General comments

The paper addresses relevant scientific questions within the scope of ACP. The paper presents novel data. Substantial conclusions are reached. Some but not all of the scientific methods are valid and clearly outlined. Some, but not all of the assumptions are clearly outlined. A better discussion is needed regarding why J and k are both likely to be in error (see below). The results are sufficient to support the conclusions, but more discussion of the results is needed (see below). The description of calculations is not complete and precise to allow their reproduction by fellow scientists. The authors give proper credit to related work and clearly indicate their own new contribution. The title clearly reflects the contents of the paper. The abstract provides a complete and concise summary. The overall presentation is well structured and clear. The language could be improved in some cases: there are run-on sentences with too many ideas. The number and quality of the references is appropriate. No supplementary material was found. No part of the paper should be reduced, combined, clarified or eliminated.

The paper presents high precision limb scattering (LS) and solar occultation (SO) observations of BrO, NO_2, and O_3. The authors have wisely chosen the azimuth difference angle in limb scattering measurements to be 90° to minimize diurnal effects along the line-of-sight. Figures 2-5 and 8 are particularly impressive. However, more should have been done along the lines of Figure 5 with the limb scattering data. As the authors appear to realize, one way to separate J and k is to exploit their different SZA-dependence, particularly at sunrise and sunset: J is dependent on SZA (or time), but k is not. Looking at the agreement of modelled and measured SCDs as a function of SZA will provide more information than the authors' approach of looking at the agreement as a function of the SCD magnitude (Figures 6-8).

In Figure 5, the 'k=0.75, J=1.3' simulation case clearly appears to best fit the data before the air mass change, which would be in agreement with the limb scattering best J/k ratio. Noticing this (and the authors should mention this), I am convinced by their data that the currently accepted values of J and k are probably both wrong, if we assume that each must lie within its respective uncertainty. The authors should explicitly state that a J/k ratio of 1.69 implies that both J and k must have opposite biases if the uncertainty on each is not to be exceeded. This would also help the reader understand why they did not try any cases with k=1 in Figure 6 and to understand the conclusion specific to $BrONO_2$ formation in the troposphere.

Ideally, different occultation data should be tried from previous and/or future flights in the hopes that the air mass will be more homogeneous. Also, I wonder if previous limb scattering campaigns would provide an additional dataset to verify the findings presented here (e.g. 23 Mar 2003, 24 March 2004 or 1 March 2006 as listed in Kritten et al. [2010], see also Weidner et al. [2006]). If so, the best J/k ratio for each sunrise or sunset could be found, and then the ratios from all twilight occultations could be analyzed for central tendency and variability to give an estimate along the lines of the J/k ratio of 1.xx ± 0.yy found in this paper. I have also found that inferred Br_y from BrO tended to be high, e.g. using SAOZ-BrO data (Figure 9 of Sioris *et al.*, 2006).

Attempting to simulate the correlation of BrO and NO_2 as a function of height with a large (satellite) dataset might provide a method to determine k because of its M-dependence (see Figure 10 of McLinden et al., 2010 and related text).

The major outcome of this submitted paper is the conclusion that the accepted value of the $\mathrm{J(BrONO_2)}/\mathrm{k_{BrO+NO_2}}$ ratio should be increased by 69±4%. The authors appear to discard the significance of a large difference between their two sets of quasi-independent measurements (limb scattering and solar occultation). They claim to merge the two sets of ratios (from LS and SO) and weight the former more heavily. However, they do not discuss the merging of the two sets of ratios, the weighting or anything else related to quantifying the uncertainty. Using the four ratios obtained from the LS measurements and averaging, I obtain 1.696±0.04 (1σ). This is almost exactly what they found in terms of the ratio (possible rounding error?) and exactly what they found in terms of the uncertainty, meaning that the SO values seem to be essentially discarded. Because this replicates their result, I presume that I have essentially replicated the method the authors used to obtain the ratio and its uncertainty. The fact that the colour coding in Figure 8 seems to be only for LS measurements points to the fact the SO measurements are ultimately discarded. If the SO data are discarded with regard to inferring the J/k ratio, this should be stated.

Because, as shown in Figures 6-7, the slope is insensitive to k_{mod}/k_{JPL} (between 0.65 and 1) and J_{mod}/J_{JPL} (between 0.9-1.4) for a near constant $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$ ratio, the authors could have essentially found an uncertainty of ~0 in the $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$ ratio, had they chosen the $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$ ratio to be identical for all 4 combinations tried in Figure 6 (if the uncertainty calculation is as I describe above).

Another method for obtaining the error on the J/k ratio from the data shown in this paper would involve positively and negatively perturbing the J/k ratio until the slope of the measured versus modelled SCDs is no longer unity within the uncertainty. If the authors did this, it should be described. This could be tried for the different four J and k combinations in Figure 6 as starting points. The work has already been done given Figure 8. To be conservative, the global maximum and minimum J/k ratios (over the four starting points) that depart from unity should serve ultimately as the uncertainty on the J/k ratio. The perturbations should proceed in four directions (from the four starting points):

1) +Δk, J constant

2) -Δk, J constant

3) –ΔJ, k constant

4) +ΔJ, k constant.

This will provide the bounds on the J/k ratio that allows the measured and modelled LS SCDs to come into agreement.

I take issue with discarding the solar occultation measurements because they should be should be more reliable because of the simpler measurement geometry. I believe the uncertainty in their current method is more appropriately the difference between the mean/median of the sets of ratios from the two measurement techniques (unless they consider limiting the SO dataset to the time period before the hump at SZA=92.5°). Otherwise, the ratio (weighting SO and LS equally) and its uncertainty would be 1.525±0.175, meaning that the uncertainty is >4 times larger than the 0.04 claimed by the authors.

The authors attribute the different $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$ ratios from LS and SO measurements on air mass differences, but make no attempt to account for air mass differences using models or other measurements. Given SLIMCAT's horizontal resolution of 2.8°, this translates to 310 km in latitude, and less in longitude, so unless tracer observations are needed, the model could be used to test the air mass difference hypothesis. I wonder if the 3 profiles from LPMA (ascent, sunset, sunrise) could be used to determine any horizontal gradients in CH_4 and N_2O or auxiliary measurements (MIPAS, etc.) and then Labmos could help with the temporal sampling.

The air mass differences are really a source of error in the $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$ ratio in this experiment. That is why J and k are historically measured in the controlled conditions of a laboratory.

Specific comments

P27823L15-16 The value of 1.4 is the combined uncertainty for the $BrONO_2$ absorption cross section and the quantum yield. The *J* value uncertainty will be slightly higher because there are several sources of uncertainty in the actinic flux (see specific comment P27826 below).

P27823L17 The authors have done a nice job of surveying the chemical kinetics literature and pointing out that low temperature data is not available for the rate of Reaction 1.

P27824L20 It should be noted that all SZAs are defined at the sensor.

P27825 McArtim claims to be accurate for limb radiance to SZAs of 90-91° but not for larger SZAs, yet the model appears to be used at even larger SZAs at dusk on the first day of the flight. Also, if BrO SCDs from SZA>93° are used to determine the J/k ratio, a method to correct for stray light should be probably be included in the data analysis.

P27826 Labmos assumes 0.3 for the surface albedo, but with overcast cloud, the effective albedo might be 0.8, which could affect bromine partitioning, particularly at smaller SZAs (<80 deg). This should be tested given that $BrONO_2$ dissociates at long wavelengths, if a suitable literature reference is not available.

P27827L11-13 Is diurnal variation of BrO along the incoming solar path taken into account in both solar occultation and limb scattering simulations? It sounds like the capability exists with McArtim, but this needs to be stated explicitly. Note that limb single scattering consists of two directions for photon transport, the incoming solar path and the path along the line-of-sight, whereas in solar occultation, these two paths are one and the same. Diurnal variation is an issue for solar occultation because only the tangent layer is at SZA=90°.

P27827L17 "…elevation angles and tangent heights…" -> "…tangent heights…". Also the minimum tangent height of all BrO SCDs during the flight should be stated. I calculate it to be 7.8 km assuming the sensor is at 31 km and an elevation angle of -4.88.

P27828L1-2 "…samples are always taken at SZA=90°, i.e. at the tangent height from…" -> "tangent layer is always at SZA=90°, and this is…". Also, Reaction (1) should be omitted from "…less sensitive to Reactions (1), (2a), …" since Reaction (1) is not SZA-dependent. Also, the authors should move the reference to Fig. 5 to follow immediately after "sunset" on P27827L26.

P27829L1-2. I disagree with this statement. The best range of k_{mod}/k_{JPL} is stated to be 0.65 to 0.85, but this experiment is only truly sensitive to the $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$ ratio and the values of k_{mod}/k_{JPL} are somewhat arbitrarily chosen. For example, the combination k_{mod}/k_{JPL}=0.9 and J_{mod}/J_{JPL}=1.5 would also be expected to produce a slope of 1 for LS measurements, based on Figure 8, but exceeds the uncertainty of J_{JPL}. The stated, best k_{mod}/k_{JPL} and J_{mod}/J_{JPL} should not be outside the uncertainty of k_{JPL} and J_{JPL}, to respect those uncertainties and thus the case with k_{mod}/k_{JPL} =0.65 should be omitted from Figures 6 and 8 and any subsequent calculations regarding $(J_{mod}/J_{JPL})/(k_{mod}/k_{JPL})$. This sentence should be changed, otherwise the authors' approach becomes difficult to comprehend.

P27829L7 The tangent points for limb scattering are much farther than "30-70 km" at low tangent heights.

P27829L24-27 I suggest this sentence is omitted since the normalized limb radiance is not as sensitive to the surface albedo as the flux, partly because of the normalization and partly because the former is not a large-solid-angle quantity, whereas the latter is (including the hemisphere below, see comment above regarding scene albedo). Furthermore, I understood that McArtim is not used to provide the fluxes to SLIMCAT or the Labmos facsimile. The argument in the next sentence regarding the direct beam is sufficiently convincing anyway.

Figure 1 – Is it possible to include the SZA as an alternate x-axis (along the top of this figure)? At the very least, it would be helpful to know the SZA at 3:55 UT and I assume the SZAs for the first day can be read from Figure 5. Note that the SZA at the tangent point would be more helpful in Figure 1 because as the authors have noted, most of the absorption occurs in the tangent layer and not at balloon altitude. Also, the insetted text refers to dSCDs of BrO, O_3 and NO_2, but the SCD appears in the y-axis title and the caption. This should be cleared up (i.e. is a model estimated dSCD for the reference spectrum added to each underlying dSCD to make it an SCD?) Or, if measured dSCDs are really used throughout the paper, the McArtim model should also calculate dSCDs by subtracting off the SCD for the simulated reference spectrum.
Why is the LS time series longest for ozone and shortest for BrO on the first day? In a way, it is good that the BrO SCD time series cuts off since data in the lower stratosphere for SZA>90 should be discarded anyway if there were high tropospheric clouds. Do the authors have info on this from nadir observations? It seems that the last BrO measurement in Figure 1 occurs at 17:49 UT, but the last measurement shown in subsequent figures was at 17:48 UT. At this time, given an estimated latitude of 67.9° and a longitude between Kiruna and the Finnish/Russian border, the SZA would equal or exceed

91.8°. If the SZA≥91.8°, the measured BrO SCD becomes sensitive to high tropospheric clouds, because the sun appears to be below them at the balloon and the radiative transfer becomes very complicated. For overcast cloud with top at 8 km (within 4 degrees of latitude of the sensor), the height of the cloud's shadow at the tangent point can reach 11.2 km at SZA=91.8° and 22.8 km for SZA=93.9°. With the lowest tangent height (TH) at 7.8 km, clearly, the authors need to be careful about data at SZA>90 deg. The LS measurements on the next morning appear to be after sunrise and do not need to be filtered. However, the diurnal variation along the line-of-sight, should be considered in the modelled SCDs even if the authors have wisely chosen an azimuth difference angle of 90°, I estimate the SCD error at SZA=89° may be a few percent at the lowest THs based on McLinden et al. [2006] (relates to specific comment re: P27827L11-13).

Figure 2- Why are there missing points in only the 350 nm radiance time series? Figure 1 shows BrO measurements that are not discontinuous, which seems impossible given discontinuous 350 nm radiance measurements. Perhaps the lines should not be included in Figure 1 (i.e. point-markers only). Why is only 350 nm affected? Are the missing points due to anomalous data caused by shadowing of the tangent point by broken high tropospheric clouds?

Figure 4- dSCDs in Figure 1 are larger than SCDs shown in Figure 4. This probably means that some SZA cutoff or other filter has been applied. Please describe.

Figure 8- State in the caption that the colour coding is appropriate to limb scattering measurements.

Technical corrections

p27822L4 "turn-over" -> "turnover"

p27822L7 "…indicates that,…"-> "…indicate that…"

p27822L10 "…reasons likely…" -> "…reaons are likely…"

p27822L25 "…with the amount…" -> "with the underestimated amount…" (see p27832L14-5 as well, where I suggest: "Also the overestimate of stratospheric Br_y from the inorganic method due to… models by an underestimate of reactive bromine.")

p27823L14 (and elsewhere) "uncertainty" -> "uncertainty factor"

p27824L10 "skylight"-> "sunlight"

p27824L25 "Finish" -> "Finnish"

p27825L28 "…evaluated." -> "…evaluated, respectively."

p27825L29 "…both,…" -> "…both…"

p27826L9 "…lab-owned 1-D Facsimile code…" -> "…1-D facsimile code…"

p27826L10 "neccessary" -> "necessary"

p27826L21 “are” -> “is”

p27827L22 (and throughout) “higher” -> “larger”

p27828L21 “size” -> “magnitude”

p27829L1 “…regression measured vs. …” -> “…regression of measured versus …”

p27829L24 “Incorrect modelled…” -> “Incorrectly modelled…”

Figure1 It would help the reader to note (somewhere, perhaps in the 3rd paragraph of Section 2) that the malfunctioning of the scanning telescope only affects the limb scattering observations.

Figure1 “commenced” -> “continued”

Figure1 “…down…” -> “…down to…”

Figure 3 What is meant by “local angles”? Please state whether these are at the balloon or the tangent point.

Figure 5 caption: “k=0.075” -> “k=0.75”

References

McLinden, C. A., et al. (2010), Odin/OSIRIS observations of stratospheric BrO: Retrieval methodology, climatology, and inferred Bry, J. Geophys. Res., 115, D15308, doi:10.1029/2009JD012488.

McLinden, C. A., C. S. Haley, and C. E. Sioris (2006), Diurnal effects in limb scatter observations, J. Geophys. Res., 111, D14302, doi:10.1029/2005JD006628.

Sioris, C. E., et al. (2006), Latitudinal and vertical distribution of bromine monoxide in the lower stratosphere from Scanning Imaging Absorption Spectrometer for Atmospheric Chartography limb scattering measurements, J. Geophys. Res., 111, D14301, doi:10.1029/2005JD006479.