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Lidar measurements of atmospheric ozone

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1. Introduction

The first lidar measurements of atmospheric ozone were performed back in the late 1970's, using tunable dye laser sources which provided by frequency doubling techniques the appropriate wavelengths required for a DIAL measurement in the Huggins absorption bands of ozone (280-320 nm). These pioneering works [1, 2] were followed by the advent of powerful laser sources, i.e. exciplex lasers, providing a direct emission in the UV-wavelength range [3,4]. Furthermore, the possible generation of frequency shifted laser lines using stimulated Raman scattering in hydrogen or deuterium cells, allowed the emissions of appropriate wavelengths for ozone measurement in the UV-wavelength range, from the various harmonics of solid state lasers such as the Nd:Yag laser, or exciplex lasers [5,6]. These developments led to the replacement of the tunable dye laser sources, due mainly to their lake of reliability for field experiments. Developments were also performed in the IR wavelength range where ozone presents an absorption band at 9.6 µm, based on the CO_2 laser technology [7]. Their field of applications is however mainly restricted to measurements in polluted aeras, and will not be further developed in the present paper. Considering the UV-wavelength range, from 266 nm to 355 nm, the fifteen years of development as referenced above led to the present availability of reliable lidar systems for deployment in field experiments, which have largely contributed to the overall research on atmospheric ozone.

2. Progress in methodology of DIAL ozone measurements

The methodology of DIAL measurement for application to atmospheric ozone has been assessed very carefully over the past 25 years [8]. First of all, the optimization of the DIAL measurement in terms of the statistical uncertainties related to the signal to noise ratio, implies an appropriate choice of the absorbed and reference wavelengths [9]. Taking into account the average ozone altitude distribution, this optimization is quite different for measurements below and above the ozone maximum. In the first case, i.e for tropospheric and lower stratospheric ozone, shorter wavelengths have to be selected, mainly below 300 nm. For measurements above the bulk of the ozone layer, i.e. in the altitude range from 25 km to 50 km, the optimized wavelength for the absorbed emission line corresponds to the emission of the XeCl exciplex laser at 308 nm. Therefore, different emitting systems have to be used for measurements in various altitude ranges, the choice of the laser source for tropospheric measurements being further determined by the exact altitude range to be probed, and the time resolution required. Boundary layer measurements led thus to different system optimization as compared to free troposphere measurements, and the same difference is to be considered between ground-based and airborne systems, for which the signal dynamics can be quite different [10].

Although the inversion of the lidar equation to retrieve the concentration of the constituant of interest, i.e. ozone, from the measurement of the atmospheric transmission can be considered as rather straightforward, a carefull analysis implies the development of appropriate algorithms to take into account the background noise and its potential variation with altitude due to signal-induced noise effects, the time sampling of the lidar backscattered signals, and the requirement for range derivation of the received signals. As part of this signal analysis, specific numerical filters have been developped, which in most cases have been tested by forward-inverse model simulations of the lidar signals for a given system.

Systematic errors have also to be taken into account which include the wavelength dependence of the backscatter and extinction coefficients between the absorbed and the reference wavelengths, and the temperature dependence of the ozone absorption cross-sections [9]. Among these uncertainties, the most important are related to the aerosol effects. In terms of backscattering, large errors can be induced by a rapid altitude variation of the aerosol scattering, which occurs at the edge of intense scattering layers. In background aerosol conditions, such interferences are mainly restricted to the boundary layer. However, measurements in the stratosphere can also be affected in the presence of dense stratospheric aerosol layers, resulting from major volcanic eruptions such as El Chichon in 1982 and Mount Pinatubo in 1991. Furthermore, the large value of the wavelength separation between the absorbed and reference wavelengths, i.e. 50 nm, adopted in order to increase the differential absorption cross section of ozone for stratospheric measurements, can in turn lead to large errors due to the differential extinction of the aerosol particles. Although such biases are restricted to the altitude ranges where aerosols are present, they have to be corrected. Specific algorithms have been developped based on ancillary lidar measurements of the aerosol backscatter, usually from the reference wavelength signal or from a distinct aerosol lidar operating in the visible. Nevertheless, assumptions have then to be made on the wavelength dependence of the aerosol scattering and extinction, i.e on the granulometry, nature and shape of the particles [11]. For high aerosol scattering coefficients however, these corrections are far from being reliable, especially when lidar measurements are used for the analysis of the interannual natural variability or for the determination of long-term trends. In order to avoid such corrections and to take directly into account in the measurements the largest potential errors induced by the differential backscattering effects, a new method has been recently developed which makes use of the additional signal directly generated in the atmosphere from the two emitted wavelengths by spontaneous Raman scattering on the N_2 molecule [12]. By comparing the slopes of the signals measured on these particular lines, the effects of the differential backscattering are minimized and those of the differential extinction can be more easily accounted for.

These various methodological aspects have in general been tested for each lidar system at its particular site, using local intercomparison with other ozone measuring techniques, such as balloon-borne ozonesondes, satellite observations or spectrophotometers with appropriate profile retrievals provided by Umkehr type measurements [13, 14]. If the balloon sondes usually provide measurements with the appropriate vertical resolution to fully test the lidar capacity, they are limited to altitude ranges below 30 km for regular sondes launched on meteorological balloons. Large balloons flying up to 45 km can still be used, although the launching sites are in general rather distant from the lidar sites [13]. Finally, taking into account the poor vertical resolution of other high altitude ozone measuring system, with the exception of the spaceborne SAGE instruments which use the solar limb occultation technique, the problem of the validation of the lidar systems above 35 km is still open, and requires specific lidar intercomparisons. Such exercises also present the advantage to provide a frame for testing the full lidar system, including the data analysis part (see below).

3. Contribution of lidar measurements to ozone research

Lidar instruments based on the DIAL method and operating in the UV wavelength range are undoubtly recognized as mature research instruments, which can bring a determining contribution in various field of ozone research. They presently constitute the most reliable and self-calibrated ground-based instrument for high vertical resolution, long-term measurements of the ozone vertical distribution. This capacity has largely led to the concept of the Network for Detection of Stratospheric Change (NDSC) which is presently being implemented at the international level, under the auspices of the World Meteorological Organization and the International Ozone Commission, and with the involvement of several national agencies world-wide. The NDSC adresses three main objectives related to the understanding of the natural balance of stratospheric ozone and its evolution under anthropogenic influences : (1) quantification of the natural variability of the stratosphere at various temporal scales, (2) validation of satellite measurements of ozone and related constituents, (3) early detection of long term trends. Each station of the network includes state-of-the-art intruments such as lidar (ozone, temperature, aerosols), UV-visible, IR and microwave spectrometers, in addition to more classical instruments such as balloon-borne ozonesondes. In 1994, several primary and interim stations, equipped with lidar systems are operating in the Arctic (Eureka, Spitzbergen, Thule), in middle latitude of both hemispheres (Alpine Station, Lauder in New-Zealand), in the tropical regions (Mauna Loa, Ile de la Réunion) and in Antarctica (Dumont d'Urville, McMurdo) [15, 16, 17, 18]. As a complement to these sites, a permanent test site has been established at the Jet Propulsion Laboratory Table Mountain Facility, in California [19], and secondary sites are also implemented, several of which are performing routine measurements of stratospheric ozone, especially in the middle latitude regions of the northern hemisphere as part of the European project ESMOS, and in Japan [20]. Finally, in order to address the requirements for a carefull assessment of the instruments deployed in the NDSC, a travelling standard instrument has also been developped at the NASA Goddard Space Flight Center [21]. This instrument has taken part in the ozone measuring instruments intercomparison campaigns which took place in Table Mountain in 1989, and at the Observatoire de Haute-Provence in 1992. Ozone lidar operation in the NDSC have already led to important results concerning the natural intra- and inter-annual variability of stratospheric ozone [16, 22, 23], satellite instruments validation especially as part of the ground-truth programme of the UARS satellite launched in September 1991, and for such instruments as SBUV and SAGE II [14]. At the present time however, the measurement record of stratospheric ozone by lidar goes back only for 7-8 years, so that statistically significant trends cannot be inferred from such data bases.

The concept of networking can also be applied at regional scales. In Europe, the successfull coordinated developement of DIAL lidars for tropospheric ozone measurement as part of the EUROTRAC programme TESLAS, led to the implementation of several systems which are presently operating in the frame of the TOR (Tropospheric Ozone Research) network. Here again, the results have proven to be particularily usefull for process studies, mainly related to exchange processes between the various layers of the lower atmosphere : stratosphere-troposphere and boundary layer-free troposphere exchanges [24, 25].

Ground-based systems are however limited in their application, when considering the possibility to directly quantify the ozone budget at local or regional scales. The development of airborne systems for measurements both in the troposphere and lower stratosphere have led to various applications in the frame of coordinated campaigns. Examples of such applications can be found in the ABLE (Atmospheric Boundary Layer Experiment) campaigns devoted to the quantification of the ozone build-up in the boundary layer as a result of biomass burning in the tropical regions, or in the various airborne campaigns organized in the Arctic and Antarctic regions to study the evolution of the stratospheric ozone layer in polar regions [26,27].

4. Conclusion

After twenty years of experimental and methodological developments, lidar systems to measure the ozone vertical distirbution have now maturated to a state where they can be considered as reliable instruments for deployment in field experiments, both from the ground and airborne plateformes, and for long-term scientific monitoring. They constitute the basis of the presently established networks which are implemented at various spatial scales to quantify the natural variability of atmospheric ozone and to give and early evidence for anthropogenic changes, both in the troposphere and in the stratosphere. Future operation of such systems will be of particular value in the future, provided that the efficient coordination, already in place at the international level to ensure the high quality of the measurements required for trends detection, be effectively pursued.

5. References

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