Remote Sensing of Tropospheric Chemistry Using LIDARS from GEO

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Introduction

NASA's ESE has identified atmospheric composition as a crucial area of investigation within its science themes. Tropospheric chemistry is considered to be the next frontier of atmospheric chemistry, and understanding and predicting the global influence of natural and human-induced effects on tropospheric chemistry will be the next challenge for atmospheric research over the foreseeable future. In the troposphere, trace-gas species of interest include ozone, OH, NOx (NO and NO_2), CO, and some hydrocarbons; and aerosols and clouds. In particular, atmospheric ozone is one of the key tropospheric chemistry species because of its influence in many atmospheric processes. Active remote instruments that can provide vertical profile measurements of ozone can lead to better understanding of such atmospheric phenomena as: the production, distribution, and loss of ozone, anthropogenic pollution, biomass burning, atmospheric transport and dynamics, photo-chemical processes in the atmosphere, stratospheric-tropospheric exchange, atmospheric climate and radiation, and influences of atmospheric lightning. Knowledge of these phenomena is needed to evaluate the effects of chemical changes on the global hydrological cycle, the cycling of nutrient compounds through the earth environment, the accumulation of greenhouse gases, the acidity of rain and snow, and the formation of ozone in the troposphere. These are global scale problems that are particularly well suited to the use of space observations. A geostationary Earth orbit (GEO) vantage point provides an ideal location for spatially and temporally resolved distributions of trace gas species. Tropospheric chemistry poses an interdisciplinary challenge for the Earth science community and the use of such GEO observations in the study of tropospheric transport, physics and chemistry will be a frontier area of atmospheric research for the next decade and probably beyond. We investigated the capability of lidar for tropospheric profiling of chemical species.

Differential Absorption Lidar (DIAL) Technique

In the DIAL method two spectrally close laser pluses are transmitted near simultaneously with one pulse that is called the on-line, at a strongly absorbing spectral location due to the presence of an absorbing gas and another, called the off-line, at a less absorbing spectral location. In principle, concentration profiles of the absorbing gas can be retrieved using the on- and off-line lidar signals and the knowledge of the differential absorption cross-section (σ_{on}-σ_{off}) (Measures, 1984). The advantages of the DIAL method are: high vertical resolution measurements, absolute concentration profiles of constituents without a need for calibration, laser wavelength selection permits isolation from interference from other species, measurements can be made during day or night, direct inversion of species profiles with few assumptions or initial guesses, simultaneous aerosol profiles and cloud distributions are obtained. Overall, the greatest advantage of using lidar remote sensing is the capability to provide vertical profile information. In addition, active remote sensing provides an anchor and validation to total column burden retrievals from any passive measurements. The DIAL technique has been used successfully to measure a large number of trace gases in atmosphere from in the lab, ground, and aircraft (see Browell et al., 1998). The DIAL technique has been employed most successfully for atmospheric ozone and H_2O measurements in field campaigns and monitoring (Browell et al., 1997; Leblanc and McDermid, 2000).

Chemically active species in the troposphere and the feasibility of DIAL measurements

Chemical species of most interest in the troposphere are: ozone, OH, NOx (NO and NO_2), CO, and some hydrocarbons; and aerosols and clouds. The role and influence of these species in the tropospheric chemistry is given in Table 1. *We have a fairly limited knowledge on vertical distributions of any of the trace gases in the troposphere; at present, the information about the vertical distributions comes mostly from point measurements*. Information about the vertical distribution of these species is needed in Global Climate Models (GCM's).

Table 1, Chemical species that influence tropospheric chemistry

Ozone (O_3)
- Tracer of chemical and photo-chemical activity
- Influences radiation balance and climate
- Tracer of atmospheric transport
- Indicator of pollution
- Indicator of stratosphere-troposphere exchange
- Tracer of convective activity and meteorology
- Indicator of lightning
- Health hazard near surface

OH
- Highly reactive radical
- Chemical scavenger of the atmosphere
- Determines lifetime of most trace gases

NOx (NO and NO_2)
- Indicators of fast chemical processes
- Produced in urban and biomass burning events
- Indicator of lightning

Carbon monoxide (CO)
- Excellent long-term tracer
- Indicator of atmospheric dynamics and transport
- Produced in fossil fuel and biomass burning
- Urban pollutant

Hydrocarbons (non-methane); H_2CO, C_2H_6, C_2H_2, $(CH_3)_2CO$ etc
- Major component of urban smog
- Byproduct of biomass burning
- Indicators of natural (biological) processes also

NOy (HNO_3)
- Linked to NO_2 and O_3 changes
- Total nitrogen budget

In order to evaluate the optimum DIAL measurements from a GEO platform, we conducted an assessment of the DIAL measurement capability based upon the following factors.

- Absorption strength
  Optimum absorption at the target location and minimum to the target is desired (HITRAN2000 database is used). The trace species of interest and their spectral absorption regions (μm) are:

O_3	0.3, 0.6, and 10
OH	0.308/0.309
NO	0.224, 2.7, and 5.1
NO_2	0.44, 3.4, and 5.1
CO	1.7, 2.3, and 4.8
H_2CO	3.5
HNO_3	6.3
Aerosols	Nonspecific

- Species concentration
- Availability of tunable laser source
  Spectral characteristics, pulse energy, pulse repetition rate, stability, and reliability
- Intensity of atmospheric scattering
  Molecular scattering decreases by λ^{-4}, aerosol scattering $\sim\lambda^{-1}$, total scattering decreases by more than a factor of 10 from 0.3 to 2.0 μm
- Receiver efficiency
  Collection area, optical efficiency, filter bandwidth, dynamic range of signal
- Detector capability
  Quantum efficiency, detector noise, noise multiplication
- Range to observation
  Signal decreases as R^{-2}
- Day background

While the GEO platform is excellent for continuous monitoring of the evolution of atmospheric events, the R^{-2} dependence of lidar signals makes GEO observation even more challenging than from LEO (Low Earth Orbit). From the list of species in Table 1, OH was excluded in this study due to lack of any success in OH measurements in the troposphere by lidars from either the ground or aircraft even after repeated attempts. A summary of the assessment of lidar measurement potential from a GEO platform is given in Table 2.

Table 2, Measurement feasibility of GEO Chemistry Lidar profiling			
Species	Lidar Technique Demonstration	Space Tech Progress	Feasibility
O_3	routine and validated	in progress	potential
CO	no profiling	insignificant	very limited
NO_2	limited	insignificant	limited
NO	no profiling	insignificant	very limited
H_2 CO	none	insignificant	very limited

Our technology assessment was based upon the factors that affect the measurement capability that we discussed already and the success that lidar systems have demonstrated in profiling these trace gasses in the atmosphere. We also looked at the progress in technology for space-based measurement of these species. Feasibility of measurements at IR wavelengths is very limited because the very high power (>10 J per pulse) lasers and high efficiency and low noise detectors that are needed are beyond the current technology horizon (~10 years). Wavelengths beyond 5 µm are not feasible because of the very low atmospheric backscattering and the lack of desired laser and detector characteristics. Even though only limited progress has been made in profiling atmospheric NO_2, the potential exists for space-based measurements operating at 0.44 µm. However, the technologies needed for such measurements from space have not been addressed. Also information about NO_2 in itself is not sufficient when compared to the combination of NO and NO_2 for a full understanding of NOx chemistry. *From these evaluations we have concluded that amongst all the chemically important species, ozone has by far the best potential for GEO observations.*

Tropospheric ozone is expected to show considerable vertical stratification and fine horizontal structures (on the order of less than 500 meters) that contain information on the origin and age of important species. Likewise, the altitude distribution of upper tropospheric ozone is a critical parameter in climate models. Temporal and horizontal variability is also important, including significant diurnal variations. An example of the measurement of ozone in the lower atmosphere using an airborne UV DIAL system is given in Figure 1. These data are a composite of measurements taken during a series of flights over the Pacific in 1994 and they show the variation of ozone from 10^o S to about 60^o N. This figure demonstrates the capability of ozone to map many dynamical features like the latitudinal variation of the tropopause (the region that transitions from the high stratospheric ozone to lower ozone in the troposphere), low ozone in the tropical lower troposphere, low ozone transported to high altitudes by convection, and mid-latitude enhancement of ozone below 7 km.

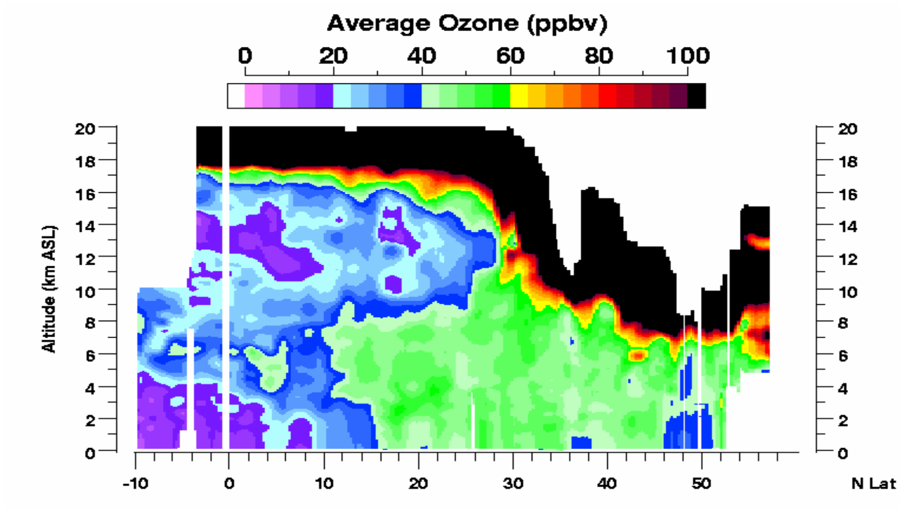


Figure 1, Latitudinal variation of ozone over the western Pacific from airborne DIAL observations during February-March 1994.

Measurement requirements

A GEO-based O_3 DIAL system will have the potential to provide the high vertical resolution measurements of O_3, aerosols, and clouds that are needed to complement other passive satellite measurements. The Total Ozone Mapping Spectrometer (TOMS), in conjunction with the Solar Backscatter UV (SBUV) and Stratospheric Aerosol and Gas Experiment (SAGE II), have provided global information on the total column of O_3 across the troposphere (Fishman et al., 1990, 1991), and this has contributed significantly to our general understanding of tropospheric processes. It is expected that the Tropospheric Emission Spectrometer (TES) will provide low vertical resolution (~5 km) O_3 profile measurements in the middle and upper portion of the troposphere. This will make an improvement in our ability to understand tropospheric O_3 and chemistry, but it is recognized that even higher vertical resolution measurements (~ 2 km) are needed to identify and resolve most of the complex tropospheric structure, chemistry, and transport issues. A joint study conducted by NASA Langley Research Center and the Canadian Space Agency in 1998 concluded that for understanding many global chemical processes ozone measurements in the free troposphere with 1-2 km vertical resolution are needed. This is consistent with NASA Post-2002 Mission Planning Workshop report (1998). The objective of a GEO lidar would be to provide such high resolution measurements as well as accurate information concerning the altitude location of atmospheric layers containing enhanced or depleted levels of O_3, aerosols, and clouds in the troposphere and stratosphere. This information can be directly assimilated into three-dimensional chemical-transport models, and it can be used to aid in the interpretation of the passive measurements. Lidar systems can also make measurements day or night to be able to address diurnally dependent boundary layer, transport, and photochemical processes. It should be noted that higher vertical resolution measurements (~ 0.5 km) are needed in the boundary layer.

The temporal evolution of atmospheric chemical processes is altitude dependent and, generally, these processes take place at a faster rate at lower altitudes. Figure 2 shows the rate of change of chemical species in the lower troposphere related to urban smog

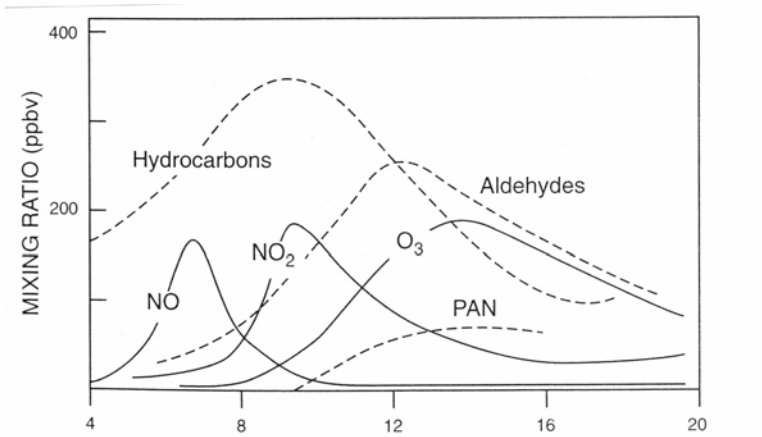


Figure 2, Evolution of urban ozone and other species during a smog episode

(Goody, 1995). Typical time scales for ozone are several hours. Lifetime of photo-chemically produced ozone depends on both altitude and season. The variation of photochemically produced ozone with altitude and season is summarized in Table 3. The measurement requirements concerning the spatial and temporal resolutions for a GEO-based DIAL are given in Table 4. The GEO platform permits observations from a fixed location to monitor evolution of events or to scan across large continental size regions. Ideally mapping to observe events over a selected region and repeating the process at intervals that are shorter than the time scales of the events would provide the best data for analysis. A pointing stability of 5 km is needed to monitor events such as development of urban pollution.

Table 3, Lifetime of O_3 depends upon altitude, and season. The following table gives model-calculated lifetime (in days) of photochemical O_3:

Altitude (km)	40^0N		20^0N	
	Summer	Winter	Summer	Winter
0	8	100	5	17
5	15	160	10	35
10	40	300	30	90

(S. Liu et. al., 1992)

Table 4, Ozone measurement requirements

-Spatial Resolution
    Boundary layer Dx=20-50 km, Dz = 0.5-1 km
    Free and upper troposphere
        (Local) Dx= 100 km, Dz = 1-2 km
        (Regional or continental scale) Dx=200 km, Dz = 2-3 km

-Temporal Resolution
    (Local in the boundary layer) hourly
    (Regional and continental scale) twice a day

DIAL modeling performance estimates

The lidar signal P (photo-electrons per range bin) is calculated using the lidar equation:

$$P = \frac{E}{h\nu} \cdot O_E \cdot Q_E \cdot \frac{A}{R^2} \cdot (B_M + B_A) \cdot \Delta R \cdot e^{-2\int_0^R (\beta_M + \beta_A) \cdot \Delta R}$$

Where E is the laser pulse energy, hν is energy per photon, O_E is the optical efficiency, Q_E is the quantum efficiency, A is the effective area of the receiver, R is the range from the lidar to the measurement location, ΔR is the range resolution, B and β backscatter and extinction coefficient profiles respectively for molecules (M) and aerosols (A). This equation relates many of the lidar and atmospheric parameters to the detected signal and is very useful in conducting some of the trade-off studies. The detector noise is calculated using:

$$D = \frac{NEP}{\sqrt{2}} \cdot \eta \cdot \frac{\lambda}{hc} \cdot \Delta t$$

Where NEP is the detector noise equivalent power, η the quantum efficiency, λ the wavelength, h the Planks constant, c the velocity of light, and Δt the sample time. Because of the long range, the lidar signals are expected to be smaller than the background for mid-day background conditions. Full-moon background was used to predict the performance for all night observing conditions. DIAL measurement simulations were conducted using the methodology given by Ismail and Browell (1989). Because of the long range, for a given set of lidar parameters, lidar signals will be smaller by a factor of about 10,000 compared to LEO. Given that the set of technologies to achieve this measurement include: LIDAR technologies in UV and IR lasers, solid-state detectors, deployable telescopes, and low bandwidth high throughput filters, the technologies that have the most significant impact to compensate for this decrease in signal are large collection area telescope technologies and high power lasers. A set of lidar parameters for predicting performance are given in Table 5 that assume large area telescopes (12 m, 35 m, and 100 m). Lidar signals, background, and detector noise for the three different telescope sizes are given in Appendix Figure A1. A mid-latitude atmosphere and US standard ozone model profile were used to compute the on- and off-line signals. Error profiles of ozone measurements for a 10 min observation period are shown in A2. These indicate large measurement errors for the 12 m telescope system in the lower atmosphere. Figure A3 shows that ozone profiling can be achieved with an observation interval of 10 min with the 35 m telescope system. Full details of this study are outlined in Appendix B (GEO Tropospheric Chemistry Study) in the form of a Power Point presentation.

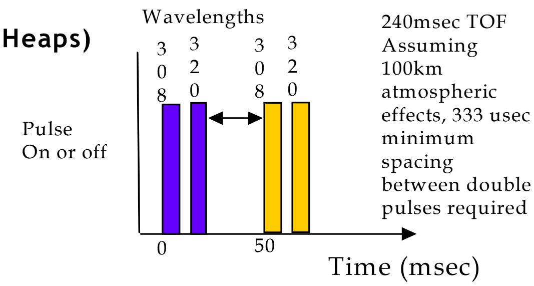
ISAL study at NASA GSFC

The Instrument Synthesis and Analysis Laboratory (ISAL) is the instrument design facility in Goddard's Integrated Design Capability (IDC). We took advantage of ISAL instrument-modeling capabilities for the GEO Tropospheric Chemistry lidar studies. The input for the ISAL study was the DIAL lidar concept with lidar parameters in Table 5 with a 35 m telescope.

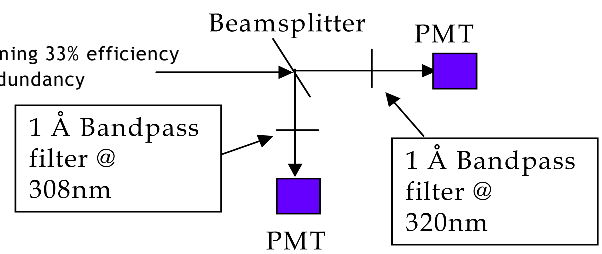
Table 5, Laser and receiver parameter of the GEO-Troposphere DIAL system

Laser/Receiver Subsystem

- **Laser Requirements (contributions from Bill Heaps)**
  - 20 Hz Pulse Rep Rate
  - 1 Joule Output Double Pulsed
    - 1 J per pulse
  - 1064nm - “mixed” to get 308-320nm
    - Challenging wavelength conversion
  - Active Q switching
  - Range -5 to 55km
    - Data 0-30 km
    - Background 30-55km
  - Up to 2Mbit/sec, will send all data during diagnostic mode



- **Laser and Detector (Receiver) Trades**
  - 1 laser vs. multiple lasers
    - 3J at 1064nm would give 1 J at 308-320nm, assuming 33% efficiency
    - Sync 4 laser systems each at 1 Joule - provide redundancy
  - PMTs
    - Redundant PMTs
    - PMT for each wavelength
    - Count rates to 10Mhz
    - On all the time



ISAL studies were conducted using the DIAL instrument concept and a systems engineering approach was used. Analysis of all subsystems was conducted including orbit; instrument /spacecraft configuration; mass power and data rate; laser, and receiver system. Full ray tracing of the optical system and design of sunshade shield for screen exposure to daylight were analyzed. Cooling of the laser system and thermal analysis was also considered. The instrument concept diagram is shown in Figure 4. The system baseline includes a large area 35-m class Gossamer optics telescope and a 1 J 20 Hz double-pulsed UV laser system. The systems engineering overview of the ISAL study is given in Appendix C (GEO Tropospheric Chemistry/GSFC). The main conclusions of this study are:

- Large aperture systems are necessary to compensate for loss of lidar signal due to the large distances
- Considerable investments are being made in large aperture (gossamer) optics by both the DoD and NASA.
- Stray light shielding of the large aperture is a requisite technology development area potentially requiring formation flying (NASA investments in next-generation space telescopes can be leveraged)

- Multiple 1-2 J lasers that are triggered together and smaller aperture receiver may offer a more cost effective design that would also reduce day background influence

Future lidar systems operating from large distances in space will require both high power lasers and large collection area receivers. With the lidar system, in general, the receiver collection area can be decreased and laser power be increased to maintain performance. However, due to material damage threshold, laser power output cannot be increased beyond 1-2 J per pulse. The ISAL study suggested that output from 4 such lasers be combined and triggered together. This has the advantage of providing a redundancy and better performance in the presence of sunlight background. Even though the cost of including a number of high-powered lasers will be high, it might still be considerably cheaper than the giant Gossamer receiver (and the background light shield) in geosynchronous orbit.

Technology Roadmap

Technology developments are in progress towards the development of UV DIAL systems from space for tropospheric chemistry. Two programs that are developing critical technologies are: the NASA LaRC/GSFC Laser Risk Reduction Program (LRRP) for developing high power UV lasers (medium pulse energy (~100 mJ) and high repetition rate (>100 Hz) and/or high pulse energy (>500 mJ) low repetition rate (20 Hz) lasers; and the Earth Science Technology Office (ESTO) Advanced Component Technology Program for a project funded at the University of Colorado to demonstrate technology leading to 2.5 m class deployable telescope. To create laser energy in the UV wavelengths requires a 1-micron laser pumping a non-linear optical component. Contemporary 1-micron lasers are available with sufficient energy, but the electrical to optical efficiency is insufficient for a space-based instrument. In addition, the non-linear optical component requires a high pump beam quality that is not typical of existing 1-micron lasers. The non-linear optical components need sufficiently high optical to optical conversion efficiency to achieve 1-2 J per pulse in the UV wavengths. Maturation of these technologies and their application to LEO space systems are essential elements to enable a GEO Tropospheric measurement. A roadmap for the enabling technologies for a GEO Tropospheric chemistry measurement using a DIAL instrument is given in Figure 5. The challenging technologies are the large collection area receiver systems, shielding system to avoid stray solar light, and high pulse power lasers. Large Gossamer optics has become an emerging technology in recent years because of DoD and NASA interest. Funding for large area optical systems as well as higher power lasers beyond what is planned under the LRRP is required for the development of these technologies. Incremental advances in other receiver optical elements including narrowband filters and more efficient solid-state UV detectors can reduce the requirements on the telescope and laser.

References

Browell, E. V., S. Ismail, T. C. McElroy, R. M. Hoff, and A. E. Dudelzak, Global measurements of ozone and aerosol distributions with a space-based lidar system, AGU Fall meeting, San Francisco, CA Dec. 8-12, 1997.

Browell, E. V., S. Ismail, and W. B. Grant, Differential Absorption Lidar (DIAL) measurements from air and space, Appl. Phys. B, 67, 399-410, 1998.

Fishman, J., C. E Watson, J. C. Larsen, and J. A. Logan, Distribution of tropospheric ozone determined from satellite data, J. Geophys. Res.-Atmos., 95, 3599-3617, 1990.

Fishman, J., K. Fakhruzzaman, B. Cros, and D. Nganga, Identification of widespread pollution in the southern-hemisphere deduced from satellite analyses, Science, 252, 1693-1696, 1991.

Goody, R, *Principles of Atmospheric Physics and Chemistry*, Oxford University Press, Oxford, UK, 1995.

Ismail, S. and E. V. Browell, Airborne and spaceborne lidar measurements of water vapor profiles: a sensitivity analysis, Appl. Opt., 28, 3603-3615, 1989.

Leblanc, T. and I. S. McDermid, Stratospheric ozone climatology from lidar measurements at Table Mountain and Mauna Loa, J. Geophys. Res. 105, 14613-14623, 2000.

Liu, S., C., M. Trainer, M. A. Carroll et al., A study of photochemistry and ozone budget during the Mauna Loa Observatory Photochemistry Experiment, J. Geophys. Res., 97, 10463, 1992.

Post –2002 Mission Planning Workshop, NASA ESE, Easton, MD, August 24-26, 1998.

Measures, R. M., Laser remote sensing fundamental and applications, Krieger Publications Co., Malabar, Fl., 1984.

Appendix A

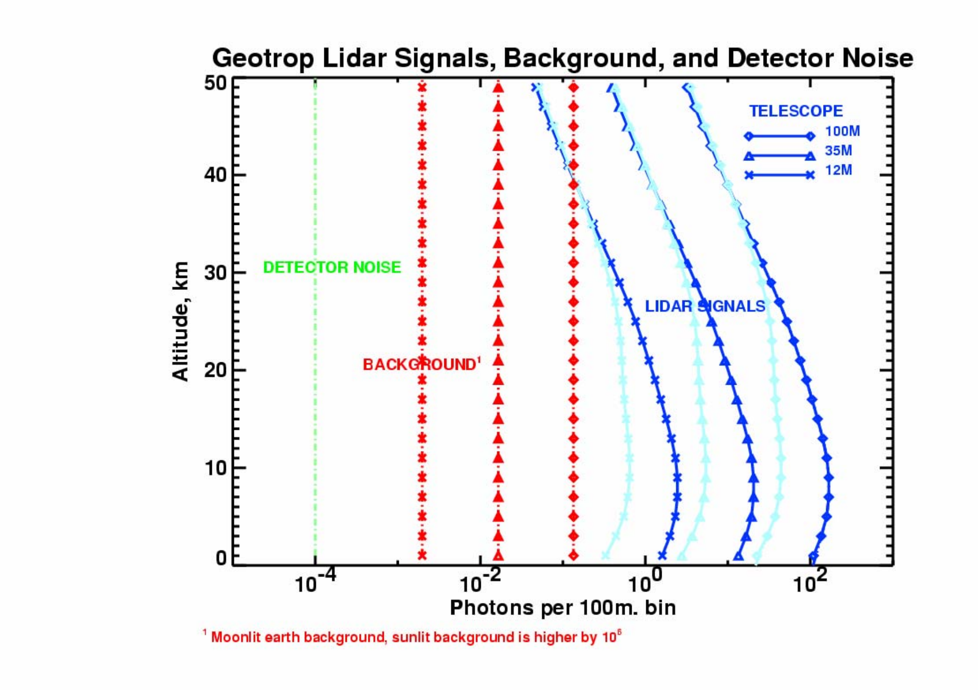


Figure A1, Lidar signals, background, and detector noise

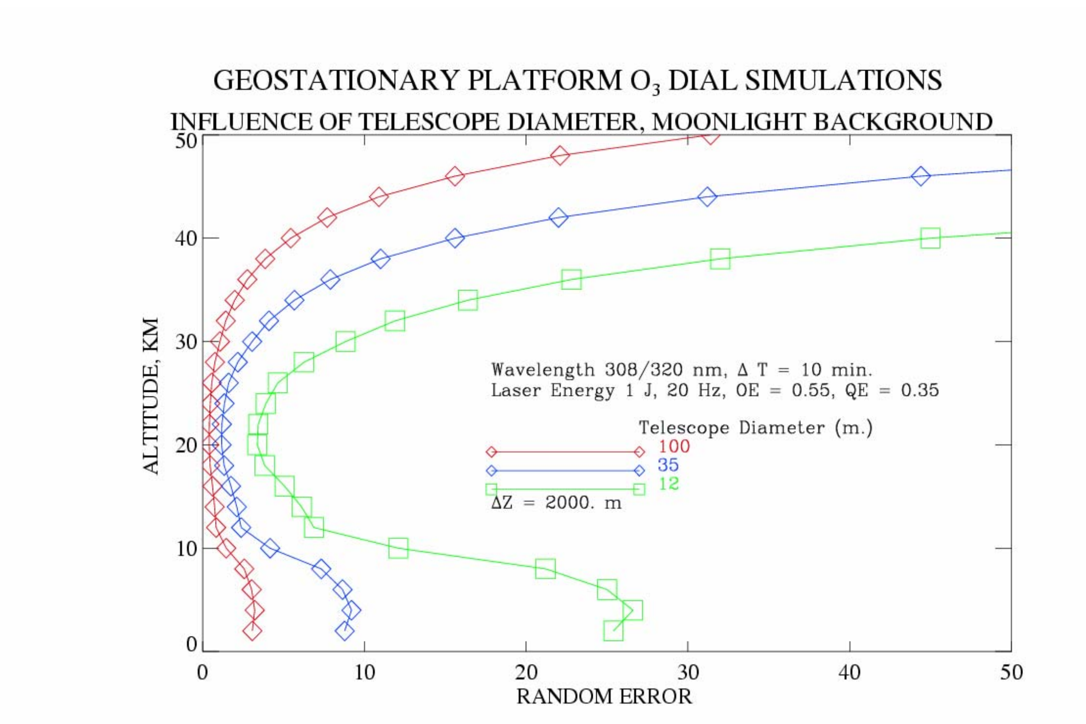


Figure A2, Performance simulations using 12, 35, and 100 m size telescopes

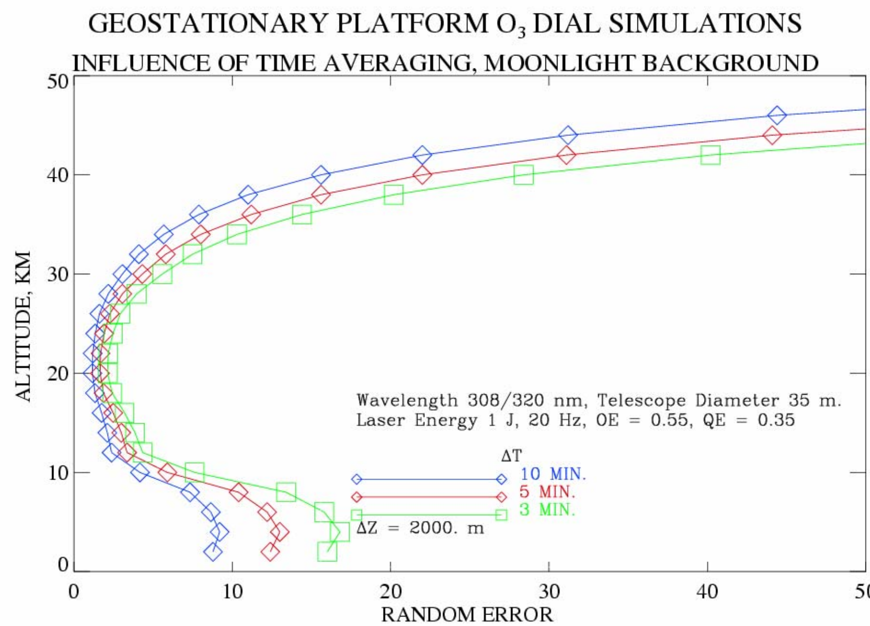


Figure A3, Influence of time averaging on DIAL measurements.

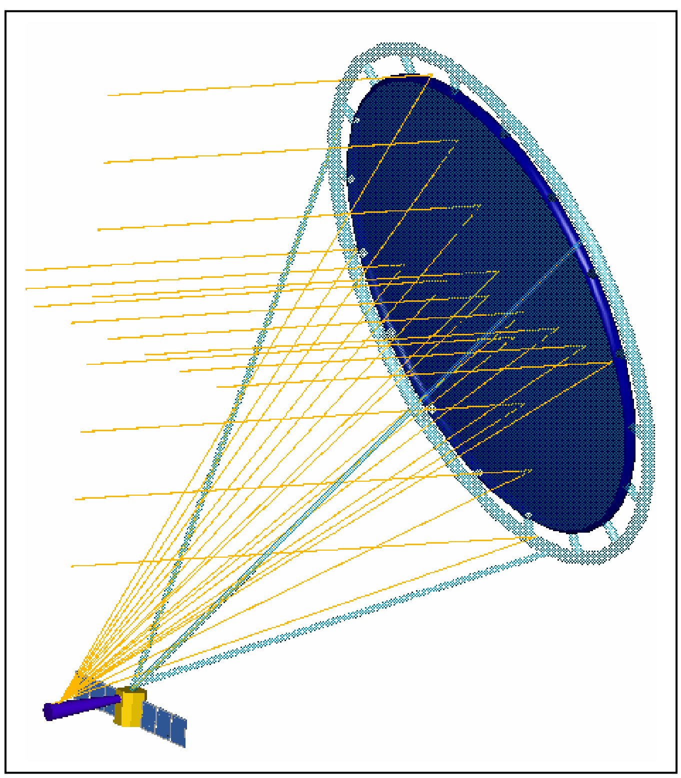


Figure A4, Instrument concept of a GEO Tropospheric DIAL instrument employing a 35-m telescope

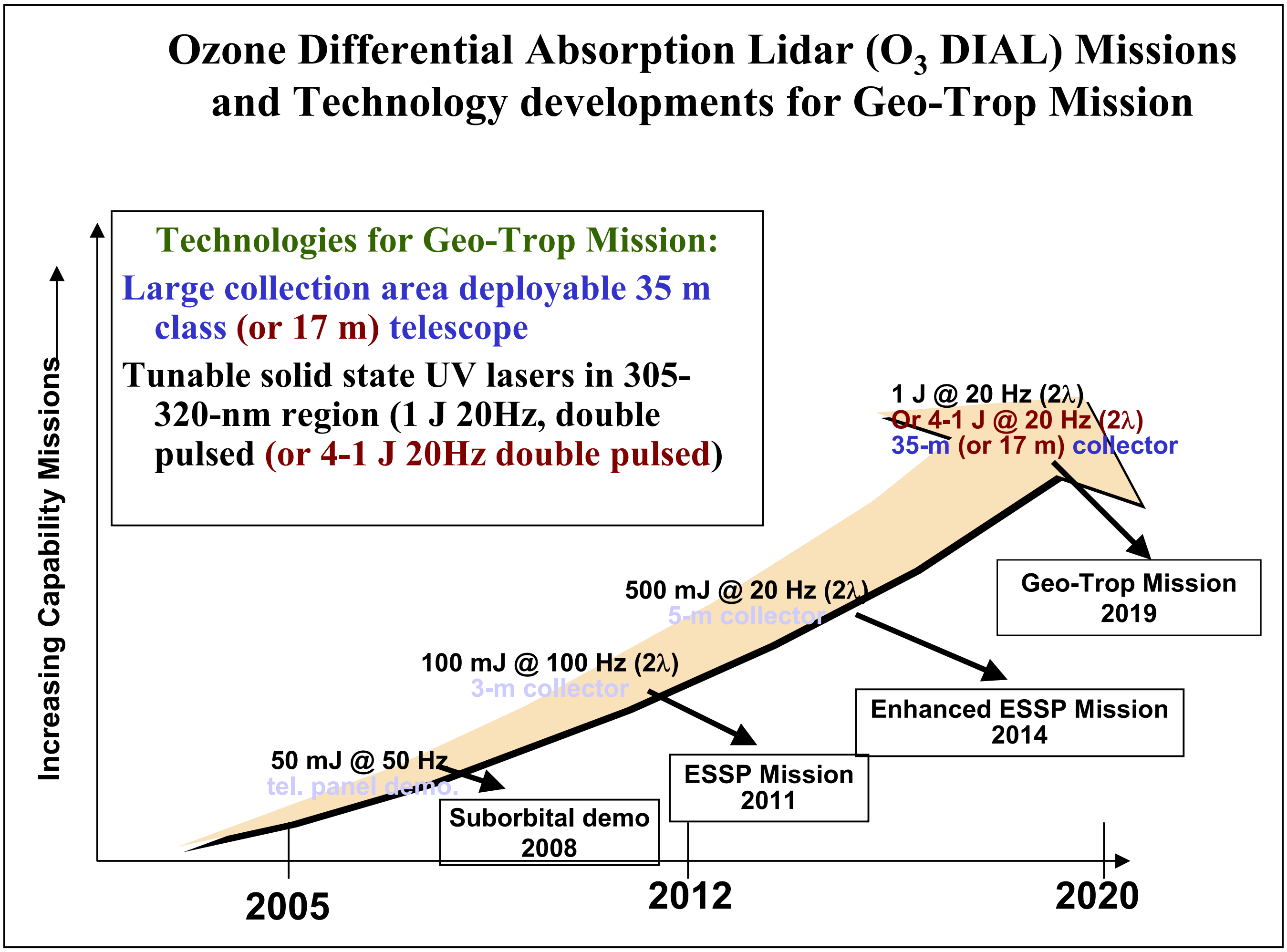


Figure A5, Technology roadmap leading to a GEO Tropospheric chemistry measurement capability using a DIAL instrument employing a 35-m class deployable telescope and a 1J, 20Hz double-pulsed laser.