# Reply to Referee #1

Dear Referee #1,

First of all, we would like to thank the referee for your helpful comments. We have studied your comments carefully and have made the necessary corrections according to the comments. In addition, wording of the original manuscript was revised by a native speaker. We feel that the revised manuscript is a great improvement on the original. We summarized points of revision below in reply to the comments.

(The referee's comments are indicated in red.)

Page 1909 line 25

Why is it important to take into account the diurnal behavior of ClO. Either cite a publication stating this or elaborate on this statement. Are there any model studies elaborating the importance of the diurnal behavior of ClO?

As mentioned in Ko and Sze (1984), one of the advantages of studying diurnal variation is that the diurnal variation can well characterize the photochemical process among the various causes. Because the diurnal variation tend to reflect the short-term properties of photochemical process rather than those of the longer-term(>1 day) dynamical process such as atmospheric transport.

In addition, detailed understanding of the diurnal variation is important to derive the long-term trend from measurements done in limited portions of the day. This is more important especially for some satellite datasets obtained in dusk and dawn. For example, in Jones et al. (2011), they estimated the ClO trend from measurement with AURA/MLS and Odin/SMR. The Odin is sun synchronous with a descending node at 06:00 h and ascending node at 18:00 h LST. However, the monthly average of ClO has been drifting because the Odin satellite has slowly descended from its initial position and the solar zenith angle (SZA) has slowly decrease over time during morning scans and increased during evening scans. As a result of the change of SZA, the monthly average of ClO has slowly increased over time during morning and decreased during evening scans. Then, they needed to correct the drifts due to the change of SZA by taking the diurnal variation into account.

We believe that the diurnal variation is one of the important factors for estimated the trend of stratospheric ClO.

We revised the text as follows.

******

"Detail study of the diurnal variation leads to well understanding of photochemical processes, because the diurnal variation tend to reflect the short-term properties of photochemical process rather than those of the longer-term(>1 day) dynamical process such as atmospheric transport (Ko and Sze, 1984). In addition, the diurnal variation is important to derive the long-term trend based on the measurements done in limited portions of the day, such as satellite measurements conducted in dusk and dawn (e.g., Jones et al., 2011)."

page 1911 line 4-6

Please cite a publication of a description of the path length modulator, i.e. the oldest one I found is by Gustincic (1977).

As the referee suggested, we will cite appropriate references for the path length modulator. The fundamental concept of Path Length Modulator (PLM) is described in Gustincic 1977, but the details of the optics are different from that in Gustincic (1977). The basic optical design of our current PLM is described in Mizuno et al. (2002). Therefore, we will cite both Gustincic (1977) and Mizuno et al. (2002).

page 1911 line 11-12

Please give the frequency of the LO. Also specify how the contribution from the mirror side band is dealt with.

The frequency of LO is 202.446 GHz. For spectral line, there is no significant spectrum at mirror side band by using the forward model calculation with molecular line parameters listed in JPL Submillimeter, Millimeter, and Microwave Spectral Line Catalog (Pickett et al.,1992). For the continuum emission that we use for intensity calibration, we assume the equal response for both signal- and mirror-bands, and this equality has been confirmed by observing ozone spectrum at 203.453159GHz that is close enough to assume the same side band ratio as that of ClO at 204.346GHz.

We added explanation as follows.

****

"The frequency of the local oscillator (LO) is 202.446 GHz. There is no significant spectral lines in both signal- and image-bands confirmed by using the forward model calculation with molecular line parameters listed in JPL Submillimeter, Millimeter, and Microwave Spectral Line Catalog (Pickett et al.,1992). For the continuum emission that we use for intensity calibration, we assume the equal response for both signal- and mirror-bands, and this equality has been confirmed by observing ozone spectrum at 203453.159GHz that is close enough to assume the same side band ratio as that of ClO at 204.346GHz."

page 1911 line 17

Please give more information on the type of the FFT spectrometer. Is there an apodization function applied? How is the apodization dealt with?

We used the Hanning window function. The frequency resolution of spectrometer is reduced by Hanning apodization function from 61 kHz to ~70 kHz. We will mention this in the text. Thank you for your comment.

page 1911 line 19

Why is beefing up the cooling system relevant for this publication? I see the importance for the operation of the instrument, but this is a technical detail which I do not consider interesting.

As the referee says, this may be a technical detail, but we think the fact that the commercial DFS (Acqiris AC240) cannot be used as it is at high altitude place is useful information, because we know that other groups are using or intend to use the same DFS.

page 1911 line 21

Please give a citation for the Allan variance time and its usage for millimterwave radiometry or describe more in detail e.g. Schieder, R. Kramer, C. (2001).

What is meant by spectrometer, the whole instrument of just the FFT? For the measurement the Allan variance time of the FFT is not relevant, relevant is the Allan variance time of the whole instrument, which may also be limited by 1/f noise. Please give some more details.

For the first point of the comments, we will cite Schieder and Kramer (2001) as the referee suggests, and we will explain the Allan minimum time briefly as "The stability of spectrometer for microwave receiver is commonly characterized by Allan minimum time (see more detail e.g., Schieder and Kramer 2001), and this value is defined by the minimum point of Allan variance plot.".

For the second point, as the referee mentions, the Allan minimum time of the whole instrument is important issue. Therefore we will mention about this in the revised text. The Allan minimum time of whole instrument is ~ 1 minute after we use the FFT spectrometer. It is improved compared with the previous minimum time using AOS. That was ~ 10sec.. This means that the stability of the whole instrument is limited by AOS for the previous system but that the new system is limited, if we take account that the Allan minimum time of the FFT spectrometer itself is ~ 1,000 sec.

page 1912 line 23

What temperature is meant here? Physical temperature, Brightness temperature, Raleigh-Jeans-Temperature?

"T" in "T(1-exp(-tau))" denotes the Brightness temperature of blackbody at physical temperature of T, and "T" is also equal to the Physical temperature of the translucent medium in thermal equilibrium because the Raleigh-Jeans approximation is well fitted to the Planck's law of radiation for 200GHz and 300K.. The Brightness temperature of the translucent medium is "T(1-exp(-tau))".

page 1912 line 25 ff

Please be more exact here. Give the definition of the Brightness temperature you use.
I recommend Janssen (1993) for the differences and finer issues of the definitions of Brigthness temperatures.

We defined as "the Brightness temperature is an equivalent temperature of blackbody that emits the same amount of radiation energy at the observing frequency". This definition is written in our original manuscript in page 1912 line 25. This is more general definition of the Brightness temperature.

Janssen defined the "microwave" Brightness temperature or Rayleigh-Jeans brightness temperature in page 6 of Janssen (1993). That is the special case of Brightness temperature applicable for microwave. In this study, i.e., under the condition of

200GHz and 300K, the Rayleigh-Jeans approximation can be applied well, because the ratio between the plank-law and Rayleigh-Jeans law, $\frac{kT}{h\nu}\left(e^{\frac{h\nu}{kT}}-1\right)$, is ~1.02. Therefore, we can regards as the general Brightness temperature is equal to the Rayleigh-Jeans brightness temperature and also equal to the physical temperature.

We revised from page 1912 line 20 to 1913 line 1 as follows;

********

“Atmospheric gases (and, more generally, translucent media like lossy dielectric plates) not only absorb incident radiation but also emit thermal radiation simultaneously. The thermal radiation intensity is expressed as a brightness temperature or an equivalent temperature of a black body that emits the same amount of radiation energy at the observing frequency. At millimeter wavelengths, thermal radiation intensity is approximated to proportional to the black-body temperature, owing to the Rayleigh-Jeans law of radiation. The transmission coefficient of the translucent medium is given by $e^{-\tau}$, and the thermal radiation intensity, i.e. brightness temperature is expressed as $T(1-e^{-\tau})$ where ($\tau$) is the optical depth of the translucent medium. The “T” corresponds to the brightness temperature of black body at physical temperature of T, and is considered to be equal to the physical temperature of the translucent medium, because the Rayleigh-Jeans law can be applied in this study, i.e., under condition of ~ 200 GHz and ~300 K.”

page 1915 line 22

What is mean by "a little bad"? In the figure 2 I cannot see much of a difference to the time between 5th and 17th of December.

As the referee says, the difference of optical depth is not so significant. The average for December 1 to 5 is 0.8, and that for December 5 to 16 is 0.7. However, the rms noise on the spectra was different, 0.12mK for December 1 to 5 but 0.8mK for December 5 to 16. The baseline flatness was worse in December 1 to 5. In addition, we would like to minimize the averaging period of spectral data to avoid the contamination due to other non-diurnal dynamical factors such as atmospheric transportation. From these reasons, we finally considered that it is not better to use the data between December 1 to 5 for retrieval analysis.

We changed the expression of this sentence as follows;

*****************

"…., the sky optical depth was a little bad and the rms noise on the spectra was worse than that obtained after December 5."

page 1916 line 5 to 25 page 1917 line 1 to 3
The averaging kernels depend on the signal-to-noise ratio and this certainly changes if the noise on the spectrum is constant (as it can be made by changing the integration time) as the signal itself changes. I am not convinced that a trick like this can be used to overcome the problem of the changing information content of the spectrum. I would therefore suggest to prove this by calculating the averaging kernels for the minimum and maximum values of ClO and show both averaging kernels.

In Rodgers (2000), the averaging kernel "A" is expressed as follows.

$$\mathbf{A} = \left(\mathbf{W}^T\mathbf{S}_R^{-1}\mathbf{W} + \mathbf{S}_{sp}^{-1}\right)^{-1}\mathbf{W}^T\mathbf{S}_R^{-1}\mathbf{W}$$

where W is weighting function, $S_R$ is covariance of a priori, $S_{sp}$ is covariance of spectrum measured. According to the definition of covariance, the each element of covariance is difference between the measured value and the expected value. Thus, $\mathbf{S}_{sp}$ is related with only noise but not signal strength. This means that mathematically the averaging kernel does not depend on signal-to-noise but only noise. The noise levels of day-time and night-time data are almost the same, and the averaging kernels can be regarded as the same.
In connection with this matter, the description "the vertical resolution is broad above 50km as seen in Fig.5 due to the limitation of the signal-to-noise of the spectra" in page 1918 at line 15-16 is not correct. We correct "signal-to-noise" as "noise".

page 1916 line 16
The authors claim, that there is no significant channel-to-channel correlation. On page 1917 line 5 they explain, that the spectrum is smoothed by a moving average. As I understand this method, it is introducing a channel to channel correlation. Please explain and/or prove that this is not relevant.

The effect due to the channel-to-channel correlation can be neglected in this study because of the following reasons.
(1)In case the correlation exists between the distant channels separated far-away, long

wavy patterns will appear on the spectrum. But such wavy features were already subtracted during the data reduction of the spectral lines.

(2) In case the correlation exists between the nearby channels, fine periodical ripple will appear on the spectrum. But these fine ripples were removed by the channel binding up.

For these reasons, there is no feature showing the channel-to-channel correlation on the final form of the spectral data that proceeds to the retrieval analysis, Therefore, only random noise should be considered in the retrieval analysis.

We changed the expression of line 16 as

*********

"…. and the channel-to-channel correlation in the spectrum is negligible in this study (Nagahama et al., 1999)."

page 1918 line 5

I guess the authors mean the standard deviation of the residuum. The absolute values of the residuum in figure 3 seem larger than 1.8 mK.

Yes, we calculated the rms of the residuum except for the signal frequency range from 204.306 to 204.386 GHz. The rms value is 1.84 mK. We confirmed this.

page 1918 line 13

Why was the baseline ripple not removed, e.g. by retrieving a set of sinusoidal functions?

What happens if the range is chosen wider than 150 MHz?

As the referee says, the ripple may be removed by sinusoidal functions, if the ripple is produced only by the standing waves in the optics of the instrument, But in practice, at an order of a few mK level, non-sinusoidal artifacts or spurious appears on the spectrum, and the base line was not fitted only by simple sinusoidal functions. It was not able to fit with the forward model for broarder frequency range. Since the half width at half maximum of the Voigt profile is ~ 10 MHz at 40km, we think that 150MHz is enough for retrieving the ClO mixing ratio at 40 km.

We added more explanation;

".….150 MHz frequency range to avoid the contamination of the baseline ripple and spurious that cannot be removed simple sinusoidal functions."

page 1920 section 4.2.3.

The random noise generated by random noise on the spectrum can directly be calculated (refer to Rodgers, 2000, page 46, the retrieval error.)

As the referee mentioned, the random error generated by random noise on the spectrum can directly be calculated by Rodgers (2000) (and also Conner et al., (1995 )).

In Conner et al., 1995, the retrieval error due to random noise on the spectrum is calculated by the following equation.

$$\mathbf{S}_m = \mathbf{D}_y \mathbf{S}_\varepsilon \mathbf{D}_y^T$$

where $\mathbf{S}_\mathrm{m}$ is the covariance of retrieval due to the random noise on the spectrum, $\mathbf{D}_\mathrm{y}$ is the contribution function, and $\mathbf{S}_\varepsilon$ is the covariance of measurement. In Conner et al., 1995, $\mathbf{S}_\varepsilon$ is determined by the residual of the calculated spectrum to the measured spectrum. In case of Conners' measurements, the residuals were larger than the expected noise from the theory, except in the narrowest channels for a short observation. The residuals also show features of correlation between nearby channels, although the features of the residuals is by no means periodic. As a result, they must consider the off-diagonal elements of $\mathbf{S}_\mathrm{m}$.

On the other hand, in this study, the noise on the spectrum is regarded as random noise, because the rms noise decreased as proportional to square root of integration time. Therefore, we can put the off-diagonal element of $\mathbf{S}_\varepsilon$ to be 0. This is the reason why we calculated the retrieval error due to the random noise on the spectrum by using the method of Nagahama et al., 1999. In case $\mathbf{S}_\varepsilon$ is diagonal, both methods give the same result.

page 1922 section 4.2.6.

Apart from the uncertainties mentioned, the uncertainties caused by spectroscopic data (pressure broadening, line intensity etc.) may cause large systematic errors. Why have they not been dealt with?

As the referee suggested, the molecular spectroscopic parameters are possible causes of systematic errors. Probably the line intensity is most important. But in the catalog and references therein, we did not find the description about the errors of the ClO

molecular parameters. Thus, we cannot discuss the error quantitatively. We will mention the possible systematic error due to the molecular parameter but not deal with quantitatively. This is common problem for spectroscopic measurements of ClO.
We added the following text at the end of section 4.2.6;
*****:
"For the spectral line parameters, we cannot evaluate qualitatively in this study, because no error information are found in JPL Submillimeter, Millimeter, and Microwave Spectral Line Catalog (Pickett et al.,1992). But they are potential causes of systematic error."

page 1922 lines 23ff
Why does the spectral signal come from only one side band? Acc. to figure 1 there is no side-band filter. Hence, what one sees is a mixture of the spectral signal from both side-bands, the signal band and the mirror band. This may lead to artifacts which resemble baseline features or even contain signatures of trace gases which emit in the mirror band.

Our SIS receiver is double-sideband mixer receiver and has sensitivity for both sidebands. There is no significant spectrum signal other than ClO for both sidebands by using the forward model calculation with molecular line parameters listed in JPL Submillimeter, Millimeter, and Microwave Spectral Line Catalog.. The continuum signals of hot- and cold-loads equally come into both sidebands, and we correct this for intensity calibration.
In the revised manuscript, this matter is already mentioned in the "Instruments" section, as described in the reply to the comment concerning with page 1911 line 11-12.

page 1923 line 15 to 20
Nedoluha et.al. (2011) and Ricaud et.al. (1997) also used ground based data and have also a rather coarse resolution. They would probably not see a peak at 50 km altitude. The argument that MLS is unrealistically large at 50 km because it is not seen in other ground-based MW measurements is therefore not convincing. Because of the strong dependency of ClO on the solar zenith angle it would seem natural to ensure that the MLS data are taken at the same SZA as the NATAOS data and not the same local time of the NATAOS position. The 60 degree longitudinal coincidence criterion cover a range of 8 hours solar time. Please correct or be more precise. The precision of the MLS data is also influenced by the convolution with the NATAOS

averaging kernels. Has this been taken into accout? If not, please refer to Rodgers and Connor (2003) on how to compare data sets with different altitude resolution and how to calculate the error on the comparison.

We agree with the referee's first point. We cannot exclude the possibility of 50km peak based on the coarse resolution ground-based data, as the referee says. We deleted the unconvincing description. But we will not use MLS data at 50km for comparison with NATAOS because of the following reason. Santee et al. (2008) discussed the precision of the MLS measurements, and they mentioned that ClO profile of AURA/MLS (v2.2) is unsuitable to use for science above 1.0 h Pa.. In Atacama, 1.0 hPa is corresponding to ~ 50 km.
For the second point, the referee misunderstood maybe due to our insufficient description. We searched MLS data over 60 deg in longitude, but we selected the data coinciding with the same local solar time-slot at "each" MLS position. Thus the SZAs are almost the same for all selected MLS data within the 3-hours time-slot. We corrected the text to avoid misleading statement.
For the third point, we did not take account of the influence of convolution on the MLS precision. Thank you for the comment. We calculated the precision for the convolved AURA/MLS with our averaging kernel, and revised Fig 11.

page 1923 line 25
I cannot see why the measurements at a completely different latitude by Nedoluha et.al. (2011) and/or time by Ricaud et.al. (1997) can serve as an argument, that NATAOS measurements are more correct than the MLS measurements.

We agree the referee's point that the argument is not convincing based on the comparison between the measurements done at completely different latitudes. We do not have sufficient ground to discuss quantitatively. Therefore, we removed the ambiguous speculation.

Section 4.3
The comparison to MLS leaves the impression, that the comparison is restricted to the region where it fits alright. Please consider this and correct or explain more conclusive. I would also suggest to show a time series of the MLS and NATAOS measurements.
Given the deficiencies in the comparison the impression that NATAOS is measuring correct

cannot be regarded conclusive and convincing.

As the referee recommends, we will exclude Neduluha's and Ricaudo's ground-based data for the comparison with NATAOS because of the difference of observing regions. We made plots of time series of the MLS and NATAOS measurements (Fig A). The MLS data points are daily average value at 40km, and the NATAOS data point is average over 5 to 16 Dec, and that duration is indicated by an arrow.
The both ClO mixing ratio show good agreement within errors. We won't claim that NATAOS is more correct than MLS. We will describe only objective results and will delete the speculative sentence "Thus the positive bias to the MLS mixing ratio is likely, although we cannot directly compare them because of the difference of the observing site location and observing time".

page 1924 section 4.4.
I would recommend to remove this section. This is not part of the description of the measurement but an application and is not in the scope of AMT. The data compared are more than 10 years apart and are made in different parts of the world. The authors should explain the aim of the comparison and what one would expect given the fact, that the Montreal protocol led to a significant decrease of anthropogenic chlorine emissions.
The results are only descriptive and the authors themselves point out some major problems encountered by the measurements (resolution, averaging kernels). They also suggest a solution (comparison with model runs) and refer to a later publication.

We agree that detail and long explanation of background and previous studies are beyond the scope of AMT, and some speculative discussions should be removed. However, our main aim of this ClO measurement including instrumental setup is to detect the diurnal variation from Atacama at 204GHz. We stated that the total random error of NATAOS is ~20% at 40km, and this is enough to detect the diurnal variation with only 12 days integration from the ground. We think that the result of the diurnal variation is an appropriate example showing the performance of the NATAOS instrument and data reduction in this study. In this sense, we think that the result of diurnal variation is not mere application but is a part of the measurements. Thus, we believe that showing the diurnal variation is not beyond the scope of AMT.

We will delete large part of this section and summarize to be more concise as follows;

***

The present study shows the ClO diurnal variation in the Southern Hemisphere obtained by the ground-based millimeter-wave measurements over Atacama, Chile. Figure 13 shows the diurnal variation of ClO at 40, 45 and 50 km, averaged over 5–16 December 2009 over Atacama (23° S). The horizontal axis is the local time, LT, in Chile. The vertical profiles are derived from the averaged spectra for every 3-h bin from 00:00 to 24:00 over the 12 days. During this period, the sunrise time and the sunset time were 05:30 and 19:30, respectively, and the culmination time was 12:30 at Aacama. At 40 km, the mixing ratio of ClO quickly increases after the sunrise and reaches up to the daytime plateau level at the time slot of 06–09 LT. The increasing rate at 45 km is slower than that at 40 km, taking more than 3 h to increase up to the daytime plateau level. There is no difference of increase rate depended on altitude in Ricaud et al. (2000).

For the clarified the reason of this difference, we plane to run a model calculation that is fitted for the Atacama site to compare with the observational results. We intend to make such a comparison with model calculation in the next paper.

Figure 6 to 10

I would suggest to combine figure 6 to 10 into one figure. I would be much easier to actually compare the contributions of the different noise sources.

As the referee suggested, we combined Fig. 6 to 10 into one.

Figure 12

Why is there only one measurement from NATAOS. I understood measurements have been taken from 5th to 17th of December 2009. Is this a mean of all profiles? Please explain.

Yes, it is average as the referee think. The marked points show the mixing ratio of ClO retrieved from the spectrum measured 12:00 – 15:00 LT averaged from December 5 to December 16.

We revised this figure caption.

***

ClO mixing ratio at 40 km (orange) and 50 km (purple) which are retrieved from the average from 12:00 to 15:00 over observation period (right-left arrow) at NATAOS and ClO mixing ratio of JEM/SIMLS measurements at 40 km (red open circle) and 50 km (blue open circle) near NATAOS from November 2009 to February 2010. Red and blue closed circles show the ClO daily averaged profiles of JEM SMILES measurements at

40 km and 50 km, respectively.

Figure 13

What denote the error bars? The standard deviation of all measurements during this time?

The error bars denote the total of random error which is described in section 4.2.5.

We revised the figure caption.

Technical issues:

page 1911 line 2:

add an ’A’ at the start of the sentence before Path Length Modulator (PLM)

page 1911 line 3: ".. between the parabolic mirror." There seems a part of the sentence missing.

page 1912 line 20

Either "a translucent medium" or "translucent media"

Figure 12

I cannot find left-light arrows.

Figure 13

Please mention in the caption that this is a mean of all measurements between 5th and 16th of December.

Figure 14 caption

please replace "out" by "our"

in the reference section

what are the numbers behind the reference entries?

We will revise the sentence which is noted at technical issues in you comment.

Reference List

Ko and Sze (1984), Diurnal variation of ClO: Implications for the stratospheric Chemistries of ClONO2, HOCl, and HCl, J. Geophys. Res., 89, 11,619-11,632, 1984.

Janssen, M. A. (ed.) Atmospheric Remote Sensing by Microwave Radiometry John Wiley Sons, Inc., 1993

Jones et al (2011), Analysis of HCl and ClO time series in the upper stratosphere using satellite data sets, Atmos. Chem. Phys., 11, 5321.5333, 2011

Gustincic (1977), J. A quasi-optical reciver design Proc. IEEE. conf., 1997

Mizuno (2002), Millimeterwave radiometer for the measurement of stratospheric ClO using a superconductive (SIS) receiver installed in the southern hemisohere, Int. J. Infrared Milli., 23, 981-995, 2002.

Pickett et al (1992), Submillimeter, millimeter, and microwave specral line catalog, Tech. Rep., 80-23, Rev. 3, Jet Propulsion Lab.

Nagahama et al (1999), Ground-based millimeter-wave observatins of ozone in the upper stratosphere and mesopshere over Tsukuba, Earth Planets Space, 51, 1287-1296,1999

Conner et al (1995), Error analysis for the ground-based microwave ozone measurements during STOIC, JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 100, NO. D5, PP. 9283-9291, 1995

Santeee et al (2008), Validation of the Aura Microwave Limb Sounder ClO measurements, J. Geophys. Res., 113, D15S22, 2008.

Ricaud et al (2000), Temporal evolution of chlorine monoxide in the middle stratosphere, J. Geophys. Res., 105, 4459-4469,2000.

Schieder, R. Kramer, C. (2001) Optimization of heterodyne observations using Allan variance measurements Astronomy Astrophysics, 2001, 373, 746-756

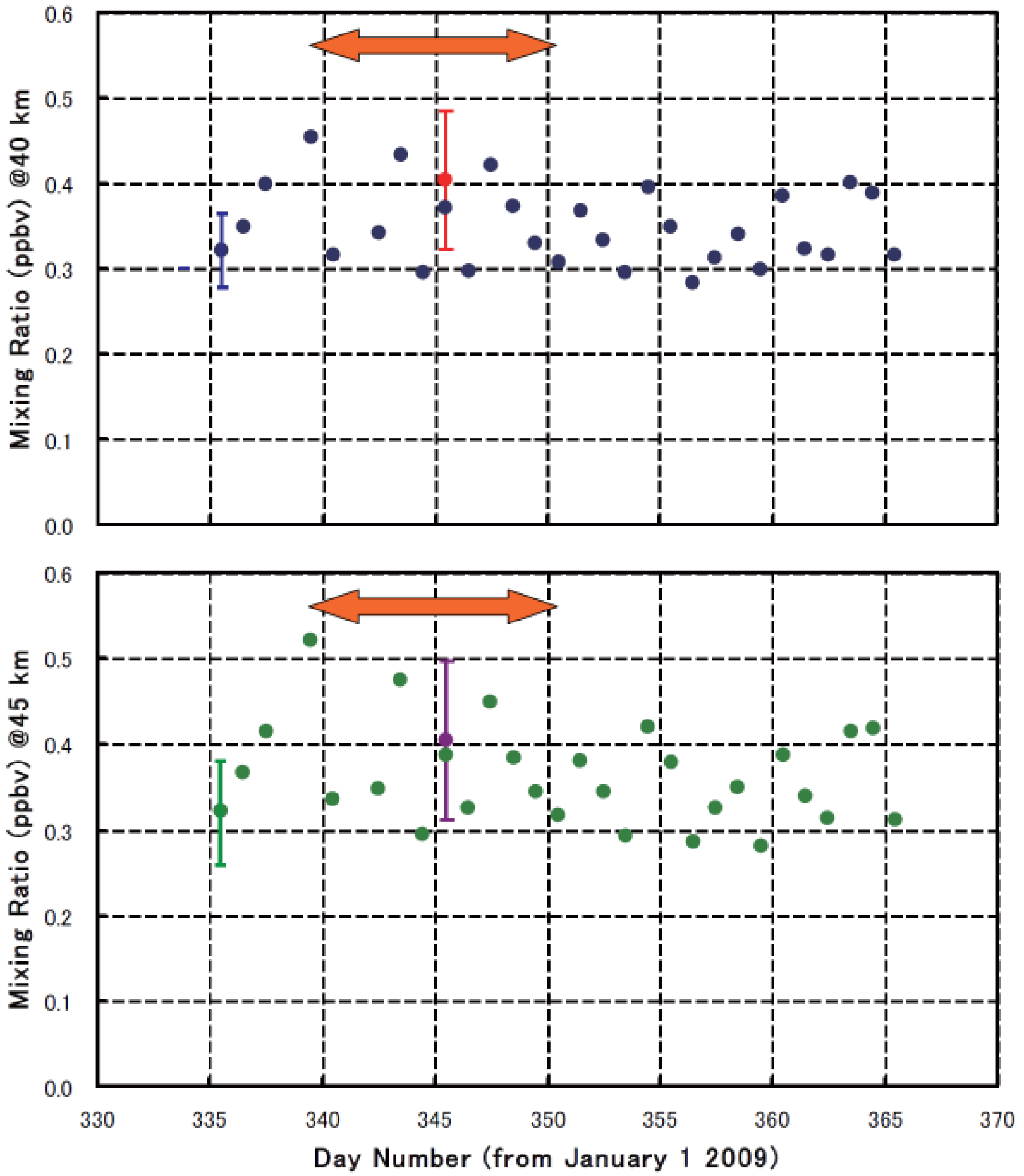

Fig A: The time series of day time ClO at 40 km and 45 km of daily average of AURA/MLS data (blue, green) and that of mean average (12/5 -16) NATAOS data (red, purple). The error bar of AURA/MLS is shown the 1 σ of time variation. The error bad of NATAOS is shown the retrieval error (Section 4.2). The right-left arrow shows the period of observation at NATAOS