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A review of remote sensing techniques and related spectroscopy problems

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Abstract

Remote sensing based on quantitative spectroscopy is a powerful tool for precise measurements of atmospheric trace species concentrations, through the use of characteristic spectral signatures of the different molecular species and their associated vibration–rotation and electronic bands in the microwave, infrared, and UV-visible domains. A reliable retrieval of the concentration profiles requires a good characterisation of measurement and spectral fitting errors. This includes an accurate knowledge of spectroscopic parameters of all transition lines or absorption cross sections of interest since uncertainties lead to systematic retrieval errors. ***To cite this article: S. Payan et al., C. R. Physique 6 (2005).***

Résumé

Une revue des techniques de mesure à distance et des problèmes de spectroscopie associés. Les mesures à distances qui s'appuient sur la spectroscopie quantitative sont un outil puissant pour la mesure précise des concentrations d'espèces à l'état de trace dans l'atmosphère par l'utilisation des signatures spectrales caractéristiques des différentes espèces moléculaires, qui sont associées aux bandes de vibration–rotation et aux bandes électroniques dans les domaine micro-onde, infrarouge, et ultraviolet ou visible. Une inversion fiable du profil de concentration nécessite une bonne caractérisation des erreurs de mesure et d'ajustement des spectres. Ceci implique une connaissance précise des paramètres spectroscopiques de toutes les raies ou des sections efficaces d'absorption dont les incertitudes ont un impact direct sur les erreurs résultant de l'inversion des spectres observés. ***Pour citer cet article : S. Payan et al., C. R. Physique 6 (2005).***

Keywords: Atmosphere; Remote sensing; Spectroscopy

Mots-clés : Atmosphère ; Télédétection ; Mesures à distance ; Spectroscopie

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1. Introduction

In the broadest sense, *remote sensing* is the measurement or acquisition of information of some characteristics of an object or a phenomenon, by a recording device that is not in physical or intimate contact with the object or phenomenon under study.

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For instance, it can be the long-range utilization, from the ground, a balloon, an aircraft, a spacecraft, or a ship, of a device for gathering information pertinent to the environment, such as measurements of force fields, electromagnetic radiation, or acoustic energy. By extension, the term *remote sensing* refers to any technique making measurements on large scale systems without the use of in situ instruments. This review will cover remote sensing techniques used to observe the Earth atmosphere and will be focused on measurements using wavelengths ranging from microwave to ultraviolet regions such as gas measurements for meteorology, ozone layer, climate and pollution related studies. Reviews of remote sensing techniques for ocean, archaeology, urban and suburban land and vegetation can be found in [1].

Remote sensing involves the collection of information, carried by electromagnetic radiation, about the Earth surface or about the atmosphere. A broad classification of remote sensing systems distinguishes passive systems that detect a naturally emitted radiation, and active systems that emit a specific radiation and analyse the corresponding backscattered signal. Passive systems can be further subdivided into those detecting radiation emitted by the Sun, the Moon or a star (or any source) and those detecting thermal radiation emitted by the Earth surface or the atmosphere. For sources at typical terrestrial temperatures, the thermal emission occurs mostly in the infrared range of the spectrum, at wavelengths around 10 µm (the so-called thermal infrared region), although measurable quantities of radiation are also detectable at longer wavelengths, as in the microwave domain of the spectrum.

The sensor, either of a passive or an active instrument, detects electromagnetic radiation after interaction with the atmospheric components. Three variables describe the radiation that is received: its intensity, its wavelength and its time dependence.

In most cases, the information available is the intensity and the wavelength. For thermal emission, the intensity (or radiance) is determined by the temperature and the emissivity of the background, and by the atmospheric species along the path to the instrument. In the cases of active systems (measuring retro-reflected radiation), the received signal is determined by the amount of radiation exciting the atmosphere, and by the corresponding backscattering properties. Thus, one realises that the information on the atmosphere that is directly observable from remote sensing observations is actually rather limited: we can measure its range-resolved backscattering properties (active methods), and a combination of temperature and emissivity. However, these parameters can be measured at different locations and times, over a large range of wavelengths and, sometimes, at different polarisation states. This diversity of available observables is responsible for the large range of indirectly derived atmospheric parameters.

Once the sensor has collected data, they must be analysed and geophysical quantities retrieved. The major goal of data processing is the extraction of useful information from the measurements, based on the radiance values (probably in a number of spectral bands, at a number of different dates, in different polarisation states, etc.).

The atmospheric transmission spectrum is of fundamental importance for the design of remote sensing instruments. In general, an instrument designed to look at the Earth surface, clouds, or aerosols, will operate in an atmospheric window (visible and near infrared, around 10 µm in the thermal infrared or in the microwave window), whereas a device designed to measure atmospheric temperature or composition will work at wavelengths that are absorbed by the atmosphere. General techniques for atmospheric remote sensing are described in [2].

2. Microwave remote sensing of the atmosphere

2.1. Introduction

Microwave remote sensing techniques are now playing an important role in probing the terrestrial atmosphere either from the ground or from space for acquiring a better knowledge of atmospheric chemistry and physics as well as for understanding changes caused by anthropogenic activities. In addition, they have specific advantages compared to other wavelengths ranges such as smaller sensitivity to cloud contamination, no need for external source, almost linear dependence of thermal emission on temperature as opposed to a non-linear relationship in other spectral regions. An accurate retrieval of the quantities of interest requires an accurate knowledge of the spectroscopic parameters, such as line strength, line position, pressure broadening parameters and pressure shift. An uncertainty in the spectroscopic parameters will lead to a systematic retrieval error. This is why a careful investigation of the current accuracy of the spectroscopic parameters and their impact on the retrieval is absolutely necessary.

2.2. Microwave remote sensing measurements

There exists a large number of ground-based microwave radiometers all over the world belonging to the Network for the Detection of Stratospheric Change (NDSC: http://www.ndsc.ws), an international network to provide a consistent standardised set of long-term measurements of atmospheric trace gases, particles and physical parameters through a set of globally distributed sites [3]. Such microwave radiometers are generally operating at frequencies between 20 and 300 GHz to measure essentially stratospheric water vapour, ozone, and chlorine monoxide.

Examples of airborne measurements are the German Airborne SUb-millimeter Radiometer (ASUR) (either aboard the NASA DC3 or the DLR Falcon), the Swiss Airborne Measurements Of Stratospheric water vapour (AMSOS) [4] aboard the Swiss Air Force Learjet T-781 and the ESA/MARSCHALS experiment [5] aboard Geophysica. Flights of such instruments allow the use of frequencies higher than 300 GHz for stratospheric platforms, since absorption by H_2O (lines and continuum) is strongly reduced above the tropopause. Balloon measurements are also performed as the Balloon OH (BOH) of JPL with a 2.5 THz limb sounder.

For about 15 years, the development of satellite-borne radiometers using limb sounding and technologies allowing increased sensitivity, accuracy and precision, favoured new generations of instruments operating at higher and higher frequencies up to several THz, from the millimeter-wave to the sub-millimeter-wave range. The Upper Atmosphere Research Satellite (UARS) launched in 1991 was the first satellite with a Microwave Limb Sounder (MLS) in the millimeter-wave region. Then the Odin satellite, a Sweden–France–Finland–Canada collaboration, was launched in February 2001 with a Sub-Millimeter Radiometer (SMR) operating at higher frequencies in the range 480–580 GHz. It was followed in July 2004 by the new Earth Observing System EOS/MLS instrument aboard the NASA spacecraft Aura with frequencies in the range 190–640 GHz and a receiver at 2.5 THz. Eventually, the Sub-Millimeter Limb Emission Sounder (SMILES) will be placed in the Japanese Experiment Module (JEM) aboard the International Space Station in 2008, with a sensitivity increased by the use of a SIS junction cooled down to 4 K in the frequency range 620–660 GHz.

Consequently, at sub-millimeter wavelengths, a much larger number of molecules can be observed with increasing line intensity and thus sensitivity, such as H_2O and isotopes HDO, $H_2^{17}O$ and $H_2^{18}O$, O_3 and isotopes, H_2O_2, N_2O, HNO_3, HCN, CO, H_2CO, ClO, HOCl, BrO, HOBr, HO_2, SO_2 and some others.

2.3. Spectroscopic parameters

The spectroscopic line parameters necessary to interpret the observations are essentially line intensity, line position, air and self-broadening parameters and their temperature dependence, and pressure shift. Some theoretical considerations can be found in [6]. Data provided by laboratory measurements are crucial to the determination of concentration profiles through limb sounding measurements. Temperature and pressure variations of the line widths are compounded with the actual temperature and pressure variations within the stratosphere and the upper troposphere. In order to minimize errors associated with the inversion of atmospheric limb measurements, laboratory measurements of pressure broadening are done throughout the temperature range of atmospheric conditions, i.e., 190–300 K.

Line parameters can be found in different catalogues. The JPL Sub-millimeter, Millimeter, and Microwave Spectral Line Catalogue [7] provides data for more than 340 species (http://spec.jpl.nasa.gov) of atmospheric and astronomical interest. Concerning rotational frequencies in the sub-millimeter domain, the JPL database is generally recommended. It was found quite reliable except for HNO_3 for which a new calculation was based on the most recent spectroscopic analyses [8]. The Cologne database for Molecular Spectroscopy [9] covering more than 120 species (http://www.ph1.uni-koeln.de/vorhersagen/) and the NIST Recommended Rest Frequencies for Observed Interstellar Molecular Microwave Transitions [10] (http://physics.nist.gov/PhysRefData/micro/html/contents.html) are dedicated to molecules of astrophysical interest. Finally, the Smithsonian Astrophysical Observatory Database SAO92 [11] contains data for about 35 astrophysical and atmospheric molecules.

The increasing number of radiometers operating at sub-millimeter wavelengths creates new needs for precise molecular parameters. Some of them have been measured by different teams, but they can present discrepancies in their values and accuracies as quoted by [12], providing a critical review of spectroscopic parameters. In the ro-vibrational spectral region, multiple measurements are often available, thus necessitating a critical evaluation and subsequent recommendations for the best values. In contrast, for the rotational bands very few re-measurements of parameters are available and careful evaluation is recommended prior to introduction in the database.

2.4. Recent measurements

Frequency and pressure/temperature dependent line width measurements have been the recent focus of JPL [13], as new frequency measurements have been made for the species $HClO_4$, $H^{15}NO_3$, CH_3CN, $(CH_3)_2CO$, $(CH_3)CO(CH_2OH)$, HO_2NO_2 and HO_2NO. In addition, new pressure/temperature dependent line width measurements have also been made for O_3, O_2, HCl, HO_2, BrO, SO_2, HO_2 and OH.

In the framework of the ESA study of the ACECHEM mission (Atmospheric Composition Explorer for Chemistry and Climate Interactions), laboratory measurements linked to the ESA/MASTER instrument have been carried out in the 294–626 GHz spectral region in which O_3, H_2O, CO, N_2O, HNO_3, ClO, BrO and HCl are observable species [6,14].

A strong effort has been made to determine accurate pressure broadening coefficients and their temperature dependence for molecules to be monitored with JEM/SMILES as BrO at 624.77 and 650.18 GHz, O_3 at 544.86 and 625.37 GHz, and the O_3 isotope $^{16}O^{18}O^{16}O$ at 647.69 GHz. Broadening coefficients are consistent between various groups while temperature exponents are not quite as accurate because the measured temperature is often not indicating the true gas temperature.

A millimeter-wave spectrometer is devoted to laboratory measurements in the range 50–630 GHz [15] providing N_2-, O_2-, and air-broadening coefficients for three O_3 lines at 301.8, 317.2 and 320.0 GHz and for five HNO_3 lines. Another low-noise heterodyne spectrometer was also used at JAXA/CRL [16] to study the non-linear behaviour of the line shape in, for instance, $H^{37}Cl$ and $H^{35}Cl$. Finally, absorption profiles have been recorded with the AIST THz spectrometer [17] to provide accurate spectroscopic parameters for pure rotational transitions of N_2O at 628 and 653 GHz, $H^{35}Cl$ and $H^{37}Cl$ at 625 and 626 GHz and SO_2 at 702.6 GHz.

2.5. Error analysis

Some discrepancies have been found between intensity values quoted in the HITRAN and JPL databases. New intensity calculations have been performed in [8] for different molecules based on the recalculation of the total partition function. For example N_2O values quoted in the JPL database are overestimated by 13%, this difference being due to the neglect of the vibrational part of the partition function.

Comparisons of measurements made in parallel in Lille and in Bologna for O_3 (two lines) and HNO_3 (one line) revealed that collisional broadening parameters are consistent within 2%. Such comparisons indicate that systematic errors are about 2%. They are generally greater than the statistical errors. Therefore the estimated accuracy of the temperature exponent is of the order of 10–15%.

A study made in [18] has been made in three cases: first, errors of 5% were assumed for the line broadening parameters of target and interfering lines; second, a 10% uncertainty was assumed for the temperature dependence exponent and third, 30% errors were assumed for the line broadening parameters in order to simulate a worst case scenario. Results of this study confirm that errors in the foreign-broadening parameters are more critical than uncertainties of the temperature dependence exponent. Foreign-broadening parameters should be measured with an accuracy of 5% or better in order to avoid that systematic spectroscopic errors dominates the statistical retrieval error. It has been also found in [6] that an uncertainty in the intensity of the strong lines has a direct impact on the retrieval error. For instance, a 10% uncertainty in the intensity of the strongest N_2O line at 301 GHz generates a 10% error on the retrieved N_2O concentration. The same applies for O_3 and H_2O retrievals with a 2% and 5% uncertainty, respectively. Uncertainties in the central frequency have a negligible impact on the retrieval. However air-broadening parameters and their temperature exponents dominate the error budget, mainly for the strongest lines. However, such parameters of strong lines are found to have a much higher impact on the retrieval of weak lines in the same frequency band. Self-broadening parameters are found to have a small impact on the retrievals even in the case of species with high self-broadening coefficients and high volume mixing ratios, whereas pressure shifts have a rather limited impact on the retrievals, except in the case of HNO_3 lines.

2.6. Conclusions

The millimeter and sub-millimeter wavelength regions cover a large number of transitions of numerous species of major importance for the ozone chemistry of the middle atmosphere and for the greenhouse effect of the upper troposphere/lower stratosphere (UT/LS). Accurate retrievals of the concentration profiles require a good characterisation of measurement and retrieval errors. This includes a precise knowledge of spectroscopic parameters of all lines of interest since uncertainties lead to systematic retrieval error. Intense efforts are made by different groups to develop new instrumentation in order to produce improved laboratory measurements as well as performing critical evaluation of the results before introducing new parameters in the spectroscopic databases. Such studies are quite important for improving concentration profiles retrieved from observations carried out by ground-based, balloon-borne, airborne and satellite experiments. In addition, the results will contribute to select the most adequate lines to be used for species of interest in order to insure the protection of the corresponding frequencies, an item that will be discussed and decided in the next World Radio Conferences to be held in 2007–2008 and 2010.

3. Infrared remote sensing

Infrared spectroscopy is a powerful tool for precise measurements of atmospheric trace species concentrations thanks to the use of characteristic spectral signatures of the different molecular species and their associated vibration–rotation bands in the far-, mid- or near-infrared [19]. A large number of methods based on quantitative spectroscopy permit tropospheric, stratospheric or mesospheric measurements with instruments on the ground, airborne, balloon-borne or satellite-borne.

As mentioned previously, instruments may be classified into two types: active ones that contain both a transmitter and a receiver, and passive ones which rely on a natural source of radiation (Sun, Moon, star...) or on thermally emitted atmospheric radiation. A further distinction can be made between:

(i) upwards viewing instruments which look (at the Sun mainly) from the ground [20] or balloon-borne [21];
(ii) nadir viewing instruments which look straight down or scan around this direction from aircraft, balloon or spacecraft [22]; and
(iii) limb viewing instruments which look toward the limb or the edge of the Earth atmosphere from aircraft, balloon or spacecraft [23].

In the nadir direction, the atmosphere is seen against the background of the Earth surface or cloud, while looking toward the limb or upwards, the background is the cold space or the Sun, depending on the experiment.

3.1. Ground measurements

Some remote sensing measurements are performed from the ground. The instruments are generally Fourier transform spectrometers in the infrared (FTIR) or in some cases heterodyne spectrometers. Spectrometers aim at a natural source (generally the Sun, but also the Moon or selected stars) when they are operating in absorption (they can also collect the radiation emitted by the atmosphere in the thermal infrared or radiation backscattered in the visible-UV domain). Such measurements are generally performed at observatories or dedicated stations of the Network for Detection of Stratospheric Change (NDSC) [3].

The main advantages of such measurements are the wide spectral regions covered (allowing simultaneous measurements of a large number of species), the high signal to noise ratio (achieved by using the Sun as source), and the possibility to obtain long time series of measurements to establish mid- and long-term trends of stratospheric or tropospheric species [20].

Ground-based observations are hindered by two main factors: the vertical resolution is rather low (even if a vertical profile can be retrieved with high spectral resolution instruments resolving the line profiles, like for O_3, CH_4, HCl, ...) and for stratospheric species measurements, the useful radiation must go through the lower atmosphere limiting measurements in the most transparent windows.

3.2. Aircraft and balloon-based observations

Airborne platforms carrying high resolution spectrometers benefit from a better viewing geometry for remote sensing measurements than from the ground. Let us consider the case of solar occultation from a balloon [21]; two factors enhance the possibility to derive useful information on the vertical distribution of target species:

(i) the variation of the balloon altitude during the ascent, which creates a variation of the observed column along the line of sight; and
(ii) the variation of the tangent height during the solar occultation at sunset or sunrise (or of a star in the case of a stellar occultation).

There exist several powerful FTIR experiments using the solar occultation technique like the Limb Profile Monitor of the Atmosphere (LPMA) [21], and the MkIV experiment onboard both balloon and aircraft [24].

A constraint of the solar occultation technique for atmospheric measurements is the local time of the observation, always around sunset or sunrise. Using the limb thermal emission technique is relaxing this experimental constraint. This is the case of the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) experiment, either when balloon-borne (MIPAS-B [23]) or airborne (MIPAS-STR [25]) platforms are used.

However, the methods described previously do not permit global measurements because they are limited in space and time.

3.3. Satellite experiments

Remote sensing from satellites is ideally suited for monitoring the Earth atmosphere [26] in order to better understand atmospheric processes as a whole. A global and continuous observation is possible including inaccessible regions for which very few data are available. As an example, the Upper Atmosphere Research Satellite (UARS) launched in 1991 whose instruments were designed in the 1980s has paved the way for observing the middle atmosphere of the Earth [27] by combining several remote sounders on the same platform.

Limb sounding from space has proved to be a valuable technique for atmospheric composition measurements in the stratosphere and mesosphere. With this technique, one obtains a good vertical resolution while taking advantage of the optical path amplification factor in the vicinity of the tangent height, providing a higher sensitivity to low concentration species. Stratospheric composition measurements, which have contributed to better understand the photochemistry of this region of the atmosphere, are best made by the limb sounding technique. As an example, the Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment is a good illustration of the solar occultation technique. ATMOS has been accommodated onboard the American space shuttle (4 flights between 1985 and 1994). Taking advantage of a high spectral resolution ($\sim 0.010\ \text{cm}^{-1}$), of the coverage of a wide spectral region and of a good signal to noise ratio, vertical profiles of many species have been measured [28]. Vertical profiles of abundant species like CO and CO_2 have been retrieved up to the thermosphere where non-local thermodynamic equilibrium effects become important [29]. The occultation technique has also been used to measure atmospheric aerosol concentrations. The limb geometry is also used in thermal emission with cooled spectrometer like MIPAS onboard the ENVISAT platform [30].

By comparison with limb sounders, nadir sounders benefit of a good horizontal resolution but with a degraded vertical resolution [22]. They followed the imagers used initially for meteorology or Earth observations. They have a better spectral resolution (increasing the vertical resolution for some species). Compared to UV-visible nadir sounders, infrared ones are not limited to diurnal measurements. The new generation of infrared sounders has a better spectral resolution: about $1\ \text{cm}^{-1}$ for the Atmospheric Infrared Sounder (AIRS) and $0.5\ \text{cm}^{-1}$ for the Infrared Atmospheric Sounding Interferometer (IASI) [31]. They provide the tropospheric temperature (using CO_2 as a probe of its environment) and the water vapour profile with a better precision (1 K and 10% respectively) and with a vertical resolution of 1 km. In the case of O_3, infrared sounders should permit to derive information on the vertical distribution by separating at least the tropospheric from the stratospheric contribution. The IASI or AIRS like sounders make only possible the retrieval of a vertical column for tropospheric species like CO, CH_4 and N_2O.

Only instruments having a spectral resolution of the order of $0.1\ \text{cm}^{-1}$ in the thermal infrared are able to obtain a better vertical resolution [32] by using the information contained in the shape of pressure broadened lines. An optimal use of the information contained in the measured spectra is a current priority of the atmospheric remote sensing community.

3.4. *Data analysis*

The analysis of remote sensing atmospheric data is a multi-dimensional problem. It requires being able to accurately simulate atmospheric spectra [33]. The measured signal is the result of the convolution of vertical profiles of geophysical parameters (species concentration, temperature…) with resolution functions that are usually quite broad, often covering several kilometres [34]. The detailed structure is lost and any attempt to recover it only results in spurious structures arising from measurement noise (noise resulting from unknown variability in properties of the scene and noise arising within the instrument). A method for finding a solution to this under-constrained problem is to combine the measurements with the most likely atmospheric profile and its statistical properties (optimal estimation method). Recently, neural networks have been developed to perform real time retrievals [35]. This type of inversion or retrieval problem has been widely studied in geophysics. A recent summary of inverse methods for atmospheric sounding is given by Rodgers [34].

When an instrument covers large spectral regions, information redundancy makes it possible to select a set of narrow spectral intervals containing the most useful information on the target parameters, whereas the intervals containing little or no information can be ignored [36]. The use of selected spectral intervals, called micro-windows, is reducing the number of spectral elements to be considered and is avoiding the analysis of spectral regions that are affected by interfering species or by nonlocal thermal equilibrium [29]. More generally, the priority is given to the analysis of spectral elements containing most of the information on the target species and which are less affected by systematic errors.

3.5. *Spectroscopy problems*

The analysis of atmospheric spectra requires a preliminary knowledge of absorbing and emitting properties of the target molecules. These properties are tabulated as spectroscopic parameters (line positions, intensities, widths, …) included in databases such as HITRAN [37] or GEISA/IASI [38], which are derived from experimental and/or theoretical spectroscopic studies. These data are used in a 'forward model' (or radiative transfer algorithm) that simulates the propagation of the radiation in the atmosphere [39]. The accuracy of the retrieved profiles depends highly on the quality of the spectroscopic parameters and, given the improved capabilities (higher spectral resolution, better signal to noise ratio…) of the new instruments, it is clear that new laboratory studies using the best experimental techniques and/or sophisticated theoretical models are required. However these studies can present many difficulties. The case of the ozone molecule for which simultaneous measurements in two different spectral regions were required to derive reliable intensities is a good example of the current spectroscopic problems pertaining to atmospheric sounding [40]. Let us quote in particular the difficulty of measuring and/or generating accurate line intensities for unstable molecules or of modelling the spectrum of heavy species or accounting for realistic line shapes.

For heavy molecules, the individual lines cannot be resolved either because of the lack of resolution and/or because of the extreme density of their vibration–rotational spectra. Thus, the absorption of these molecules appears rather continuous in the atmospheric spectra, and there are two approaches to model their absorption: either to measure all desired absorption cross sections for the whole range of atmospheric pressures and temperatures or to model and generate a synthetic vibration–rotation spectrum. As an example, the case of the $ClONO_2$ molecule can be mentioned [41].

Atmospheric continua observable in the IR spectral region can be divided into gaseous continua and continuous spectra caused by liquid or solid particles. The physical origin of the N_2 and O_2 continua is collision-induced absorption [42], while the continua of H_2O [43,44], and CO_2 are due to deviations of their line shapes from the commonly assumed Voigt line shape resulting from collisional broadening. Liquid or solid particles in the stratosphere can cause a continuum-like contribution to radiance spectra due to absorption and scattering (the simulation of the absorption coefficient for liquid and solid particles can be performed by a Mie code in the case of spherical particles on the basis of their spectral refractive index and particle size distribution).

The influence of line mixing effects can be significant in the infrared region. This influence has been demonstrated for the absorption or emission of the CO_2 Q branch and models have been developed [45] to be included in radiative transfer algorithms. Indeed, the shape of the CO_2 Q branch (sensitive to pressure and temperature) can have a significant influence in the inversion of atmospheric spectra to infer vertical profiles of temperature, pressure as well as concentrations of molecules that absorb in the same spectral regions as CO_2 [46]. Significant line mixing effects in methane atmospheric absorption have been demonstrated [47] and modelling is currently under study.

The duration of collisions leads to the well-known sub-Lorentzian nature of the far wings of CO_2 lines [48]. Two types of calculations can be performed: taking into account collisional narrowing [49] or modifying the simple Lorentzian wings of the lines by a factor (usually called χ) in order to reproduce their sub-Lorentzian behaviour.

Because atmospheric sounders are now operating at significantly higher resolution than previous instruments, their sensitivity to the spectral line shape has increased. Thus, effects that have been neglected in the past must now be considered such as collisional narrowing and line mixing in methane.

4. UV-visible remote sensing

UV-visible spectroscopy allows measurement of trace species by both passive and active techniques. Ozone is a strong absorber in the UV. It prevents solar light below 300 nm to reach the surface and it is the main absorber in the visible spectrum. It has been used for ozone monitoring since the beginning of the ozone layer study. Several other minor constituents involved in stratospheric ozone chemistry or in air pollution can be observed by UV-visible spectroscopy due to their absorption features in this wavelength domain, among them NO_2, SO_2, OClO, BrO, HCHO. These techniques are used in ground-based, balloon and satellites passive instruments and in active lidar systems.

4.1. Ground-based passive

The Dobson spectrophotometer is a ground-based instrument that measures the amount of ozone present in the atmosphere [50]. Gordon Dobson designed the Dobson spectrophotometer in the 1930s. It measures ultraviolet light from the Sun at 2 to 6 different wavelengths from 305 to 345 nm. By measuring UV light at two different wavelengths, the amount of ozone can be calculated. One of the wavelengths used to measure ozone is strongly absorbed by ozone (305 nm), whereas the other wavelength is not absorbed (325 nm). Therefore the ratio between the two light intensities is a measure of the amount of ozone in the light path from the Sun to the observing spectrophotometer.

The Differential Optical Absorption Spectroscopy (DOAS) UV-visible instruments are broad-band (300–600 nm), medium resolution (0.5 to 1.0 nm) spectrometers [51], looking at the sky spectrum illuminated by the Sun. Spectra are recorded during twilight up to 94° solar zenith angle (SZA) when the contribution of the stratosphere to the absorption is at its maximum. Spectra are analysed by differential absorption spectroscopy relative to a single reference spectrum taken with the same instrument on a clear day at small SZA. Vertical columns of O_3, NO_2, H_2O and O_4 are retrieved at sunrise and sunset when the atmospheric slant path is amplified.

4.2. Satellite

UV-visible remote sensing from space permits to perform a global monitoring of ozone and related species and to detect possible long-term changes. The development of the Antarctic ozone hole is monitored every austral spring since its discovery in 1985 using nadir viewing observation of scattered UV solar light by the series of American TOMS [52] satellites and more recently by the European GOME instrument [53]. Limb viewing and occultation instruments retrieve the vertical profile of constituents in order to better understand the atmospheric chemistry and improve chemistry-transport models.

Nadir viewing space-borne spectrometers provide vertical columns of constituents with a horizontal resolution of a few tens of km. The principle is to observe the UV visible spectrum of the solar light reflected by the atmosphere and the surface. The concentration of absorbers is obtained using the DOAS technique. The DOAS principle is to determine the slant column density of trace gases by measuring their specific narrow band absorption features [54]. Nadir observations were first used for the global stratospheric ozone monitoring and they are now also used for the observation of minor species in the stratosphere (NO_2, OClO, BrO) and for the survey of tropospheric pollution arising from industrial activity and biomass burning (NO_2, SO_2, HCHO). GOME2 onboard ERS2 and SCIAMACHY onboard ENVISAT are good examples of such instruments [55].

Limb viewing instruments observe the spectrum of the solar light scattered by the atmosphere in a narrow vertical field of view. The restitution of the tangent altitude is coming from the knowledge of the satellite attitude and a good accuracy is needed in this parameter (1 arc-min represents about 1 km on the limb). A good description of the effect of multiple scattering is also important for the correct determination of atmospheric air mass factors for the different species. As for nadir observations, the DOAS inversion is applied. This technique is used for the vertical profiling of ozone and minor species (NO_2, BrO, OClO) by limb measurement of SCIAMACHY onboard ENVISAT or of OSIRIS onboard Odin [56].

Occultation instruments observe the Sun, the Moon or a star when these natural sources are located on the other side of the atmosphere with respect to the remote sensing instrument. The light path is much better determined than for instruments based on scattered solar light due to the much simpler geometry of observation and a reference spectrum without absorption can always be taken outside the atmosphere. These instruments are then very well adapted for the long-term monitoring of the vertical profile of ozone and other constituents. On the other side, they do not provide a geographical coverage as complete as nadir instruments. The solar occultation instrument SAGE2 [57], launched in 1984, is often used as a reference for ozone trend studies over the last 20 years. It has been complemented in 2001 by SAGE3 with improved performances. POAM2 [58] and POAM3, also based on the solar occultation technique, provide vertical profiles of O_3 and NO_2 at polar latitudes and are used for the study of ozone destruction in both hemispheres.

Occultation measurements can also be performed using stars. Compared to solar occultation, the advantages are the point source nature of stars, which improves the knowledge of the light path and the number of available stars, allowing a better latitude coverage. GOMOS onboard ENVISAT is the first space instrument dedicated to the study of the atmospheric composition of the Earth using the technique of stellar occultation [59].

4.3. Balloon-borne observations

UV-visible spectrometers are often embarked under stratospheric balloons to measure vertical profiles of chemical species in specific locations (polar vortex, tropics...) during scientific campaigns or for the validation of satellite sensors. They are based on principles similar to those of satellite instruments, i.e., solar [60], lunar [61] and stellar occultation [62].

4.4. Ozone lidar

Ozone is measured using a technique called Differential Absorption Lidar or DIAL [63]. In this technique, two different pulsed laser beams are transmitted vertically into the atmosphere. One is emitted at a wavelength absorbed by ozone (i.e., 308 nm for stratospheric ozone lidars), and the other one at a wavelength not absorbed (i.e., 351 nm). These two beams are scattered elastically by molecules and particles, and the backscattered light is collected using a telescope. The absorbed signal falls off much more quickly than the non-absorbed one. Ozone concentration as a function of the altitude can be extracted from the ratio of the two backscattered signals.

This technique fails, however, in situations of high aerosol burden in the atmosphere, as this occurred after the eruption of Mt. Pinatubo in 1991. With such heavy aerosol loading, a technique known as Raman DIAL is used to measure ozone in the presence of aerosols [64]. In this technique, the two laser beams are scattered inelastically by atmospheric N_2, which is a purely molecular scattering process. The Raman backscattered signal from the shorter wavelength laser still retains the ozone absorption signature, and the Raman backscattered signal from the longer wavelength laser is essentially a purely dry air signal. Ozone extracted from these signals is much more reliable when aerosols are present.

The altitude range for active ozone measurements is typically from 10 to 50 km for a stratospheric lidar, and 2 to 15 km for a tropospheric lidar.

4.5. Spectroscopy problems

The main uncertainties in UV-visible spectroscopy lie in the uncertainties in absorption, cross sections and in their temperature dependence. UV-visible absorption spectra are often too complex to be calculated from theoretical methods. They are usually measured in the laboratory with an uncertainty of a few % and at only some temperatures. Differences of up to 3% can be obtained in retrieved ozone concentration depending on the choice of ozone cross section dataset. Vandaele et al. [65] report up to 7% differences between NO_2 cross sections measured by different groups and [66] mentions up to 10% differences for NO_3. Additional uncertainties can occur from species not taken into account in the retrieval or from unknown weak absorption bands.

5. Conclusion

Many techniques are usable for the measurement of the components or pollutants of the atmosphere. Among those which are most powerful and which allow simultaneous measurements of a great number of species, quantitative molecular spectroscopy is emerging quite markedly. In this paper, we have more particularly insisted on remote sensing measurements using microwave, infrared, and UV-visible spectroscopy. Constant improvement of spectrometric instruments (used currently from the ground, onboard aircraft, balloon and satellite) makes them very effective sounders by the number of simultaneously measurable species (resulting from their broad spectral coverage) and by their sensibility/selectivity (resulting from optimisation between spectral resolution and signal to noise ratio).

The principle of remote sensing measurements requires a precise knowledge of the spectroscopic properties of the probed molecules. Experimental work for recording high resolution laboratory spectra as well as theoretical studies for their detailed analyses makes it possible to extend and improve the spectroscopic data banks used for modelling atmospheric spectra. Thus, they are essential for increasingly reliable and precise inversions of the spectral signatures in order to retrieve the atmospheric distributions of the corresponding constituents. The future will see a more frequent use of active techniques (lidar) or heterodyne receivers (with embarked laser local oscillator) for the measurement of atmospheric composition from space, but the corresponding instruments are not yet completely ready for space applications.

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