Comment on:

Werner, B., Stutz, J., Spolaor, M., Scalone, L., Raecke, R., Festa, J., Colosimo, F., Cheung, R., Tsai, C., Hossaini, R., Chipperfield, M. P., Taverna, G. S., Feng, W., Elkins, J. W., Fahey, D. W., Gao, R.-S., Hintsa, E. J., Thornberry, T. D., Moore, F. L., Navarro, M. A., Atlas, E., Daube, B., Pittman, J., Wofsy, S., and Pfeilsticker, K.: Probing the subtropical lowermost stratosphere, tropical upper troposphere, and tropopause layer for inorganic bromine, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-656, in review, 2016.

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Citations of the manuscript are marked blue.

p.13, line 15ff: Our study on the sensitivity of the O_2-O_2 absorption measured in limb direction as function of the cloud cover underneath (see sections 4.2 and Fig. 7 in Stutz et al. (2016)) as well as the results presented by Volkamer et al. (2015) (in their Fig. 3) clearly demonstrates the limitation of the O_2-O_2 method to constrain the radiative transfer for UV/vis studies above an altitude 10 km, mostly because the bulk of the O_2-O_2 collisional complex is located near the surface. Therefore, any skylight analyzed for the O_2-O_2 absorption in limb direction may carry additional, or even predominantly information on the radiative transfer of lower atmospheric layers (see Figure 7 in Stutz et al. (2016)), rather than of the targeted atmospheric layers.

We generally agree that "skylight analyzed for the O_2-O_2 absorption in limb direction may carry additional, or even predominantly information on the radiative transfer of lower atmospheric layers", but want to point out that BrO profiles published in Volkamer et al. (2015) and Wang et al (2015) were neither affected by underneath cloud cover, nor by cirrus above. Sections 2.10 and 3.1 in Volkamer et al. (2015) discuss explicitly the effect of aerosol and clouds, and make fully transparent that the presented RF12 and RF17 case studies are not affected by clouds. Furthermore, we show below HSRL data from these two flights (Fig. 1) that make transparent that no aerosols or thin cirrus layers were present above the aircraft. Moreover, the authors are referred to Fig. 2 and Section 2.1 in Dix et al. (2016a), where it is shown that in cloud-free conditions measurements of O_2-O_2 are suitable to constrain RTM up to 15 km. The statement by the authors is too broad, and certainly does not apply to the Wang et al. and Volkamer et al. case studies. This should be corrected.

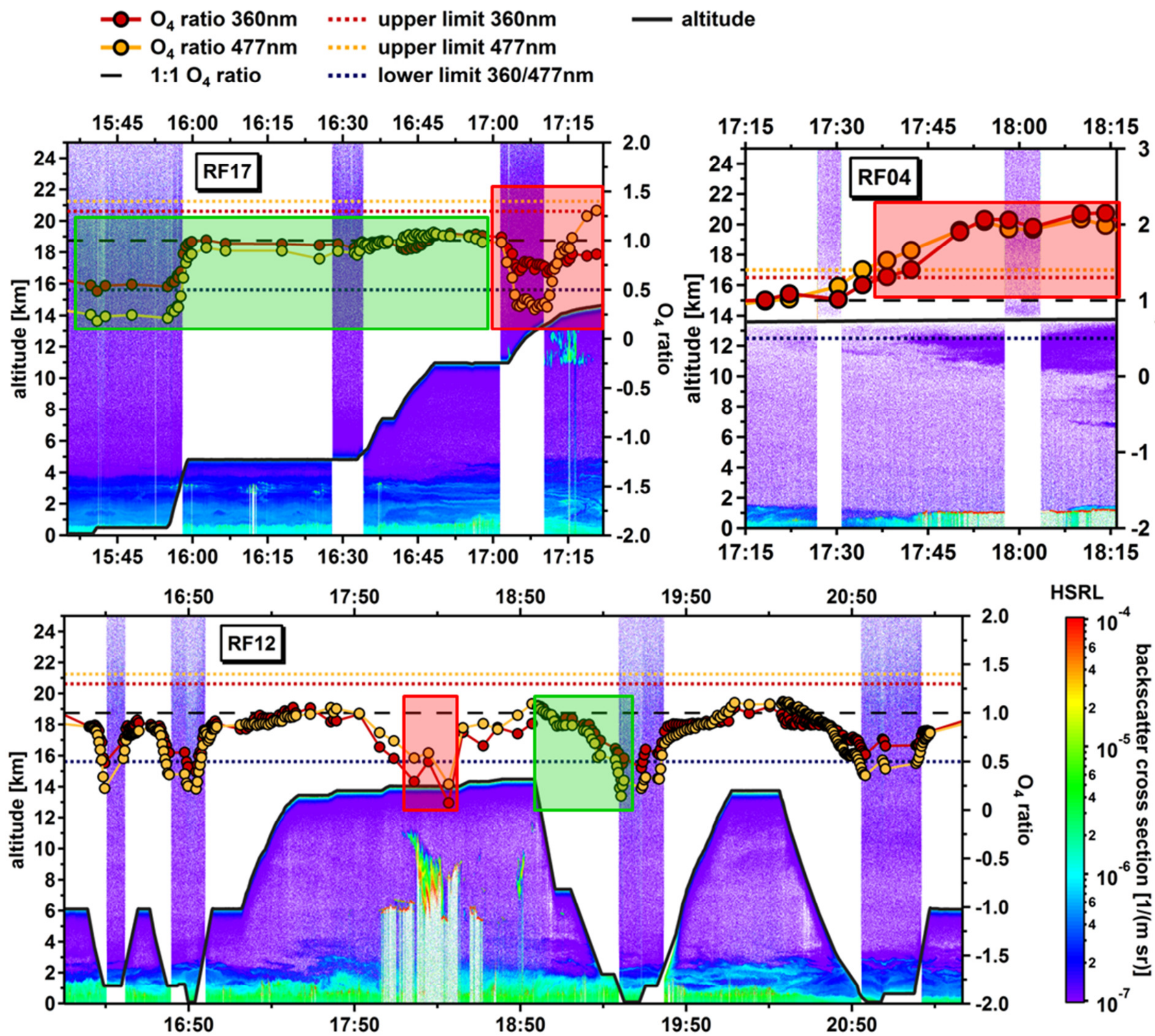

Figure 1. *Comparison of O_4 ratios at 360 nm and 477 nm with HSRL particulate backscatter cross section data for TORERO RF04, RF12 and RF17 (Dix et al. (2016a); Volkamer et al. (2015); Wang et al. (2015)). Altitude resolved HSRL backscatter data is plotted and color coded along the flight track. Larger signals denote the presence of aerosol/clouds. HSRL is either measuring above or below the aircraft. The shading directly around the flight track seen in part of RF12 and RF17 is a near field effect that leads to erroneous large back scatter signals by HSRL. Green boxes in RF12 and RF17 mark data periods that were used for BrO, IO and NO_2 optimal estimation profile retrievals as published in Volkamer et al. (2015). Regular HSRL upward scans show that for these time periods no aerosol or cloud layers were present above the aircraft. For more information see Dix et al. (2016a and b).*

p.13, line 22ff: Furthermore, Wang et al. (2015), and Volkamer et al. (2015) did not use a stratospheric CTM to study the potential influence of changing overhead BrO concentrations on their results. As result, the predominant occurrence of atmospheric BrO in the stratosphere at daytime, and its potential column changes mostly due to a changing tropopause height (e.g., at the subtropical or polar jet) may mimic the presence of BrO in limb the direction, or at flight altitude.

This is incorrect, and a misleading reflection of the literature. First, Volkamer and Wang et al. (2015) used a stratospheric model (RAQMS) to study the influence of changing BrO concentrations above, and show that potential changes in the stratospheric BrO VCD, or apparent changes in the measured limb dSCDs due to a changing tropopause altitude do not affect the results. Second, the authors are referred to section 2.10 in Volkamer et al. (2015), and Fig. S4 in the SI text of Wang et al. (2015) for the excellent agreement with the aircraft microwave temperature profiler measurements and the location of the thermal tropopause in the model. Third, the supplement of Volkamer et al. (2015) shows that the stratospheric profile above the aircraft is accurately corrected. Finally, Dix et al. (2016a) used RAQMS BrO profiles for the correction of stratospheric BrO contributions to the limb dSCD measurement, and confirms excellent agreement with the optimal estimation case study profiles from Wang et al. (2015) and Volkamer et al. (2015) using a parameterization method within low error bars.

p.13, line 27ff: In conclusion, even though the reported TORERO flights 12 and 17 were performed under clear-skies (Volkamer et al., 2015), it is unclear the extent to which unaccounted scattering due to aerosols and (probably) optically thin upper tropospheric clouds, lower level clouds, or changing overhead stratospheric BrO contributed to the inferred (or by error attributed) elevated BrO in the UT, and around the bottom of the TTL.

This conclusion is incorrect in all aspects listed. See our above responses. TORERO flight RF12 and RF17 are neither affected by aerosol/cloud extinction above, nor lower level clouds below, nor changing stratospheric BrO.

Caption of Figure 3b: …, the unexpected kink around 12 to 13 km in the inferred BrO profile when the inversion is constrained to the Wang et al. (2015) BrO profile indicates that our and the Wang et al. (2015) BrO profiles are not compatible … .

We respectfully disagree, and show below that the results presented in Fig. 3b of Werner et al. (2016) and Wang et al., (2015) are in fact quite compatible.

Werner et al. show that optimal estimation (OE) profile retrievals in Figs. 3a and b yield within error bars the same results for the altitude range between 14.5 and 18.5 km, regardless of a priori profile choice. This shows that the OE inversion is well constrained by measurements for these altitudes. However, below 14.5 km, the measurements by Werner et al. are not well constrained, and essentially follow the a priori in both cases shown. The “unexpected kink around 12 to 13 km” is therefore not unexpected at all, but to the contrary, it is the expected result of the OE solution that transitions from ‘constrained by measurements’ (above 14 km) to reproducing the a priori profile at lower altitudes (below 12 km). This behavior is likely reflected in the averaging kernel that are not shown, and should be included in the manuscript. Also, is the OE based on limb spectra only or are downward scans included? This information is missing in the paper.

Furthermore, section 4.4 in Stutz et al. (2016) states that GH measurements during SF3 are compatible with up to 1.5 pptv of BrO directly below flight altitude. This is quite compatible with the TORERO campaign average BrO vertical profile, which shows a significant decrease of BrO above 14 km, with a mean of 1.86 ± 0.16 pptv at 13.5 km, and 1.38 ± 0.16 pptv at 14.5 km (Dix et al., 2016b). The TORERO average profile is compared with model predictions in Fig. 5 in Schmidt et al., (2016), and is shown with a better resolution in Fig. 10 of Dix et al. (2016b) (included as Fig.2 here). Notably, the case studies in Volkamer et al. (2015) and Wang et al. (2015) are 100% consistent within low error bars (5%) with the parameterization retrieval (Dix et al., 2016a), if the same data subsets are compared. These case studies had probed primarily air masses influenced by convection over oceans. The lower mean BrO for the complete TORERO data set is mainly reflecting different air mass histories, consistent with the variability in Br_y noted in Wang et al. (2015), and our hypothesis that sea-salt derived Br_y is a source for BrO in the upper free troposphere downwind of marine convection.

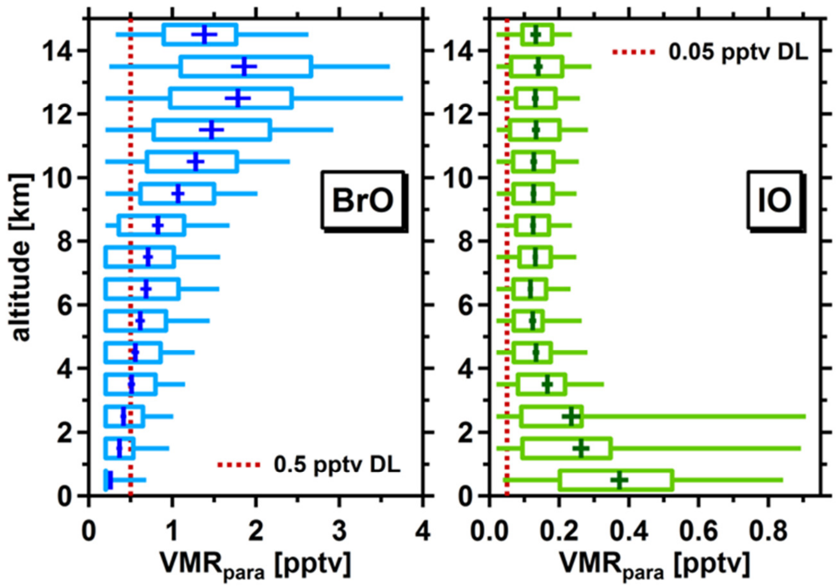

Figure 2. *TORERO AMAX-DOAS BrO (left) and IO (right) volume mixing ratio data derived by parameterization method (VMR_{para}) for the complete campaign. Plotted are 1 km altitude means with their 95% confidence interval as uncertainty in darker colors, while the lighter colored whiskers denote 5, 25, 75 and 95 percentiles.*

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