

MICROWAVE LIMB SOUNDER MEASUREMENT OF STRATOSPHERIC SO₂ FROM THE MT. PINATUBO VOLCANO

W. G. Read, L. Froidevaux, and J. W. Waters

Jet Propulsion Laboratory, California Institute of Technology, Pasadena, Ca

Abstract. The Microwave Limb Sounder (MLS) experiment on the Upper Atmosphere Research Satellite (UARS) began atmospheric measurements on September 19, 1991. Residual SO₂ from the Pinatubo eruption was detected immediately after instrument activation. The SO₂ was located in a tropical band with peak mixing ratios of ~15 ppbv near 26 km altitude. The observed decay of SO₂ has provided an estimate of the OH concentration in the plume.

Introduction

The large Mt. Pinatubo eruption is expected to exhibit detectable climate forcing [Hansen et al., 1992]. The injected SO₂ into the stratosphere is oxidized to sulfuric acid [McKeen et al., 1984] then forms aerosols through heteromolecular homogeneous nucleation of H₂SO₄ and H₂O vapors [Turco et al., 1982]. This aerosol layer scatters a fraction of the incident solar radiation to space causing surface cooling. Satellite based measurements of SO₂ (TOMS [Bluth et al., 1992] and SBUV/2 [McPeters, 1992]) and aerosols (SAGE II [McCormick and Veiga, 1992] and AVHRR [Stowe et al. 1992]) have provided spectacular images of atmospheric perturbations following the eruption.

The Upper Atmosphere Research Satellite (UARS) [Reber, 1990] was delivered into a 585 km, 57° inclined orbit by Space Shuttle "Discovery" (STS-48) on 12 September 1991. The UARS instruments have provided vertically resolved (2.5–5.0 km) daily global maps of volcanic SO₂ and aerosols. Presented here are SO₂ measurements from the UARS Microwave Limb Sounder (MLS) which began observations three months after the eruption when concentrations were below TOMS and SBUV/2 detection limits. MLS contains 3 heterodyne radiometers at 63, 183, and 205 GHz and a vertically scanned field-of-view [Barath et al., 1993]. The daily global coverage is 34°S–80°N or 80°S–34°N depending on the UARS yaw state. The technique used for constituent retrievals is summarized in (J. W. Waters, submitted for publication in *Nature*, 1993).

MLS SO₂ Data

Beginning with the first day of data collection on 19 September 1991, a significant spectral feature located in a channel near the edge of the ClO band was evident in the equatorial radiances and had no diurnal variation. Figure 1 shows the feature, which is assignable to the SO₂ 18₃16 → 18₂15 rotational transition at 204247 MHz, and the fit quality. Confirmation of this assignment comes from the difference spectrum between the modelled and measured radiances in the MLS 205 GHz ozone band which reveals a feature assignable to the SO₂ 24₃21 → 24₂22 ro-

tational transition at 200287 MHz. The line positions and strengths used for SO₂ profile retrievals are from [Pickett et al., 1992] and line broadening parameters are from [Meier, 1978]. MLS measures the ³²S¹⁶O₂ isotope and all abundances have been increased 5% to yield total SO₂.

Figure 2 shows an average of 95 individual vertical profiles between 10°S and the equator on 21 September 1991. The SO₂ mixing ratio retrieval coefficients shown in the figure are amplitudes of triangular functions in logarithm of atmospheric pressure with peaks located at ~21 km, ~26 km, ~31 km, ~36 km, and ~41 km, and having a full width at half maximum of ~5 km. The right and left axes of Figure 2 show the corresponding atmospheric pressure and altitude scales. The figure shows the vertical range of MLS SO₂ where the random error exceeds systematic errors for an individual profile retrieval. The error bars show the individual profile random error component which varies between 3.0–7.0 ppbv for mixing ratio or 8 × 10¹⁵ molecules / cm² for the column above 21 km. An MLS SO₂ map at 26 km on 21 September 1991, is shown in Figure 3.

Temporal Decay

Figure 4 shows the time evolution of zonally averaged SO₂ as a function of latitude at three heights. Background concentrations which do not decay, caused by systematic errors in the MLS retrieval model, have been subtracted. The subtracted background is less than 20% of the initial data retrievals for the 26 and 31 km layers and approximately 50% of the 21 km layer. The 21 km SO₂ layer is displaced north of the equator. A layer at 26 km containing most of the SO₂ is initially displaced south (~ 10°S) but moves northward with the approaching northern hemi-

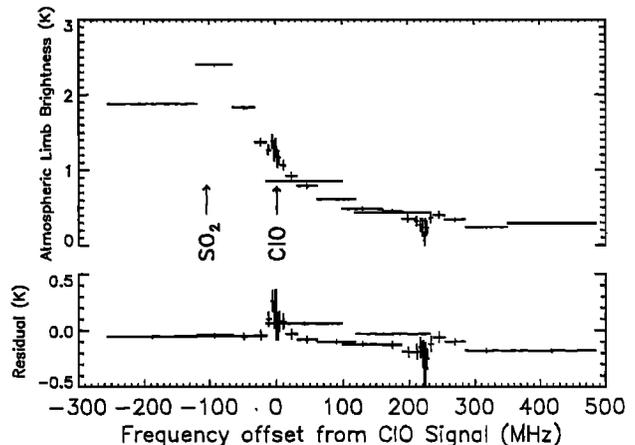


Fig. 1. Top: A 10°S–5°S zonal average of MLS nighttime radiances at 26 km showing the SO₂ line at 204247 MHz in the edge of the bands used to measure ClO. Bottom: The radiance residuals after fitting SO₂ and ClO. The ClO signal does not appear prominently at night because ClO is a photolytic product.

Copyright 1993 by the American Geophysical Union.

Paper number 93GL00831
0094-8534/93/93GL-00831\$03.00

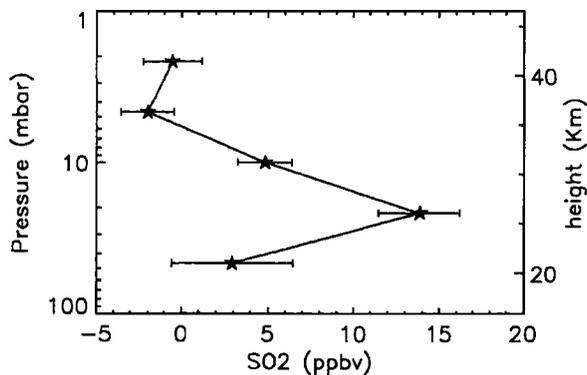


Fig. 2. Average of 95 individual SO_2 profiles between 10°S — 0° latitudes on 21 September 1991. Individual profile error bars are shown.

sphere winter. SO_2 at 31 km is peaked near the equator, and spreads into both hemispheres with a majority of material going south. When the 31 km layer is integrated to total molecules, there is no decay (in fact a slight increase) for ~ 30 days following initial MLS measurements, which would suggest that chemical removal of SO_2 is balanced by vertical transport during this time. These results appear consistent with other observations and dynamical expectations. SAGE II observes aerosols spreading north at altitudes below 21 km and south above [McCormick and Veiga, 1992] which correlates with MLS SO_2 at 21 km and 26 km. The upwelling SO_2 at 31 km might be caused by diffusion and aerosol heating [Kinne et al., 1992]. The latter dynamical effect is expected to lower tropical ozone and has been observed by TOMS [Schoeberl et al., 1993], [Chandra, 1993], ozonesondes [Grant et al., 1992], and MLS [Read et

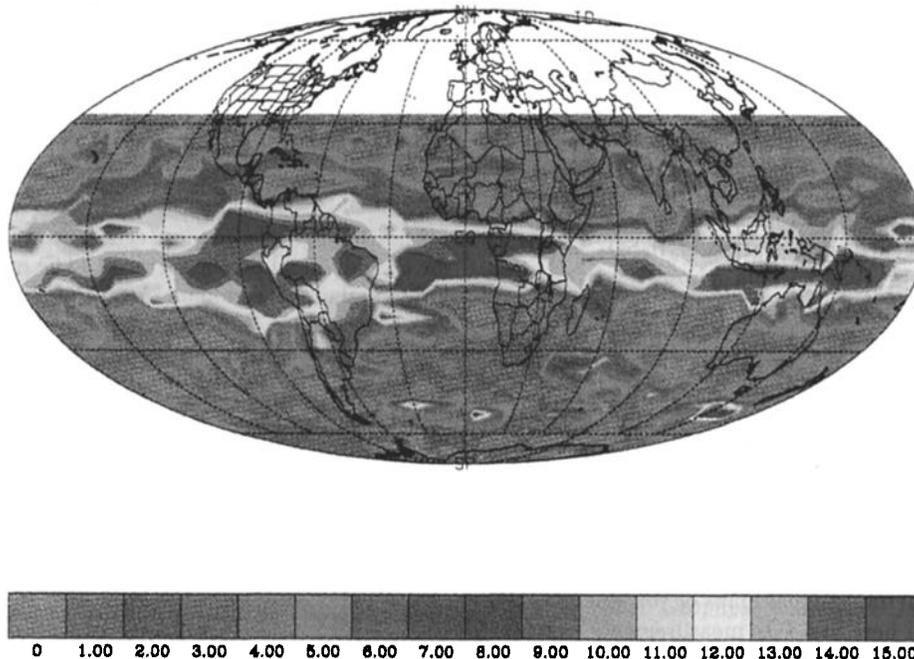


Fig. 3. Distribution of SO_2 at 26 km on 21 September 1991. MLS was viewing towards the south on this day, and no measurements were obtained north of 34°N . The color bar units are ppbv.

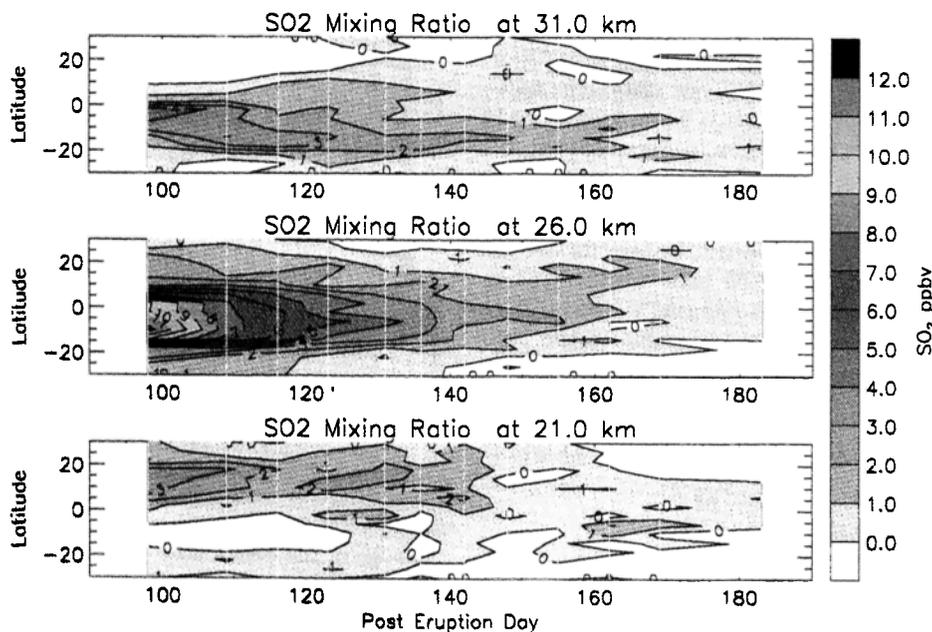


Fig. 4. Temporal evolution of zonally-averaged SO_2 between 32°S and 32°N at 31, 26, and 21 km. Contour units are ppbv, and the vertical lines indicate the days for which data were used to make this figure.

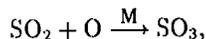
al. 1992a, 1992b).

The SO₂ removal rate and initial mass injection are relevant to OH chemistry and aerosol formation. Figure 5 compares SO₂ mass estimates from TOMS, SBUV, aircraft COSPEC, and MLS. Also shown in Figure 5 are the contributions by altitude to the MLS total mass estimate. TOMS obtains its value from integrating the SO₂ column within the cloud immediately following the eruption, and in principle detects all the SO₂ injected into the atmosphere. The aircraft measurement is a column value above its flight track, which has been zonally extrapolated to yield a total estimate [Mankin et al., 1992]. Two values extracted from NOAA/11 SBUV/2 column measurements are included for comparison [McPeters, 1992]. A best fit decay line to the MLS total SO₂ mass is shown in Figure 5. The slope is proportional to the e-folding decay rate and the SO₂ mass-axis intercept is the extrapolated initial injection. The greatest error source for the mass estimates in Figure 5 is the accuracy of the background removal applied to figure 4. This is estimated to be 50% of the subtracted background (shown in figure 5) and used for error weighting the data in the linear regression analysis. The ~21–31 km column e-folding decay of 33 days with an initial input of 17 Mtons compares well with TOMS 35 days and 20 Mtons [Bluth et al., 1992]. The measurement points from the various experiments are near the extrapolated MLS decay line.

Sulfur dioxide is catalytically converted to sulfuric acid by hydroxyl (OH) [McKeen et al., 1984]. The conversion rate is controlled by the reaction



A competing reaction,



which is roughly 300–1000 times slower between 21–26 km is neglected [Golombek and Prinn, 1993]. If all the SO₂ is processed by the former reaction and neglecting transport, an approximate OH profile can be inferred from the altitude dependent SO₂ decay rates using

$$\frac{d[\text{SO}_2]}{dt} = -k[\text{SO}_2][\text{OH}][M],$$

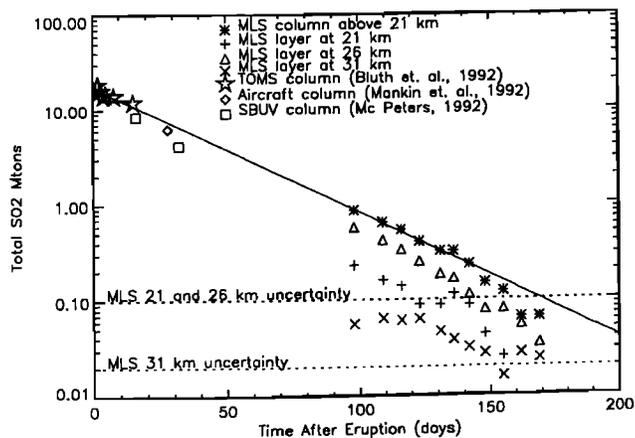


Fig.5. SO₂ from TOMS, aircraft, SBUV, and MLS.

where $[X]$ indicates the number density of X (M is total number density) and k is the pressure and temperature corrected reaction rate constant [DeMore et al., 1992]. Solving for daytime $[\text{OH}]$ gives

$$[\text{OH}] = \frac{2}{\Delta t k [M]}$$

where Δt is the SO₂ e-folding time and the factor of 2 approximately accounts for the diurnal behavior of OH. Table 1 provides the computational parameters and resulting OH profile. Both values are approximately 50% larger than a typical equinox model [Brasseur and Solomon, 1984] which is acceptable agreement considering the simplicity of this analysis and existing differences among models [Jackman et al., 1987]. The small SO₂ amounts in the 31 km layer are clearly perturbed by transport, invalidating a similar analysis at that altitude.

Table 1: MLS inferred monthly daytime tropical OH profile for October

height km	T K	[M] cm ⁻³	Δt days	$k[M]$ cm ³ sec ⁻¹	[OH] cm ⁻³
21	210	1.6×10^{18}	41	4.6×10^{-13}	1.2×10^6
26	230	6.7×10^{17}	29	2.4×10^{-13}	3.3×10^6

Conclusions

Initial results from the UARS/MLS experiment have shown that 100 days after the Pinatubo eruption, the atmosphere contained ~0.9 Mt of volcanic SO₂. The data indicate the majority of the residual SO₂ was in a layer near 26 km between 30°S and 15°N latitudes. A 33 day e-folding decay time with an extrapolated initial SO₂ injection of 17 Mt are estimated, and are similar to the analogous values from TOMS. The observed SO₂ decay is consistent with catalytic OH chemical removal; inferred zonal daytime average OH concentration at 21 and 26 km which are reasonably consistent with model predictions.

Acknowledgements. The research described herein was carried out by the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration.

References

- Barath, F., M. Chavez, R. Cofield, D. Flower, M. Frerking, M. Gram, W. Harris, J. Holden, R. Jarnot, W. Kloezeman, G. Klose, G. Iau, M. Loo, B. Maddison, R. Mattauch, R. McKinney, G. Peckham, H. Pickett, G. Siebes, F. Soltis, R. Suttie, J. Tarsala, J. Waters, and W. Wilson, The Upper Atmosphere

- Research Satellite Microwave Limb Sounder Instrument, *J. Geophys. Res.*, in press., 1992
- Bluth, G. J. S., S. D. Doiron, C. C. Schnetzler, A. J. Krueger, and L. S. Walter, Global Tracking of The SO₂ Clouds from the June, 1991 Mount Pinatubo Eruptions, *Geophys. Res. Lett.*, vol. 19, pp. 151—154, 1992.
- Brasseur G. and S. Solomon, *Aeronomy of the Middle Atmosphere*, D. Reidel Publishing Co. pp., Dordrecht, 441, 1984.
- Chandra, S., Changes in Stratospheric Ozone and Temperature Due to the Eruptions of Mt. Pinatubo, *Geophys. Res. Lett.*, vol. 20, pp. 33—36, 1993.
- DeMore, W. B., S. P. Sander, D. M. Golden, R. F. Hampson, M. J. Kurylo, C. J. Howard, A. R. Ravishankara, C. E. Kolb, and M. J. Molina, Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling, *JPL Publication 92-20, Evaluation No. 10*, 185 pp., Jet Propulsion Laboratory, Pasadena, Ca., 1992.
- Grant, W. B., J. Fishman, E. V. Browell, V. G. Brackett, D. Nganga, A. Minga, B. Gros, R. E. Veiga, C. F. Butler, M. A. Fenn, and G. D. Nowicki, Observations of Reduced Ozone Concentrations in the Tropical Stratosphere After the Eruption of Mt. Pinatubo, *Geophys. Res. Lett.*, vol. 19, pp. 1109—1112, 1992.
- Golombek, A. and R. G. Prinn, A Global Three-Dimensional Model of the Stratospheric Sulfuric Acid Layer, *J. Atm. Chem.*, vol. 16, pp. 179—199, 1993.
- Hansen, J., A. Lacis, R. Ruedy, and M. Sato, Potential Climate Impact of Mount Pinatubo Eruption, *Geophys. Res. Lett.*, vol. 19, pp. 215—218, 1992.
- Jackman, C. H., R. K. Seals, M. J. Prather, editors, Two-Dimensional Intercomparison of Stratospheric Models, *NASA CP-3042*, 608 pp., National Aeronautics and Space Administration, Washington D. C., 1989.
- Kinne, S., O. B. Toon, and M. J. Prather, Buffering of the Stratospheric Circulation by Changing Amounts of Tropical Ozone: A Pinatubo Case Study, *Geophys. Res. Lett.*, vol. 19, pp. 1927—1930, 1992.
- Mankin, W. G., M. T. Coffey, and Aaron Goldman, Airborne Observations of SO₂, HCl, and O₃, in the Stratospheric Plume of the Pinatubo Volcano in July 1991, *Geophys. Res. Lett.*, vol. 19, pp. 179—182, 1992.
- McCormick, M. P. and R. E. Veiga, SAGE II Measurements of Early Pinatubo Aerosols, *Geophys. Res. Lett.*, vol. 19, pp. 155—158, 1992.
- McKeen, S. A., S. C. Liu, and C. S. Kiang, On the Chemistry of Stratospheric SO₂ from Volcanic Eruptions, *J. Geophys. Res.*, vol. 89, pp. 4873—4881, 1984.
- McPeters, R. D., Global Measurements of SO₂ From Pinatubo, the Conversion to Aerosol, and the Resulting Ozone Effects, presented at 1992 Fall Meeting, Am. Geophys. Union, San Francisco, Ca., Dec. 7 to Dec. 11, 1992.
- Meier, D., Linewidth and Intensity Measurements of SO₂ Lines at 94 GHz with Self-Broadening and Broadening by H₂O, and N₂, *J. Quant. Spectrosc. Radiat. Transfer*, vol. 19, pp. 323—329, 1978.
- Pickett, H., R. Poynter, and E. Cohen, Submillimeter, Millimeter, and Microwave Spectral Line Catalog, *Technical Report 80-23, Revision 3*, 217 pp., Jet Propulsion Laboratory, Pasadena Ca., 1992.
- Read, W. G., J. W. Waters, and L. Froidevaux, UARS MLS Measurement of SO₂ From Mount Pinatubo Volcano, presented at Climate, Volcanism, and Global Change, AGU Chapman Conf., Hilo, Hawaii, Mar.23 to Mar.27, 1992a.
- Read, W. G., L. Froidevaux, and J. W. Waters, UARS MLS Measurements of SO₂ and O₃ in the Plume of Mount Pinatubo Volcano, presented at 1992 Fall Meeting, Am. Geophys. Union, San Francisco, Ca., Dec. 7 to Dec. 11, 1992b.
- Reber, C. A., The Upper Atmosphere Research Satellite, *EOS Trans. AGU*, vol. 71, pp. 1867, 1990.
- Schoeberl, M. R., P. K. Bhartia, E. Hilsenrath, and O. Torres, Tropical Ozone Loss Following the eruption of Mt. Pinatubo, *Geophys. Res. Lett.*, vol. 20, pp. 29—32, 1993.
- Stowe, L. L., R. M. Carey, and P. P. Pellegrino, Monitoring the Mt. Pinatubo Aerosol Layer With NOAA/11 AVHRR Data, *Geophys. Res. Lett.*, vol. 19, pp. 159—162, 1992.
- Turco, R. P., R. C. Whitten, and O. B. Toon, Stratospheric Aerosols: Observations and Theory, *Rev. Geophys. Space Sci.*, vol. 20, pp. 233, 1982.

W. G. Read, L. Froidevaux, and J. W. Waters Jet Propulsion Laboratory, California Institute of Technology, Pasadena, Ca. 91109.

(Received: February 2, 1993;
Accepted: March 1, 1993.)