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Atmospheric Measurement Techniques
Discussions

Interactive comment on "A new differential absorption lidar to measure sub-hourly fluctuation of tropospheric ozone profiles in the Baltimore–Washington DC region" *by* J. T. Sullivan et al.

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The authors would like to thank the referees for their comments and suggestions. Please find below the comments/answers for each item posted by each referee. The author response/comments are initialized with an "AR -" in front of the text. A revised version of the paper has been prepared.

Author Response for Anonymous Referee #1

1. As already mentioned, the DIAL technique used in this paper is not new and similar systems based on the same technique are currently operated in other part of the world. Although some previous literature is cited (e.g. Megie et al. 1985; several Browell et al. publications), the paper does not sufficiently acknowledge previous work on tropospheric ozone lidar measurements, e.g. regarding the optimum choice of wavelength for tropospheric ozone lidars, the previous use of dye lasers or the optimization of the Raman cell gas mixture.

AR - Additional references have been added based on the referee's request. Wavelength selection is mentioned with appropriate references for previous dye laser systems. References have also been added in the Raman cell discussion for optimization of the Raman cell gas mixture.

2. Similarly, the authors do not mentions lidar systems in parts of the world other then the US, although such lidars exist in Europe (e.g. in Bavaria and Southern France) or in Japan. The most striking example of this lack of reference to previous work is page 4326 line 12, where it is mentioned that the described system is the only one using SRS cells for wavelength generation aside from another system at Jet Propulsion Laboratory. A system using similar wavelengths has been operated for 20 years at Haute Provence Observatory in France (Ancellet et al., JAOT 1989). In Japan, the AMT paper of Uchino et al. (2014) also describes a system based on the Raman shifting technique. In general, the references strongly focus on the lidar work done in the US, while similar work in other part of the world is neglected.

AR - Additional references have been added based on the referee's request. The context of page 4326 line 12 was meant for the lidar with respect to TOLNet only. We understand that this may have led to some confusion and it is now much more clearly stated.

3. I strongly recommend the use of the International System of Units, especially in the section devoted to the optimization of the Raman cell gas mixture. The use of psi

prevents comparison with other published results.

AR - All pressure units have been changed to conform with those used in the international lidar literature.

4. The complete development of the lidar equation for ozone measurements in section 2.1 is not necessary. This has been mentioned in a lot of publications dating back in the eighties, which could just be cited here.

AR - The development of the DIAL equation from the general lidar equation has been abbreviated and additional references have been added.

5. Only one measurement profile is shown in the paper (Figure 5), a part from the measurement time series in Figure9. For clarity, the figure should display the ozone retrieval from the various channels of the receiving system. An estimate of the bias with respect to ozone sonde measurements should be given.

AR - Figure 5 has been altered to show the join regions of the upper and lower altitude retrieved ozone profiles. This gives the reader a much better idea of how much overlap exists between the channels and emphasizes the advantages of having multiple channels to cover the entire troposphere. As a side note, the retrieved profile data that was used was changed from 17:44 (average from 17:39 to 17:49) UTC to 17:54 (average from 17:49 to 1759) UTC based on the ascent rate and coordinates of the sonde as it is much more representative of the atmosphere that is actually being resolved. An estimate of the bias with respect to the ozonesonde is also given in this section.

6. Also for clarity, I recommend to display in a figure the various error sources of the measurements (including aerosols and other absorbers error sources as well as the error due to photon noise) and the vertical resolution as a function of altitude.

AR - Because the statistical noise is the dominant source in the uncertainty analysis of this instrument, we feel it is not necessary to have an additional figure based on the other, smaller, sources of uncertainty. The coupling of the total uncertainty bars

in Figure 5 and all of the sources of uncertainty in Table 3 (described in detail in the Accuracy section of the paper) provide a quantitative description of this uncertainty. For perspective, most of these errors are on the order of 1-2% as compared to 10-25% for statistical uncertainty. During this analysis, the vertical resolutions were constant for each channel and we do not feel that this information in a separate Figure would yield much more information than simply stating it in the text (P4342L18-19).

Specific comments Page 4322, abstract: It is not necessary to explain the DIAL technique in the abstract (lines 12-18).

AR - This portion of the abstract has been abbreviated.

Page 4324, l12: Tropospheric ozone profile measurements from the IASI instrument should be mentioned.

AR - The IASI instrument has now been mentioned along with the other satellites/platforms that have been mentioned.

Page 4326, l14: Here again reference to important publications from Europe on the performance of ECC ozone sonde uncertainty is missing, e.g. Smit et al., JGR 2007.

AR - This reference has been added within the context of current ECC uncertainty.

Page 4326, l25: 1st DIAL ozone measurements were made in 1977 (Megie et al. Nature, 1977). For stratospheric ozone trend measurements, records exist in other stations then in the US (e.g. Godin et al., GRL 1989; Steinbrecht et al., JGR 2006).

AR - Additional references have been added based on the referee's request.

Page 4327, l26: what is the difference between routine monitoring and continuous monitoring?

AR - This instrument has been designed to have the ability to run continuously during an air quality event of interest. When there is no significant event, there have been routine observations (i.e. several hours in the morning or afternoon). This has been changed to emphasize the continuous monitoring ability of the instrument and deter confusion.

Page 4328, l16: Cite previous reference on the optimization of tropospheric DIAL ozone measurements.

AR - Additional references have been added based on the referee's request.

Page 4329, l18: it sounds strange to cite a reference for the lidar equation prior the invention of the lidar and the laser!

AR - The reference given for the lidar equation was indeed prior to the invention of the laser. Middleton and Spilhaus were able to determine the cloud base height by using high voltage sparks to produce a short (one microsecond) light pulse at the focus of a large searchlight mirror and detected the light pulse from the clouds by a photocell at the focus of a second mirror. This has been removed during the abbreviation of the lidar theory section.

Page 4329, l22: from equation 1, P(r) is the energy and not the power. The lidar signal depends on the wavelength, so it should be added in the equation as an index.

AR - This equation has been removed during abbreviation of the lidar theory section of the paper based on the referee's comments.

Page 4329, l23: the receiver efficiency and background signal also depend on the wavelength.

AR - This portion of the text has been removed during abbreviation of the lidar theory section of the paper based on the referee's comments.

Page 4330, l9: the extinction coefficient cannot be considered as absorption coefficient only. What about Rayleigh and Mie scattering?

AR - This portion of the text has been removed during abbreviation of the lidar theory section of the paper based on the referee's comments. Rayleigh scattering is extremely

important and is essential to the correction terms C and D for the DIAL equation. This is now more clearly emphasized. Mie scattering has now been mentioned.

Page 4332, l19: provide earlier reference for dye laser sources used for DIAL measurements (e.g. Pelon and Megie, JGR, 1982)

AR - Additional references have been added based on the referee's request.

Page 4334, l21: There are other references for the optimization of the Raman cells for DIAL ozone measurements, e.g. Papayannis et al., Appl. Opt. 1990; Ancellet and Ravetta, Appl. Opt. 1998; de Schoulepnikoff, App. Opt., 1997; Heese et al., JGR, 2001.

AR - Additional references have been added based on the referee's request..

Page 4339, l24: It is not the right reference for dead time correction. A better reference is Donovan et al., Appl. Opt., 1993.

AR - We have found that correcting for the nonparalyzable dead time correction of the PMTs using equation 9 has led to satisfactory results when comparing the final retrieved ozone profiles derived from the corrected photon counts and the ozone sonde profiles.

Page 4342, l1: A reference is necessary for the assumed value of the lidar ratio.

AR - The value of S= 60 sr was chosen based on the work by Sawamura et. al (AMTD, 2014). These results show comparisons of the lidar ratio for an analysis done on 11 separate days at 355 and 532 nm and show that 60 sr is a representative value of the aerosol loading in the Baltimore-Washington region. A reference has been included.

Page 4347, equation 16 is incorrect: the error on the ozone number density is the quadratic sum of the error of both lidar signals.

AR - This is now clearly stated in the text that the total uncertainty is the quadratic sum of the statistical uncertainty of both lidar signals.

Page 4348, l15: the estimation of the error linked to NO2 and SO2 absorption should be better explained. Why only one value is given and not a range of values? The error depends on the assumption made on NO2 and SO2 profiles.

AR - Although the vertical profile is taken into account for this calculation, the values stated in Table 2 and in the text represent the largest uncertainty from each of the trace gases listed (without a large pollution or volcanic event occurring). This has been changed to reflect that these are approaching the maximum uncertainties. Author Response for Anonymous Referee #2

P4324L5-6 Clarify that while CO2 is measured in the IR, measurement of O3 is generally carried out in the UV. The current text makes it seem that CO2 is also measured in the UV.

AR - This has been changed.

P4325L11-14 There have been several O3 lidars developed using Raman shifted laser sources in the past. It may be appropriate to mention some. A few other systems include those described in the literature (separately) by E. Uthe, G. Ancellet, Y. Zhao, or R. Alvarez.

AR - Additional references have been added based on the referee's request.

P4326L25-29 Consider including reference to the stratospheric ozone lidar work (including long-term measurements) by Langford and Proffitt.

AR - Additional references have been added based on the referee's request.

P4327L12-16 The paper cited (Banta) includes ozone lidar and Doppler (wind) lidar profiles in the lower two kilometers of the troposphere. It is not clear what the author means regarding the lack of information in the first few kilometers.

AR - This has been clarified based on the referee's suggestion.

P4333L10 The length of the Raman cell is stated as 76 cm here, but is given as 1.8

m later in the text and in Figure 3. Is the 76 cm referring to the effective interaction length?

AR - This has been changed to 1.8 m, which is the actual interaction length of the cells.

P4337L17-19 The numbers of telescopes (4 vs 3) and wavelengths (2 vs 3) described in the text do not agree with Figure 3.

AR - This figure has been altered to meet the referee's request and the 266 nm detector has been removed.

P4337L21-22 It may be clearer to state tha the wide FOV channel allows collection of signals from nearer ranges so that, in conjunction with the larger, narrow FOV channel, the lidar can accommodate a wider dynamic range of signals.–The wide FOV itself causes an increase in the dynamic range of the signals reaching the detector (by allowing the very large signals from the near-field into the receiver in addition to the existing weaker signals from the far-field) rather than accommodates it.

AR - This has been changed to meet the referee's request.

P4339L5-9 How does interleaving the pulses cause an offset in the signal? Is it a temporal offset or a signal level offset?

AR - This offset is simply a temporal artifact of the gating scheme that we have put into our system in order to interleave the pulses. The offset and method of determining the offset are important so that we truly know where our first altitude bin begins. This has been removed because of the confusion over such a minor issue.

P4339L15-17 The background correction is described as linear. Is it a constant value, or does it have some slope over the range of data? If so, over what range is the linear fit calculated?

AR - This has been changed based on the referee's request. The background subtraction is a constant background subtraction. For this to be a safe assumption it needs to be done in a region where the slope of the background is close to zero (i.e. no counts from the actual laser light). For the data presented in this paper, this region is between 30 and 45 km.

P4339L23-25 Is this correction for just the PMT pulse "pileup" or does it include the counter dead time as well? i.e. would a faster counter improve this result?

AR - Currently our correction is for the convolution of the pulse "pileup" and counter dead time. A faster counter might be somewhat better, but we are really trying to correct two effects with one equation. The result is a correction that works for some of the non-linear range but not all. Because of this, we have had to determine an empirical value of maximum counts/shot that will still conform to this correction. We have had satisfactory results in correcting the raw signals for the final ozone profile when we are below this saturation threshold and it dictates the altitude join regions.

P4341-4342 What is the typical reference height used for the aerosol profile calculation? Also, generally, these inversions are sensitive to the direction that the iterations are carried out (from far to near ranges or from near to far ranges), so a comment on the stability of the inversion and how it influences the choice of reference range is useful.

AR - The typical reference height is mentioned in P4341-L22. A comment has also been added about the stability of the iterative technique.

P4343L13 The humidity and temperature profile information mentioned here should refer to Figure 7 as well.

AR - This has been changed based on the referee's request. This information has been moved further down to where it is more applicable with Fig. 7.

P4345L13 On 25Oct2013, 1519-2218 UTC would be 1119-1818 EDT or 1019-1718 EST.

AR - This has been corrected.

P4348L28-P4349L1 It would be good to describe the saturation correction and how it is determined. What saturation effects have been included? (Note that this is one of the two larger corrections that cannot be reduced by additional averaging.) Also, see earlier comment regarding dead time correction.

AR - Additional comments have been added to the discussion on this correction based on the referee's comments.

Table 2. Add the diameters of the transmitted beams and the focal lengths of the telescopes to the specifications. This allows readers to more fully understand the overlap ranges for the various receivers.

AR - These parameters have been added to the table based on the referee's request.

Figure 3. What is the beam that passes through the angled mirror/beamsplitter on the left side of the figure? It currently seems to just pass out of the detection system.

AR - This has been removed based on the referee's request. This beam was used in previous missions to detect 532 nm. It has not been used with this system and is therefore of no interest to the reader.

Figure 6. It would very useful to add a mark on the map to indicate the location of the TROPOZ lidar.

AR - An annotation to the map has been added based on the referee's request.

AR - All of the following technical corrections have been implemented P4324L6 "ultraviolet" does not need a hyphen. P4325L23-25 This is a run-on sentence. Consider replacing first "and" with a comma (and add a comma before the second "and"). P4328L22 "above" is repeated P4331L2 Equation (5) has an extraneous comma that should be removed. P4335L1-3 The use of "internally" may be ambiguous here (could refer to the design and fabrication of the cell interior or the location of the cell construction). Perhaps "in-house" is clearer? P4336L21 "comprised" should be "composed" P4336L24 "optic" should be "optical" P4339L12-14 This is a run-on sentence and seems to say the same thing twice. P4340L20 “of” should be “or” P4345L21 “maintaing” should be “maintaining” P4346L12 “small yield” should be “small to yield” P4350L1-3 The use of the word “obtainable” is ambiguous here. It seems to suggest the use of the ozonesonde to collect the proposed lower altitude ozone data (rather than the lidar data being extended to lower altitude ranges).

Author Response for Anonymous Referee #3

GENERAL COMMENTS: The paper presents a DIAL system for ozone profiling in Baltimore-Washington DC region. In fact the system presented here, is based on previously developed DIAL systems during the end of ‘80s and early ‘90s. Therefore, in no case it can be regarded as “new”. As a consequence the title of the paper has to be changed to ”An ultraviolet differential: : :”. The authors present their O3 DIAL system, in detail, presenting experimental SRS data using H2, D2 as active Raman gases with He as a buffer gas. Based on previous published work on existing UV DIAL systems and the corresponding SRS technique at 266 nm (using H2, D2 as active Raman gases and He as a buffer gas) the methodology and data presented here are not at all new, they are of low scientific significance, and therefore, they are not of any interest to publish them again in a Journal as the AMT, which promotes new work and new ideas. As a conclusion, the paper should not be published in AMT but in another relevant journal and should focus more on data case study analysis. Previous work on DIAL and SRS systems: Browell et al., A.O., 1985 Ancellet et al., JAOT, 1989 ,Hanner and McDermid, IEEE JQE, 1990 Papayannis et al., A.O., 1990 Tzortzakis et al., Appl. Phys. B, 2004

AR - We feel as authors that it is important to publish it as a new instrument based on the fact that it will be the first ozone lidar within the U.S. and many other countries to use Raman cells for wavelength generation, have transportable capabilities, and have the ability to profile ozone from near the surface to beyond the tropopause with multiple receivers during the daytime. The emphasis in the title was not meant to suggest that it is the first ozone dial instrument, but rather to let the air quality community know that our instrument is now operational (along with several other lidars within the network),

that we can provide quantitative ozone profiles (with good agreement with co-located ozonesondes) and the instrument has captured a very interesting regional ozone test case.The title has been changed to ”A mobile differential. . .”. For the United States to begin to routinely monitor tropospheric ozone within a network, it is important for the instruments to be validated, published, and discussed.

SPECIFIC COMMENTS P4322 L15: Replace : : :”deuterium” with “deuterium, using helium as buffer gas”.

AR - This change has been made based on the referee’s request.

L22-23: The uncertainties estimated here are not at all realistic ones; they are optimized. It is well known that aerosol concentrations in the lower troposphere, alone, can induce very large errors in the retrieval of the ozone profiles (if not corrected) of the order of 25%. If they are corrected by using the Klett or the Raman technique they can be lowered to 15 and 5%, respectively (cf. relevant papers by Browell et al., Applied Optics, 1985; Papayannis et al., Applied Optics, 1990). In the paper the authors do not specify how they correct the aerosol induced errors. Therefore, the errors presented in the abstract should specify this. Also, after revising the retrieved uncertainties, the authors should add “according to the relevant aerosol concentration aloft” after the “: : :to 12 km.”

AR - The abstract has been changed to briefly describe the aerosol correction in place. We have implemented an aerosol correction using an iterative method (based on Browell et al., 1985) and it has been described in detail throughout P4340 – 4342 of the current text. Additional references and explanations have been added based on the suggestions of the previous referees. The co-located sun photometer measurement of the AOD (500 nm) for this day was on average 0.07 during the cloud free portions of the day. This, as compared to the regional aerosol loading in the summer is on the order of a factor of 10 less AOD. We understand that these uncertainties will be much larger during aerosol and air quality episodes. The analyzed data set is much more focused on the large intrusion of ozone in the upper free troposphere and lower stratosphere in which aerosols have not played a large role. Because of this, we intentionally shifted the focus of the analysis and discussion to the controlling meteorology and evolution of the large anomalous ozone reservoir and it's associated intrusion (at 6.5 km) from the stratosphere. We do agree that an aerosol uncertainty analysis with respect to the DIAL equation and ozone lidar is an extremely important and difficult challenge. We feel that this would be out of the breadth of this publication and could be the topic of a completely separate publication. Because the AOD was fairly low and the aerosol aloft was not very concentrated, the corrections we have in place are actually very useful in quantifying the uncertainty due to aerosols in a typical "clean" environment and can be the foundation of another discussion.

P4323 L17: Add after Langford, 1999" also "Stohl et al., 2003)

AR - This reference has been added based on the referee's request.

P4325 The section in L12-14 is confusing. It should be stated in the text that "Aside from the site at: : :..this is the only instrument implemented in the Mid Atlantic currently utilizing high-pressure: : :"

AR - This has been changed based on the previous referee's (Anonymous #1) request. We understand that this may have led to some confusion and it is much more clearly stated that this is the first in the Mid Atlantic region.

P. 4326 L6: cite here all relevant work. L22: cite here work by Pelon and Megie (JGR, 1982), Trickl et al. (ACP, JGR: : :), Galani et al. (JGR, 2003)

AR - These references have been added based on the referee's request.

P4328 L14: Replace "This method: : :" by "The DIAL method: : :" L16: at the end of the phrase cite : Pelon and Megie, JGR, 1982; Measures, 1983. L25: After ": : : 300nm" add a citation (Pelon and Megie, JGR, 1982). P4329 L4: Cite also (Ancellet et al., 1989; Papayannis et al., 1990; Tzortzakis et al., 2004)

AR - These references and corrections have been added based on the referee's request.

Section 2.1 is well known and detailed presented in the literature. The authors could just provide a short description of the DIAL equation referencing all relevant papers (by Measures, 1983; Browell et al., 1985; Papayannis et al., 1990).

AR - Section 2.1 has been abbreviated based on the referee's suggestion.

P4331 Section 3.1. This is very well known. This section has to be shortened, as it has already been published (Measures, 1992; Tzortzakis et al., 2004 and references there in).

AR - This sectioned has been abbreviated and cited based on the referee's suggestions. Additionally, Table 1 has been removed, as it does not add anything new to the current discussion from previous literature with respect to the vibration Raman frequencies.

P4335-4336 All pressures have to be reported in bar. L20. The authors present a photon conversion efficiency of 10% or less, without the use of a buffer gas in SRS generation. The use of He as a buffer gas increases the photon conversion efficiency up to 53% (P4336, L6) for H2 and up to 27% (P4336, L10) for D2. However, it is well known these efficiencies are quite lower than the relevant ones reported in the literature (cf. >70% for H2 and >45% for D2, in Tzortzakis et al., 2004). Therefore, section 3.2 is not worth publishing and can be combined with section 3.1, by just mentioning, in a couple of lines, the obtained conversion efficiencies at H2 and D2 using the He as a buffer gas.

AR - All pressure units have been changed to conform with those used in the international lidar literature. Although a similar discussion exists with a fixed instrument detailed in Tzortzakis et al., (2004), almost all of the beam parameters (diameter, repetition rate) and Raman cell characteristics (confocal beam parameter, photon interaction volume) differ. For example, Tzortzakis et al., (2004) concludes that pumping 10 mJ of 266 nm at 10 Hz into a single 60 cm (2 cm inner diameter) Raman cell is the preferred method for future research. With this described hardware setup and photon efficiencies (70% w/ H2 and 45% w/ D2), the lidar would be profiling the atmosphere with 7 and 4.5 mJ of energy at the Raman wavelengths. For our research applications, especially in our interest in monitoring stratospheric tropospheric exchange of ozone with a mobile system, we have had to use a pump source with a much higher pulse energy and repetition rate, that have yielded different results than the previous literature. We did notice a much larger amount of thermal blooming and activation of higher Stokes orders at 50 Hz than Tzortzakis et. al (2004) has shown at 10 hz, and because of this we carefully made a quantitative assessment of the addition of Helium as our buffer gas to optimize our DIAL wavelengths. We have presented results that indicate that although our Raman photon conversion efficiencies are less, we have still produced a much more energetic pulse of light (17mJ and 11 mJ), which ultimately will allow us to resolve much more of the atmosphere. Especially for future instruments that may be portable and/or are interested in characterizing the ozone in the upper free troposphere, this discussion is pertinent has remained in the manuscript.

P4336 L26: The emitted laser energy per pulse at 266 nm has to be mentioned.

AR - This has been mentioned based on the referee's request. It was already labeled in the Hardware Specifications (table 2), but it does need to also be clearly stated in the text.

P4342 L1: How the S=60sr is chosen? The authors should make a sensitivity analysis about the possible values of the lidar ratio for the Baltimore-Washington region. L1: Please correct the units of S. It is in steradians (sr).

AR - The value of S= 60 sr was chosen based on the work by Sawamura et. al (AMTD, 2014). These results show comparisons of the lidar ratio for an analysis done on 11 separate days at 355 and 532 nm and show that 60 sr is a representative value of the

aerosol loading in the Baltimore-Washington region.

P4347 This section can be put in an Annex, as it is well known in the published literature. To my opinion, this section could focus on a discussion of the different error sources in the retrieval of O3 by DIAL in the lower atmosphere. More excessively, the authors should focus on the role of aerosols in this uncertainty of the retrieval of ozone (the work of Browell et al., 1985 and of Papayannis et al., 1990 could be the source of the literature used). It is important that the authors show how they correct the aerosol interference and how they estimate the relevant errors due to the presence of aerosols (see also my comments previously provided in for P4322, L22-23).

AR - Please see our previous comments.

P4350 L9: The use of 266 nm in the retrieval of O3 by DIAL inide the PBL (together with 289 nm) is already presented and used in the literature (cf. Papayannis et al., 1990, as well as other later papers of the same author).

AR - We understand that this has already been done and believe that this may be useful in the future to more accurately determine the regional aerosol influence on the retrieval. We have cited Papayannis et al.,1990.

P4358 The vibrational Raman frequencies of S1 for H2 and D2 have to be updated. The values given by Measures 1983, have been updated in the later years. The authors are invited to use the new values, referring ONLY to papers dedicated in the Raman spectroscopy and not to a book (Measures, 1983) which is just reporting old values.

AR - Based on previous abbreviations to the section on SRS theory, this table has been removed. This would not have been an appropriate publication source for these values alone.

P4360 Table 3. The aerosol uncertainty has to be more precisely estimated. Please update and see my previous comments.

AR - We believe that this discussion has been addressed and updated in our response to comments by the first two referees. Please see our previous comments.

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 4321, 2014.

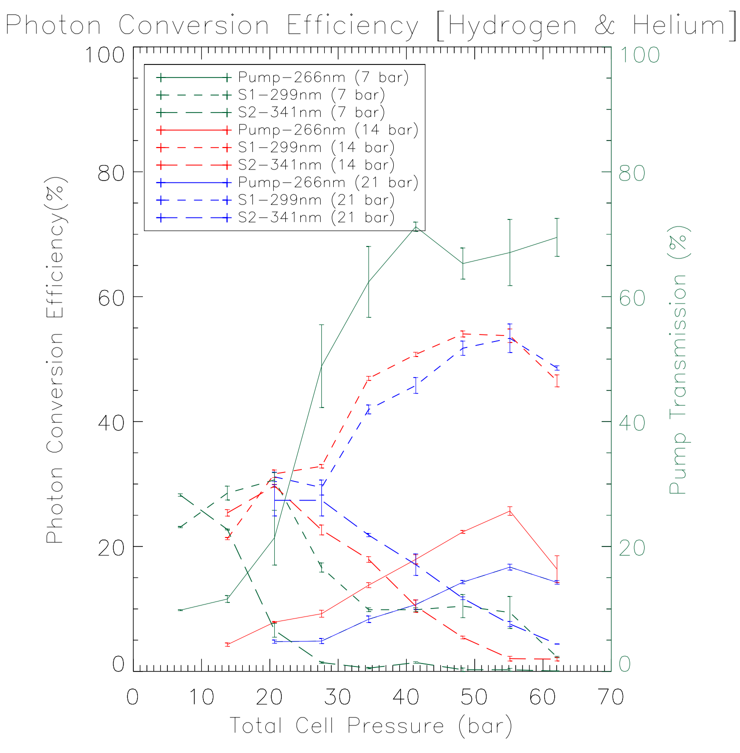

Fig. 1. Revised Raman Cell units (H2)

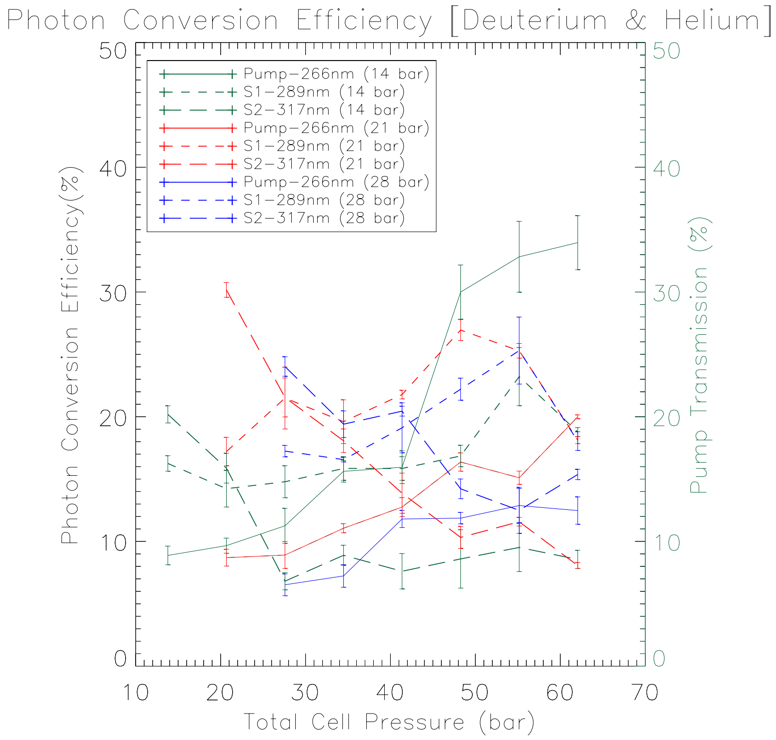

Fig. 2. Revised Raman Cell units (D2)

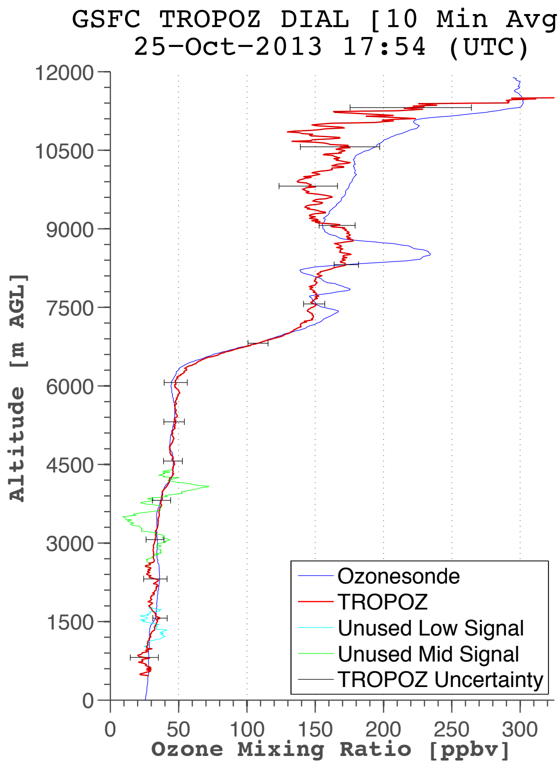

Fig. 3. Revised Ozone Profile with Channels

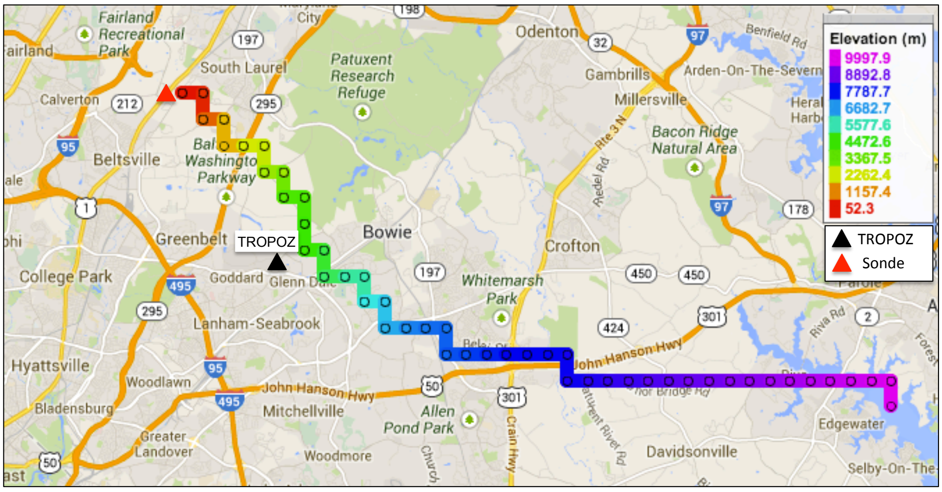

Fig. 4. Updated Sonde Map Location

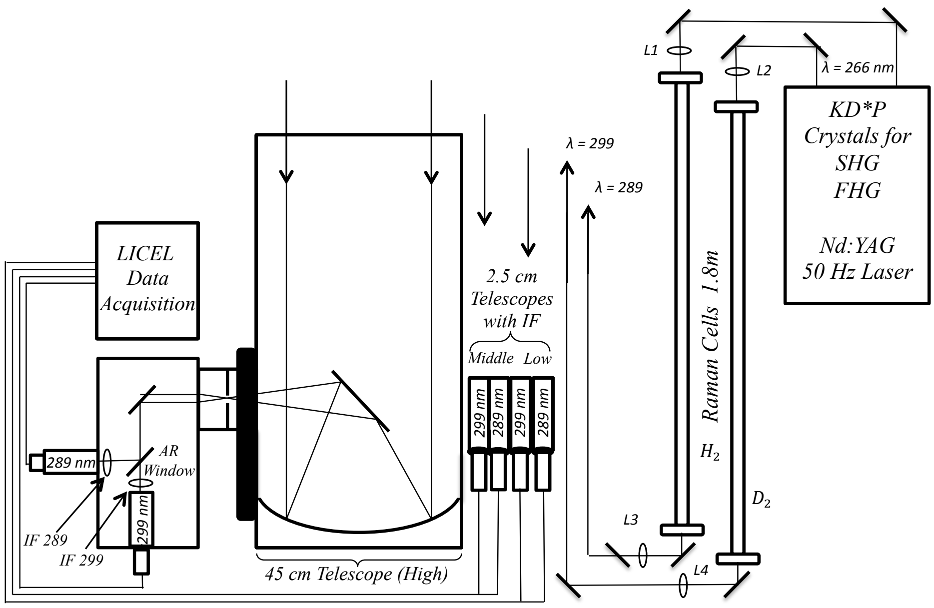

Fig. 5. Updated Hardware Schematic