Reports

Measurements of the Stratospheric Plume from the Mount St. Helens Eruption: Radioactivity and Chemical Composition

Abstract. *Gas measurements made in the stratospheric plume from the eruption of Mount St. Helens on 18 May 1980 were not consistent with a reported large injection of radon-222 into the atmosphere. No enrichment in the volatile element polonium was found in filter samples, and the ratio of polonium-210 to lead-210 was not different from background values. Data obtained with an experimental impactor, flown shortly after the eruption, showed an increase of 10^3 in the stratospheric number concentration of submicrometer sulfate particles compared to concentrations before the eruption.*

The High Altitude Sampling Program (HASP) of the U.S. Department of Energy provides data on the distribution of radioactivity and various nonnuclear compounds in samples of the upper atmosphere obtained from aircraft and balloons. In Project Airstream (*1*), the aircraft part of HASP, a WB-57F aircraft is used to sample in the Northern Hemisphere stratosphere and troposphere three times a year (April, July, and October). Data have been reported on the distribution of naturally occurring radionuclides (^{210}Pb, ^{7}Be), fission products (^{90}Sr, ^{137}Cs, ^{144}Ce, ^{95}Zr), and transuranics (^{239}Pu) (*2*); of trace gases such as CCl_3F, CCl_2F_2, CCl_4, N_2O, and SF_6 (*3, 4*); and of sulfate, nitrate, and ammonium ions (*5*). For these measurements two types of samples have been obtained.

A filter sampler containing IPC-1478 filter paper is inserted into the airstream to obtain aerosol samples for radioactivity and nonnuclear chemical analyses, and a high-pressure gas-sampling system (P-system) collects more than 2000 standard liters of air for gas analysis. On 13 May 1980, 5 days before the Mount St. Helens eruptions began, an experimental four-stage impactor called the Environmental Measurements Laboratory impactor (EMLI) was successfully tested and provided stratospheric samples for analysis by electron microscopy (*6*).

On 18 May 1980 the first eruption of Mount St. Helens injected large quantities of aerosols and gases into the atmosphere. On two separate flights, 50 and 75 hours after the first eruption, the WB-57F aircraft left Ellington Air Force Base, Houston, Texas, and attempted to intercept the volcanic plume in the upper troposphere and lower stratosphere as it moved across the United States. It was expected that measurements of ^{222}Rn and the daughters ^{210}Po and ^{210}Pb would show that large quantities of radon gas were injected into the atmosphere (*7*) and that ^{210}Po was enriched with respect to ^{210}Pb, as reported for other volcanoes (*8*). In addition, the experimental impactor, previously shown to collect particles with diameters as small as 0.05 μm, would provide information on the size and chemical composition of the aerosols from the eruption.

Pressurized gas samples were ana-

Table 1. Background and plume measurements on 13, 20, and 21 May 1980.

Altitude (km)	Location	Sulfate* (μg/m³ STP)	Ash* (μg/m³ STP)	^{210}Po* (fCi/m³ STP)	^{210}Pb* (fCi/m³ STP)	^{210}Po/^{210}Pb	^{222}Rn*,† (pCi/m³ STP)
			13 May 1980‡				
9.2	South Texas			1.2 ± 0.2	5.7 ± 0.3	0.21 ± 0.04	
12.2	South Texas			1.9 ± 0.2	4.8 ± 0.4	0.40 ± 0.05	
13.6	South Texas			3.7 ± 0.9	6.1 ± 0.8	0.61 ± 0.17	
15.2	South Texas			3.3 ± 0.2	5.2 ± 0.3	0.63 ± 0.05	
16.8	South Texas			3.7 ± 0.2	6.4 ± 0.4	0.58 ± 0.05	
19.2	South Texas			6.5 ± 0.2	6.5 ± 0.7	1.0 ± 0.1	
			20 May 1980§				
9.4	South-central Kansas	3.3 ± 0.7	47 ± 10	2.2 ± 0.2	4.4 ± 0.8	0.5 ± 0.1	8.7 ± 0.1
9.4–10.7	Central Kansas	1.8 ± 0.4	6.9 ± 1.0	2.9 ± 0.3	7.1 ± 0.5	0.4 ± 0.1	
12.4	Central Kansas	0.9 ± 0.2	<0.6	3.0 ± 0.3	6.7 ± 0.9	0.5 ± 0.1	0.30 ± 0.01
12.4	Southwest Kansas	1.0 ± 0.2	<0.6				
15.2	West Kansas	32 ± 6.0	710 ± 140	5.2 ± 0.4	8.4 ± 1.4	0.6 ± 0.1	1.1 ± 0.1
15.2	West Kansas–Nebraska	1.6 ± 0.3	<0.6	5.8 ± 0.6	4.4 ± 1.3	1.3 ± 0.4	
16.7	West Colorado	1.3 ± 0.3	<0.6	4.8 ± 0.6	6.9 ± 1.7	0.7 ± 0.2	BLD‖
16.7	West Colorado–Kansas	1.5 ± 0.3	<0.6	4.0 ± 0.9	9.1 ± 1.5	0.4 ± 0.1	
			21 May 1980¶				
15	Southeast Colorado	2.0 ± 0.4	<0.6	4.0 ± 0.3	7.2 ± 0.3	0.6 ± 0.1	0.62 ± 0.09
15	North-central Colorado	1.2 ± 0.2	<0.6	4.0 ± 0.7	5.9 ± 1.0	0.7 ± 0.2	0.37 ± 0.09
15	South-central Wyoming	2.7 ± 0.5	40 ± 8	4.6 ± 0.4	6.4 ± 0.9	0.7 ± 0.1	
18.2	South-central Wyoming	5.1 ± 1.0	47 ± 9	4.1 ± 0.9	6.8 ± 5.6	0.6 ± 0.5	0.54 ± 0.09
18.2	Northwest Colorado	40 ± 8.0	110 ± 22	5.2 ± 0.8	5.0 ± 1.9	1.0 ± 0.2	
18.2	Southwest Colorado	0.7 ± 0.1	15 ± 3.0	1.2 ± 0.3	7.8 ± 1.5	0.15 ± 0.09	0.47 ± 0.09
18.2	Northwest New Mexico	44 ± 9.0	37 ± 7.0				

*Mean ± standard deviation; STP, standard temperature and pressure. †All gas samples, except the sample from southeast Colorado on 21 May, spanned two filter samples. ‡Average tropopause height, 13.6 km. §Average tropopause height, 11.0 km. ‖Below limit of detection. ¶Average tropopause height, 12.5 km.

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lyzed for ^{222}Rn by techniques developed at the Environmental Measurements Laboratory (*9*); the lower limit of detection for these measurements was 0.27 pCi (0.6 dis/min). Filter samples were analyzed for ^{210}Po and ^{210}Pb as described by Harley (*10*); the lower limits of detection for these nuclides are 0.05 and 0.33 pCi, respectively.

Radiochemical results from the 13 May preeruption flight and the 20 and 21 May posteruption flights are shown in Table 1. The gas samples were collected over a longer period than the filter samples, and in most cases they overlap two filter samples. The sulfate and ash data (mass remaining after ashing) (*11*) are included to show which samples were taken within the plume. The overall accuracy of the concentration data is better than 20 percent (*1*). The error associated with the radionuclide data reflects the counting errors and filter blank variability.

In contrast to the results of another study (*7*), the radon analyses did not show any evidence of a large release of radon gas from Mount St. Helens. Only one of the eight whole air samples in Table 1 (the 15.2-km sample of 20 May) indicates a slightly elevated radon concentration coinciding with the plume. This value is within a factor of 2 of the concentration in the 15-km nonplume sample collected on 21 May and is in agreement with the 1962 profiles measured at the Environmental Measurements Laboratory (*12*) and the profiles published by Moore *et al.* (*13*). The vertical profiles are in reasonable agreement with the atmospheric radon model of Jacobi and André (*14*). The radon concentration measured in the plume in the troposphere (first sample of 20 May at 9.4 km) is similar to upper tropospheric data reported elsewhere (*8*).

Table 1 also indicates that the volcanic plume was not rich in ^{210}Po and ^{210}Pb. On the 20 May flight at 15.2 km over western Kansas, where the ash content of the filter reached 710 $\mu g/m^3$, the ^{210}Po and ^{210}Pb concentrations were 5.2 and 8.4 fCi/m^3, respectively. The concentration range from the preeruption flight included similar values. Positive verification that the plume was intercepted is give by the ash and sulfate concentrations from the 21 May flight (15 and 18.2 km). The ^{210}Po and ^{210}Pb concentrations do not differ from those in the preeruption or nonplume samples. Lambert *et al.* (*8*) concluded that the concentrations of radon daughters in the atmosphere are higher than would be expected from known rates of exhalation of radon by soils. They attributed this to volcanic emissions of radon and its daughters. The average $^{210}Po/^{210}Pb$ ratio in the present samples was 0.6, and the highest value (1.3 ± 0.4) was measured outside the plume.

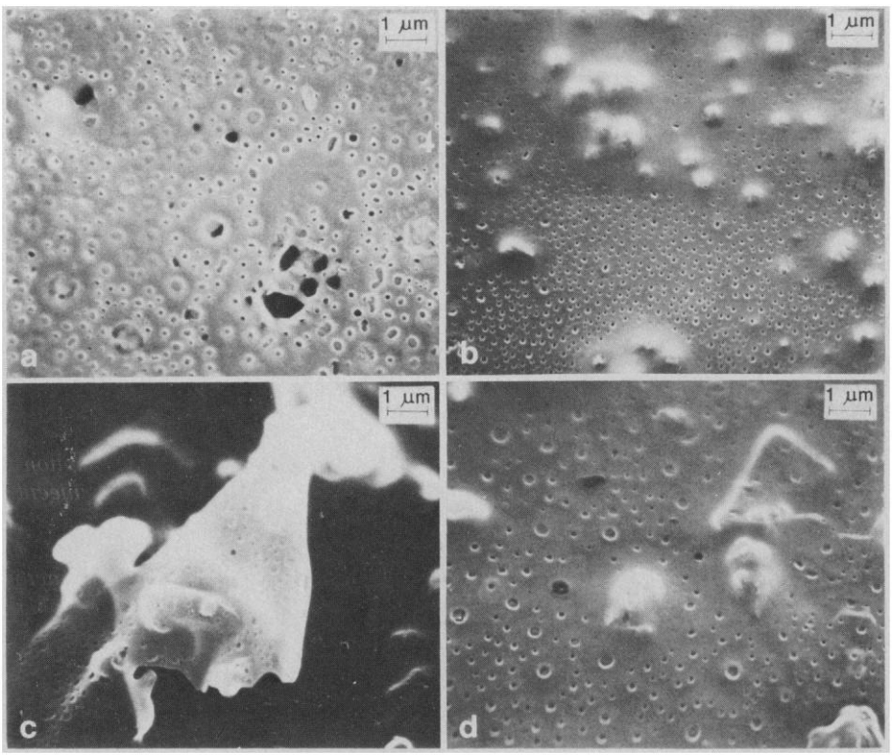

Fig. 1. Electron micrographs of (a and b) preeruption and (c and d) posteruption stratospheric aerosols. (a) Sulfuric acid. (b) Pitting left by volatile sulfur particles; larger particles are stable in the scanning electron microscope and contain sulfur. (c) Ash particles showing extensive pitting. (d) Pitting left by volatile sulfur particles; larger particles are ash and contain sulfur.

A stratospheric sample collected with the EMLI on 13 May at 19.2 km (28°30′N, 95°30′W) provided information on the aerosol composition before the eruption. On 21 May at 18.2 km we intercepted the plume from the eruption (42°15′N, 110°W) and collected sufficient samples for analysis by electron microscopy and proton-induced x-ray emission analysis (PIXE). All four stages of the posteruption sample collected large particles (*15*). Since the first stage does not contain the kinds of particles found on the other stages, the particles on the latter stages are not attributed to particle bounce. The average particle characteristics (morphology and elemental composition) were different from stage to stage. The number concentrations of large particles were: stage 1, 30 cm^{-3}; stage 2, 50 cm^{-3}; stage 3, not measurable because of stage overloading; and stage 4, 25 cm^{-3}.

The first stage was dominated by ash particles with smooth protrusions. The second stage contained large flaky ash particles with extensively pitted surfaces (Fig. 1c); the surface pitting indicates that the ash provided sites for the growth of volatile sulfur particles. The third stage was overloaded with ash particles which obscured a major part of the Mylar surface; volatile material associated with these particles is indicated by their surface pitting. Black areas, which were found only on this stage, are unexplained (Fig. 1d). It appears as though volatile material released from the particles did not completely evaporate in the electron beam. The last stage contained particles composed predominantly of silicon and iron.

In addition to ash, the eruption increased the number of volatile small particles in the stratosphere (*16*) by three orders of magnitude (from 10 cm^{-3} on the preeruption flight to 10^4 cm^{-3}), as indicated by pitting of the silicone oil film caused by evaporation of volatile sulfur compounds (compare b and d, Fig. 1). At such small sizes, present analytical techniques do not allow the differentiation of sulfate compounds. Laboratory studies of ammonium sulfate and sulfuric acid aerosols showed similar pitting patterns in the silicone film. At present, the small particles observed are identified as volatile sulfur compounds. The preeruption sample contained large liquid droplets (Fig. 1a) that evaporated

Table 2. Proton-induced x-ray emission analysis of a posteruption sample collected on 21 May 1980 between 39°04'N, 108°48'W and 37°20'N, 108°32'W at 18.2 km.

Element	Concentration ($\mu g/m^3$ STP)*			
	Stage 1	Stage 2†	Stage 3	Stage 4
Sodium	1.1 ± 0.4			
Aluminum	0.9 ± 0.4	0.79		
Silicon	4.2 ± 0.4	0.74	7 ± 2‡	
Sulfur	0.26 ± 0.01	0.64	1.7 ± 0.6	0.9 ± 0.6
Potassium	0.24 ± 0.02		0.2†	0.23 ± 0.07
Calcium	0.35 ± 0.11	0.36	0.4 ± 0.2	0.22 ± 0.07
Titanium	0.03 ± 0			0.11†
Iron	0.35 ± 0.04	0.14	0.13 ± 0	0.4 ± 0.2
Copper	0.02 ± 0			
Rubidium	0.04 ± 0.02			

*Mean ± deviation from the mean of duplicate analyses. †Based on a single analysis. ‡Unexplained high concentration.

in the electron microscope, leaving overlapping satellite patterns around a residue. This pattern is characteristic of sulfuric acid (*17*). Similar patterns were seen in the plume sample. The observation of large numbers of volatile small particles after the eruption may be explained by either direct injection of sulfate particles during the eruptive phase of the volcano or gas-to-particle conversion during plume transit.

Another sample from the EMLI was used for PIXE analysis; the sample was collected at the same altitude (18.2 km) as the sample analyzed by microscopy and within 5° latitude of that sample. An independent filter sampler provided measurements of the ash and sulfate concentration (Table 1) in the plume near where the PIXE sample was collected.

Table 2 shows the elemental analysis of the aerosols collected on the four impactor stages from the volcanic plume. The errors shown are deviations from the means of duplicate analyses and reflect interference from the background silicone oil and Mylar film. The elemental composition in the plume is highly variable (*11*), and it is not useful to compare it with other posteruption measurements. The peak in the sulfur concentration on the third stage indicates that different mechanisms are involved in the production of sulfur particles. Not all parts of the plume were measured within 1 day of the eruption, but some data indicate absence of sulfuric acid in the early stages (*18*).

The high concentration of iron on stage 4 (Table 2) is consistent with microscopic data. Apparently, fractionation of the iron occurred in the early stages of the eruption. Microscopic examination of the particles from stage 4 shows that they are flaky, predominantly silicon and iron, and not spherical as would be expected if they were meteoritic.

Except for the high silicon concentration on stage 3, which is unexplained, most of the ash mass was found on stage 1, consistent with the impactor collecting the largest particles on the first stage. In the filter collections from the flight of 21 May the sulfate concentration varied from 0.7 to 44 $\mu g/m^3$ and the ash concentration varied from 15 to 110 $\mu m/m^3$ (Table 1). The total ash and sulfate concentrations estimated from the elemental concentrations measured on the impactor (Table 2) and from the ash composition in the filters (*7*) are 49 ± 23 and 11 ± 4 $\mu g/m^3$, respectively. These concentrations do not differ from those found in samples collected independently (Table 1).

The absence of chloride on the impactor samples is unusual, since large quantities were detected in the first 24 hours in the plume (*19*). This could reflect a significant loss of volatile chloride to the atmosphere during transit. Also, because there were large inhomogeneities in the composition of the plume, one must be cautious in comparing early and late samples.

The eruption of Mount St. Helens injected large quantities of ash and gases into the atmosphere. Extremely high but variable quantities of ash were measured in different parts of the spreading plume after the initial eruption. The elemental composition of the ash varied by factors of ~2, depending on where in the plume measurements were made. The ash provided excellent surfaces for gas-to-particle conversion and formation of sulfuric acid. From the samples collected it is not possible to determine whether this conversion occurred during the initial eruption or at a later time. The low $^{210}Po/^{210}Pb$ ratios in the plume indicate that the ash from Mount St. Helens was not enriched with respect to polonium, as previously reported (*7*). No excess ^{222}Rn was found in the plume on two successive days, and therefore no large injection of radon into the atmosphere from the volcanic eruption is indicated.

R. Leifer, L. Hinchliffe
I. Fisenne, H. Franklin
E. Knutson, M. Olden
Environmental Measurements Laboratory, Department of Energy, New York 10014
W. Sedlacek, E. Mroz
Los Alamos National Laboratory, Los Alamos, New Mexico
T. Cahill
Crocker Nuclear Laboratory, University of California, Davis 95616

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6. To efficiently collect stratospheric samples on the EMLI, a Mylar film covering a rotating drum is coated with Dow silicone oil 200. Laboratory studies have shown that the diameters at which 50 percent of the particles are collected on the four stages of the EMLI are stage 1, 0.13 μm; stage 2, 0.1 μm; stage 3, 0.07 μm; and stage 4, 0.05 μm. These diameters are referenced to a laboratory-simulated atmosphere (density equivalent altitude, 12 km; ambient flow rate, 7 liter/min; and sulfuric acid and ammonium sulfate aerosols). The actual cut diameters vary with altitude, shifting to slightly smaller values at higher altitudes. A scanning electron microscope with an x-ray spectrometer was used to examine the particles. Substrate preparation was carried out in a class-100 laminar flow bench, providing extremely clean surfaces for aerosol collection. Microscopic examination of areas away from the impactor zone show no contamination. Part of the sampling system for the impactor consists of a large bottle of specially prepared nitrogen, free of ammonia, that is used to purge all sampling lines at the ground and during aircraft ascent and descent. After the last sample has been collected, the nitrogen is used to pressurize the impactor and keep sampling lines free of contamination. Two electrically operated ball valves are closed to protect the stratospheric collections. At the laboratory the samples are prepared for microscopic examination and PIXE analysis.
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9. Air from the pressurized P-system tanks was passed through sulfuric acid and triethanolamine solutions into a 180-g charcoal trap (at dry ice temperature) at a flow rate of 10 liter/min. The volume of air collected (500 to 1000 liters) in the trap was measured with a wet test meter. The charcoal traps were heated to 350°C for 1 hour and the ^{222}Rn was transferred into the EML 2-liter pulse–ionization chambers. The ^{222}Rn contamination from the steel tanks was determined 1 month after the sample measurements.
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12. The ^{222}Rn concentrations in the southwestern United States during spring 1962 ranged from 5.4 to 6.2 pCi/m^3 at 9.4 km, 0.1 to 0.6 pCi/m^3 at 12.5 km, 0.03 to 0.4 pCi/m^3 at 15.2 km, and 0.1 to 1.3 pCi/m^3 at 18.3 km.
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15. Large particles, as defined for this report, have diameters > 0.2 μm.
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20. We would like to thank the NASA, Johnson Space Center pilots, scientific equipment operators, and ground support personnel who made the measurements of these data possible. NASA/JSC operates the aircraft for the Department of Energy, Office of Health and Environment. We would like to thank our colleagues Dr. K. Tu, V. Negro, and S. F. Guggenheim for their work on the impactors and P. Perry for her help in the radiochemical analysis. We would also like to thank B. Vaughan for her help in preparing the preliminary manuscript and N. Chieco for the editing and typing of the final manuscript.

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Measurement of Solar Radius Changes

Abstract. *Photoelectric solar radius measurements since 1974 at Mount Wilson show no change in the solar radius, with a limit of about 0.1 arc second (1 standard deviation), over the interval. The limit is set by residual systematic effects.*

Several studies of possible secular and periodic variations of the solar radius have yielded conflicting results (*1–7*). The data analyzed have been almost entirely historic measures from meridian circles, heliometers, transits of Mercury, and eclipse timings. All of these data are based on visual estimates. None of the daily measurement programs are currently obtaining data. We present here results of daily photometric measurements of the solar radius over the past 7 years at the Mount Wilson Observatory.

Measurement technique. One part of the reduction of the Mount Wilson daily full-disk magnetograms is a determination of the radius of the solar image. Although high precision is not required for the magnetic analysis, the formal error of the daily values is about 0.1 arc second. These data have been taken for a number of years, and we have examined them for a secular variation in the solar radius. Our measurements are not absolute, as the focal length of the telescope is not known to sufficient precision; a nominal radius of 960.0 arc seconds is used to convert our scale to arc seconds.

The Mount Wilson 150-foot Solar Tower Telescope and magnetograph have been described in detail by Howard (*8–10*). The daily observation is a boustrophedonic scan of the full disk in the wings of the 5250.2-Å FeI line. The objective, polarizing optics, and spectrograph aperture (12.5–arc second square) are fixed. Scanning is done by moving the guider sensors located at the prime focus image; the guider error signals drive the second flat mirror, moving the solar image across the aperture. The (x, y) position of the guider sensors is recorded with each data sample. One x, y encoder unit corresponds to 0.28 arc second. Scan lines are aligned perpendicular to the solar pole. The observation is started in the solar north or south by random selection. Each scan line extends off the disk and onto the sky at both ends. There are about 150 scan lines in each observation.

The average intensity of the disk is computed from the data, and a linear trend of intensity as a function of time is removed from the data. This linear trend averages $\lesssim 4$ percent, so systematic nonlinearity of our photometric scale may be ignored. The limb is defined as the steepest point in the limb darkening curve; at the wavelength used, this corresponds to a contour level of measured intensity of 0.25 of the disk average intensity.

The (x, y) positions of the limbs in each scan line are found by linear interpolation of the intensities. The equation $R = [(x - X_0)^2 + (y - Y_0)^2]^{1/2}$ is then solved by a least-squares method for the center of the image (X_0, Y_0) and the radius R. Separate solutions are obtained for the eastward and westward scan lines to allow for backlash in the drive mechanisms. The radius measure R_0 is the average of these two solutions. The radius solutions are done iteratively, with points far from the derived limb excluded. This rejects data where the limb is obscured by the guider sensors or a passing cloud. Typically, 20 points out of 150 are so rejected. The formal probable error of the least-squares solutions is typically 0.06 arc second, with little variation about that value. Some observations are taken under poor photometric conditions, with variable sky transparency or scattering. On these days the formal probable error is much larger than normal, ≥ 0.12 arc second. We rejected 133 days on this basis. No bias is introduced into the data set by this criterion because the formal probable error is uncorrelated with the raw radius measures or the radius residuals, and the actual errors are much larger than the formal errors and set by effects other than the internal photometric consistency of an observation.

Once the limb position is found, the scattered light of the optics plus atmosphere is determined. Scattered light is defined as the average intensity measured at a distance ($2.8 + A/\sqrt{2}$ arc seconds) outside the limb, where A is the aperture size; this guarantees that all parts of the aperture, including the corners, are off the limb, with an arbitrary 2.8–arc second allowance for seeing ripple and mechanical jitter.

Figure 1 shows the raw radius measures as a function of time. In the interval 26 July 1974 to 1 April 1981 there are 1412 measurements.

Reduction procedure. Sources of variation in the measured image size, aside

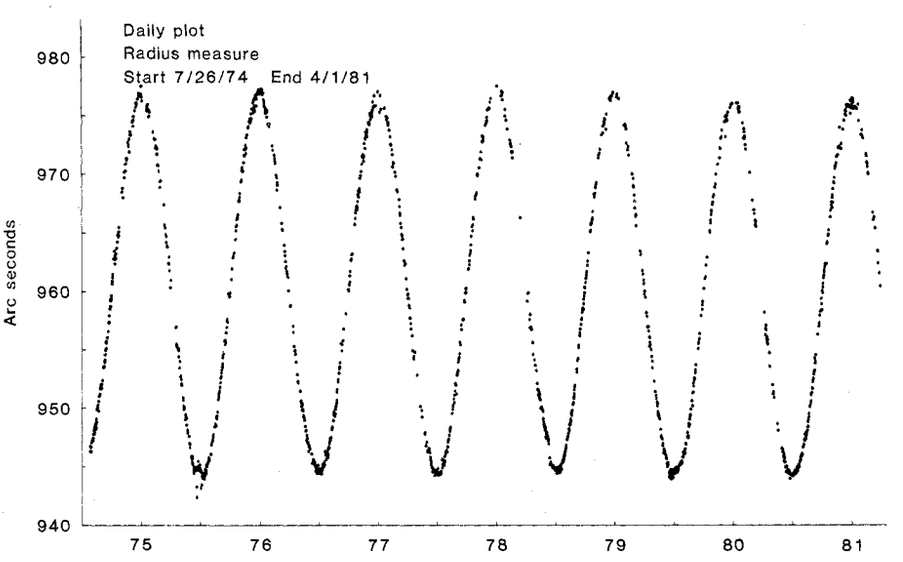

Fig. 1. Solar radius observed with the Mount Wilson magnetograph. The arc second scale is not absolute; a mean radius of 960.0 arc seconds is assumed. There are 1412 observations in this interval.

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