrO compare with the other estimates using models, such as Theys et al. 2009b and Salawitch et al., 2011? How much can the estimated tropospheric BrO abundances change along with the assumed loadings of stratospheric column BrO? These points need to be addressed in the manuscript (probably in Sect. 3.2).

We agree with the reviewer that a good estimation of the stratospheric BrO column is a prerequisite for the correct quantification of the tropospheric BrO VCD. However, as the enhanced BrO columns over the salt marsh can be expected to be tropospheric (e.g. because of the spatial correlation to the Rann surface), we applied a simple approach to remove the stratospheric BrO contribution by the empirical background correction described in Section 3.2. The advantage of this empirical correction is that it depends on few parameters for the estimation of the stratospheric BrO column and, thus, interferences are effectively avoided. Compared to the spatial extent of the Rann area, stratospheric BrO gradients over the studied area are comparatively small so that they can be modelled/fitted by a simple 2-dimensional (spatial) polynomial.

In order to make this point clearer in the manuscript, we added the following sentences to Section 3.2: "Please note that this rather simple approach can only be applied, because the stratospheric BrO gradients over the studied area are relatively small compared to the extent of the salt marsh. In general, an accurate quantification of tropospheric BrO VCDs (e.g. at high latitudes during arctic spring) needs a more sophisticated estimation of the stratospheric BrO contribution (e.g. Sihler et al., 2012)."

3) Sect 4.5: Authors used a geometrical AMF for tropospheric BrO over the Kutch of Rann using GOME-2 data. However, I do not agree with that the BrO column retrieved using nadir-viewing UV measurements and a geometrical AMF over not-so-bright surfaces (albedo ≈ 0.15) has the capability to distinguish tropospheric BrO contribution. If authors cannot prove that using a geometrical AMF has such capability or provide the GOME-2 BrO analysis using tropospheric AMF from reasonable radiative transfer calculation, the entire section need to be removed.

We agree in principle with the referee that a geometrical AMF is not well suited to quantitatively determine the BrO VCD over areas with a relatively low albedo, as a tropospheric BrO enhancement may be underestimated. However, in this subsection, the aim was to perform a qualitative comparison, since for the GOME-2 measurements, the correct calculation of a AMF is hindered by the fact that the GOME-2 cloud product is not

reliable (due to the high surface reflectivity). Also, as mentioned in the beginning of Section 4.5 of the manuscript, the GOME-2 cloud product cannot be used for sorting out measurements that were affected by clouds without losing a significant amount of measurements over the cloud free salt marsh (see also Section 3). Therefore we decided to use geometric AMFs for both satellite instruments in order to ensure a qualitative seasonal intercomparison.

To make this point more clear, we added the following sentence to Section 4.5 of the revised manuscript: "Although geometrical AMFs are not well suited to quantitatively determine the BrO VCD over the Rann, they still can be used to visualize BrO abundances exceeding the comparably smooth stratospheric background."

Minor comments:

1) Latitude and longitude of the Rann of Kutch need to be specified in the early part of the manuscript for readers who are not familiar with the area of study.

   We added coordinates at the beginning of Section 1.2 ("...about 22.5-25.5°N, 67.5-72.5°E")

2) Page 2 line 6: "an overall picture of the BrO horizontal distribution" need to be "an overall picture of BrO horizontal distribution in the troposphere" to be clear, since the mentioned satellite measurements of BrO (GOME) primarily provide the total column BrO.

   We changed the sentence to:
   "In the meantime, an overall picture of the total BrO horizontal distribution was obtained by observations from satellite instruments in the late 1990s (Wagner and Platt, 1998; Richter et al., 1998; Chance, 1998)."
   As the referee correctly mentions, the satellite measurements provided just the total column of BrO in the first place.

3) Page 2 line 8: Due to the large variations in stratospheric column BrO in high latitudes, polar tropospheric ozone depletion can be identified by satellite measurement only if stratospheric BrO loading is properly addressed, which should be mentioned here.

   We changed the sentence to read as follows: "A detailed analysis strongly indicated that the ODEs can be associated with huge tropospheric BrO 'clouds' of several thousands of km² extent, '(…) rather than by a disturbance of the stratospheric composition or a modification of the stratospheric AMF' (Wagner and Platt, 1998)."

4) Page 6 line 8: "The stratospheric BrO distribution varies little with latitude and even less with longitude (Theys et al., 2009b)." It is the case only for the low latitude regions; stratospheric column BrO has large longitudinal and latitudinal variations in middle and high latitude regions.

   We changed the sentence to read as follows: "The stratospheric BrO distribution for lower latitude regions (like the Rann of Kutch area) varies little with latitude and even less with longitude (Theys et al., 2009b)."

Sect 4.3: Choi et al. (2012) have reported the high BrO abundances associated with high planetary boundary layer height in the Arctic region, which can be a relevant reference here.

We changed the beginning of Section 4.3. as follows: "Several studies (e.g. Theys et al., 2009b, Choi et al., 2012, Sihler et al., 2012) discussed the influence of a high boundary layer height on the BrO satellite retrieval over Arctic regions. Recently, Lieb-Lappen and Obbard (2015) analysed the role of blowing snow in the activation of bromine over first-year Antarctic sea ice."