We thank both the reviewers for their helpful comments and suggestions. In the following detailed response to all reviewer comments, the Reviewer Comment is first copied using regular text in black, followed by our response using italic font in green. A copy of the text that we have changed in the manuscript is also added to facilitate a simultaneous consideration of the reviewers' comments and our replies to those comments where appropriate.

Referee #2

C2.1) Since only one case study day (9 April 2010) is presented and discussed, the data/results are for me over-interpreted in terms of both bromine content/vertical distribution for southeastern US and their impacts on Hg oxidation pathways. As Referee # 1, I wonder why the entire data set covering the May 2009-February 2011 has not been exploited in this work. Do we expect similar tropospheric BrO profiles (i.e. with only a significant content in the FT) throughout the year at this location? Is it possible to have at some period(s) of the year a more significant contribution from the marine boundary layer? If yes, what would be the impact on the bromine and Hg oxidation chemistry modelling? The authors should make clear in the text but also in the title that it is a one-day case study.

R2.1) We agree, and refer to our response to reviewer #1 (see R1.1). In general, the profile derived in this work shows some similarities to other reported profiles measured from aircraft (Volkamer et al., 2015; Wang et al., 2015; LeBreton et al., 2016), which cover large spatial gradients and probe different atmospheric conditions. While it is possible that there could exist periods where the MBL contributes more significantly to the total BrO column, several studies exist that point to a low upper limit of <0.5 pptv BrO in the MBL (Volkamer et al., 2015; Wang et al., 2015; Gomez Martin, 2013) which makes it seem unlikely that this would be the dominant scenario. If this were the case, the much warmer temperatures found in the MBL would increase the influence of the thermal decomposition of the HgBr adduct and the increased availability of additional molecules to stabilize the HgBr adduct would create a much more complex situation for mercury cycling.

We are explicit in the abstract and throughout the manuscript that this study represents a single day and further mention in the title would not add further to this already high level of transparency. The language in the conclusions has been modified to reflect the results of this study are from a certain set of atmospheric conditions.

C2.2) The error budget on the retrieved BrO vertical profiles and corresponding column densities is mainly derived through sensitivity tests using different a priori profiles in the Optimal Estimation retrieval. Nothing is said about the uncertainty related to the choice of the settings for the DOAS spectral analysis, although these settings can affect significantly the BrO slant column densities, and therefore the amount of BrO retrieved in both the FT and boundary layer. This is for me a crucial point and the authors should discuss it into more details in the revised manuscript. The main results of the sensitivity tests performed for the selection of the DOAS settings could for instance appear as supplementary material.

R2.2) Indeed, we have performed extensive sensitivity studies prior to the submission of this work. And the revised manuscript makes these now transparent. It was found that the choice of DOAS parameters (fitting window, intensity offset, and reference spectrum) impacted the resulting BrO dSCDs, but the final settings used estimate the BrO dSCDs conservatively. A section has been added to the supplemental material to provide greater detail regarding tests on the fitting window and intensity offset. The description of sensitivities towards reference

selection is found in Sect. 2.4.2. Overall, this study estimates the BrO concentration profile (and VCD) conservatively.

C2.3) Specific comment related to major concern 1: Only a day with very low aerosol content has been selected for this study. Does it mean that even for moderate aerosol contents, the methods/concepts developed in this study are not valid anymore? The presence of a moderate aerosol content could indicate a different regime where the marine boundary layer contributes more significantly to the total BrO column. If so, a different impact on the Hg oxidation pathways should be expected.

R2.3) Even moderate aerosol loads in the marine boundary layer would impact the capability of this method to retrieve profiles of trace gases located in the free troposphere. Some information could be retained, i.e. total column, but derivation of vertical profiles would most likely be significantly impacted. This has now been mentioned in the revised manuscript.

Aerosols in the MBL could indeed indicate a different air mass regime where the MBL contributes more significantly to the total column of BrO; however, there is no experimental evidence for this. The available evidence finds low BrO upper limits (<0.5 pptv) also under conditions of acidic aerosols in the tropical MBL (Gomez-Martin, 2013; Volkamer et al., 2015; Wang et al., 2015). Only if there was BrO at instrument altitude would this complicate the decoupling of the FT profile. Thus, we do not expect that moderate aerosol would fundamentally change the ability to apply this method to characterize free tropospheric BrO partial columns. At high aerosol load the view of the FT could be shielded because of the dominance of scattering at lower altitudes. We have added mentioning that this method is useful for future deployments from mountain tops, where aerosol variability and shielding of the FT can be overcome.

The ability to remotely sense the BrO partial column in the FT is an analytical question, and does not imply an impact on the conclusions drawn from this study regarding the oxidation of atmospheric mercury in the free troposphere.

C2.4) Page 28333, lines 11-21: the average tropospheric column retrieved in this study (2.3E13 molec/cm2) is in the 1-3E13 molec/cm2 range reported so far in the literature. However, it is clearly above the values published for tropical regions and which are ranging from 0.4 to 1.2E13 molec/cm2. Where does this difference come from? Why not considering in your study BrO total/tropospheric columns derived from OMI and/or GOME2A over Gulf Breeze around 10 April 2010?

R2.4) The average value found in this study is above the range currently reported for the tropics, but still within the range (albeit the upper end) reported for the mid-latitudes. The values reported by the referee seem rather low for measured profiles, which are in the range 1-1.7e13 (average 1.5e13) molec cm^{-2} for aircraft studies (Volkamer et al., 2015; Wang et al., 2015); Theys et al., (2011) reported a monthly averaged tropospheric VCD from GOME-2 of ~1.7e13 molec cm^{-2} with error bars extending above 2e13 molec cm^{-2} for the latitude range 0-30° N in using the years 2007 and 2008. While comparisons with other satellite measurements would be desirable there are few studies that report satellite derived values of tropospheric BrO at the mid-latitudes. The manuscript is already quite complex. We consider a comparison with satellite data beyond the general comparison of VCDs to be beyond the scope of this manuscript.

C2.5) Supplementary material, aerosol extinction profile retrievals: Why a extinction value of 0.01 km-1 and a scaling height of 0.6 km are used for the a priori profile? A justification is needed. Also, what would be the impact of changing this a priori profile (e.g. by using a larger scaling height value) on the retrieved aerosol extinction profiles but also on the retrieved BrO profiles? Why aerosol extinction profiles are not directly retrieved from O4 dSCDs fitted in the UV range? In other words, why retrieving aerosol extinction profiles in the visible range (483 nm) and then scaling them to 350 nm, while observations in the UV range are available? A reference/explanation should be given for Eq. S1.

R2.5) This a-priori profile for aerosol extinction was chosen as a first guess of a profile shape and magnitude that might correspond with the measured O_4 dSCDs. Changing the a-priori profile should have little-to-no impact on the derived a-posteriori profiles, as that the technique should arrive at a similar conclusion independent of a-priori. Using a larger scaling height or total AOD would only increase the computational time needed to reach the very low values found for aerosol extinction in this case study. Additionally, because such low values are found, small changes in the derived aerosol extinction at the various grid layers of the inversion would likely have little impact on the sensitivity towards trace gas retrievals.

Aerosol profiles are derived using the visible absorption features of O_4 due to the increased absorption and better signal to noise in this wavelength range. This leads to a more robust derivation of the aerosol extinction (Volkamer et al., 2015). In general, during initial sensitivity studies the aerosol extinction values derived using the O_4 dSCDs from the UV analysis show larger error bars, and have less certainty in comparing measured and forward calculated O_4 dSCDs. However, our measurements are consistent within error also at UV wavelengths. Text has been added to this section to make these decisions more transparent.

C2.6) Technical corrections: Difficult to read texts in Figs S2 and S4 (font too small).

R2.6) Text for these figures has been changed to be more legible.