Point to point response to the comments of reviewer #2 (given in italic)

We thank for the review.

This paper presents observations from 3 instruments measuring NIR, UV-Vis direct-sun and UV-Vis Limb viewing from a balloon platform conducted in Kiruna from the 6-9th September 2009 to examine the kinetics of the BrONO2 mediated ozone loss in the stratosphere. This work represents a unique data-set combined with complex photochemical/radiative transfer/chemical modeling. It is a valuable contribution to our knowledge of stratospheric chemistry, and in particular is important in the discussion concerning the total bromine loading of the stratosphere. I recommend publication, however there are some areas were the science has confusing arguments that leave the reader unconvinced of the validity of the conclusions. I believe that this can be easily addressed with the measurements that the authors have at hand, and it will significantly strengthen this paper. These are detailed below.

No analysis of the airmasses was performed to assess that indeed the assumption that the same airmass was being sampled (where the justification for the use of a 1D photo-chemical model is made – page 27826, line 11). Where the photochemical model did not match the observations then the argument that an airmass discrepancy was the cause is made. This may be the underlying cause, but as the discrepancies are being used to argue for different kinetics then the role of sampling different airmasses really needs to be eliminated. As the authors have access to N2O and O3 data (though the viewing geometry of the NIR spectrometer is not provided in the manuscript), this would be useful in assessing the airmass origin, using tracer-tracer correlations. This should be performed to assure the readers that indeed airmasses are unchanging and that discrepancies are only due to kinetic/Bry deficiencies. This will significantly strengthen the conclusions. Sampling different geometries / viewing different airmasses with the direct-sun versus the limb should not be an impediment to determining kinetic deficiencies – if the transport issues (BrONO2/Bry ratios or temperatures) are adequately dealt with in the analyses (i.e. if a large temperature range is sampled then the kinetic parameters should reflect this).

In fact, the core of this statement/concern is not well based, since prior to presenting a chemical analysis of the measurements we performed the following investigations. (More details of our investigations can also be found in the thesis of Sebastian Kreycy, see here http://db.tt/d6EsQiho

(a) **First measured and (SLIMCAT) modeled CH4, N2O (and O3) is inter-compared, where a mismatch in the profiles of all 3 gases was found. Most likely this mismatch is due to deficit in the SLIMCAT model with correctly modeling the vertical (diabatic) transport, of which the error may grow unreasonably large (some kilometers) when the model is integrated for longer times. Once the mismatch in the modeled and measured profiles for CH4 and N2O is removed (e.g. by vertically shifting both profile by the same amount) however, any discrepancy in modeled and measured O3 (both for the SC and limb measurements) is also removed. Finally, since it is know that N2O (and thus NOy) and organic bromine have a tight relationship in the stratosphere (e.g., Wamsley et al., 1998, Dorf et al., 2008), the profiles of total NOy and Bry may accordingly be changed.**

(b) **During summer to winter circulation turn-over potential horizontal (and transport related) in-homogeneities in the high latitudes air masses are known to be small, due to good**

reasons. First, during summer stratospheric air masses are excessively photochemical processed due to the high exposure to sun-light, and therefore horizontal gradients in the major gas families (HOx, NOy, Cly, and Bry) are small. Further, the still very small winds (some 1m/s as e.g. inferred by the small movement of the balloon) prevented a significant horizontal mixing of the investigated air masses with air masses of different dynamical and photochemical origin e.g., from mid-latitudes by the time of the measurements. The latter (as it is said in the manuscript) was investigated by inspecting the analysis of air mass transport as given by the MIMOSA model http://ether.ipsl.jussieu.fr/ether/pubipsl/mimosa_2009_uk.jsp **.**

(c) **The slow movement of the balloon together with the small horizontal gradients in chemical composition of the high latitude late summer air masses, guaranteed a quasi Lagrangian type investigation. This conclusion is certainly true for the limb measurements, where the investigated air masses are always located in a circle of less than 100 km (in radius) around the balloon gondola. However, this conclusion is less certain for the solar occultation measurements (as said in the manuscript), where at low sun as the solar zenith angles grow larger the tangent points (from where most of the signal is picked-up) are becoming subsequently more distant to the balloon gondola (e.g. up 1200 km at SZA = 95 degrees).**

Taking arguments (a) to (c) together and given mentioned limitations (e.g., over-interpreting the SO measurements), we strongly feel that sufficient care is taken in the study to separate the transport and photochemistry processes.

A second area requiring clarity is the selective use of sunrise and sunset data to suit the argument being made. Early in the manuscript the role of BrOH photolysis complicating the sunrise BrO polar sunrise analysis (page 27827, line 8) is made, however later this is ignored and the sunrise data is used to access the BrONO2 kinetics. This will effect both the solar occultation and limb sounding data – why is only the solar occultation data excluded from further analyses? i.e. (page 27827, line 24) where the discussion of dawn limb BrO tending to appear more rapidly that the simulations suggests could very well just be because of BrOH photochemistry inadequacies. The results section requires reworking to ensure that these apparent contradictions are clarified – both in ensuring that dynamical effects are accounted for, as well as photochemical complications due to BrOH photolysis at dawn.

Here some clarification appears to be necessary. As the reviewer correctly states for sunset, both the SO and Limb measurements were used in the analysis, but for sunrise only the SO measurements . The reason comes from (a) the photochemistry of stratospheric HOBr (see Figure 3), and (b) the timing of both (SO and Limb) measurements, i.e. in the analysis both type of sunset measurements were used since at sunset HOBr concentrations were expected to be still negligible (see Figure 3). Conversely since the SO measurements started at SZA = 95 degree at sunrise when HOBr was still present however to an unknown amount they were not considered, but the limb measurements were used since they (unfortunately) started only at SZA = 86 degree, when all HOBr was already destroyed (see the red lines in Figure 3).

Minor comments

Abstract: should contain the date range and location of the measurements

The text is accordingly changed in the manuscript (P27822L3) from
"at high-latitudes during autumn circulation turn-over. The observations are complemented by simultaneous direct solar occultation" to
"over Kiruna in northern Sweden at high-latitudes on September 7th and 8th 2009 during the autumn circulation turn-over."

Introduction:

too short, there are many examples of using atmospheric data to test laboratory kinetic data with success (for the ClO dimer cycle) [Shindell and deZafra, 1996; Solomon,2002; Avallone et al., 2003; Vogel, 2003; Stimpfle, 2004; Hobe et al., 2005; Schofield et al., 2008; Kremser et al., 2011]. It would be valuable to frame this work in this context and talk about the uniqueness of looking at BrO to do this.

We are aware that in the past many atmospheric data were used to test kinetic data, including our own (c.f., Frieß et al., 1999; Frieler et al., 2006, ….). However, we strongly feel that we should keep the introduction as tight (and readable) as possible, thus avoiding such an always incomplete review, such as they are given in many introductions found elsewhere in the scientific literature.

The first sentence is misleading since bromine has the largest effect on ozone through coupling with the ClOx cycle (which is mentioned later in this page).

We accordingly changed the text (P27823 Line 1 to 2) to "The amount of ozone destroying bromine oxide (BrO) in the stratosphere is largely dominated by the reactions 1, 2a, 2b"

How does the heterogeneous hydrolysis of BrONO2 influence conclusions drawn here? There is no discussion about the role of BrONO2 + H2O heterogeneous reaction as an important reaction in the lowermost stratosphere. Any error introduced by misrepresentation of the aerosol size distribution could potentially dramatically change the conclusions made here. Please discuss.

The BrONO2 + H2O heterogeneous reaction is not further addressed in the manuscript since it is not relevant to the core of the study (as discussed above). However, we will come back to BrONO2 + H2O heterogeneous reaction when presenting our SO sunrise measurements performed at low latitudes together sunrise measurements and mid and high-latitudes including the sounding mentioned in the manuscript.

Methods: Perhaps tabulate the instrumental descriptions (with references). i.e. Instrument, wavelength range, viewing geometry, target species, references

We feel that by providing references to our earlier studies, of which many refer to all the details of our technique, enough information is provided in the manuscript on our method.

Page 27825 line 1 and else where, please use NIR, direct-sun or limb viewing geometries to describe the spectrometers rather than a) b) and c) will stop the readers having to refer back in the manuscript.

We feel we should not change it since the spectrometers and their observation geometry are well described (and defined) on p27824l5. However, to recall this in the text at P27825L1
"Here we primarily report on the data obtained from the spectrometers (b), and (c) obtained during sunset and of spectrometer (c) during sunrise." is change to
"Here we primarily report on the data obtained from the limb observation spectrometer during sunset and sunrise and data recorded during sunset using the direct sunlight spectrometer."

Page 27825 line 21 radio-metrically calibrated – is this standard terminology for this (absolutely calibrated)?

We accordingly changed the text (P27825L21) from "radio-metrically" to "absolutely radio-metrically".

Page 27829 line 12-17 this should be earlier when 20.3ppt is described.

The paragraph P27829L12-17 is shifted to P27826L23.
In P27826L20 the text "derived from BrO observations of spectrometer (b),"is deleted

Conclusion (3) tropospheric conditions are not tested in this current paper, and this is very speculative. Due to the higher temperatures in the troposphere I am not convinced that this argument holds, as the kinetics may be well represented at these higher temperatures (closer to the laboratory tested conditions).

Not really, but please considered that J/k is also largely uncertain (according to JPL) for moderate temperatures, or consider a situation where large amounts of NO_x are emitted and plumes of BrO are present at lower than moderate temperatures (i.e. in the Arctic spring) .

Avallone, L., D. Toohey, T. Fortin, K. McKinney, and J. Fuentes (2003), In situ measurements of bromine oxide at two high-latitude boundary layer sites: Implications of variability, J Geophys Res-Atmos, 108, –, doi:10.1029/2002JD002843.

Hobe, von, M., J.Grooss, R. Müller, S. Hrechanyy, U. Winkler, and F. Stroh (2005), A re-evaluation of the ClO/Cl_2O_2 equilibrium constant based on stratospheric in-situ observations, Atmos. Chem. Phys., 5, 693–702. Kremser, S. et al. (2011), Retrievals of chlorine chemistry kinetic parameters from Antarctic ClO microwave radiometer measurements, Atmos.Chem. Phys., 11(11), 5183–5193, doi:10.5194/acp-11-5183-2011.

Schofield, R. et al. (2008), Polar stratospheric chlorine kinetics from a self-match flight during SOLVE-II/EUPLEX, Geophysical Research Letters, 35(1), –, doi:10.1029/2007GL031740.

Shindell, D., and R. deZafra (1996), Chlorine monoxide in the Antarctic spring vortex .2. A comparison of measured and modeled diurnal cycling over McMurdo Station, 1993, J Geophys Res-Atmos, 101, 1475–1487. Solomon, P. (2002), Measurements of stratospheric ClO over Antarctica in 1996–2000 and implications for ClO dimer chemistry, Geophysical Research Letters, 29(15), doi:10.1029/2002GL015232.

Stimpfle, R. M. (2004), First measurements of ClOOCl in the stratosphere: The coupling of ClOOCl and ClO in the Arctic polar vortex, J. Geophys. Res., 109(D3), doi:10.1029/2003JD003811. Vogel, B. (2003), Vertical profiles of activated ClO and ozone loss in the Arctic vortex in January and March 2000: In situ observations and model simulations, J. Geophys. Res., 108(D22), doi:10.1029/2002JD002564.