HADMÉRNÖK

IX. Évfolyam 3. szám - 2014. szeptember

Halász László - Lucas Grégory
gregory.luc4s@gmail.com

REVIEW OF AIRBORNE LASER MEASUREMENTS OF CHEMICALS AND RADIATIONS

Abstract

This article aims at reviewing the existing laser measurement technologies and methods applied in the airborne detection of radiations and chemicals. The first part introduces the main concepts and provides the reader with a general orientation. Each of the following chapters aim at describing a measurement method, its capacities and fields of application. Differential absorption LiDAR (DIAL), Tunable Diode Laser (TDL) spectrometry are successively considered with the measurement of chemicals. Concerning the ionizing radiation measurements, the bibliographic research demonstrated that active laser airborne measurements are presently not applied, neither have they been tested. Nevertheless, some stand-off measurement techniques under development are reviewed and their adaptations to airborne measurement are considered. Spectroscopic measurement of radiation-induced radical, active detection by the detection of frequency modulation and standoff detection of alpha radiation via the measurement of energy absorbed by excited state molecules are introduced.

Jelen cikk célja a meglévő lézeres mérési technológiák és módszerek bemutatása, amelyeket a vegyi anyagok és sugárzások légi felderítésében alkalmaznak. Az első rész áttekinti a legfontosabb fogalmakat, és az olvasónak egy általános orientációt nyújt. A következőkben minden egyes fejezet leírást ad egy mérési módszerről, annak teljesítményéről és alkalmazási területeiről. A vegyi anyagok mérésére áttekintjük a differenciális abszorpciós LIDAR (DIAL), állítható lézer dióda (TDL) spektrometria módszereket. Az ionizáló sugárzás méréseinek vizsgálata során a bibliográfiai kutatások kimutatták, hogy az aktív légi lézeres méréseket jelenleg nem alkalmaznak, nem is tesztelték még korábban. Ugyanakkor a kutatók egyéb fejlesztés alatt álló mérési technológiákat és azok távérzékelési alkalmazásainak lehetőségeit vizsgálják. Elsőként a sugárzás által kiváltott szabad gyök spektroszkópiai mérését, majd a frekvencia moduláció érzékelésének aktív felderítését, végül a gerjesztett molekulák által elnyelt energia mérésével az alfa sugárzás távoli felderítését mutatjuk be.

Keywords: *Airborne, laser measurement, DIAL, LIDAR, radiological material, ionizing radiation.*

INTRODUCTION

In the second half of the twentieth century, awareness arose about the presence of chemicals in the atmosphere and their consequences. Scientists discovered that heavy industries and more generally human activities hugely impact on the biotopes via the transfers of part of the chemicals in the atmosphere, thus generating air quality concerns but also environmental changes.

At the local scale for example, high concentration of particles and chemicals in the air pose seasonal pollution problems in the capitals and dense urban areas (ozone pollution peaks in winter and summer). Pollution from industrial plumes or transmission exhaust gases became a concern for the public health. The air pollution generated in the largest cities also impact at the regional scale by transfer. Last but not least, global concerns are also in scope. Greenhouse gases (like H_2O, CO_2, CH_4, N_2O, O_3) generating an increase of the average temperature on Earth. Increase of desertification, extreme meteorological phenomena, seasonal calamites in agriculture, the melting of the pole ice, the average mean sea level, the loss of arable land, etc. are nowadays proven and visible consequences of the climate change. A second example illustrates very well how a new concern can appear and how in time apprehension of the phenomenon (through adequate measurement methods and response) is useful to limit global negative impacts. In 1985 scientists discovered the stratospheric ozone depletion over the Antarctic. The various measurement methods applied demonstrated the large scale and fast dynamic of the phenomenon and allow the identification of the origin of the problem: man-made organohalogen compounds, especially chlorofluorocarbons (CFCs) and bromofluorocarbons. In 1996 all countries of the world finally agreed to ban the uses of the CFCs and industrial production of CFCs was stopped.

Worrying environmental changes are arising and incredible challenges are to come. Our ability to live in favorable environmental circumstances in the second half of the 21st century will depends on the scientific capacity to react and to which extent the society will be able to adapt itself and evolve. As a consequence, understanding how those complex mechanisms are working, how much extent and dynamics will be necessary to react. The understanding of the phenomenon whether they are local, regional or global requires advanced methods for their measurements. In the mid-60s the development of computer technologies with transistor-based machines increased the reliability. Computers were smaller, faster, and cheaper to produce and required less power. Later in the 70s, the integrated circuit technology and the subsequent creation of microprocessors further decreased size and cost and further increased speed and reliability of computers and triggered the development of airborne measurement methods. Airborne measurements are powerful tools in the sense they allow three dimensional spatial and temporal mapping.

This article aims first at considering how laser technologies can be used for the aerial measurement of chemicals. In a second part radiological airborne measurements are considered.

REMOTE SENSING OF CHEMICALS IN THE AIR

Differential absorption lidar (DIAL) technique

Short historical introduction

The methodology of DIAL has been developed in the late 1960s and 1970s. [1] DIAL was first employed in 1966 for remote measurement of water vapor (H_2O). The first aerial measurements with DIAL technique were realized by Schotland in 1974. [2] Since, differential absorption LiDAR systems have evolved significantly and have been used for the measurement of ozone, water vapor and aerosols from aircrafts for over 34 years. [3] They have yield new insights into atmospheric chemistry, composition and dynamics in large-scale field experiments. [3]. In 1994, the LITE experiment successfully managed a space-base LIDAR measurement mission completing the ground, air and space cycle. [4] A LIDAR system was carried by the space shuttle "Discovery" for 9 days. [5] The latest space development was related to the CALIPSO satellites, that carried a LIDAR system for the global measurement of clouds and aerosols. [6]

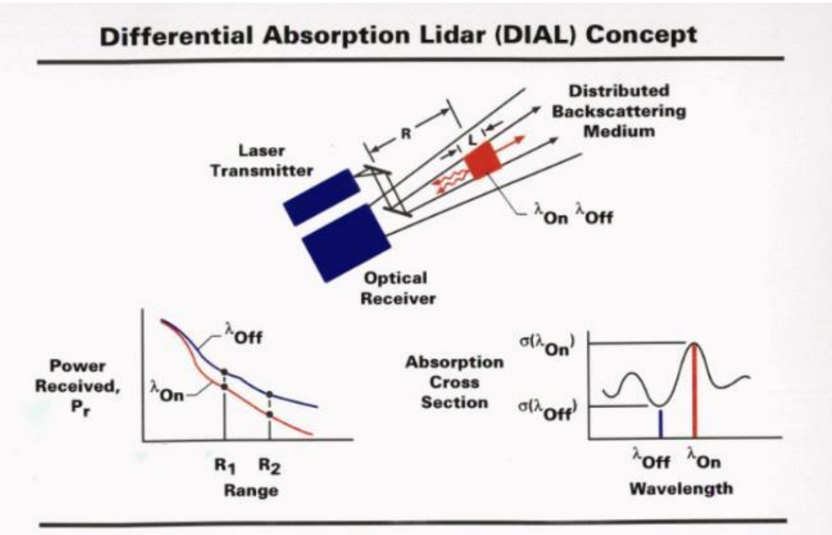
Year	Type	Measurement of species
Late 60s	Ground based	Water vapor
Late 70s early 80s	Airborne	Aerosols, clouds, winds
1978	Airborne	Tropospheric ozone
1980	Airborne UV	Ozone profile
1982	Airborne	Water vapor
1994	Space borne	Ozone, water vapor, aerosols
2006	Space borne	Cloud-aerosol

Tab.1. Summary about historical developments of DIAL

Working principle

Differential absorption LIDAR is a remote sensing technique uses two laser beams in different wavelengths that can be reflecting from any field objects and the active beam can be absorbed in the investigated gas so the type and average concentration of the gas in the air can be determined. [4] This laser based technique is employed for the measurement and mapping of concentration of various molecules and mass emission in the atmosphere. [7]

The measurement relies on the unique absorption spectrum ("fingerprint") of each type of molecules. An absorption measurement is realized by sending a dual wavelength laser pulse in the direction of a target. [8] One wavelength is tuned to a strong absorption feature of the gas of interest, generally called the 'on' wavelength (λ_{on}) and the other tuned to a nearby wavelength with weak absorption by the gas, generally called the 'off' wavelength (λ_{off}). A sensitive detector detects the light backscattered by particles at the two different wavelengths. The value of the average gas concentration, N_A, in the range interval from R_1 to R_2, can be determined from the ratio of the backscattered LIDAR signals at λ_{on} and λ_{off} , as shown in Fig.1. (Browell, 2003) In that equation, $\Delta\sigma = \sigma_{on} - \sigma_{off}$ is the difference between the absorption cross-sections at the on and off wavelengths, and Pr_{on} (R_1) and Pr_{off} (R_2) are the signal powers received from range R at the on and off wavelengths, respectively. [4]



$$N_A = \frac{1}{2(R_2-R_1)\,[\sigma_A(\lambda_{On})-\sigma_A(\lambda_{Off})]} \ln \left[\frac{P_{r_{On}}(R_1) \times P_{r_{Off}}(R_2)}{P_{r_{Off}}(R_1) \times P_{r_{On}}(R_2)}\right]$$

Fig.1. Differential Absorption LiDAR (DIAL) concept (from E.V. Browell, 2003)

This is essentially an application of the Beer–Lambert[1] law for an absorbing medium. The λ_{off} LiDAR return also provides important information on the molecular and aerosol scattering properties of the atmosphere. [4] The differential approach simplifies the calculation after the measurement process. [7]

The laser light is in short pulses and time resolution of the backscattered light (along with the speed of light) gives range resolution as in a simple LIDAR (light detection and ranging). [8]

Main differences existing with the different DIAL systems

Alvarez et al. 2011, provide a classification of the different DIAL systems based on their transmitter characteristics. [9] They mention the systems generally fall into two design classes. The first approach, pioneered by the National Aeronautics and Space Administration (NASA) Langley Research Center, uses high-power tunable dye lasers for the LIDAR transmitter. [10] The resulting system is extremely versatile, but is relatively large with high power consumption, and thus is restricted to large aircraft platforms, which are costly to operate. The second approach, which uses fixed-wavelength lasers, can be made more compact for operation on smaller aircrafts, but cannot be optimized to maximize the spatial and temporal resolution and minimize unwanted interferences. [11], [12], [13], [14] Recently, developments in tunable UV solid-state laser technology have bridged the gap between these two approaches. [15], [16], [17]

Systems can also be distinguished based on the wavelengths used. As shown in Tab. 2, many combinations are possible (UV, visible, NIR, LWIR).

Program carrier	**Circa**	**Channels**	**Laser(s) (*tunable)**	**Measurement of Species**
GND. based 48 inch	1970	2	Ruby @ 347&694 nm	Aerosols/N2
Aircraft Electra 990	1978	3	Ruby, YAG, YAG/Dye @ 1064, 720*,694,600*, 532, 347, 300* nm	Aerosols H2O/O3
LASE, ER 2	1994	3	Ti: Al2O3 @ 815 nm	H2O/Aerosols
LITE, Shuttle	1994	3	YAG @ 1064, 532, 355 nm	Aerosols/clouds Density
ESSP	TBD	3	YAG @ 1064, 532, 355 nm	Aerosols/clouds

Tab.2. Systems and wavelengths used.

Example of application of DIAL technology for chemicals detections

DIAL technology can be employed from local to global scale depending on the objectives of the measurements. Large scale applications can for example deal with water vapor and greenhouse gas distribution in the atmosphere for a better understanding of climate change. In many of these studies airborne LIDAR systems have played a key role by providing highly resolved measurements of the three dimensional distribution of ozone and aerosols. [9] Understanding the formation and transport of ozone and aerosols is of great interest because they negatively impact on air quality and also on climate. [9]

Global measurement example with ozone detection: TOPAZ is an airborne NADIR viewing system using three wavelengths DIAL system. It provides information about ozone and aerosol back scattered profiles from 400 m above airplane to near ground level (flights are generally conducted at an altitude varying from 3000 to 5000 m above sea level). Profiles are acquired at 10 sec intervals. [9]

---

1 The Beer Lamber law relates the absorption of light to the properties of the material through which the light is traveling. The law states that there is a logarithmic dependence between the transmission (or transmissivity), T, of light through a substance and the product of the absorption coefficient of the substance, α, and the distance the light travels through the material (i.e., the path length), ℓ.

Ozone concentration accuracy	Typically <5%, but can be as high as 15% under low signal-to-noise ratio conditions at ranges >2.5 km with high ozone concentrations
Ozone concentration precision	+/-(2-5) ppb (5%-8%) at close range (400-500 nm) falling to +/-(5-35) ppbv under low SNR conditions as noted above
Resolution: ozone concentration	Vertical: 90 m (with 450 m smoothing) Horizontal (time): 600 m (10 s at a flight speed of 60 $m.s^{-1}$)
Resolution: aerosol backscattered	Vertical: 18 m Horizontal (time): 600 m (10 s at a flight speed of 60 $m.s^{-1}$)
Minimum, maximum range	400m, 3000-5000 m
Laser specifications (per manufacturer) for 1000-Hz pulses	1053 nm, 527 nm, 263 nm. 283-310 nm
Power equipment	3 kW of 110 VAC
Size (volume), weight	Approximately 1.75 m^3, 400 kg total
Laser frame	1.4 m x m 0.76 m x 1.2 m, 185 kg
Cooler	0.70 m x 0.38 m x 0.59 m, 76 kg
Rack-mounted electronics and computers	Total of 1.4 m eight of rack space needed for 0.48 m wide units (unit depths range from 0.05 to 0.46 m), 83 kg
Two racks to hold electronics	0.58 x 0.51 m x 1.0 m, 16 kg each
Nitrogen cylinder	0.2 m diameter, 0.7 m tall, 18 kg

Tab.3. Summary of TOPAZ LIDAR specification (from Alvarez, 2011)

Detection and measurement of chemical warfare agents: Differential absorption LIDAR can also be applied in the detection of chemical warfare agents which constitute a potential hazard. Their release in the atmosphere could happen under different scenarios like a chemical attack, an accident during their manipulation or also during their destruction. In case of an accidental release, a remote sensing system can be used to monitor relatively large geographic areas, replacing a network of point sampling analyzers and providing information about the size, location and direction of the toxic cloud. [18]

Tests were performed using a tunable CO_2 laser designed for helicopter platform (VTB-2) measuring in the 9.2–10.8 μm range. Tab. 4. shows the sensitivity values for two different integration times of 1s and 30s at 2.5 km range. [18]

Material	**Sensitivity (mg/m3)**	
	1s integration time	**30s integration time**
Tabun	342	62
Sarin	247	45
Soman	297	54
Cyclosarin	277	51
Vx	806	147

Tab.4. Sensitivity at 2.5 km range, topographical backscattering (from Halász, 2002)

The sensibility of the detection method varies from 50 to 150 ppm with the longest integration time (30 s).

Detection and measurement of a pollutant over urban areas: Examples of small scale applications are the tracking of a pollutant in the atmosphere near a point source, plume modelling and hot spot detection. [19] Additionally the concentrations can be converted into mass emissions by making a series of scans with the DIAL along different lines within a plume and combining these with meteorological data. These measurements are then used to produce a mass emission profile for a whole site, for instance for fugitive emissions from an oil refinery. [9]

Data have been used to visualize the aerosol pollutant structure throughout the lower Fraser Valley. While the majority of the pollution in the valley is from the urbanized sector around Vancouver, the survey revealed there were at least seven additional point source emitters which impact the valley in a significant way. [20]

Validation or calibration of models: Last DIAL can be used to gather measurements useful later on for the calibration of other methods or assessing predictive pollutant models. As an example, a field experiment (Pacific '93) was carried out in Vancouver, British Columbia, in 1993. The purpose of the experiment was to provide data on the three-dimensional extent and movement of pollutants in a complex topographic regime so that predictive pollutant models could be assessed. [20] Similar measurements made with water vapor helped to improve general circulation models (GCM) and numerical weather prediction (NWP). [1] Airborne measurement campaigns were also performed in order to assess the precision of satellite measurements.

Use of high Resolution Doppler LIDAR as a complementary tool: High Resolution Doppler LIDAR is an additional active measurement tool using laser technology coupled with Doppler to measure the speed of air, convection movement etc. If such systems do not detect or measure chemicals, they help in the understanding of chemicals movement by providing information about air velocity. Such information can be used later on as input in predictive models.

Wavelength	2.0218 μm (fully eye-safe)
Pulse energy	1.5 mJ
Pulse rate	200 Hz
Frequency stability	0.2 MHz
Scan	Upper hemisphere
Range Resolution	30 m
Time Resolution	0.02 s (for 10 pulse average)
Velocity Precision	5 cm/s
Minimum range	0.2 km
Maximum range	2 - 9 km (typically 3 km)
Laser	Tm:Lu,YAG diode-pumped, injection-seeded laser
Platforms	ground, ship, aircraft

Tab.5. Characteristics of High Resolution Doppler LiDAR

Summary about DIAL detection capacities and field of use:

Species	Application	Spectral range	Uncertainties
H_2O (water vapor)	Meteorology		
CO_2 (greenhouse gas)	Global climate		
Ozone, aerosols	greenhouse gas, pollution	UV (from 283 to 310 nm)	several ppbv. or around 5% in good SNV conditions
SO_2			
NH_3	acid rain		
Hg	pollutant		
CO	greenhouse gas		
CH_4 (methane)	greenhouse gas		
N_2O	greenhouse gas		
Tabun, Sarin, Soman, Vx	Chemical warfare agents	Middle infrared tunable from 9.2–10.8 μm	30-85 mg / m^3

Tab.6. Summary table with the different species, wavelength used and the detection capacities

Tunable diode laser (TDL) spectrometry

Working principle

Tunable diode laser absorption spectroscopy (TDLAS) is a technique for measuring the concentration of certain species in a gaseous mixture using tunable diode lasers and laser absorption spectrometry. On the difference with the instruments presented above which performed standoff or remote sensing measurements, tunable diode laser instruments are designed for in situ trace-gas measurements. In the case of airborne measurements this means that air around the aircraft is sampled and measured. The advantage of TDLAS over other techniques for concentration measurement is its ability to achieve very low detection limits (of the order of ppb).

A classic TDLAS setup consists of a tunable diode laser light source, transmitting optics, a gas chamber containing the absorbing medium to be measured, receiving optics and detectors.

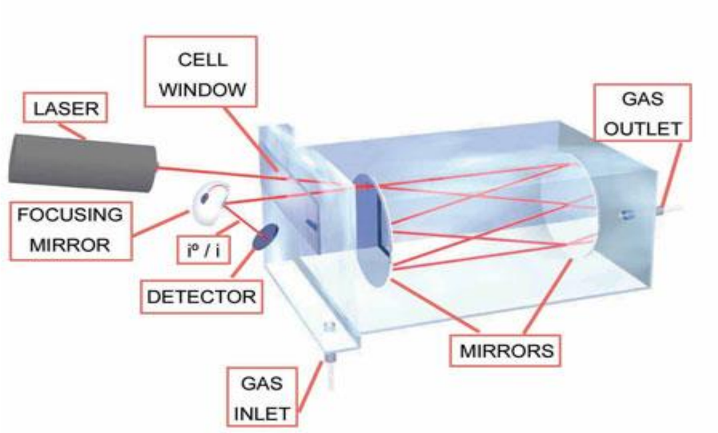


Fig. 2. Gas moisture TDLS detector (image from Servomex)

The principles are straightforward: gas molecules absorb energy at specific wavelengths in the electromagnetic spectrum. At wavelengths slightly different than these absorption lines, there is essentially no absorption. By transmitting a beam of light through a gas mixture sample containing a trace quantity of the target gas, and tuning the beam's wavelength to one of the target gas's absorption lines, and accurately measuring the absorption of that beam with a photodiode, one can deduce the concentration of target gas molecules integrated over the beam's path length. This measurement is usually expressed in units of ppm-m. [21]

The transmitted intensity is related to the concentration of the species present by the Beer-Lambert law, which states that when a radiation of wavenumber passes through an absorbing medium, the intensity variation along the path of the beam is given by:

$$\ln (I_0/I) = S*L*N$$

where I is the measured beam intensity when tuned to the absorbing wavelength of moisture; I_0 is the reference measured beam intensity when tuned away from the moisture absorbing wavelength; S is the fundamental absorption line strength and is a fixed constant; L is the path length of the beam through the sample and is a fixed constant; N is the number of molecules contained in the beam path passing through the sample.

Different variations between the instruments:

Distinctions can be done with the technology used in the laser source. Two main systems are mentioned in the literature. First, distributed feedback lasers (DFB) which is the most common transmitter type in DWDM-system. Distributed feedback diode laser serves as a spectrally bright light source having a well-defined but adjustable wavelength. The structure of a DFB laser includes a grating-like optical element that forces the laser to resonate in a single electromagnetic mode.

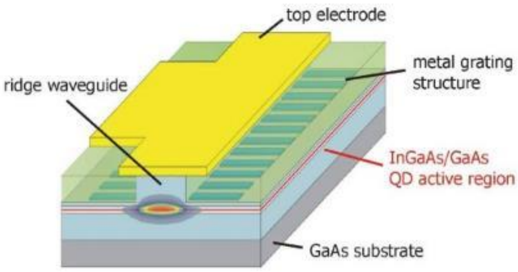


Fig.3. Structure of a distributed feedback laser

The laser emits near-infrared radiation (1.2 - 2.5 µm, or 4000 - 8500 cm^{-1}) with a line width less than 0.003 cm^{-1}, which is considerably narrower than molecular absorption line widths (typically 0.1 cm^{-1} at atmospheric pressure). By accurately controlling the laser temperature and the electrical current that powers the laser, the laser wavelength may be tuned precisely to a specific molecular absorption line that can be selected to be free of interfering absorption from other molecules. [21]

A second type, the vertical-cavity surface-emitting laser (VCSEL) is a type of semiconductor laser diode with laser beam emission perpendicular from the top surface, contrary to conventional edge-emitting semiconductor lasers (also in-plane lasers) which emit from surfaces formed by cleaving the individual chip out of a wafer.

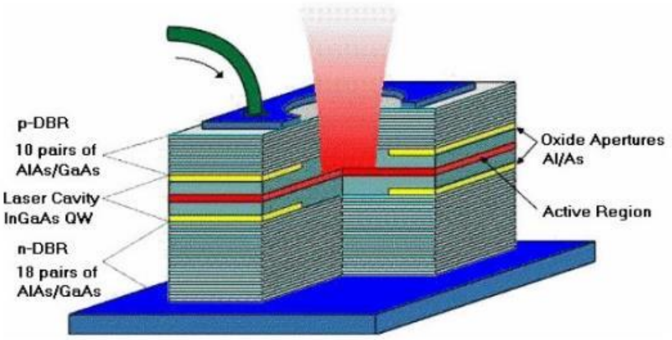


Fig.4. Structure of a vertical cavity surface emitting laser

The high reflectivity mirrors, compared to most edge-emitting lasers, reduce the threshold current of VCSELs, resulting in low power consumption. However, as yet, VCSELs have lower emission power compared to edge-emitting lasers. The low threshold current also permits high intrinsic modulation bandwidths in VCSELs.

A multipass optical cell (Herriot cell) may be utilized to provide a long optical path length within a small volume, in many cases yielding sub-ppm sensitivity with one second or faster response. [22]

Furthermore, techniques known as frequency or wavelength modulation spectroscopy (WMS) and Balanced Ratiometric Detection (BRD) are frequently employed in TDLAS instruments to make them exquisitely sensitive to even very weak absorption of the laser power. [21]

Example of application of TDLAS technology for chemicals detections

An airborne tunable laser absorption spectrometer was used in two polar ozone campaigns, the Airborne Antarctic Ozone Experiment and the Airborne Arctic Stratospheric Expedition, and measured nitrous oxide from an ER-2 high-altitude research aircraft with a response time of 1s and an accuracy ≤ 10%. Laser-wavelength modulation and second-harmonic detection were employed to achieve the required constituent detection sensitivity. [23]

Physical Sciences Inc. has developed hand-held Standoff TDLAS sensors for the inspection of municipal natural gas pipelines. In standoff devices, passive reflectance of a laser beam projected onto walls and other structures enables measurement of path-integrated target gas concentrations over distances up to a few tens of meters. These sensors can be adapted to sense any of the gases listed in Table 7. [21]

Gas	**Detection Limit(ppm-m)**
HF	0.2
H2S	20.
NH3	5.0
H2O	1.0
CH4	1.0
HCl	0.15
HCN	1.0
CO	40.
CO2	40.
NO	30.0
NO2	0.2
O2	50.
C2H2	0.2

Tab.7. Species routinely detection with stand-off TDLAS technique and detection limits

Combining EDFA and WMS provides a long range and robust and modest cost stand-off sensor. They are for example used in the aerial detection of leaks. [25]

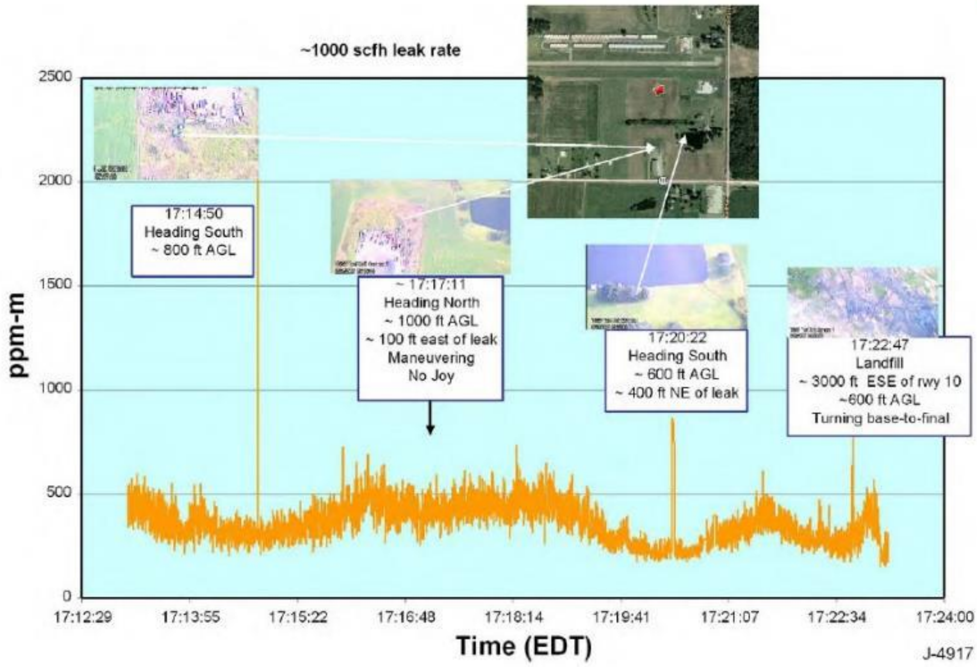


Fig.5. Results from an aerial remote methane leak detection [26]

The table below summarizes the different species measured with TLDAS technology, the accuracy of the measurement and the associated response time.

CO	precision of approximately 3.6% for the TDLAS. Precisions of 1.5 ppbv	5s response time	[28] [27]
CO and CH4	0.1% CH4, 1 % CO	5s response time	[28]
formaldehyde (CH2O)	40-60 pptv 80- 120 pptv 55 pptv	4.5 min 55 sec 20 sec	[34]
NO	measured nitrous oxide with a response time of 1s and an accuracy ≤ 10%.		
N2O, CH4, CO or O3	Accuracy: 10% (2s) ± 1ppbv (for N2O) Resolution: 1ppbv Location on ER-2: Spear Pod, right wing	1s	ATLAS
N2O, H2O isotopes, H2O, CO, CH4, CO2 isotopes, HCl	1 ppb		ALIAS
CO, CH4, N2O			ARGUS
CH3OH, CH2O			NASA Dual Channel Airborne tunable diode Laser Spectrometer [25]

Table 8. Some gases measured by TDLASM

Ammonia	Arsine	Boron trichloride
Boron trifluoride	Carbon disulfide	Chlorine
Diborane	Ethylene oxide	Fluorine
Formaldehyde	Hydrogen bromide	Hydrogen chloride
Hydrogen cyanide	Hydrogen fluoride	Hydrogen sulfide
Nitric acid, fuming	Phosgene	Phosphorus trichloride
Sulfur dioxide	Sulfuric acid	Tungsten hexafluoride

Proven
Likely
Possible
Unknown

Tab. 9. Ability of TDLAS to detect high priority TICs. [26]

RECENT DEVELOPMENTS IN THE DETECTION OF IONIZING RADIATIONS WITH LASER TECHNOLOGY AND THEIR ASSESSMENT IN THE PERSPECTIVE OF AIRBORNE APPLICATION

General orientation about radiation detection

Ionizing radiations can neither be seen by human sense, nor directly measured by instruments. Nevertheless, ionizing radiation interacts with matter, generating some changes of its physical properties (ionization, excitation). In some controlled cases the change or effect can be converted into a numerical value and it can be correlated to the quantity of energy that interacted with the matter, providing a quantitative indication about the quantity of ionizing radiation.

One could distinguish two different strategies with the detection process. In the first one the particles are the target of the detection. Particles travel from the radiological sources to the sensor where they interact with material constituting the sensor (they are trapped). The materials are chosen based on their capacities to convert the energy received from the particles into measurable physical values (light pulse or electric discharge/pulse). All the sensors belonging to this category (crystals, semiconductors and ionization chamber) are passive.

The second strategy considers the ionized molecules as the targets. It considers the quantity of ionized or excited molecules as an indicator of the importance of the ionizing radiation. Any mean estimating the concentration of excited molecules or ions falls in this category (whether they are active or passive). The measurement can be realized with the direct measurement of the molecules concentration (in a plume for example) or by remote detection. In this last case, a medium participate in the detection process. In the case of airborne laser detection, remote detection is favored with the detection of induced emission (fluorescence), reflection or absorption on specific bands of the electromagnetic spectrum.

After the extended bibliographic research we conducted, we can conclude that in the specific case of airborne detection of ionizing radiations, presently only aerial gamma spectrometry is routinely used. Until now it has also been the lonely method to have been recommended by the IAEA[2] for airborne detection. The other methods, concepts or patents we are presenting in the paragraphs below proved to be well founded by theory and experiments but where not yet adapted and applied in the airborne detection. Even airborne experimentations are not mentioned by literature.

Main form of ionizing radiations and strategies with laser detection

Several types of ionizing radiations should be distinguished depending on their characteristics.

Gamma radiation is constituted by high energy photons (energy above 100 keV). As those particles have no charge and a small size they have an important penetration range in the air and a very low specific ionization. As a consequence the ionization products are largely spread in the space and specific ionization is the lowest. Consequently it theoretically requires an important sensitivity in the detection method.

Alpha radiations are constituted by charged particles comparable to a helium nucleus. Alpha particles combine an important size with a double charge which results in the highest specific ionization and the lowest penetration range in the air (several centimeters only). As a consequence, the ionized and excited molecule density around radiological material is maximal in the case of alpha source, which theoretically result in the best opportunity for their detection. As the ionized or excited molecules usually have a particular response for emission or absorption of light, they can theoretically be detected by optical remote sensing methods.

Beta radiations also have a finite range in the air but a more important penetration range compared to alpha particles (several meters). As a consequence the specific ionization is low and the ion cloud around radiological materials has a larger footprint.

<table>
<tr><th colspan="2">Type of radiation</th><th>Ionizing radiation</th><th>Charge</th><th>Speeds</th><th>Range in the air</th><th>Specific ionization (ion pairs/cm) [35]</th></tr>
<tr><td>Electro-magnetic radiations</td><td rowspan="2">Ind. ionizing</td><td>Gamma ray</td><td>0</td><td rowspan="2">Speed of light</td><td rowspan="2">decrease exponentially, never stopped</td><td rowspan="2">5-8</td></tr>
<tr><td rowspan="4">Particles</td><td>Neutron</td><td>0</td></tr>
<tr><td rowspan="3">Directly ionizing</td><td>Electron/particle β-</td><td>-1</td><td rowspan="2">25-99% speed of light</td><td>2-8 meters</td><td rowspan="2">50-500</td></tr>
<tr><td>Positron/particle β+</td><td>+1</td><td>2-8 meters</td></tr>
<tr><td>Ion 4He/particle α</td><td>+2</td><td>3200-32000km/s</td><td>5-6 cm</td><td>20 000-50 000</td></tr>
</table>

Tab.10. Main characteristics of the ionizing radiations.

Depending on the physical characteristics summarized above, particles follows different paths and the ions and excited atoms or molecule are distributed in the space around radiologic materials very differently. Their distribution compared to the sensor sensitivity is an important point to be considered in the detection approach. The figure below proposes a schematic spatial

[2] International Atomic Energy Agency

repartition of the excited/ionized component in the space for alpha, beta and gamma radiation. Alpha radiation leads to the most dense ionization cloud. Beta is similar but more diffuse (on a bigger volume) whereas gamma is totally diffuse. In these conditions – where the goal is to capture lights affected by ionization products – alpha and beta radiation detection seems the most promising.

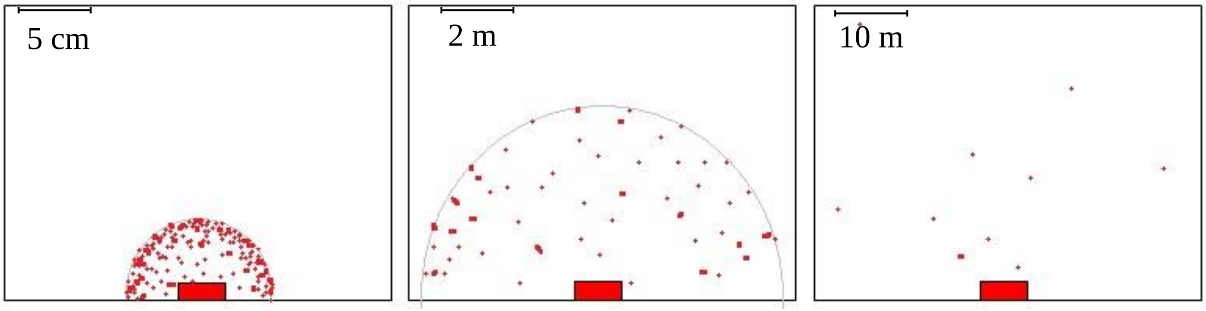


Fig.6. Schematic spatial repartition of ionized and excited molecules for α, β and ɣ radiations

Details about the ionization process and consequences for detection

$X \rightarrow X^+ + e^-$

Ionization happens when the ionizing particle free an electron from an atom. Ionization mechanism leads primarily to the production of an ion pair with a positively charged molecule (or atom) and an electron. The radical can enter secondary reactions -oxidation is the main expected reaction to be expected in the air- leading to more stability.

	N2	**O2**
Ionization	N2→N2++ e-	O2→O2+ + e- O2+ + 2O2 → 2 O3 N2O, HNO3, H2NO2 and NO2.
Excitation	N2→N2*	O2→O2*

Tab.11. Ionized and excited Nitrogen and oxygen and recombination products

Depending on the ionization and the stability of the ionization product, it could be worth not to spot to the measurement of the primary products but to also consider secondary products, whether they are more stable or whether they are more responsive to the detection process.

Importance of chemical kinetics and species' reactivity for the detection process

The detection process target specific species (excited or ionized molecules) into a gas mixture with a very low volumetric (or molar) fraction. [36] In this context the choice of the target specie (or species) should be strategically done in order to reach the highest detection threshold.

Two strategies are relevant to the detection process. The first one is selecting specie with a really specific effect on light (absorption, emission (fluorescence)). Yao has for example noticed that the reactivity of the free radicals to laser light could vary from 5% to 95%.

The second one is selecting specie with the most important population in order to again enhance the detection threshold as much as possible. This last one depends on the life time of the specific specie, which depends on its reactivity (excited and ionized species) and/or stability (mainly for excited species). This can be known from the study of chemical kinetics. Last but not least, the volumetric fraction depends on the mother molecule proportion. In the air, N_2 derivates are the most interesting followed by O_2 derivates.

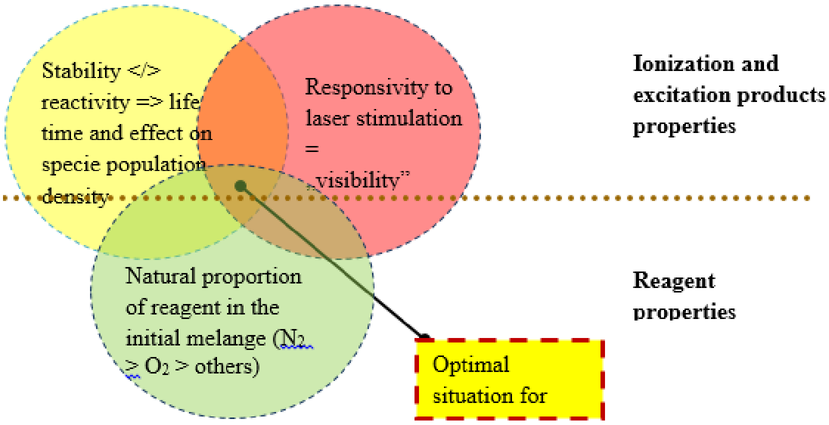


Fig. 7. Optimizing detection strategy

Application of LIDAR with the measurement of laser induced fluorescence in the surroundings of radiological materials

The system indirectly measures radiation by detecting the fluorescence UV light emitted by the ions and excited molecules primarily created by ionizing radiations and secondarily activated by the energy of the laser beam. [37]

The systems employs a DIAL technic with a pulsed laser transmitter, a telescope receiver, and associated control and acquisition systems. [37]

Light propagates out from the laser transmitter and is directed into the volume surrounding the radioactive source, or the "ion cloud." The ion cloud absorbs the transmitted light. This absorption induces otherwise undetectable, non-fluorescing ions to fluoresce. Light from the ion cloud is then backscattered and the telescope receiver subsequently collects the photons from the backscattered light. The intensity of the fluorescence (determined by the photon count) is measured, which provides an indication of the number density of the ionized atoms.

This strategy – which consists into an activation process with laser ligth of the of ions species that otherwise would not fluoresce – rise the fluorescence rate from 5% to 95%. [37]

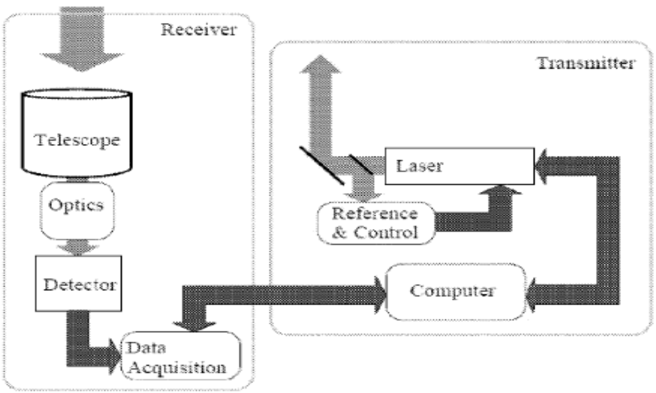


Fig.8. Picture taken from US patent US 20120112076 A1

Algorithms can then be used to relate the measured ionization rates to the source activity.

The invention use active detection of UV light. Contrary to many of the current techniques which use passive detection of ultraviolet (UV) light, the active detection can be used to detect radiation during daylight. The system enables the remote detection of radiation source ranging from 1-1000 m. The preferred wavelength for the laser light is slightly temperature dependent and varies between 390.5 nanometers (nm) and 391.5 nanometers. [37]

The detection range announced in the patent seems compatible with an airborne application under the condition that atmosphere does not absorb UV photons differently than the condition tested in the patent. Adaptation should be considered regarding two components of the system.

First the laser transmitter should be adapted to be able to scan the environment under the carrying platform. The same kind of application is routinely used in LIDAR laser scanning devices utilized in the acquisition of elevation data. Second the telescope should be replaced by an optical system covering a larger foot print on the ground. Another difficulty is to have an acquisition frequency of the detector matching with the scanning rate of the laser transmitter. The last aspect to be considered is the flight speed (and associated platform to be used) to have the required accuracy. [37]

Radiation remote-sensing method based on laser spectroscopic measurement of radiation-induced radicals

Laser spectroscopy could constitute a solution for the measurement of intense radiation fields such as around nuclear reactors, high energy accelerators or nuclear disasters. Tomita et al developed a reliable radiation sensing method with high radiation resistance. They proposed a novel radiation remote-sensing method based on high sensitive cavity ring-down (CRD) laser spectroscopic measurement based on the detection of radiation induced radicals. To verify the detection principle they first have made basic experiments on the CRD spectroscopic measurement of the radiation induced ozone concentration in the air irradiated by ^{60}Co gamma-rays. Secondly they have developed a calculation model to estimate the yields of radiation induced radicals by solving simultaneous rate equations numerically. Through comparison between the experiments and the calculations, they have confirmed the detection principle and the validity of the calculation model, where the results show that the detectable range for the absorbed dose rate range from 4.8×10^{-2} to 3.2 Gy/s with time resolution of 35 sec by controlling the flow rate of the irradiated air. [38]

Such a system could find application on UAV because such platform can first go where radiation level is quite important without risking human life and secondly it can fly at lower speed, even making stationnnary flight (a condition necessary to have sufficient integration time). Two aspects should be considered in the specific case of UAV application. First the energy consomption as laser system is an important energy consumer. Secondly the load of the detection system plus its energy supply should be compatible with available carriage capacity of current UAV. Nethertheless, it should be noticed that in the case of intense radiative environment, aerial gamma spectrometry already fullfil the requirements.

Active remote detection of radioactivity based on the frequency modulation of a probe beam by the rise of electron density induced by laser radiation

The concept uses a laser radiation as a photo-detaching beam and a probe beam to detect electromagnetic signatures in the vicinity of radioactive material. [39]

Radioactive materials emit gamma rays that ionize the surrounding air. The ionized electrons rapidly attach to oxygen molecules, forming superoxide (O_2^-) ions. The elevated population of O_2^- extends several meters around the radioactive material. Electrons are photodetached from O_2^- ions by laser radiation and initiate avalanche ionization, which results in a rapid increase in electron density. The rise in electron density induces a frequency modulation on a probe beam, which becomes a direct signature for the presence of radioactive material. Gamma rays emitted by radioactive material will increase the free electron density as well as the O_2^- density. The concept makes use of laser beams to photoionize the O_2^-, thus providing the seed electrons for air breakdown. [39]

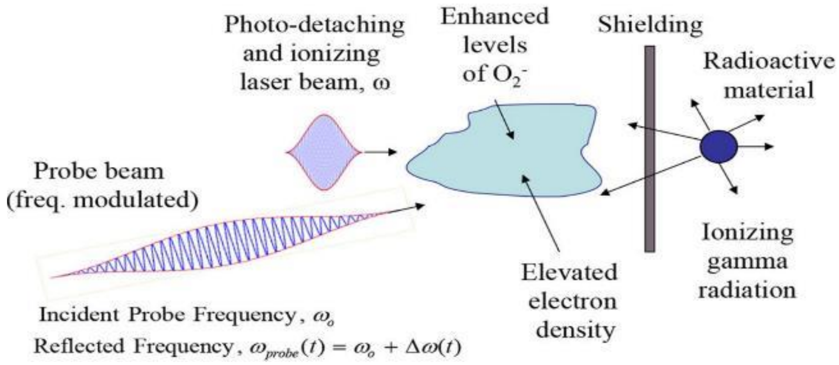


Fig. 9. The effect of laser beam on the ionized air by gamma radiation

As an example of this method of detection, the case is considered where the ionizing laser has a peak intensity of 160 GW/cm^2 and pulse duration of 1ns. The probe beam is a millimeter wave source of frequency 94 GHz. In the absence of radioactive material there is no frequency modulation of the probe. For $\alpha_{rad} = 10^3$ and a probe-beam interaction distance of 10 cm, the fractional frequency modulation is significant, around 5%, which is readily detectable. In other words, the frequency shift is the sought-for electromagnetic signature of radioactive material and can be measured. [39]

The author stated that standoff detection can be done from distance geater than 100 m. In the experiment presented in the paper, the distance probe source is 10 cm and allow a detection of 5 percent of fractional frequency modulation. [39]

The author considers the detection of enhanced levels of O_2^- generated by a shielded gamma radiation source. If considering Alpha or Beta source without shielding, the α_{rad} would be much more favorable for the detection in the volume where alpha and beta ray are ranging.

Standoff alpha radiation detection via the measurement of energy aborbed by excited state molecules.

Yao emphasizes the fact that methods employing the detection of faint light (fluorescence) or backscattered laser light (in DIAL application) have limited detection capacities when the distance between the laser source and the target is increased. [40] The reason is that spontaneous emission radiates uniformly within an entire 4p solid angle, so its intensity drops rapidly according to 1/r^2 law, where r is the standoff distance. The same intrinsic problem of propagation loss happens with backscattered laser light and still limits the distance of the standoff detection. [40]

To overcome these limitations, Yao proposes to base the alpha radiation detection on the measurement of the transmitted laser energy in order to determine (by subtraction) the quantity of absorbed energy at specific wavelength. Since the probe beam is a collimated beam, its propagation does not suffer the fundamental limitation of 1/r^2 propagation loss. [40]

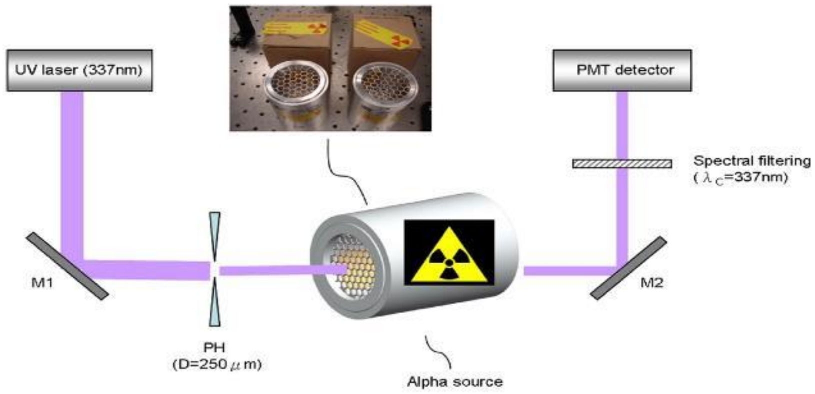


Fig. 10. Schematic illustration of the experimental setup

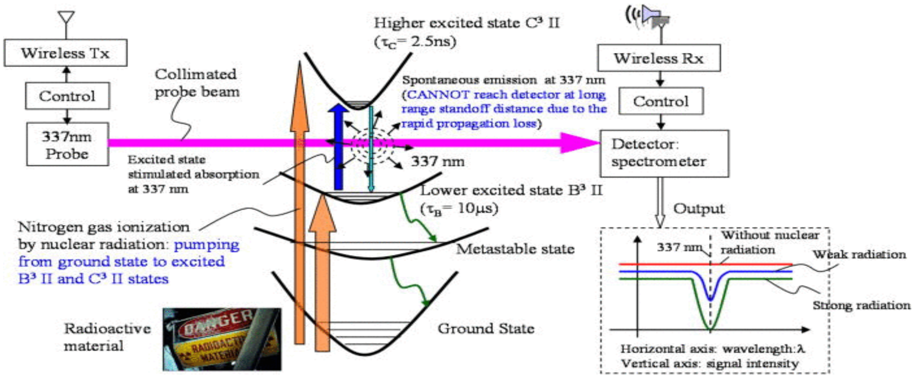


Fig. 11. Conceptual illustration of standoff detection of radiation via excited state absorption of radiation excited/ionized air

Experiments were done at distance of 0.5 and 10 m with a 337 nm UV probe beam through a 40 mCi Po-210 alpha source. The detected signal as a function of time is not sensitive to the separation distance between the light source and alpha radiation source. [40]

This technology has a potential to realize long range standoff detection but because of the bi-static measurement dispositive – in which the UV radiation source and the detector are on the

opposite sides of the beam path – this technique is applicable only for stationary measurements. Consequently application in airborne detection and measurement is excluded.

CONCLUSION

The review of airborne applications of laser measurement methodologies demonstrated both the numerous specialties where it is applied – like environmental monitoring, chemical warfare protection, pollution detection and monitoring, meteorology – and the diverse species of chemical measured like water vapor, aerosols, nerve agent, greenhouse gases. The technics introduced deserve different goals with different detection capacities. DIAL technology range from several hundred ppm to ppt detection capacity and allows an integrated measurement approach (layer or profile approach). TDLS provides punctual but very precise measurements (ppb or ppt detection measurements).

The extended bibliographic researches conducted about the airborne radiation detection first of all highlighted that laser detection is not currently employed neither airborne experimentation was mentioned in the literature. Only aerial gamma spectrometry is routinely employed for this purpose. The reasons are twofold: gamma radiations are the most penetrating radiations so they better allow the detection from distance and most of the radiological materials can be detected from the gamma radiations they or their daughter products emit. Nevertheless methods employing active laser technology were developed and patented for the standoff or remote detection of alpha, beta and gamma radiations. With further developments or adaptations they could potentially find application in airborne detection. Important concepts emerged from our analysis. Only systems employing light backscattering have the potential to be adapted to airborne measurement. This results in a propagation loss that follows a $1/r^2$ law which limits the range for detection. Expectations for alpha and beta airborne measurement should be based upon that. Then several innovative ideas could be extracted from the measurement systems:

- The possibility to push target molecules to fluoresce by pumping electrons to an energy level that naturally does not occur.
- Exploring further the effect of ionization on the fractional frequency modulation.
- Targeting free radical species which have longer lifetime and associate denser population.

References:

[1] M. Wirth, A. Fix, P. Mahnke, H. Schwarzer, F. Schrandt, G. Ehret: The airborne multi-wavelength water vapor differential absorption lidar WALES: system design and performance. Applied Physics B. Volume 96, Issue 1 , pp 201-213. 2009-07-01. DOI 10.1007/s00340-009-3365-7

[2] R.M. Schotland, J. Appl. Meteor. **13**, 71–77 (1974)

[3] E.V. Browell, S. Ismail, W.B. Grant: Differencial Absorption Lidar (DIAL) measurements from air and space. Applied Physics - Laser and optics. p. 399-410 (1998)

[4] E.V. Browell, S. Ismail and W.B. Grant: LIDAR | DIAL, In Encyclopedia of Atmospheric Sciences, edited by James R. Holton, Academic Press, Oxford, 2003, Pages 1183-1194, ISBN 9780122270901, http://dx.doi.org/10.1016/B0-12-227090-8/00204-9.

[5] Space lidar http://www.nasa.gov/centers/langley/news/factsheets/LITE.html

[6] COLIPSO http://www.nasa.gov/pdf/137028main_FS-2005-09-120-LaRC.pdf

[7] http://www.spectrasyne.ltd.uk/html/about_dial.html

[8] http://www.spectrasyne.ltd.uk/html/technique.html

[9] R. J. Alvarez II, C. J. Senff, A. O. Langford, A. M. Weickmann, D. C. Law, J. L. Machol, D. A. Merritt, R. D. Marchbanks, S. P. Sandberg, W. A. Brewer, R. M. Hardesty, R. M. Banta: Development and Application of a Compact, Tunable, Solid-State Airborne Ozone Lidar System for Boundary Layer Profiling. *Journal of Atmospheric and Oceanic Technology* **28**:10, 1258-1272. Online publication date: 1-Oct-2011. L.

[10] E. V. Browell, A. F. Carter, S. T. Shipley, R. J. Allen, C. F. Butler, M. N. Mayo, J. H. Siviter Jr. and W. M. Hall: NASA multipurpose airborne DIAL system and measurements of ozone and aerosol profiles. Appl. Opt., 22, (1983) 522–534.

[11] E. Uthe , J. Livingston, and N. Nielsen: Airborne lidar mapping of ozone concentrations during the Lake Michigan ozone study. J. Air Waste Manage. Assoc., 42, (1992), p.1313–1318.

[12] R. J., II Alvarez, C. J. Senff, R. M. Hardesty, D. D. Parrish, W. T. Luke, T. B. Watson, P. H. Daum, and N. Gillani: Comparisons of airborne lidar measurements of ozone with airborne in situ measurements during the 1995 Southern Oxidants Study. J. Geophys. Res., 103, (1998), 31 155–31 171.

[13] G. Ancellet, and F. Ravetta: Compact airborne lidar for tropospheric ozone: Description and field measurements. Appl. Opt., 37, (1998), p. 5509–5521.

[14] M.H. Proffitt and A.O. Langford: Ground-based differential absorption lidar systemfor day or nightmeasurements of ozone throughout the free troposphere. Appl. Opt., 36, (1997) p. 2568–2585.

[15] Coutts, D., and A. J. S. McGonigle: Cerium-doped fluoride lasers. IEEE J. Quantum Electron., 40, (2004) p. 1430–1440.

[16] K. A. Elsayed, S. S. Chen, L. B. Petway, B. L. Meadows, W. D. Marsh, W. C. Edwards, J. C. Barnes, and R. J. DeYoung: High-energy, efficient, 30-Hz ultraviolet laser sources for airborne ozone-lidar systems. Appl. Opt., 41, (2002) p. 2734–2739.

[17] A. Fix, M.Wirth, A.Meister, G. Ehret, M. Pesch, and D. Weidauer: Tunable ultraviolet optical parametric oscillator for differential absorption lidar measurements of tropospheric ozone. Appl. Phys. B, 75, (2002) p.153–163, doi:10.1007/s00340-002-0964-y

[18] L. Halász: The role of remote sensing equipment in air monitoring systems. In: NATO Series of Disarmement Technologies, Vol. 13, Kluwer, Dodrecht, 1997, p. 241–253

[19] L. Halasz, I. Pinter, A. Solymar Szocs: Remote sensing in the biological and chemical reconnaissance. AARMS, vol 1. Issue 1 (2002), 39-56.

[20] R.M. Hoff, M. Harwood, A. Sheppard, F. Froude, J.B. Martin, W. Strapp: Use of airborne LiDAR to determine aerosol sources and movement in the Lower Fraser Valley (LFV), BC, Atmospheric Environment, Volume 31, Issue 14, July 1997, Pages 2123-2134, ISSN 1352-2310, http://dx.doi.org/10.1016/S1352-2310(96)00302-0.

[21] http://www.tdlas.com/theory.shtml

[22] http://www.tdlas.com/applications.shtml

[23] J. Podolske, M. Loewenstein: Airborne tunable diode laser spectrometer for trace-gas measurement in the lower stratosphere, Appl. Opt. 32, 5324-5333 (1993) http://www.opticsinfobase.org/ao/abstract.cfm?URI=ao-32-27-5324

[24] Physical Inc. http://www.psicorp.com/pdf/library/VG07-187.pdf

[25] https://airbornescience.nasa.gov/instrument/DCALS

[26] M. B. Frish, R. T. Wainner, M. C. Laderer, B. D. Green and M. G. Allen: Standoff and Miniature Chemical Vapor Detectors Based on Tunable Diode Laser Absorption Spectroscopy. http://www.psicorp.com/pdf/library/SR-1343.pdf

[27] J. S. Holloway, R. O. Jakoubek, D. D. Parrish, C. Gerbig, A. Volz-Thomas, S. Schmitgen, A. Fried, B. Wert, B. Henry, J. R. Drummond: Airborne intercomparison of vacuum ultraviolet fluorescence and tunable diode laser absorption measurements of tropospheric carbon monoxide. Journal of Geophysical Research: Atmospheres (1984–2012). Volume 105, Issue D19, pages 24251–24261, 16 October 2000.

[28] G. W. Sachse, J. E. Collins, Jr., G. F. Hill, L. O. Wade, L. G. Burney et al.: Airborne tunable diode laser sensor for high-precision concentration and flux measurements of carbon monoxide and methane, Proc. SPIE 1433, Measurement of Atmospheric Gases, 157 (May 1, 1991); doi:10.1117/12.46162; http://dx.doi.org/10.1117/12.46162

[29] A. Fried, P. Weibring, D. Richter, J. Walega, C. Roller et al.: tunable diode laser and difference frequency generation absorption spectrometers for highly sensitive airborne measurements of trace atmospheric constituents, Proc. SPIE 6378, Chemical and Biological Sensors for Industrial and Environmental Monitoring II, 63780F (October 17, 2006); doi:10.1117/12.691318; http://dx.doi.org/10.1117/12.691318

[30] B. P. Wert, A. Fried, and J. R. Drummond: Airborne measurements of tropospheric formaldehyde by tunable diode laser absorption spectroscopy, Proc. SPIE 2834, Application of Tunable Diode and Other Infrared Sources for Atmospheric Studies and Industrial Process Monitoring, 175 (October 21, 1996); doi:10.1117/12.255323; http://dx.doi.org/10.1117/12.255323

[31] https://www.servomex.com/Servomex/web/web.nsf/en/delta-f-moisture-technology

[32] http://212.201.48.1/course/c320352/Presentation/NS%20Vertical%20Cavity%20Surface%20Emitting%20Laser.pdf

[33] Aerial Methane Leak Detection. Asel-Tech http://asel-tech.com/documents/Asel-Tech%20aRMLD.pdf

[34] A.n Fried, B. P. Wert, B. Henry, J. R. Drummond: Airborne tunable diode laser measurements of formaldehyde, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, Volume 55, Issue 10, September 1999, Pages 2097-2110, ISSN 1386-1425,
http://dx.doi.org/10.1016/S1386-1425(99)00082-7
(http://www.sciencedirect.com/science/article/pii/S1386142599000827)

[35] specific ionization of particles
http://www.fas.org/nuke/guide/usa/doctrine/dod/fm8-9/1ch2.htm

[36] C. E., Moss, R. M. Goeller, D. F. Milligan, J. E. Valencia, J. Zinn: Remote sensing of radiation. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 422(1-3) (1999) 832 – 836.

[37] GEORGIA TECH RESEARCH CORPORATION, GEORGIA : Remote detection of radiation, US Patent US 20120112076 A1, issued 2012.01.11.

[38] H. Tomita, K. Watanabe, J. Kawarabayashi, T. Iguchi: Development of novel radiation remote-sensing method based on laser spectroscopic measurement of radiation-induced radicals, Proc. SPIE 5198, Hard X-Ray and Gamma-Ray Detector Physics V, 281 (January 20, 2004); doi:10.1117/12.506447; http://dx.doi.org/10.1117/12.506447

[39] P. Sprangle, B. Hafizi, H. Milchberg, G.S. Nusinovich, A. Zigler: Active remote detection of radioactivity based on electromagnetic signatures. 30 October 2013, SPIE Newsroom.

[40] J. Yao, J. Brenizer, R. Hui, S. S. Yin. Standoff alpha radiation detection via excited state absorption of air. Appl. Phys. Lett. 102, 254101, (2013); doi: 10.1063/1.4812338