AC: We thank Robyn Schofield for her constructive comments and the appreciation of our work.

RC: General Comments:

This well structured paper presents a very nice methodology to retrieve BrO using remotely sensed UV-Vis spectra taken from an aircraft in the Arctic as part of the ASTAR campaign. The use of the regularization formalization is a nice advancement in the tropospheric trace-gas remote sensing retrievals. It is beyond the scope of this paper but I wonder what difference one might expect from the use of this approach over using a correlation length to account for smoothing in the vertical?

AC: Indeed, using a correlation length operator and using a derivative operator in the Phillips-Tikhonov side constraint are conceptually similar. The first order derivative operator explicitly relates neighboring layers of the profile to be retrieved. Therefore, the similarity to the correlation operator approach depends largely on the design of this operator, in particular on the actual correlation length. The derivative operator, in contrast to a correlation length approach, provides a constraint on smoothness only, not on the absolute value of the state vector.

RC: The retrievals are performed in two steps with the non-linear aerosol retrieval being conducted initially as this is a major sensitivity in tropospheric trace-gas retrievals using scattered light. Using the radiance to constrain this presents also a methodological advancement which, I am sure, will be utilized in future trace gas retrievals of this nature.
The second retrieval step involves the familiar trace-gas retrieval with characterization. This is a nice paper, I do have some technical and semantic comments, which I discuss below and need to be addressed, but I unreservedly recommend publication in AMT.

AC: We appreciate the reviewer's recommendation. The following changes have been made after the reviewer's comments:

Specific Comments:

RC: Page 3929, line 22 HOx and NOx are chemical families not species, I would suggest naming the specific species.

AC: We agree. Therefore "HOx, NOx" is changed to "HOx (= H+OH+HO_2), NOx (=NO+NO_2)". Additionally, "hydrocarbons" are also included.

RC: Page 3930, line 3 Add references for the satellite measurements that show the BrO horizontal extent.

AC: In Page 3930, line 3, the text "(e.g., SCIAMACHY, GOME, OMI)" is exchanged by "(e.g., Richter et al., 1998; Wagner and Platt, 1998; Wagner et al., 2001;Theys et al. 2010)", and they are added in the bibliography, i. e.,

Richter, A., Wittrock, F., Eisinger, M., and Burrows, J. P.: GOME Observations of Tropospheric BrO in Northern Hemispheric Spring and Summer 1997, Geophys. Res. Lett., 25(14), 2683-2686, doi:10.1029/98GL52016, 1998.

Wagner, T. and Platt, U.: Satellite mapping of enhanced BrO concentrations in the troposphere, Nature, 395, 486–490, 0028-0836, doi:10.1038/26723, 1998.

Wagner, T., Leue, C., Wenig, M., Pfeilsticker, K. and Platt, U.: Spatial and temporal distribution of enhanced boundary layer BrO concentrations measured by the GOME instrument aboard ERS-2, J. Geophys. Res., 106, 24 225–24 235, 0148-0227, doi:10.1029/2000JD000201, 2001.

Theys, N., Van Roozendael, M., Hendrick, F., Yang, X., De Smedt, I., Richter, A., Begoin, M., Errera, Q., Johnston, P. V., Kreher, K., and De Maziere, M.: Global observations of tropospheric BrO columns using GOME-2 satellite data, Atmos. Chem. Phys. Discuss., 10, 28635-28685, doi:10.5194/acpd-10-28635-2010, 2010.

In addition, the reference "Deutschmann et al. (2010)" (Pag. 3935, line 11) is corrected:

Deutschmann, T., Beirle, S., Friess, U., Grzegorski, M., Kern, C., Kritten, L., Platt, U., Prados-Roman, C., Pukite, J., Wagner, T., Werner, B., and Pfeilsticker, K.: The Monte Carlo atmospheric radiative transfer model McArtim: introduction and validation of Jacobians and 3-D features, J. Quant. Spectrosc. Ra., 112, 6, 1119-1137, ISSN 0022-4073, doi:10.1016/j.jqsrt.2010.12.009, 2011.

RC: Page 3931, line 24 What is the field of view in the horizontal? i.e. How wide is the slit?
AC: In the text, "of 0.2° in the vertical" is exchanged to "of 0.19° in the vertical and 2.1° in the horizontal".

RC: Page 3935, line 23 How can 4% for the measurement error really cover all the errors of the measurement? I would expect that dark current and calibration errors (i.e. systematic) would result in an absolute error quantity. Using a percentage error everywhere means that where the measurements are close to 0 (i.e. close to the reference) the errors are also small and therefore in the retrieval these low measurements are (falsely) regarded as being of a higher quality than measurements with a higher signal to noise.
AC: Before addressing any retrieval, each measured spectrum is corrected for electronic dark current and offset. This is stated in Page 3932, line 14 (DOAS). Additionally, in the corrected manuscript "where Li/ref are the measured radiances at a certain geometry with index i related to the reference geometry ref." (Page 3935, line 8) reads now as "where L represents the measured radiances, corrected for electronic dark current and offset. The sub-index i and ref stand for a certain geometry index and for the reference geometry, respectively."

Regarding the uncertainties, the 4% error is not assumed for the radiances ratio, but for every measured radiance (i.e., for L_i and for L_{ref}, Eq. 2). The error of $y_i=ln(L_i/L_{ref})$ is estimated by error propagation. This is now clarified in the text (Page 3935, line 21):

"The relative error of the measured radiances L is chosen as 4% in order to account for systematic RT uncertainties such as the Ring effect (e.g., Landgraf et al., 2004; Langford et al., 2007; Wagner et al., 2009a), the used trace gas cross-sections, etc. The diagonal covariance matrix Se contains the squared error of the normalized radiances y, calculated through error propagation."

RC: Page 3936, line 1 The Levenberg-Marquardt approach ensures that convergence takes into account the distance of the forward model results to the measurements – a note to this effect may add to the discussion here.
AC: Lines 1-2 of Page 3936 is changed to: "Equation (3) is then minimized following a standard Levenberg-Marquardt approach (Levenberg, 1944; Marquardt, 1963). The convergence criteria of the method assure the minimal distance of model and measured quantities in each iteration (e.g., Rodgers, 2000)." Accordingly, the following references are added in the bibliography:

Levenberg, K.: A Method for the Solution of Certain Non-Linear problems in Least Squares, Q. Appl. Math., 2, 164–168, 1944.

Marquardt, D. W.: An algorithm for the least-squares estimation of nonlinear parameters, SIAM, J. Appl. Math., 11, 431–441, doi:10.1137/0111030, 1963.

RC: Page 3938, line 5 here and elsewhere there is not a clear distinction made between the forward model error and the forward model parameter (FMP) error. Forward model (FM) error includes all errors in the forward model approximation of the true atmosphere (i.e. the physics of the atmosphere can not be always completely described i.e. refraction, Mie scattering approximation) and it includes (but is not limited to) errors of the forward model parameters (such as temp, press, aerosol profile etc). The forward model error unlike the forward model parameter error often cannot be quantified (you can change the temp profile and see its effect on the retrieval (i.e. FMP error), but often you can't know what implications the true physics have over using a Mie scattering approximation in your RTM (FM error)). I would argue that "efrw, the error in the forward model" does not originate from uncertainties of each of the forward model parameters, but rather the FMPs contribute significantly to efrw.
AC: We agree. The forward model error *efrw* comprises the error in the model approximation and the error in the forward model parameters. However, the parameterization and approximation of the physics of the atmosphere with the McArtim RTM used in this work are assumed to be a fair representation of the reality (McArtim has been previously validated with other RTM and with measurements, Deutschmann 2011). Page 3937 (line 5) now reads as:
"This error (the error in the forward model) comprises the errors in the forward model approximation, and the uncertainties of each of the forward model parameters b. As demonstrated through its validation with measurements and other RT models (Deutschmann et al., 2011), the McArtim RT model provides a fair representation of the true atmosphere. Thus, the errors in the forward model approximation are assumed negligible. In the following, the error in the forward model (efrw) stands for the error in the forward model parameters."

RC: Page 3940, line 11 more than 40% of the forward model error – I think the absolute FM error is unknown, you only know the absolute FMP error.
AC: See above.

RC: Page 3943 to 3944 'Validation using O4', I just caution here the use of validation, I do not dispute the use of O4 as a great method for obtaining aerosol and cloud scattering information – but it is not as useful where there is less O4 (i.e. free troposphere and above). This is stated by the authors for the UT/LS region last sentence of section 3.3 but then in section 4 the sentence 'confidence is gained in the novel method to retrieval vertical profile distribution of trace gases in the troposphere' is too strong.
AC: We agree in the limitation of the O_4 for retrieving scattering information. Therefore, as stated in Pg 3934 (lines 22--27) the "O_4 method" is not used in this work for the retrieval of the aerosol extinction coefficient (i.e., is not used for the characterization of the scattering processes). The O_4 is used in Sect. 3.3 only for the self-validation of the trace gas mathematical inversion since O_4 is a fairly constant and known trace gas in the atmosphere. Results of the self-validation (given at the end of Sect. 3.3 and in Fig. 7) indicate that in regions where the instrument is close to its detection limit (e.g. in the UT/LS for O_4 measurements), the error of the trace gas retrieval is dominated by the measurement error. On the other hand, the limit of the trace gas retrieval in atmospheric regions where measurements are above instrumental detection limit (e.g. in the BL in the case of O_4), the error of the trace gas retrieval is dominated by the error in the forward model parameters. For clarity, the authors will change "validation" for "Self-consistency" in the title of Sect. 3.3.

RC: Page 3945 on – why is ppt chosen as the retrieval unit? Is the retrieval of BrO really conducted in ppt? Does this not make the FMP sensitivity to temperature and pressure higher than if the retrieval and AKs were reported for molec.cm-3? Please describe how the retrieval of the trace-gases are performed exactly.
AC: The retrieval of BrO provides concentration (molec/cm^3). Considering the air density (measured pressure and temperatures), the units are then transformed into volume mixing ratios (ppt).

RC: Page 3947, line 23. It would be great if the example of the stratospheric influence was tied more closely to the figure 9. As the paragraph stands it is just stating what one would expect to see if a stratospheric folding event was present, but doesn't commit to whether the authors consider it has occurred here.
AC: Studies based on the correlation of O_3-CO, not shown in this work for simplicity (refer to Prados-Roman, 2010, Dissertation), indicate that the UT/LS transition layer (e.g. Pan et al. 2004) in the selected passage extended from 7-9.5 km (thermal tropopause at 8.5 km). In addition, the O_3 vmr vertical profile (in situ measured during that ascent) shows a 'sudden' peak at 8-8.5 km. This observation suggests that, indeed, the aircraft crossed a tropopause fold which was captured by the in situ and by the DOAS instruments. However, further investigations would require additional information such as PV maps across the flight track. Since this falls out of the scope of the paper, no further discussions are made in that direction. Indeed, we believe this is a very interesting topic that will be particularly address in campaigns such as TACTS on board the HALO aircraft.

RC: Page 3948, 3 didn't Fitzenberger et al. see much higher free tropospheric values in the Arctic than are reported here?
AC: In the submitted manuscript the mentioned line reads as "there are reports of some pptv of BrO detected in the free troposphere during similar conditions (e.g., Fitzenberger et al., 2000)". Indeed, for measurements performed in Kiruna during summer 1998 and winter 1999 in Kiruna, Fitzenberger et al. (2000) reported on positive detection of tropospheric BrO (0.4-2.3 pptv). In their work, a BrO concentration of 1-3 $\cdot 10^{13}$ molec/cm2 was shown in the free troposphere (Fig. 2, Fitzenberger et al., 2000). If uniformly mixed in the troposphere, it would correspond to about 0.5-2 pptv. Those and other values were later discussed by e.g. Schofield et al. 2004 (0.9 pptv as an upper limit of tropospheric BrO), and von Glasow and Crutzen (2007). In our work, the BrO mixing ratio in the free troposphere is estimated as $\leq$1.5 pptv. However, due to instrumental limitations „we cannot conclude that BrO was unequivocally detected in the free troposphere during the ASTAR 2007 campaign" (Page 3948, line 13-15).

RC: Page 3951, _ 20 could it be that the satellite retrievals do not systematically underestimate BrO because in the study here rather a cloud-free scene is selected for – to reduce complication in the RT?
AC: Indeed the passages used for satellite comparison are chosen to be cloud-free in order to reduce uncertainties in such an inter-comparison exercise. For issues concerning the satellite retrieval, please refer to the response to Salawitch's review.

RC: Page 3952 line 27 on, split the long sentence into two distinct points.
AC: The line is now changed into "The inferred BrO profiles generally show large and heterogeneous mixing ratios within the BL (8–30 pptv), and small mixing ratios within the free troposphere (1.5 pptv). In the upper troposphere and lowermost stratosphere, the inferred variable mixing ratios (1–4 pptv) increase with height. The latter two findings can be explained by the known atmospheric photochemistry of bromine and by the transport of stratospheric air masses to

tropospheric altitudes (as seen by simultaneous O_3 and CO measurements). The former finding points to halogen activation within air masses of so-called ozone depletion events (e.g., Simpson et al., 2007)."

RC: Page 3953, line 17, in the outlook part, some references for the proposed work using DOAS for aerosol retrievals such as Friess, 2006, Wagner 2004, 2009 (referenced earlier in paper but appropriate here too) Langford et al. Atmos. Chem. Phys., 7, 575–586, 2007 and cloud retrievals Daniel et al. JGR, 111, D16208, doi:10.1029/2005JD006641, 2006 and Schofield et al. JGR. 112, D21203, doi:10.1029/2007JD008737, 2007.

AC: In the new manuscript, lines 17-20 (Page 3935) read as "In addition, the retrieval of aerosol and cloud particle optical properties simultaneously measured can improve the accuracy of the key trace gas retrievals (e.g. Wagner et al., 2004; Friess et al., 2006; Wagner et al., 2009b; Vlemmix et al., 2010). Furthermore, the retrieval of optical properties of particles - such as an extinction coefficient profile - represents a research field with great potential for, e.g., radiative forcing and climate feedback investigations (e.g. Daniel, 2006; Schofield et al., 2007)."

Additionally, and also as references for line filling-in of the Fraunhofer lines (Ring effect), the work of Langford et al. (2007) is now referred to in Page 3935, line 24.

Hence, the following references are added to the bibliography:

Daniel, J. S., Portmann, R. W., Miller, H. L., Solomon, S., Langford, A. O., Eubank, C. S., Schofield, R., Turner, D. D. and Shupe, M.D.: Cloud property estimates from zenith spectral measurements of scattered sunlight between 0.9 and 1.7 μm, J. Geophys. Res., 111, D16208, doi:10.1029/2005JD006641, 2006.

Schofield, R., Daniel, J. S., Portmann, R. W., Miller, H. L. R., Solomon, S., Eubank, C. S., Melamed, M. L., Langford, A. O., Shupe, M. D. and Turner, D.D.: Retrieval of effective radius and liquid water path from ground-based instruments: A case study at Barrow, Alaska, J. Geophys. Res., 112, D21203, doi:10.1029/2007JD008737, 2007.

Langford, A. O., Schofield, R., Daniel, J. S., Portmann, R. W., Melamed, M. L., Miller, H. L., Dutton, E. G., and Solomon, S.: On the variability of the Ring effect in the near ultraviolet: understanding the role of aerosols and multiple scattering, Atmos. Chem. Phys., 7, 575-586, doi:10.5194/acp-7-575-2007, 2007.

RC: Technical Corrections and Typographical Errors:

Page 3928, line 9 delete 'amount of'
AC: Deleted

RC: Page 3928, line 23 delete 'and rare'
AC: Deleted

RC: Page 3929, line 1 replace 'just' with 'only'
AC: Line corrected as "here not **only** the total aerosol optical thickness is inferred, but **also** the vertical profiles…"

RC: Page 3929, line 24 ‘eventually yield scavenge of Hg’ should be ‘eventually scavenge Hg’
AC: We agree and change accordingly.

RC: Page 3929, line 25, ‘toxic’ could be replaced by bio-accumulative or bioactive, because in the polar ecosystem it accumulates but is toxic to humans
AC: “Toxic” exchanged by “bio-accumulative”

RC: Page 3930, line 5 replace ‘trigger’ with ‘motivation’
AC: Replaced.

RC: Page 3938, line 17 and 22 here and elsewhere forward parameter should be forward model parameter
AC: See above.

RC: Page 3939, line 14 were measured in situ
AC: Corrected.

RC: Page 3939, line 18 The aircraft ascent considered here : : :(14:30 UT), flying
AC: Corrected.

RC: Page 3940, line 9-10 deployment studied herein,
AC: Corrected.

RC: Page 3940, line 11 more than 40% of the forward model parameter error
AC: Changed.

RC: Page 3940 line 28 particles in the size
AC: Changed.

RC: Page 3948 line 13 aply -> apply
AC: Corrected.

RC: Page 3950 line 29 heights
AC: Corrected.

RC: Page 3952 line 5 large -> high
AC: Changed.

AC: Note that new affiliations have been added for the following co-authors:

Dr. André Butz: now at the Institute for Meteorology and Climate Research -- Atmospheric Trace Gases and Remote Sensing, Karlsruhe Institute of Technology, Germany.

Dr. Lena Kritten: now at the Institute for Space Sciences, Freie Universität Berlin, Germany.

AC: “this temperature” in Page 3932 (line 26) and Page 3933 (line 2) is changed to “the temperature”.

AC: “of” (Page 3939, line 5) is corrected to “in”.

AC: Page 3941, line 19: “(i.e., 1.54+0.0*i* is assumed)”.

AC: Page 3941, line 25: “then” is erased.

AC: “herein” is erased (Page 3946, line 1).

AC: Page 3947, line 7: “result (3)” is changed to “finding (3)”.

AC: “in” is deleted from Page 3953 (line 22).