Supercontinuum laser sensing of atmospheric constituents

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ABSTRACT

Extending our developments of a previously reported supercontinuum lidar system has increased the capability for measuring long path atmospheric concentrations. The multi-wavelength capability of the supercontinuum laser source has the advantage of obtaining multiple line differential absorption spectra measurements to determine the concentrations of various atmospheric constituents. Simulation software such as MODTRAN™ 5 has provided the means to compare and evaluate the experimental measurements. Improvements to the nanosecond supercontinuum laser fiber coupled transceiver system have allowed open atmospheric path lengths greater than 800 m. Analysis of supercontinuum absorption spectroscopy and measurements utilizing the updated system are presented.

Keywords: DIAL, DOAS, MODTRAN, supercontinuum laser, differential absorption

1. INTRODUCTION

The Supercontinuum Absorption Spectroscopy system (SAS) at Penn State was developed as a way to utilize a broad band source to conduct multiple-line differential absorption spectroscopy (DAS) for measurements of atmospheric trace species. While there are various approaches and techniques for implementing DAS, a supercontinuum laser provides the capability to simultaneously probe the atmosphere with a very large number of wavelengths. Absorption of various trace species are able to be observed, together with any interfering species contributions when the supercontinuum is spread across many wavelengths in the optical spectrum. By implementing a multiple-line (hundreds of lines) differential optical absorption spectroscopy (DOAS) analysis methods are applied to analysis of the atmospheric path return measurements to quantify absorption and converge to the species concentration values. Since the system reported utilizes a quasi-continuous wave (CW) supercontinuum source and the measurements are path integrated, it is not range-resolved and therefore does not fit the true definition of a lidar system (light detection and ranging). The system also has an advantage over DIAL techniques where the laser line shape must be matched against the spectral line to obtain accurate concentrations. Its' advantage over the DOAS approach lies in the narrow beam laser properties. This approach is a variation that extends the capabilities of three techniques that are long established for optical absorption of chemical species; DIAL, DOAS, and white cell spectrometry.

The SAS system at Penn State is improved over previous versions[1,2] to operate over horizontal path lengths approaching 1 km. The configuration reported operates on an 890 m path across the downtown State College area (previous configurations operated over 150, 300, 540 m paths across the Penn State University campus). The advantage for utilizing the longer path is twofold. The extended path length provides the opportunity to test the SAS system over ranges where added atmospheric effects such as turbulence and scattering are considered. Also, the longer path is more useful for real world applications for pollution monitoring and mapping. Second, larger path integrated absorption values of lower concentration trace species are obtained. This leads to greater ability for detection of atmospheric species that exist at lower background absorption levels; effectively increasing the signal to noise (SNR) and the ability to obtain greater accuracy in concentration measurements.

To demonstrate the benefits of the longer path absorption configuration, measurements of atmospheric oxygen were conducted and compared with MODTRAN™ 5 simulation data over the 890 m path. These results are further compared with previously reported measurements at shorter path lengths and additional measurements conducted on various absorption features shown in the simulated MODTRAN transmittance plot in Fig. 1. A similar inversion algorithm to that which was used on the previously reported measurements[1] was applied to analyze the measurements presented here.

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2. MOTIVATION FOR SINGLE SOURCE BROADBAND DETECTION OF ATMOSPHERIC SPECIES

There are several reported systems of various types that implement differential absorption techniques for detection and quantification of atmospheric species. Differential absorption lidar (DIAL) and many DAS systems are optimized for detection of a single or only a few species. These systems are often bulky and possess complicated optical layouts in order to operate at two or more wavelengths. On the other hand, broadband DOAS approaches implementing a single incoherent light source are inherently less complicated and often are capable of sensing several species with little or no modification to their current operating state. However, DOAS systems tend to experience difficulties with extended range and limited sensitivity. The accuracy of long path measurements using any one of these techniques is highly dependent on the configuration of the source(s), detector(s), the absorption path, and the method of analysis for quantification of atmospheric species. Motivation for a supercontinuum source for performing absorption spectroscopy, detailed in Section 3, is based in optimizing long path analysis techniques, reducing error sources, and improvements in sensitivity. A comparison to previous systems discussed in this section.

2.1 Long path absorption analysis and sources for error

Long path absorption spectroscopy applying DIAL and DAS techniques are implemented today using either multiple light sources, tunable sources, or a single broadband source. Each technique has its own inherent strengths and weaknesses for detection of atmospheric species. Common across all systems are the basic methods used in differential absorption analysis.

Concentration path lengths for absorption species are calculated by modifying the lidar equation for elastic scattering and long path implementation of a system operating at two or more wavelengths. Differential absorption analysis is then applied to the ratios of the measured receive energies at the operating wavelengths from a topographical or gaseous target. Several methods exist for implementing this technique; typical DIAL systems select operation wavelengths that are located on (λ_{on}) and slightly off (λ_{off}) an absorption feature to maximize the cross sectional difference (σ_{diff}) of the DIAL wavelength pair. DOAS techniques operate at multiple wavelengths spanning several absorption features to perform multiple differential absorption calculations. Each method utilizes the modified equation for differential absorption analysis,

$$\int_0^{R_T} N(R)dR = \frac{1}{2\sigma_{diff}^{A}}\left(\ln\left[\frac{E_{rec,off}(\lambda_{off},R_T)}{E_{rec,on}(\lambda_{on},R_T)}\right]\right), \tag{1}$$

where the differential absorption cross section is,

$$\sigma_{diff}^{A} = \sigma^{A}(\lambda_{on}) - \sigma^{A}(\lambda_{off}), \tag{2}$$

and R_T is the range to the topographical or gaseous target, $E_{rec,on}$, $E_{rec,off}$ are the respective energies received at online and offline wavelengths, per pulse [J], and $N(R)$ is the number density of the absorbing species [$\#m^{-1}$].

This modification and simplification of the lidar equation for calculating concentration path length limits the long path absorption approach to only three parameters that vary based on system configuration and atmospheric conditions. These parameters are the range to the target or length of absorption path, the differential cross section of the spectra, and the ratios of the received energies. However, there are several additional sources of error that may exist in systems implementing differential absorption techniques. In Brown[3], five major contributions to noise and fluctuation in measured return signals are outlined and are; uncertainty in energy measurements including electronic noise and statistical error, effects of ground or target reflectivity variations, uncertainty in atmospheric optical depth, match of the online laser to the selected absorption feature, and interference from additional atmospheric phenomena such as transmission scattering from the path or absorption by other species. Several of these contributions to error were analyzed in system and algorithm development for the Airborne Natural Gas Emission Lidar (ANGEL) system operated by ITT Space Systems.[4] ANGEL is a three wavelength, DIAL system based on sensing natural gas pipe leaks and quantification of the concentration path length values. A number of advanced hardware and software techniques were developed to specifically address several of the error sources outlined by Brown[3] including contributions from beam mismatch on an absorption feature, mismatch in beam overlap on the hard target or target volume in space, and timing of transmitter/receiver pulse emission/detection.

2.2 Applications of long path absorption spectroscopy and multiwavelength, multiple species detection systems

The ANGEL system mentioned previously implements a true DIAL configuration for atmospheric species detection and overcomes many of the shortcomings of traditional DIAL systems by implementing various error source correction techniques. Analysis of the current system has helped to form the foundation of methods for detection of several hydrocarbons utilizing a multiwavelength approach.

Several additional multi-wavelength approaches exist for simultaneous measurement and detection of multiple species. DOAS systems as mentioned previously operate using high power, incoherent light sources for measurement of several pollution contaminants such as ozone, nitrogen dioxide, and sulfur dioxide. DOAS systems typically focus on detection at ultra-violet and visible wavelengths due to the nature of their light source. The incoherent source provides a smooth, continuous spectrum across these wavelengths allowing for simultaneous detection of the various atmospheric species.

The terawatt supercontinuum lidar[5,6] offers another approach utilizing similar supercontinuum generation similarly to that implemented in the Penn State SAS system. Operating at extremely high power levels, the Teramobile White Light Lidar transmits 5 TW (350 mJ, 70 fs pulses) at a 10 Hz repetition rate for generating supercontinuum in air[7]. Absorption measurements of several atmospheric species are obtained using this high power lidar configuration.

3. SUPERCONTINUUM ABSORPTION SPECTROSCOPY

A supercontinuum laser source offers several advantages over other multiwavelength sources in obtaining differential absorption measurements of atmospheric species. A supercontinuum source provides spectrally broadband emission in the visible and infrared that is essentially continuous across several hundred nanometers in wavelength. Absorption features of multiple atmospheric species and overlapping interferents are observed simultaneously, when coupled with a high resolution spectrometer or optical spectrum analyzer. Absorption spectroscopy implementing the supercontinuum source is being developed at Penn State for obtaining concentration values of various chemical species in the atmosphere over several hundred meters path lengths. Supercontinuum generation and the development of the sensor at Penn State are discussed in this section.

3.1 Supercontinuum generation

A supercontinuum fiber laser source is the transmitter implemented in the Penn State SAS system. The broadband supercontinuum is generated by coupling a 40 mW average, pulsed sub-nanosecond 1064 nm laser (JDSU NP-10620-100) into a 18 m photonic crystal fiber (SC-5.0-1040, Blaze Photonics), which generates about 18 mW supercontinuum output at the fiber end. The supercontinuum is generated as the extremely intense pulses undergo a process of self-phase modulation (for a thorough discussion of supercontinuum generation in photonic crystal fibers, see Alfano[8]). The supercontinuum output is spread across the visible and near-infrared spanning continuously in wavelength from 500 nm to approximately 1650 nm. Fig. 3 reveals several of the atmospheric species capable of detection within the bandwidth of the supercontinuum laser. The simulation provides MODTRAN™ 5 calculations for transmittance across an 890 m path that is utilized in the current configuration of the Penn State SAS system.

3.2 Development of SAS at Penn State

The SAS system at Penn State has undergone several phases of development in acquiring the capability to measure atmospheric species. Initial proof of concept studies were conducted in a laboratory environment for measurements of indoor humidity across a 20 m path. These measurements were captured using an optical spectrum analyzer at 0.4 cm^{-1} resolution and correlated well with simulated transmittance calculations for water vapor from 1380 to 1420 nm. The calculated water vapor concentrations also agreed well with a sling psychrometer used at the time of measurement[3,9].

The next phase of SAS development relocated the transmitting and receiving system to a roof-top location and utilized the supercontinuum laser for atmospheric path measurements. Atmospheric path configurations of 150 m and 300 m across the Penn State campus and transmission of 10-15 mW supercontinuum provided successful measurements of water vapor that again compared well with local MET station humidity measurements. Strong correlation with simulated transmittance spectra was achieved and reported in Brown et al.[2]. Modifications to the path and location of the retro-reflector target extended the path to 540 m, where analysis of atmospheric oxygen was performed. A Princeton Instruments SP-2500 CCD Spectrometer with a 1200 g/mm grating provided high resolution measurements of the

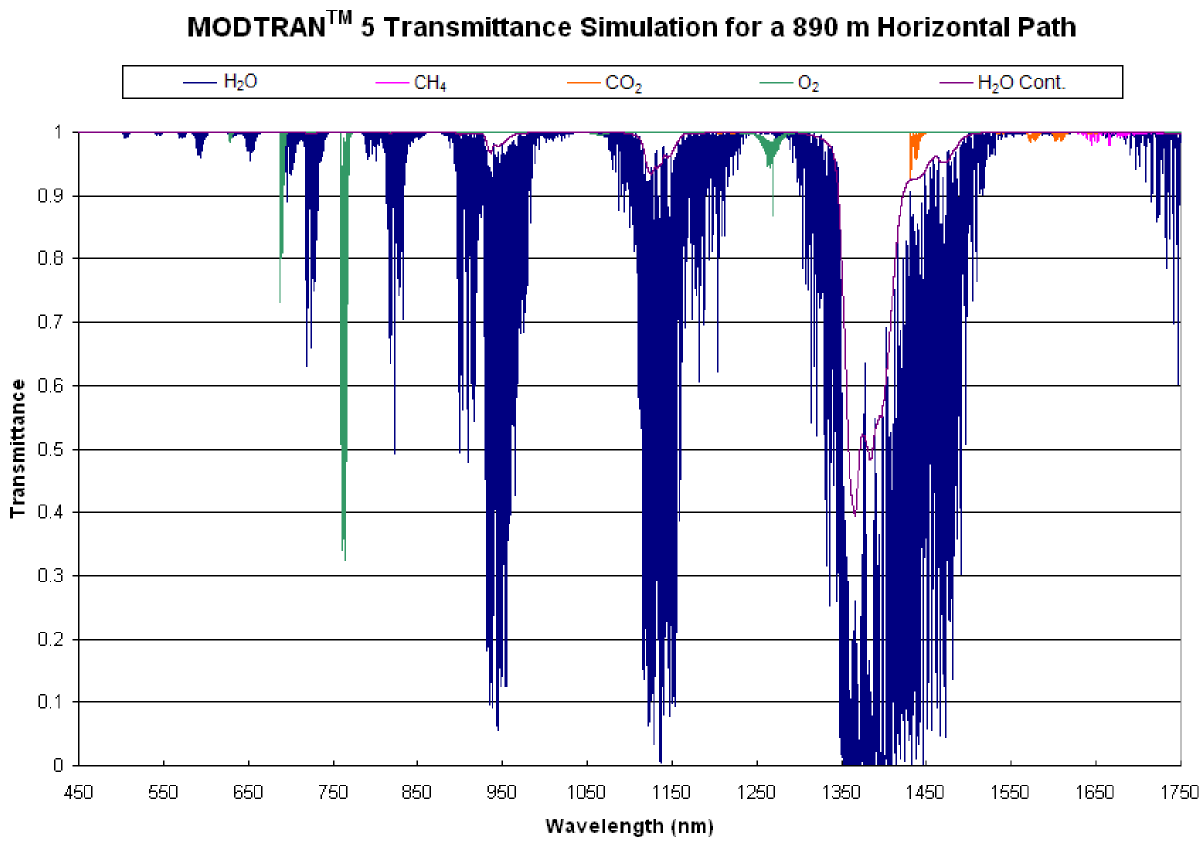

Fig. 1. MODTRAN™ 5 simulation for a horizontal path of 890 m corresponding to the path used in the reported SAS configuration. The simulation provides the transmittance spectra for several atmospheric constituents that exist within the spectral bandwidth of the supercontinuum laser.

absorption band of oxygen centered at approximately 763 nm on the optical spectrum. Inversion of the measurement resulted in a calculation of atmospheric oxygen concentration within 0.2% of the 20.95% standard[10].

The measurements for normal constituents in the atmosphere that possess strong absorption bands in the visible and near-infrared demonstrate the capability of the SAS system and are used to characterize the system for measurements of trace species and atmospheric contaminants. The detection capabilities of the configurations at 150 m, 300 m, and 540 m work well for the strongly absorbing species present; however a longer path length is desired for improved detection of the addition species and the first phase of this configuration is presented in the following section.

3.3 Configuration for long path measurements

The SAS system at Penn State is built primarily from commercial off the shelf components with only a few customized optic mounts build in-house. Improvements to the hardware over the previous system include a 10-inch receiving telescope which offers a 6x increase in receiving area over the previous 4-inch telescope configuration. Beam steering capability is added to the system via placement of a 14-inch turning mirror along the optical path of the sytem. A 6-inch gold flashed corner cube retro-reflector is used as a means to maintain the co-axial alignment between the transmitted and received beams (with only a small lateral shift); thus allowing for co-location of the transmitter and receiver as in previous configurations of the system. The retro-reflector is placed on the roof-top of a downtown area five-story parking garage and the transceiver system is located on the roof-top laboratory of the Penn State Electrical Engineering East building.

A depiction of the operation of the SAS system is shown in Fig. 2. The supercontinuum fiber laser described previously is located in a third floor laboratory and is coupled to the roof-top laboratory through approximately 30 m of

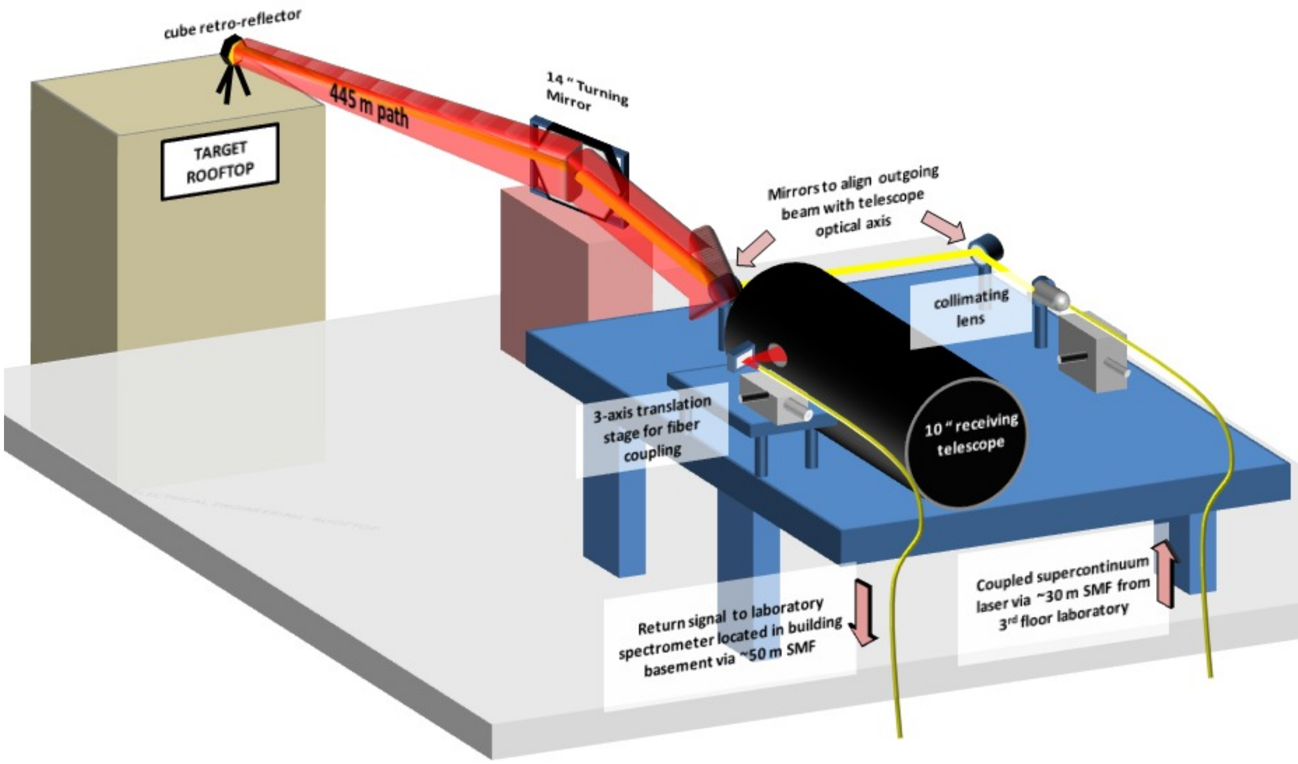

Fig. 2. Configuration of the Penn State SAS transmitter, receiver, and optical path.

Corning SMF-28 single mode fiber. Approximately 3 mW of average power is transmitted through the fiber for the initial measurements used to verify the operation of and characterize the updated SAS system. The fiber output is then collimated through an objective lens that closely matches the numerical aperture of the fiber and is aligned along the optical axis of the telescope via two micro-adjust mirrors. Alignment of the system is done by viewing the laser spot on an adjacent building's rooftop and adjusting until centered on the axis of the telescope. The turning mirror placed in the path of co-aligned transceiver system turns the beam towards the retro-reflector located at the parking garage at approximately 445 m distance. The telescope receives the retro-reflected return and the received light is focused into another SMF-28 single mode fiber that is coupled to the high resolution CCD spectrometer used in previous configurations of this system. In Fig. 3, part (a) provides a map of the transmission and return path between the two buildings with respect to their locations on the Penn State campus and downtown State College area. Fig. 3 (b) is a view of location of the retro-reflector from the roof-top laboratory, and Fig. 3 (c) is a picture of the roof-top transceiver system during one of its' various configuration states.

4. RECENT MEASUREMENTS USING THE LONG PATH CONFIGURATION OF SAS

The SAS system was used to perform measurements over the extended long path configuration of 890 m during an April 2009 demonstration of the system. Measurement of atmospheric oxygen was conducted to verify system performance and for comparison to previous measurements along shorter paths.

4.1 High Resolution Oxygen Measurement

The spectral region from approximately 750 - 775 nm was examined for measurement of the atmospheric oxygen absorption feature. Within this region there exist only a few water vapor absorption bands which are easily filtered out providing an analysis of the concentration path length value by use of the multiwavelength maximum likelihood estimatation (MLE) inversion algorithm used in analysis of previous measurements[1]. Fig. 4 shows the measured absorption spectra captured by the high resolution CCD spectrometer configured for an effective resolution of 1.43 cm^{-1}. Comparison is made to a MODTRAN™ 5 simulation by adjusting the simulation for vacuum-air index differences at the recorded temperature during measurement. A fifteen measurement average with 0.0117 1σ deviation across average is also over-layed in Fig. 4 to demonstrate consistency in the measurement. The measurements were recorded during a 10 minute interval and consistent mode-filtering techniques were applied to the fibers used for coupling the laser source and the spectrometer to the roof-top transceiver configuration. The measurement to simulation comparison shows good correlation across several of the "double-peaked" sections of the absorption feature as well as within the densely peaked feature centered at approximately 760.5 nm. The differences of several of the peak values compared to simulation may be attributed to losses in the fiber coupling, although this needs to be further examined.

Fig. 5(a) shows the measured absorption spectra conditioned for processing through the MLE inversion algorithm. The measured spectra are convolved with a 6.4 cm^{-1} slit width window to smooth the data for better convergence from the MLE. The same window is convolved with high resolution HITRAN data, which provides the absorption cross-sections utilized in the calculation of the concentration path length. The MLE inversion is provided in Fig. 5(b). The calculated concentration value from the measured absorption spectra of atmospheric oxygen across the 890 ± 5 m path

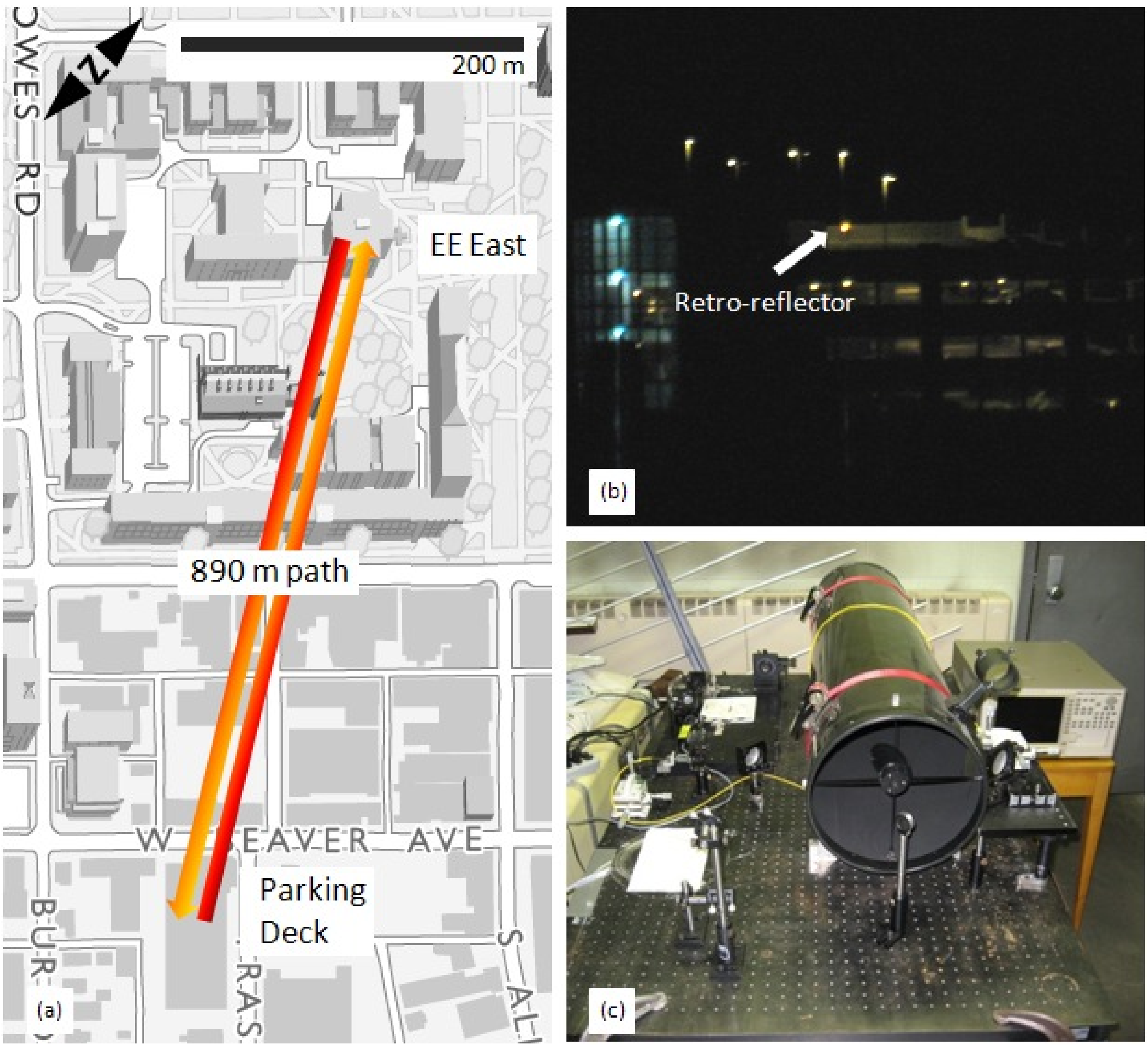

Fig. 3. (a) Map of the long path configuration of the Penn State SAS system, (b) view of the return from the retro-reflector (located on parking deck) before the turning mirror, (c) configuration of transmit and part of the receive portions of the Penn State SAS system on the roof of Electrical Engineering East.

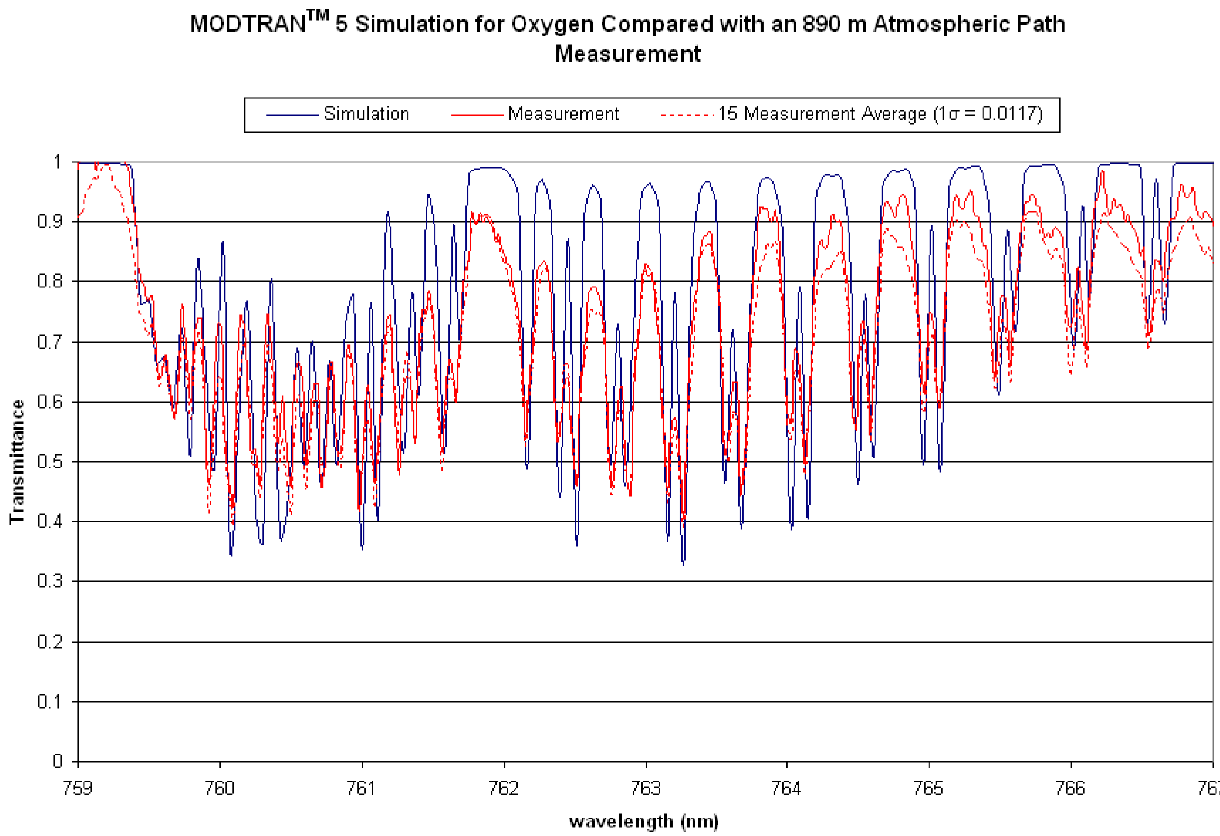

Fig. 4. Simulation and raw measured data adjusted for vacuum-air index of refraction differences for the oxygen absorption feature centered at 763 nm; the fifteen measurement average is taken during a 10 minute period.

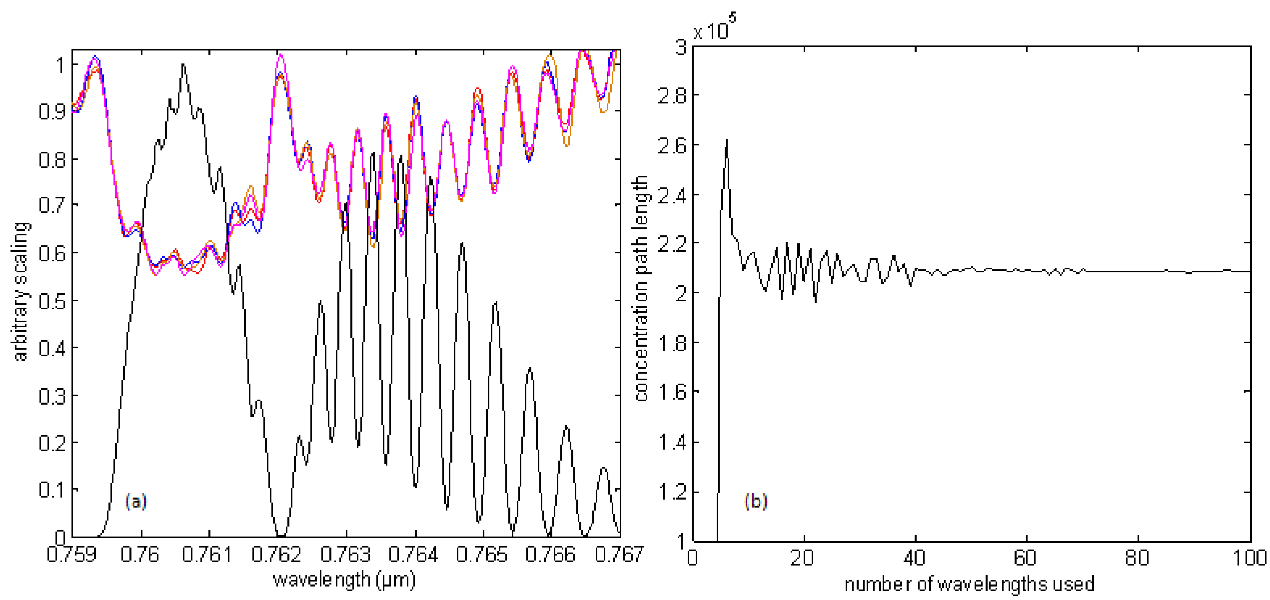

Fig. 5. (a) HITRAN lines (blue) and measured oxygen absorption data (overlapping multiple colors) convolved with a 6.4 cm^{-1} slit width to improve the convergence performance of the inversion algorithm, (b) inversion calculations for concentrations (in ppm) for each of the 100 wavelengths used (200 total in calculation) in the MLE algorithm applied to the data set shown on the left.

was 208640 ± 63 ppm which corresponds to approximately 20.86%, which is about 0.4% lower than the nominally accepted dry atmosphere value. The quick convergence of the inversion algorithm and its' precision suggests that error in this calculation is most likely attributed to path length measurement discrepancies. The 890 m measurement has an error bar of ± 5 m. There are additional airy portions of the system path not included in the measurement.

5. CONCLUSIONS

The SAS system at Penn State has progressed from laboratory to open air atmospheric measurements of various constituents at path lengths of 300, 540, and 890 m. A supercontinuum source transmitting only 3 mW of average power was used to make differential absorption measurements of atmospheric oxygen over the 890 m horizontal path. Strong correlation of measured and simulated data and convergence of the concentration of the measured spectra compared to atmospheric oxygen levels was achieved. This provides as an effective demonstration for the capability of the updated SAS configuration for measurements of additional species at long path lengths. Further improvements to the SAS system will be made to provide better signal to noise ratios for low level absorption measurements of additional atmospheric species. The techniques demonstrated are planned to lead to applications of supercontinuum sources for trace species and contaminants in the mid-infrared (MWIR).

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