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OPTICAL REMOTE SENSING FOR AIR POLLUTANTS–REVIEW

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Air monitoring techniques need to be simple, unobtrusive, acquire real-time data, have increased sensitivity, and the ability to analyze many compounds simultaneously. Optical remote sensing techniques address many of these criteria for air monitoring methods. Optical remote sensing uses light energy between ultraviolet and midinfrared to detect and measure contaminants in situ. *While optical remote monitoring techniques provide similar monitoring accuracy compared to conventional monitoring methods, they also provide certain unique advantages. Advantages and disadvantages of specific optical remote sensing techniques is discussed in this review, which focuses on the techniques most useful to the industrial hygiene profession. Detailed information about every technique will not be supplied; rather the review attempts to provide an understanding of the development of the remote sensing techniques, and briefly explains typical applications, so that readers can understand and evaluate applications appropriate to their work.*

Air monitoring plays an important role in industrial and environmental health science. It is an essential ingredient that has helped to make modern industrial hygiene a profession that not only prevents acute forms of occupational disease, but also protects workers against long-term adverse effects from chemical exposure.(1) Quantitative and qualitative information from air monitoring programs enable the industrial hygienist to predict the effects of the work environment on a worker. For the environmental scientist it elucidates the complex interactions between the emission source, the pathway, and exposures in surrounding communities.

Currently, air sampling strategies involve general area (point and multi-point) and personal sampling. General area sampling evaluates occupational exposure to workplace contaminants in the vicinity of workers at approximately breathing zone elevation. This introduces some uncertainty when evaluating the precise exposure of each worker.(2) Direct reading instruments have the temporal resolution necessary to measure sudden departures and short-term exposures. However, these instruments normally operate by monitoring for a specific chemical, which means if other hazardous chemicals are present they will not be detected, and in some cases may interfere. Often direct reading instruments do not have the specificity and sensitivity needed for monitoring in a complex work environment.(3)

Personal sampling tracks exposure as workers move and engage in a variety of operations producing varying amounts of air contaminants. The main advantage of personal sampling is that air contaminants in the breathing zone are measured directly, giving better spatial resolution than area monitors. A disadvantage is that it requires a worker to wear a sampling device throughout the sampling period. Another drawback is that personal sampling methods require numerous steps before obtaining analytical results. These steps may include collection of an air sample onto a medium, shipment to a laboratory, desorption of an analyte(s), and analysis by proper instrumentation. Not only does such a complicated procedure increase the chance of biased results due to contamination and loss, but also it usually results in significant delays in acquiring results. Concerns over exposure to acutely toxic chemicals, and evaluation of equipment or process areas for leaks or fugitive emissions, cannot be addressed with personal sampling because these devices are almost always optimized for time-weighted average (TWA) and not short-term exposure or ceiling exposure monitoring.

Improvements in air monitoring techniques in the past decade have focused on combining the advantages of personal and area sampling. Techniques need to be simple, less obtrusive, acquire real-time data, have increased sensitivity, and have the ability to analyze many compounds simultaneously. Optical remote sensing techniques address many of these criteria for air monitoring methods.

The establishment of the Clean Air Act Amendments (CAAA)(4) in 1990 has increased interest in new monitoring technologies. Some of the requirements made with these amendments include implementing maximum achievable control technology (MACT) standards and promulgation and setting of threshold quantities for air pollutants that pose the greatest risk of accidental release.

In the environmental arena continuous real-time monitoring is essential for industry to monitor emissions along fence

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TABLE I. Optical regions and remote sensing techniques

Optical region	Wavelength limits	Method	Detection principle	Target compound	Comment
Ultraviolet	200–400 nm	UV-DOAS DIAL	Absorption	SO_2, H_2S, OH, NO_x, Hg, Cl_2, Aromatic compounds	Interference from O_2
Visible	400–700 nm	UV-Vis	Absorption	SO_2, NO_2, O_3, H_2O	Only a few compounds have absorption in this region
Near infrared	700–2500 nm	DIAL, LEL Monitor	Absorption	CO_2, CO, H_2O O_2	Not for trace species due to weak absorption
Middle infrared	2500–25000 nm	IR, FTIR, DIAL, Laser, Gas correlation	Absorption Emission, Fluorescence	Most organic vapor, gases	Interference from H_2O and CO_2
Far Infrared	0.025 to 0.5 mm		Absorption Emission	H_2O, O_3, N_2O, NO_2	Not useful due to strong interference from H_2O absorption

lines or to track sudden releases. Fast changes in atmospheric contaminant composition and concentration depending on current meteorological conditions make real-time monitoring a requirement not a choice. Spellicy et al.[5] have addressed the role of remote sensing in association with the CAAA. It appears that remote sensing techniques may provide a complimentary approach to traditional single or multi-point monitoring of the workplace and environment.

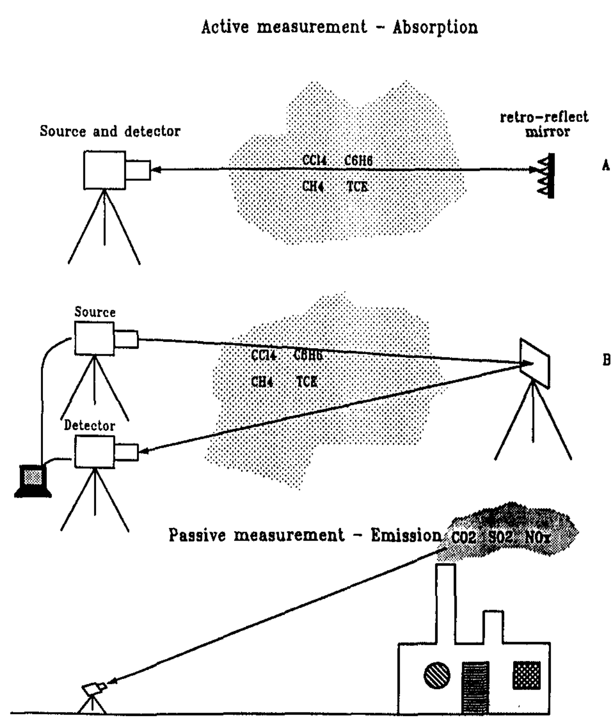

FIGURE 1. Schematic diagram of configurations of remote sensing measurements.
(A) Monostatic with retro-reflector that returns the beam to its point of origin (B) Bistatic configuration. Active (absorption) method shown (middle) and passive (emission) method shown (bottom).

OPTICAL REMOTE SENSING

Overview

The terminology of remote sensing has been compiled by Vaughan,[6] and Russwurm and Childers have developed a guidance document that includes a glossary of terms used in open path Fourier transform infrared (OP-FTIR) spectroscopy.[7] In a review paper by Grant, et al.,[8] the application of optical remote sensing technologies to environmental health have been discussed extensively. Minimum detectable concentrations (MDC) of various volatile organic compounds by optical remote monitoring methods were listed. However, these MDC values were calculated assuming no interferences from water or carbon dioxide lines, which is unlikely.

Remote sensing is defined as the measurement of gases, solids, and liquids *in situ* without the use of a closed sample cell in the instrument, or a sample storage medium.[7,9] Optical remote sensing uses light energy between ultraviolet (UV) and mid-infrared (IR) to detect and measure contaminants *in situ*. Contaminants interact with the beam of light causing absorption or scattering of the beam. Absorption of light by contaminants in specific spectral regions can be used both for qualitative and quantitative analysis. Scattering can be used to detect the presence of aerosols or gases, and give ranging information. Table I summarizes the optical regions used for optical remote sensing, along with typical compounds measured and applications in each optical region.

Optical remote sensing systems operate in either an active or passive mode, although there are some that do both, as shown in Figure 1. Active systems operate with a controlled source of light, like a lamp or a laser, and a telescope and detector. Passive systems are similar, but use uncontrolled light sources such as the sun, emission source heat, or ambient heat. Passive monitoring is generally performed only when active methods cannot be used, due to difficulties in determining source temperatures. It is necessary to know the source temperature, or any change in temperature, because it directly effects spectra.

While optical remote monitoring techniques provide similar monitoring accuracy compared to conventional monitoring methods, they also provide certain unique advantages. There are applications that can be achieved only with remote measurement. Optical remote sensing offers the following advantages over traditional monitoring:

(1) Remote measurement—Unlike traditional methods, with remote sensing there is no contact during sampling. Consequently, sampling errors introduced during collection are minimized. Adsorption to tubing, canisters, or sampling bags is eliminated. In comparison, many traditional methods are not suitable for measurement of polar and labile compounds because of the high and unpredictable losses in the sampling system.

Remote measurement also allows for monitoring of inaccessible or isolated areas. An example of this is the monitoring of stack emissions from a location on the ground outside the fence line of a plant.

(2) Speed—Most remote sensing measurements are made in real time or near real time.

(3) Multi-compound analysis—Some systems, such as Fourier transform infrared spectroscopy (FTIR), measure numerous compounds simultaneously.

(4) Path integrated measurement—Long path measurements average emission fluxes that are affected by wind or a localized source. Point source monitoring may miss plumes or emissions that wander, while a beam path is more likely to capture the emission.

Disadvantages common to optical remote sensing instruments include:

(1) Collection of background spectra—The primary problem facing users of open-path optical methods is the acquisition of a clean background spectrum. This clean air background spectrum is used to remove, through ratioing with the sample spectrum, spectral features due to alignment, source characteristics, and interferences. After ratioing, the remaining spectral features are due to the target analytes. With closed path systems it is easy to obtain a clean background by using a separate gas cell or collecting a background before or after the sample. With open path systems used in industrial or environmental settings, it is difficult to collect a clean air background because the target analyte cannot be removed.[7,9]

(2) Instrument cost—Most optical remote sensing instruments are in limited production. Cost can range between $70–170,000.

(3) Technical expertise—Instrument operation and analysis of data requires an operator with some technical background and specific training.

(4) Path averaging—Open path systems integrate concentration along the beam path. By dividing by the total path length, a path averaged concentration is obtained. There is no difference between 10 ppm of a contaminant well-mixed along a 10 m beam path, or 100 ppm contained within 1 m of a 10 m beam path and 0 ppm in the remaining 9 m. This decrease in spatial resolution needs to be addressed if remote sensing is to be used to determine actual exposure concentrations. An exception is the light detection and ranging (LIDAR) technique, discussed in this review.

Advantages and disadvantages of specific optical remote sensing techniques will be discussed further in the review.

This review will focus on the techniques that are the most useful to the industrial hygiene profession. Some techniques are still in the research stage. However, the authors feel it is important that the industrial hygiene community be made aware of these developing technologies. Detailed information about every technique will not be supplied, rather, the review attempts to provide an understanding of the development of the remote sensing techniques and briefly explains typical applications, so that readers can understand and evaluate applications appropriate to their work.

The review begins with a short discussion of spectral analysis methods. This is followed by a section on laser optical remote sensing methodologies that rely on scattering or absorption of laser light, or a combination of both, to analyze samples. Next, methods utilizing the UV optical region are discussed. The third topic covered is infrared (IR) optical remote sensing. This section includes discussions of gas filter correlation, long path gas cell, FTIR spectroscopy, and near-IR for sensing of conditions approaching the lower explosive limit (LEL). Finally, tomography is discussed as it applies to industrial hygiene.

Spectral Analysis

Spectral analysis plays a very important role in acquiring quantitative information from data obtained when using an optical remote sensing instrument. Various methods have been used for spectral analysis that utilize characteristic responses to light to identify and measure properties of analytes present in an air sample. In general, remote sensing measurement yields an absorbance or transmittance spectrum. The relationship between absorption peak intensity of a species in the spectrum and concentration is expressed by the Bouguer-Beer-Lambert law as follows:

$$A(v) = -\log(T) = a(v)cl$$

where A(v) and a(v) are the absorbance and absorbtivity at wavenumber v, respectively; T is transmittance; c is the concentration of the sample; and l is the path length. Absorbance is a unitless quantity. Therefore, a(v) is expressed in reciprocal units of concentration and path length. The quantity cl can be calculated using the absorbance spectrum generated by the remote sensing instrument. The units used are concentration times length, usually ppm-m or ppb-m.[6] The path averaged concentration is obtained by dividing the path integrated concentration by the total path length.

Multiple component quantitative analysis is usually performed using classical least squares fit,[10] partial least squares fit,[11] and principle component regression techniques.[12] The CLS method is the most robust of these analytical tools. In this context, "robust" means that these methods can be used when spectral bands to be analyzed are overlapped or affected by baseline variation. It is necessary for target compounds to be defined before these methods are used.

Spectral data obtained with FTIR spectroscopy can also be used to identify unknown compounds in the sample. Compounds are identified based on peak locations and intensities using the relationship between the chemical structure of compounds and their molecular spectra. Identification methods try to mimic human logic to determine components of spectra. Some of the techniques used are pattern recognition, expert systems, and neural-networks.[13-16]

LASER OPTICAL REMOTE SENSING TECHNOLOGY

Lasers possess many properties that make them ideal spectroscopic tools. Specificity in spectroscopy depends on the spectral width of radiation being narrower than the absorption lines of a sample. Lasers can produce highly monochromatic and collimated light with very narrow line widths (< 0.001 cm−1), providing greater specificity than is found in conventional spectroscopy. Other important laser properties include tunability over wide spectral regions, and high intensity so that measurements can be made over long distances. Laser techniques tend to be more applicable to environmental monitoring because of the high power necessary for long path monitoring. The main concern in using these methods in the workplace is the issue of eye safety.

Laser optical remote sensing techniques depend on the mode in which laser radiation interacts with the target gas or aerosol. In general, the interaction can be characterized into two categories: absorption and scattering. Specific information regarding chemical composition, content, and distribution in the atmosphere can be determined by observing absorption bands and scattering phenomena.

Absorption Technique

Direct absorption is a common technique used to obtain concentration information in laser remote sensing. This technique relies on observing the attenuation of a laser beam when the laser beam frequency is matched to an absorption band of a specific compound. To detect the absorption of a spectral line, and thereby determine the concentration of a target compound, differential absorption is usually employed.

Figure 2 illustrates the use of differential absorption. Two laser beams are alternately emitted by the instrument and measured at slightly different wavelengths (v_{on}, v_{off}). One wavelength (v_{on}) is selected "on" the absorption line of a target compound, and the other (v_{off}) is located "off" the absorption. The attenuation of the light by the target compound at the absorption wavelength (v_{on}) is compared with the attenuation at the neighboring wavelength (v_{off}) to determine the concentration. The "on" and "off" wavelengths must be carefully selected to avoid interferences from absorption lines of other species. Inaccurate measurement of the absorption coefficient of an absorption line invariably results in erroneous quantitative information.

A gas cell is ordinarily used to determine the absorption coefficients of various gases. Different techniques, such as carbon dioxide (CO_2) laser and high resolution FTIR, have been used to calculate absorption coefficients. Low ppb minimum

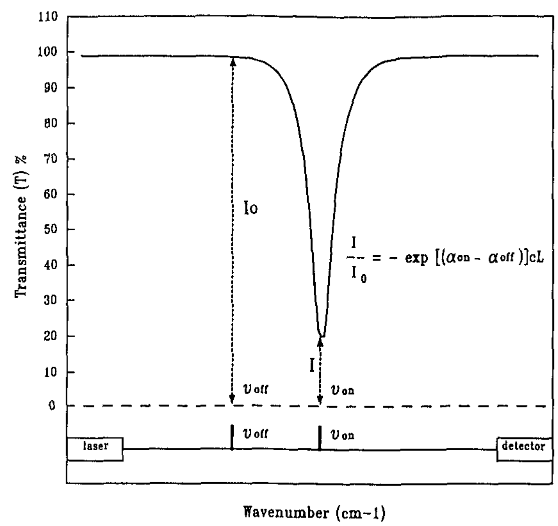

FIGURE 2. Schematic representation of the differential absorption technique in determination of a gas concentration ("c" in the equation on the figure). I_0 and I are the incident and transmitted intensity respectively. a_1 and a_2 are absorption coefficients at v_{on} and v_{off} respectively. L is the total path length.

detectable concentrations of certain investigated gases and vapors have been demonstrated. Results of studies have been used to estimate sensitivities for field measurements of gases and vapors using CO_2 and tunable diode lasers.[17-19]

Real time measurement of methanol in air has been demonstrated using a 1.66-μm diode laser operating at room temperature. The minimum detectable concentration of the method was 0.3 ppm for a signal averaging time of 1.3 seconds.[20] Kreuzer et al. reported that CO and CO_2 lasers were used to determine 10 gaseous pollutants at the ppb level.[21] Laser spectroscopy has also been applied to the determination of sulfur dioxide concentrations in estimating oil consumption at steady-state automobile engine operating conditions.[22]

Several remote sensing systems based on CO_2 laser long path differential absorption have been built for workplace exposure measurements. These systems have short response times and give path averaged concentrations of measured gases such as ammonia, ethylene, and chlorine-dioxide.[23-25] A compact, fully automatic CO_2 laser long path absorption system was specifically built to monitor workplace gaseous pollutants.[26] In this system the laser beam was transmitted via a periscope at a height of 2.1 m to avoid interruption in measurements from people blocking the beam. A low-power laser was used to ensure complete eye safety, even when eyes were directly exposed to the output beam. The system has been used to measure several gaseous pollutants such as ammonia and methyl ethyl ketone in the work environment.

Future Direction

Remote sensing laser absorption spectroscopy will find increasing applications to workplace air monitoring in the future. As lasers become cheaper and more stable, they will al-low greater accuracy and precision for monitoring results at a lower price. However, the method will almost always be limited to monitoring of species that have an absorption peak that is not overlapped with peaks of other compounds. Thus, this method will be more applicable to monitoring of workplaces in which a single air contaminant is present than for workplaces in which a mixture of air contaminants is present.

Light Detection and Ranging (LIDAR) Technique

LIDAR is widely used in laser remote sensing for detection of pollutants in the atmosphere. It is the optical equivalent of radar, except for the use of an optical laser rather than a microwave source.

In this technique, short wavelength laser radiation is transmitted through the atmosphere where gas molecules and particles along the beam path cause absorption and scattering of the laser beam. The absorption and/or scattering is then detected. A small fraction of the radiation is backscattered from molecules and aerosols in the direction of the LIDAR system. Topographical targets, such as hills, buildings, and walls, can be used as radiation retro-reflectors. The detected signal is used to identify and measure chemical concentrations based on either differential absorption or scattering.(27)

Since light travels at a known velocity, the time interval between sending the light out and receiving the signal from back-scattering can be used to obtain ranging information. The direct observation of the backscattered laser beam provides spatially resolved concentration data of a target compound in the atmosphere. Differential absorption measurements in combination with LIDAR make it possible to determine the presence and concentration of air pollutants using selective wavelengths (v_{on}, v_{off}), specific to the compound of interest.

This method was first suggested by Schotland(28) to remotely evaluate water vapor content in the atmosphere. Most researchers refer to this technique as DIAL, which stands for differential absorption LIDAR, although it has also been called DAS (differential absorption and scattering).

DIAL systems have gained more attention in environmental monitoring in the past decade. A critical review by Zanzottera(29) thoroughly discusses the theory and applications of the DIAL technique to determine trace pollutants in air. The gases and vapors measured using various DIAL techniques include water, carbon monoxide, ozone, nitrogen monoxide, nitrogen dioxide, sulfur dioxide, chlorine, mercury, and hydrocarbon compounds.(30–37) Sources of analytical error associated with DIAL measurements due to aerosols, temperature, and interferent gases also have been investigated.(38,39)

As mentioned, the other interaction that takes place when light is propagated through the atmosphere is scattering. There are different scattering processes. These include (1) Rayleigh and Mie scattering, (2) Raman scattering, and (3) resonance scattering and fluorescence. Since measurement of scattering alone provides limited information in remote sensing, only a brief discussion is given here.

Rayleigh and Mie scattering is used to provide a backscattered signal in LIDAR to obtain spatial information. In addition, Mie scattering can be applied to dust analysis to determine the size distribution and mass concentration of atmospheric aerosols. Remote sensing analysis of aerosols based on Mie theory has been published by several researchers.(40–43) Raman scattering does not have a return signal sufficient for analysis of trace species in the atmosphere. Consequently, it is most likely to be employed in remote monitoring of effluent plumes, where concentrations can be quite high (hundreds of ppm),(44) and when range is limited to about 1 km.(45)

A laser-based system has been developed by the Lawrence Livermore National Laboratory that can be used to locate leaks and fugitive emissions. The system is called backscatter/absorption gas imaging (BAGI).(46) The system does not quantify gas leaks. It locates gas releases and estimates relative concentrations.

BAGI uses backscattered laser light from surfaces to create a video image. The reduction in light due to a gas is shown on the video image in contrast to the gas free background.

BAGI systems operate over short ranges (30 m) with IR laser sources. Research is continuing to extend ranges to up to 300 m. A shoulder mountable system is available that can be calibrated for up to 77 gases. The sensitivity of the BAGI system for these gases has been reported.(47) Workplace applications of this technology include locating cylinder and pipeline leaks in real time from a distance.

In summary, laser remote sensing techniques provide high resolution, specificity, and sensitivity. Compounds that have absorption in optical regions ranging from the UV to the IR can be detected if a suitable laser is available and if an interference-free absorption band can be found. The DIAL system can provide 2D or 3D information of air pollutants.

Very powerful lasers are required to provide a sufficient signal to measure trace pollutants in the atmosphere since most systems are designed for long-distance measurements and little backscattering signal is received. The main concern with laser systems is the risk of damage to the eye. Modification of systems to use a low power or eye safe laser source and reduction of cost is necessary before these techniques will be applied to workplace monitoring.

Future Directions

It appears that in the near future this method will be used primarily for environmental atmospheric composition, concentration, and dispersion modeling studies, including fenceline and stack monitoring to characterize the distribution of pollutants, and for process leak detection.

High pulse power is required for the laser because the returning signal is so weak. This will continue to limit severely the applicability of this method in the workplace. The BAGI system seems to be the most useful laser system for the workplace, although it operates with a class IV laser. LIDAR systems are large and complex, and currently not easy to operate. Thus, if future applications to the workplace are to take place, these problems need to be overcome.

LONG PATH ULTRAVIOLET (LPUV) SYSTEM

LPUV systems use a classic analytical technique for concentration measurement. The technique involves measuring the attenuation of a beam of UV radiation that has been directed through the atmosphere, in which chemical species absorb UV radiation at characteristic wavelengths. Usually, the UV system uses least-squares fitting or spectral differentiation to analyze spectra.

Many gases and vapors such as sulfur dioxide,[48] ozone,[49] hydrogen sulfide, hydroxide, chlorine, nitric oxide,[50] nitrogen dioxide, and aromatics[51] have strong absorption features in the UV region. However, the region is limited by the absorption of oxygen[52] below 220 nm. UV-DOAS (UV-differential optical absorption spectrometry) is commonly used for long-path measurements.[3,53-55] UV-DOAS uses the same method to determine a target concentration as differential techniques mentioned earlier. Several studies have been published describing the application of long path UV-DOAS to measure ambient concentrations of nitrous acid, ozone, sulfur dioxide, nitrogen dioxide, formaldehyde, nitrate radical, and ammonia.[56-58] Currently, up to 20 compounds have been detected by using UV systems.

Future Directions

LPUV systems will see future use for workplace air monitoring for certain specific classes of compounds, such as aromatics. Users of remote sensing instruments will have to evaluate each situation for performance and cost of both LPUV and IR systems to see which best meets the needs of the specific project.

INFRARED TECHNIQUES

Monitoring techniques that rely on infrared (IR) light have many advantages over other methods. Measurements can be made in a passive mode using the sun or hot body radiation as an IR source, and there is little chance of eye injury when using an IR light source in an active mode. Most importantly, almost all gases and vapors can be measured by infrared techniques, with the exception of homonuclear diatomic molecules or monatomic gases (nitrogen, oxygen, and argon).

Unfortunately, atmospheric water vapor and carbon dioxide absorb over broad IR regions making it difficult to observe some compounds. In gas cell measurements, the effect of water and carbon dioxide are minimized or eliminated by introducing contaminant-free air into the gas cell as a background. Nevertheless, there are windows of the IR spectrum that have little or no water and carbon dioxide absorption. It is possible to identify and measure concentrations of compounds using these windows even if they are not the major absorption bands of the analyte.

Absorption is the most common method of detection in IR spectroscopy, although emission characteristics can also be measured (Figure 1). The IR absorption of several gases and vapors is shown in Figure 3. Each compound has a unique signature which can be identified. The aggregate absorption due to all the compounds is shown in the composite at the bottom of Figure 3. Spectral windows for IR detection are generally limited to the 720-1300, 2000-2224, and 2390-3100

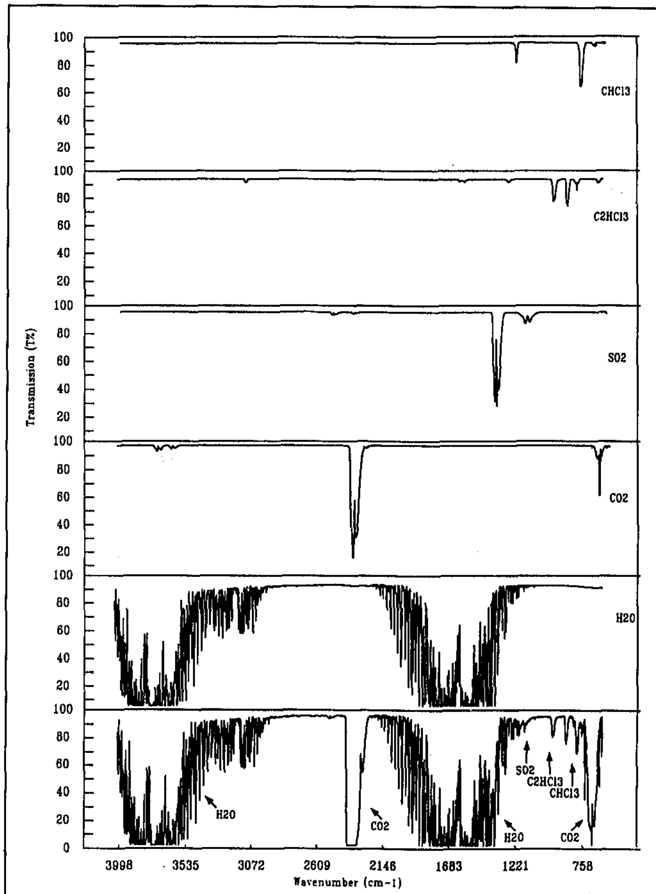

FIGURE 3. Infrared absorption by gases and organic vapors, individually and in a mixture in ambient air.

wavenumber (cm-1) regions because of the previously mentioned water and CO_2 interference. For indoor monitoring with shorter path lengths windows will be wider. Most environmental and occupational pollutants have been identified with this technique.(59)

Gas Filter Correlation Technique

In gas filter correlation techniques, an optical filter is used to limit the spectral regions used in measurements. In a typical system the IR beam emitted from a source is transmitted through a sample, which can be in a gas cell or an open path. A beamsplitter is used to divide the beam into two paths, each going through a correlation cell and a reference cell to reach detectors. The correlation cell contains the gas of interest and the reference cell contains a spectrally inactive gas such as nitrogen. An optical chopper is run at a certain frequency to provide modulation.

Quantitative information is obtained based on a comparison of the signal from the correlation cell with that from the reference cell. The sensitivity and selectivity of the method depends on how strong the absorption peak chosen is, and how free from interference.[60] Cross-stack measurement of pollutants was an early use of gas cell correlation methods.[61]

This technique also has been used to measure tailpipe CO emissions from vehicles. The device consists of an IR light source, a gas filter radiometer, and two detectors. A germanium beamsplitter was utilized to divide the sampling beam into two bandpass filters used to isolate CO and CO_2 spectral regions.[62] Studies have evaluated the accuracy of the remote sensing system, which is also called fuel efficiency automobile test (FEAT), for measurement of CO concentrations from vehicles that were driven on the road,[63] and emissions directly from in-use vehicles.[64] Inter-comparisons also have been made between FEAT measurements and laboratory emission measurements.[65] The aim of these studies has been to improve vehicle fuel economy. Carbon monoxide was monitored as an indicator of incomplete fuel combustion.

General Motors (GM) conducted a similar study to help understand the impact of vehicle emissions on air quality.[66] The continued failure of urban areas to meet clean air standards due to CO emissions has prompted the investigation of more effective control strategies. GM collaborated with the FEAT group at the University of Denver to evaluate a remote CO emission monitor (filter band pass remote sensing device). The main difference between the FEAT instrument previously described and the GM instrument was the use of a spinning gas filter correlation cell in the FEAT. GM data demonstrated a clear trend of higher emission rates from older model vehicles.

Methane leaks along gas pipelines have been measured using a novel remote sensor based on gas filter correlation.[67] The gas pipeline leak sensor, GASPILS, was laboratory and field tested for sensitivity and false alarm characteristics. A second gas channel was used to discriminate interferences caused by temperature fluxes due to seasonal changes, and to differentiate from natural background methane.

Future Directions

The gas filter correlation method may see very widespread use as a real-time enforcement measurement method for tailpipe emissions from cars on the road. In the workplace the applicability of the method to detect methane leaks around pipelines and related facilities will come into competition from the near-IR method discussed in the "Multiple Beam Systems" section of this review. It also may find some use in monitoring emissions from fork lift trucks in the workplace.

Fourier Transform Infrared Spectrometry

The discovery of the fast Fourier transform algorithm and the advances of computer technology have pushed FTIR spectroscopy to the forefront of monitoring technology. Griffiths and de Haseth and others have published texts that comprehensively introduce and discuss the theory, instrumentation, and applications of FTIR spectrometry.[68,69] Russwurm and Childers have written a comprehensive guidance document for open-path FTIR monitoring.[7] The authors discuss the optical system and issues pertaining to data analysis, obtaining backgrounds, selecting path length, and quality control. A glossary of terms and a bibliography are included.

The major advantage of FTIR spectrometers over dispersive IR instruments is the use of an interferometer to achieve high-resolution spectral data. FTIR spectrometers are faster, have greater sensitivity, and greater throughput than dispersive IR spectrometers. In theory an FTIR spectrometer needs only one moving part, the moving mirror in the interferometer, while dispersive IR instruments have several moving parts including filters, slits, and a beam chopper.

FTIR spectrometers are capable of monitoring more than one compound at once. Advantages and disadvantages of FTIR versus filter infrared (FIR) for industrial hygiene applications were summarized by Levine, et al.[70] Ying has evaluated the applicability of FTIR for quantitation of airborne solvent vapors.[71] Detector performance, including the sensitivity and linearity of detectors, are discussed in a paper by Roush.[72]

Measurement Using a Long-Path Gas Cell

The earliest applications of IR spectroscopy for the identification and quantitation of pollutants in air were made using a long-path gas cell into which an air sample was drawn. Initial studies by Stephens et al. measured atmospheric chemistry related to smog pollution. These studies employed a prism spectrometer (equipped with thermocouple detectors) interfaced to a White-type multiple reflection cell operated at pathlengths between 40 and 400 m. Ozone formation in photochemical smog and the discovery of peroxyacetyl nitrate (PAN) were made using long-path infrared techniques.[73,74]

Other spectroscopic applications included studies of auto exhaust,[75] reactivity of hydrocarbons,[76] and composition of polluted urban air.[77] Some later studies, employing grating monochromators and high-performance cooled detectors, dealt with calibration techniques for ozone.[78]

Hanst, et al.[79] developed multiple reflection systems with greater path lengths and replaced grating systems with interferometer-based systems that had the ability to do fast Fourier transforms. An FTIR long-path system with an eight-mirror multiple reflection cell operating over 2 km distances has been used to study trace pollutants in ambient air and synthetic atmospheres.[80] A four-year study utilizing a long-path FTIR spectrometer was initiated to study noncriteria pollutants.[81, 82] Results furnished a database for testing of chemical kinetic models, reported detection of trace levels of formaldehyde and nitric acid, and detected levels of ammonia at ppb levels.

Herget and Levine proposed the use of an FTIR spectrometer with a 20 m gas cell as a near-real time monitoring method for semiconductor process gas emissions in 1986.[83] Compounds measured were arsine, diborane, phosphine, silane, dichlorosilane, silicon tetrafluoride, boron trifluoride, and hydrogen cyanide. This paper provided experimental details that allowed the user of an FTIR to choose optimum optical absorption peaks for the monitoring of semiconductor manufacturing gases in the workplace in the presence of potential interferences. Some research also has been carried out using an FTIR for measurement of mobile source emissions in real time.[84]

The long-path gas cell FTIR has been applied successfully to the analysis of various chemicals in the workplace.[85–87] Computer simulated spectra of chemical mixtures used in paints have been generated to test the accuracy of FTIR quantitative analysis.[88] FTIR limits of detection were expected to be high when the spectra of mixtures were analyzed in comparison to single compounds. However, studies demonstrated that the sensitivity of the FTIR method was sufficient to detect most compounds well below current federal standards.[88,89]

FTIR Remote Sensing Systems

Remote sensing FTIR has evolved logically from the gas-cell FTIR. In remote systems the gas cell is replaced by telescopes that are used to collimate, send, and receive IR light. One telescope is used to collimate the IR light from the interferometer to send the beam across space. Another receives the light at the detector. The IR source may be at one end of the optical path and the detector at the other, or the source and detector may be together at one end, with a mirror or multiple mirrors that relay the beam to the detector. Optical remote sensing FTIR may operate in active or passive mode (Figure 1).

As discussed earlier, the main drawback to remote sensing FTIR currently is obtaining a clean background spectrum. Various methods to overcome this problem have been introduced, including promising results from a method in which the sample spectrum is used without a background spectrum.[90] A summary of the advantages and disadvantages of various methods can be found in the Russwurm and Childers guidance document.[7] Other issues involving the effects of beam blocking,[91] particulates in the air, and weather conditions need to be addressed more fully.

As more field work with open-path FTIR systems is done, issues of stability over time and ruggedness of instrumentation in hostile environments will be addressed more comprehensively. A study performed at an aluminum smelting facility has evaluated the performance of an open-path FTIR in one type of hostile environment.[92] Real-time fluoride measurements were taken during the baking-in of a smelting pot. The instrument was located away from the strong magnetic fields that might have disrupted instrument operation. One of the motivations of the field study was to determine the effect of dense particulates on instrument operation. However, results concerning this were not discussed.

Active methods—A remote optical sensing of emission (ROSE) FTIR system was developed in 1972 by the Environmental Protection Agency (EPA) to measure environmental pollutants over km distances. Initial experiments indicated that although the measurement concepts were valid, the system was limited in sensitivity due to lack of sufficient spectral resolution (max. 1 cm^{-1}) and energy throughput. The original grating system was replaced by 1977 with an FTIR system. This increased maximum resolution of the ROSE system to 0.06 cm^{-1}. It provided an increased sensitivity of about two orders of magnitude. The entire system was housed in a mobile bookmobile van and could be operated in both active and passive modes.[93–95]

Calibration was done with an enclosed gas cell.[93] The ROSE-FTIR was used to measure contaminants at hazardous plant sites and lagoons, and measured emissions from stacks.[94,95] Levine, et al. applied this method at a hazardous waste site, but with limited success due to difficulties in access to site emission sources, aiming the beam and evaluating the data.[70]

Gosz et al. have applied long-path FTIR spectroscopy to concentration measurements of multiple gases (carbon monoxide, nitrogen dioxide, ozone, and sulfur dioxide) simultaneously over spatial scales of up to 1 km. Long-path beam information with micrometeorology data was used to assess flux (mass per unit area per unit time) between aquatic or terrestrial ecosystems and the atmosphere.[96]

An attempt also has been made to compare data from a remote sensing FTIR with data obtained simultaneously with canisters. Results have indicated that remote sensing technology can provide reliable air pollution information.[97–99] Open-path FTIR has been applied to air pollution monitoring of trace gases in ambient air. Concentrations ranging in ppb were measured for ethylene and ozone over 24-hour time periods to gain an understanding of atmospheric processes.[100]

Remote optical sensing of volatile organic compounds (VOCs) for Superfund activities is discussed by Minnich, Scotto, and Pritchett.[101] Wastewater treatment facilities have been monitored for VOC emissions, and using long-path FTIR spectroscopy batch dumping was observed at two treatment facilities.[102] An open-path FTIR system also has been used to monitor ammonia, methanol, methylene chloride, and sulfur hexafluoride at a chemical wastewater treatment site.[103] In one demonstration test a remote sensing FTIR system has been applied to fence-line monitoring at a petrochemical plant in Houston, Texas.[5]

A transportable open-path FTIR system has been developed to quantitatively monitor gases and vapors in the workplace.[104] Studies have shown that results obtained with this prototype instrument are well correlated to multiple point samplers set up along the beam path.[105] Testing was performed using a straight-line beam path in a corridor, a multiple-pass beam path in front of a filter press, and a beam path with several mirrors in the vicinity of a reactor complex. This system was tested in the workplace to monitor numerous organic vapors and to obtain TWA concentrations for the evaluation of process emissions and worker exposure.

Passive methods—The first remote sensing FTIR built with a commercial Fourier transform interferometer system employed telescopic optics to make long-path absorption and single-ended hot stack gas IR emission measurements of gaseous pollutant concentrations ranging from 10 ppm to 1 ppb over a 1-km path length.[94] The previously mentioned EPA ROSE system was developed and used to measure both the emissions of hot stack gas plumes in a passive mode and emissions from lagoons and processes in an active mode.[95] Persky et al. reported that IR methods were applicable to combustion efficiency monitoring and, in many instances, indicated substantial advantages over currently available monitoring methods.[106]

One of the more practical extensions of laboratory studies on IR emission spectra of gases is the determination of temperature, composition, and concentrations of chemical species in plumes of gas stacks using IR emission measurements at a distance. Studies have indicated that errors in emission measurements can be much larger than those in absorption measurements, due to uncertainties in temperature and concentration gradients from the source.[95]

Many other studies have utilized passive remote sensing FTIR to measure effluents from stacks. One research group has detected emissions from small building smoke stacks.[107] They developed a method that distinguished natural gas from fuel oil as the fuel being burned in a building's furnace. The method was based on the H_2O/CO_2 concentration ratio in exhaust gases determined by observing several prominent H_2O and CO_2 transitions (peaks). In another application a double-beam interferometer system with background suppression was used to analyze stack effluents. Evaluation of the instrument for measurement of atmospheric pollutants was carried out both in the laboratory and in the field.[108]

A test of a passive FTIR spectrometer for measurement of gaseous pollutants on the ground was carried out on with a helicopter.[109,110] In this study the issue of background measurements was addressed. A method was proposed to eliminate the problem of irreproducible backgrounds by directly analyzing the FTIR instrument signal (the interferogram) prior to conversion to a spectrum. The principal application of this instrument was for use on the "chemical warfare" battlefield to detect organophosphate nerve gasses and sulfur mustard gas.

A unique IR system, a gas imaging IR camera, has been developed to remotely visualize and detect toxic and flammable gas leaks. The system enables one to actually "see" gas clouds in real-time by observing the IR radiation emitted by the gas.[111] Here the emitted IR radiation is used as the signal, not as a source. The BAGI system, discussed above in the laser section, used IR radiation as a source to image gas releases.

Future Directions

The FTIR-based methods are expected to be used increasingly in the future. However, significant advances in software, and continued price decreases, will be necessary. The first operating systems are already being designed for use in large chemical complexes in which the real-time output of the FTIR will be tied into an alarm system through a multi-point source dispersion modeling and real-time computerized meteorology system.

MULTIPLE BEAM SYSTEMS

For almost all of the applications described in this review article, the investigators envision improved performance from their systems when multiple beams are used. "Multiple beams" may mean either multiple beams used at the same time, or a single beam rapidly moved from location to location. Multiple beams increase spatial and temporal beam density and thus reduce the chance of undetected dangerous concentrations of hazardous and/or explosive gasses. Two applications of open-path methods that have actually utilized multiple beams are given below.

Near-IR Flammable Gas Monitor

A system that uses near-IR light has been built to detect concentrations approaching the lower explosive limit (LEL).[112] The system is designed to replace the traditional catalytic LEL sensor, especially in off-shore oil and gas platforms where the catalytic sensor frequently fails due to fouling by salt spray. This system operates in the near-IR spectral region, utilizing simple glass optics, which allows a multiple beam system to be built at a very low price. Sensitivity is more than adequate for LEL applications, but would be inadequate for monitoring of toxic gases.

Multiple beams are used to "fill" a space with beams, thus reducing unmonitored areas and increasing confidence in measurements. Also, when the system fails due to, for example, the failure of the light source, an alarm is automatically sounded. With the catalytic LEL system, this may not be the case, and thus LEL monitor failures might go undetected.

Limitations of the flammable gas detector under various conditions have been evaluated, and maximum path lengths for operation in all weather conditions have been determined.[113] In heavy rain the transmittance falls, but most instruments have a dynamic reserve to compensate for the loss. When wind speed reaches 37 ms^{-1}, attenuation by dust or sand becomes severe and path lengths must be restricted to 100 m or less. Maritime aerosols appear to have little effect on transmittance. Fog can reduce atmospheric transmittance and may also scatter radiation. Humidity and variations in temperature were found to have little effect on transmittance. However, changes in temperature could effect the sensitivity of the detector.

Future Directions

It is reasonable to predict that the near-IR flammable gas monitor will replace the traditional catalytic LEL sensor for many applications. This is already occurring in Europe, especially in off-shore oil platforms.

Application of Tomography in Air Monitoring

The technique of reconstructive tomography (RT), also known as computed tomography (CT), has been discussed in detail in a review by Brooks and Chiro as it applies to medical imaging.[114] There have been many attempts to employ tomographic techniques outside of medical applications. For example, tomography has been employed in laboratory gas concentration measurements and remote air pollution monitoring.[115,116]

Problems encountered in laser absorption computed tomography over kilometer ranges, performed in order to map pollution concentrations, have been addressed.[117] Construction of a system that can generate pollution maps with ranges of 6 km and with detection sensitivities of fractions of ppm to ppb is said to be within the current state of the art. A fan-beam configuration has been used already in optical tomography for mapping iodine vapor density in a plane.[118]

The theory and concepts of tomography have been discussed for use in industrial hygiene.[119,120] A computer using tomographic techniques may be able to reconstruct the chemical concentration distribution in a room and produce a map of this distribution. Successful work comparing FTIR use in CT in comparison to point samples has been accomplished on the scale of a full-sized room.[121] The most recent work by the current authors has evaluated the ability, on the scale of a full-sized room, to acquire spectra from 55 beams in less than four minutes for calculating time-resolved concentration maps using the tomographic technique. Although remote sensing FTIR provides a minimally obtrusive way to monitor air contaminant concentrations across space, many technical problems must be resolved before the system can be used with tomographic techniques to qualitatively and quantitatively assess exposure, ventilation, and room air mixing.

Future Directions

The potential of multiple-beam FTIR and LIDAR-based systems to map the concentration profiles of multiple components under varying conditions through entire work areas will be an incentive to continue development of this method. If sufficient time and resources are available, tomographic methods may become a powerful tool for both ventilation engineering and health hazard evaluation studies. This method may not come into routine use during this decade.

CONCLUSIONS

In this review optical remote sensing techniques have been discussed that are currently available or are being developed. The methods most relevant to industrial hygiene have been discussed in some detail in order to supply the industrial hygienist with a guide to remote monitoring technologies.

Some techniques, such as DIAL, are more applicable to environmental monitoring. However, the industrial hygienist should at least be aware of these methods since hygienists are increasingly responsible for environmental monitoring and issues involving SARA III and CAAA.

Open-path FTIR spectroscopy appears to be the most promising technique for monitoring of toxic air contaminants in the workplace. Open-path FTIR systems are being tested as fenceline monitors and are being developed for leak or fugitive emission detection. The ability of open-path FTIR instruments to monitor multiple compounds in real time appears to make it an ideal industrial air monitoring instrument.

However, it is important to remember that widespread acceptance of open-path instruments still depends on advances in hardware that may make these instruments more portable and inexpensive and on advances in software that will make optical remote sensing instruments more useable by industrial hygienists without extensive spectroscopic knowledge.

Issues of how to optimize the use of a beam path average measurement in comparison to traditional point sampling techniques, and how to obtain backgrounds in industrial environments need to be addressed if optical remote sensing instruments are to become practical air monitoring instruments.

Furthermore, rigorous laboratory and field evaluation tests must be performed and published in the peer review literature. For each new open-path instrument, these studies must answer the questions of the relationship of open-path concentration data to point sampling and breathing zone concentration data. In addition, questions must be answered surrounding the meteorological aspects of the performance of these new open-path instruments.[122]

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