Ground Based, Millimeter Wave Measurement of Ozone in the Middle Atmosphere

Summary of Research

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Principal Investigator: Dr. Alan Parrish

Department of Astronomy
University of Massachusetts
Amherst, MA 01003

Introduction and Overview:

There is a need for highly reliable measurements of stratospheric ozone. Policy makers worldwide concerned with public health rely on a clear consensus from the scientific community as a basis for ozone-related environmental policy that has a significant impact on national economies. The latest such consensus was presented in WMO [1999], and used in a 1999 meeting of the parties considering amendments to the Montreal Protocol on Substances that Deplete the Ozone Layer. The scientific community, in turn, needs highly precise and accurate measurements of ozone levels, and small time derivatives of these levels, both in continued development of its understanding of the physical and chemical processes involved and as clear evidence that these processes are occurring as stated. Over most of the world, changes in ozone levels are small. For example, over the heavily populated northern midlatitudes, the linearized rate of ozone decline is between 0.2% per year and 0.7% per year, depending on altitude. These values are small enough to make measurement requirements technically challenging. Data quality may suffer from imperfections in individual instruments. In one instance, early results from a satellite-borne ozone sensor were later found to be invalid because of calibration drift. Even in the absence of drift, the absolute calibration of a new sensor may differ slightly from that of its predecessor in service. Most ozone remote sensing instruments operate at ultraviolet or infrared wavelengths where scattering from dust and aerosols must be taken into account; results from these systems may be or are affected following a major volcanic eruption, such as the one at Mt. Pinatubo in 1991. Given these difficulties, a consensus of measurements from several independent systems is required to insure a reliable understanding of stratospheric ozone levels.

Because of the above-described need for highly precise and accurate ozone measurements using several independent techniques, there was interest in developing several techniques which were known but not highly developed in the 1980's into systems capable of being used in an operational manner to make measurements with the level of quality needed to detect small trends in ozone levels. A ground-based microwave instrument capable of remotely sensing stratospheric ozone had been designed by the Principal Investigator of the present project. This instrument was built at the Millitech Corporation in South Deerfield, Massachusetts before the present work began. (Funding for design and construction of the instrument came from sources other than the present grant.) The instrument measured the spectrum of one of the many emission lines produced by purely rotational transitions of ozone, one at a frequency of 110.8 GHz. The altitude distribution of ozone can, in principle, be retrieved from the details of the pressure-broadened spectrum of the ozone transition. However, the level of contamination of the spectral measurement by instrumentally induced artifacts must be very low in order to retrieve a ozone profile of useful quality from it. The Millitech instrument demonstrated spectral purity at an adequate level, and there were promising ideas for instrumental improvements and for further development of the technique.

The initial objectives of the present project, then, were to develop techniques for calibrating the Millitech instrument, to minimize artifacts in the spectra it produces, to optimally retrieve ozone profiles from the spectra, to test the quality of the microwave profiles by comparing them with profiles obtained using several other, independent techniques over both short and periods of time, and to perform research using the ozone data gathered with the instrument. This work was carried out with two principal collaborators. One was Dr. Brian J. Connor, who was at the NASA-Langley Research Center from the beginning of this work in 1989 until 1995, and since has been at the National Institute of Water and Atmospheric Research (NIWA) in Lauder, New Zealand. The other was Dr. J. J. Tsou, who has been variously affiliated with the Science Applications International Corporation (SAIC) and Gordley Applied Technology Software, Inc. (GATS), both in Hampton, Virginia.

Course of the Work:

Following initial development of calibration and retrieval software, we installed the Millitech instrument at the Table Mountain Facility (TMF), in Wrightwood, California in July, 1989. (TMF is a field station of the Jet Propulsion Laboratory (JPL).) This site was chosen for several reasons: First, the site had been chosen as a test and development site for the Network for Detection of Stratospheric Change (NDSC). The NDSC is an international project that had been initiated by the NASA Office of Earth Science, Upper Atmosphere Research Program at about that time. Second, a JPL group headed by Dr. I. S. McDermid was developing a lidar system for ozone measurements at the site, and ongoing intercomparisons of profiles produced by the microwave and lidar instruments would lead to improved understanding of both. Third, the site is at an altitude of 2500 meters. The high altitude minimized both the tropospheric attenuation of the stratospheric ozone signal and the variability of this attenuation. Because error in the measurement of the tropospheric attenuation is an important component of the microwave measurement error budget, the qualities of the site were expected to improve the precision and accuracy of the measurements.

The initial activity at TMF was centered around the NDSC-sponsored Stratospheric Ozone Intercomparison Campaign (STOIC), which took place in July and August, 1989. The campaign was intended to test the performance of both newly developed and existing ozone measurement systems. The microwave system, two lidar systems, two sets of balloon-borne, electrochemical concentration cell ozonesondes, the satellite-borne Stratospheric Aerosol and Gas Experiment (SAGE-II), one set of rocket-borne ozonesondes, and ground-based Dobson and Brewer total column ozone instruments all participated in the campaign. The campaign was carried out "blind"; that is, all data were privately submitted to, and evaluated by, an independent referee. Results from the campaign were published in *Margitan, et al,* [1995]. While the agreement between the microwave profiles and the average of all profiles measured during the campaign was within 5% from 22-50 km, we had some concern that these results might not be repeatable. Initial microwave measurements made at Table Mountain were seriously compromised by interference from nearby radar transmitters. Measures hastily undertaken before the STOIC campaign began temporarily shielded the instrument from the interference but also introduced some low level systematic effects in the spectra, which could, under certain circumstances, cause significant errors in the retrieved profiles. The only method that we believed would effectively eliminate interference from nearby transmitters but not compromise the spectral quality involved substantial modifications to the microwave receiver. These modifications were carried out at the University of Massachusetts during the first several months of 1990, and have been proven effective.

The microwave instrument remained at Table Mountain until June, 1992, taking data nearly continuously. During this period we made a number of improvements to the instrument itself that further improved the quality of the spectra. We also upgraded calibration procedures, and simplified operational procedures. We published a thorough description of the instrument, calibration and measurement techniques, profile retrieval method, and displayed some early results from intercomparisons made with the collocated JPL lidar over a period of several months in *Parrish et al.* [1992]. We subsequently published a detailed, formal error analysis for our measurements in *Connor et al.* [1995], and results from a long term (1989-1992) intercomparison between the microwave profiles and those produced by the collocated JPL lidar and those measured during overpasses by the SAGE-II satellite-borne instrument (*Tsou et al.* 1995).

Most ozone-measuring techniques are not capable of making good measurements in the upper stratosphere and mesosphere. The microwave technique is an exception. We used the Table Mountain data in a study of the diurnal ozone variation in the upper stratosphere and mesosphere which resolved some issues regarding measurements made with the satellite-borne Limb Infrared Measurement of the Stratosphere (LIMS). The discrepancy between models and LIMS daytime measurements and models was larger during the day than it was at night, and a suggestion had been made that this was due to failure to take into account in data analysis departures from local thermodynamic equilibrium in the energy levels of the molecular transitions used in the LIMS measurements, rather than problems with the models that were compared with the LIMS measurements. Our work showed that the non-LTE extended to lower altitudes than previously expected, to about 55 km. Our work was reported in *Connor, et al.*, [1995]. A companion model-data comparison using our mesospheric data was reported in *Siskind, et al.* [1995].

Powerful volcanic eruptions introduce large amounts of sulfur dioxide into the stratosphere, which reacts with water vapor to form sulfuric acid aerosols, The scattering of light by the aerosol droplets hampers ozone measurements made with techniques that employ infrared, visible, or ultraviolet light. However, the scattering is negligible at the much longer wavelengths in the microwave region. Microwave measurements are therefore useful for detecting changes in ozone levels during periods of high aerosol loading. We used our Table Mountain data in a study that demonstrated ozone depletion of up to 15% in the year following the eruption of Mount Pinatubo in the Phillipines compared to previous years. This work was reported in *Parrish, et al.* [1998].

The performance demonstrated by the microwave instrument while it was at Table Mountain led to acceptance as a primary instrument in the Network for Detection of Stratospheric Change, and an agreement that the instrument would be moved to the southern midlatitude station of the NDSC at Lauder, New Zealand. We completed the installation at Lauder in November, 1992, following participation in a NDSC intercomparison campaign at l'Observatoire de Haute Provence (OHP) in France in the summer of that year. The good performance also led to funding, from other sources, of a second instrument. The new instrument was constructed at Millitech during the second half of 1993 and the first half of 1994. The new instrument is identical to the original one in design, except for minor modifications made to correct problems discovered in field use of the original instrument and to simplify manufacturing. The work also led to an invitation to prepare a paper for a special issue on remote sensing of the *Proceedings of the Institute of Electrical and Electronic Engineers,* which appeared in December, 1994 (*Parrish* [1994]).

The new instrument was installed at Table Mountain for initial testing in August, 1994. The original instrument was temporarily returned from New Zealand to Table Mountain for several months during the winter of 1994-1995 in order to perform a direct comparison between the two instruments. This work was hampered by an unusually stormy winter and a series of equipment failures. We found that the small differences between the two instruments that affected the key measurement of tropospheric attenuation could be precisely identified and measured using our calibration techniques. However, small differences between the profiles produced by the two instruments were noted, despite our high level of confidence in the principal calibration parameters. A likely cause of these profile differences, involving a weak spurious signal generated in the intermediate stages of the receiver electronics, was identified.

The original instrument was returned to New Zealand and reinstalled at the Lauder site in March, 1995, in time to participate in a NDSC intercomparison campaign, "Ozone Profiler Assessment at Lauder" (OPAL), which took place that April. Results from the campaign were published in two papers, *McDermid et al.* [1998a] and *McDermid et al.* [1998b]. The microwave profiles were found to be about 10% higher than the consensus of all instruments. This discrepancy was larger than we expected, based on the excellent measurements previously obtained with the same instrument at Table Mountain. The discovery of this discrepancy triggered a major reanalysis of our Lauder data set which principally centered on two issues. First, we needed a more sophisticated model in the analysis that determined the tropospheric attenuation of the ozone signal, because that attenuation is higher by more than a factor of two at Lauder than it was at Table Mountain. Also, particularly at night in certain seasons, the attenuation measurement was affected by an inversion in the temperature profile near the surface. We developed a new model which incorporated climatological data based on a large number of radiosonde temperature profiles measured by others at Lauder. Second, we found that our measurement of the signal beam elevation angle, which is important to the calibration, was slightly in error because either the concrete pad under the instrument shelter, or the one supporting the calibration target used for elevation angle calibration had heaved or settled by about a centimeter since the pads were installed and initially surveyed. We then developed an alternate technique for performing the calibration, one which did not require physical survey information, and which therefore could be used retrospectively. A new intercomparison, based on the revised data as analyzed using the new techniques, showed performance comparable to our earlier experience at Table Mountain, despite the poorer tropospheric conditions at the Lauder site. Results from the new intercomparison were published in *Tsou et al.* [2000].

Meanwhile, the second Millitech instrument was installed at the NDSC Hawaii station in the summer of 1995. Because construction of the permanent building for NDSC instruments on Mauna Loa had been delayed, we designed and procured construction of a small building to house the microwave instrument to start acquiring Mauna Loa data without delay. With installation complete, we participated in the initial "blind" intercomparison campaign (known as the MLO3 campaign) with excellent results as published in *McPeters et al.* [1999]. Preliminary results from an analysis of the data recorded between August, 1995 and the end of 1998 are presented in *Parrish, et al.* [2001].

Summary of Performance:

We summarize the results presented in the long term intercomparison papers referred to above as follows: At both Lauder and Mauna Loa, the profile differences are generally less than 5%, particularly in the middle stratosphere. In the lower stratosphere, the average differences at Lauder are less than 5% down to our 56 mbar lower limit. At Mauna Loa, there is a considerable range of differences in the lower stratosphere. Generally, the Mauna Loa measurements appear to be of the order of 5% too high in the lower stratosphere. The profile comparisons in the upper stratosphere indicate that the microwave accuracy is good at both sites up to at least 0.6 mbar. (We have not extended comparisons to higher altitudes because of the difficulty in accounting for the effect of photolysis-induced (i.e. diurnal) changes in ozone levels along the line of sight of the satellite-borne, occultation type instruments.) In general, the average differences between the microwave measurements and those made with other instruments are biased slightly more positively at Mauna Loa than at Lauder. Except for this, there appears to be no characteristic shape that is consistent across the difference profiles. We are aware of differences between the sites in both the data analysis procedures and the instruments themselves that could cause small differences between the systematic errors in the profiles. We have not yet analyzed these differences thoroughly enough to comment on them further.

We have also performed regression analyses to calculate relative trends between time series of our measurements and others, such as SAGE-II, lidar, and ozonesondes. In the absence of systematic drift in one or both of the compared data sets, the relative trends should be zero within statistical limits set by noise on the data. To summarize the Lauder relative trends data, we find that about two thirds are smaller than 0.5% per year, 9% are significant at the two standard error level and between 0.5 and 1.5% per year, and one point (2%) is at 1.9% per year and significant. In the lower stratosphere, the trends between the microwave and the other instruments at 56 mbar are consistently positive, but only one is significant. At this altitude, the most sensitive comparisons suggest a trend of about 1% per year. No trends are indicated in the upper stratosphere. There may be a small trend in the microwave data in the middle stratosphere, but an analysis based on a longer record is needed to draw a firm conclusion on this point. Using the present, preliminary Mauna Loa data set, we find statistically significant, altitude dependent relative trends between the microwave and lidar measurements in the lower and middle stratosphere. Typical values are 1-2% per year in the middle stratosphere. The uncertainty in the lower stratosphere trend data is too large to draw any useful conclusions, given the short length of the presently available microwave data set. We expect an improvement in this result when a uniform reprocessing of the Mauna Loa data, being done with funding from other sources, is completed.

To provide some perspective on the relative performance of the microwave technique to that of other new techniques, we have compared the SAGE-II microwave relative trend figures from our long term comparisons with those derived by *Russell and Schmidt* [1998] from comparisons between several lidars and SAGE. (The lidar technique is thought to be particularly promising because very little external calibration information is required in the analysis of its data.) We find that the distribution of SAGE-microwave trends is no larger than the SAGE-lidar trends, indicating that the performance of the two is comparable.

Conclusions:

We have developed the technique of measuring stratospheric ozone profiles by means of ground- based microwave remote sensing from a promising experimental technique to an operational system having precision and accuracy that is both sufficient for measurement of small ozone changes (if deployed on a sufficiently large scale) and competitive with other modern techniques. We have demonstrated advantages of the microwave technique, such as the upper stratospheric and mesospheric capability, and the insensitivity to aerosols in research projects. We have deployed two instruments and, with them, accumulated a substantial beginning to a long term record of high quality ozone measurements.

Inventions:

No inventions were developed during the course of this work.

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