Reports

Volcanically Related Secular Trends in Atmospheric Transmission at Mauna Loa Observatory, Hawaii

Abstract. *Twenty years of atmospheric transmission data from Mauna Loa Observatory show secular decreases at irregular intervals. In addition, a regular annual variation is present during unperturbed as well as perturbed periods. These variations in transmission can be measured to a few tenths of a percent from the data record. Transient decreases in transmission are strongly correlated with explosive volcanic eruptions that inject effluent into the stratosphere. Recovery from these ejections takes as much as 8 years and the recovery curve is linear. Observations in 1977 at Mauna Loa show that, for the first time since the Mount Agung eruption in 1963, the atmospheric transmission of direct-incidence solar irradiation at Mauna Loa returned to values measured in 1958 to 1962.*

The direct component of solar irradiance has been measured at Mauna Loa Observatory, Hawaii (19.53°N, 155.58°W; elevation, 3.5 km), since 1958 (*1*). From these measurements daily determinations of atmospheric transmission have been made for clear sky conditions over a 20-year period. Ellis and Pueschel (*2*) reported long-term secular changes in atmospheric transmission observed at Mauna Loa that were unrelated to anthropogenic sources. They also deduced that the short-term fluctuations in the record were the result of a natural annual cycle in tropospheric aerosols. Other reports have documented such secular changes in atmospheric turbidity on a global scale (*3–7*). Atmospheric transmission data are important in climatology research. For example, attempts by climatologists to link worldwide atmospheric cooling to secular decreases in atmospheric transmission require accurate long-term records of transmission data such as those obtained at baseline stations. It has been known that a decrease in global temperature could result from an increase in the concentration of aerosols in the atmosphere (*8–11*).

We report here on a more recent analysis of the solar transmission record at Mauna Loa Observatory that shows several transient variations in transmission. The transmission record is compared with the record of episodic volcanic activity after the eruption of Mount Agung.

Solar irradiance was measured over a broad spectral band, using a normal-incidence pyrheliometer with a quartz window transmitting between 2,000 and ≥30,000 Å (*12*). A possible source of uncertainty is the variation of the transmittance of the quartz window with ambient temperature changes under field observation conditions. Another source of uncertainty is instrumental errors and drift, which is particularly problematic because different pyrheliometers were used for the measurements over an extended period.

To minimize these uncertainties, atmospheric transmission factors (q) for the period of record were obtained as described in (*2*). From Bouquer's law, $q^n = I_n/I_0$, where I_0 is the solar constant and n is the secant of the zenith angle. Measurements of n were made at unit intervals (from 5 to 2) during the morning and afternoon. We then computed two daily values of

$$\bar{q} = \frac{1}{3}\left(\frac{I_5}{I_4} + \frac{I_4}{I_3} + \frac{I_3}{I_2}\right)$$

where the exponent of $\bar{q}$ is always unity and the subscripts of I are the secants of the zenith angle. These values represent the transparency of the atmosphere in the morning and in the afternoon of each clear day. The computation of $\bar{q}$ by this method provides a way of getting around the problem of drift in the absolute calibration of the instruments used (*7*). The method also has the advantages that the values of $\bar{q}$ obtained are more representative of the average transmission than are values obtained from any one pair of secant angles, and that it compensates for seasonal changes in the sun-earth distance. Our data consisted of all measurements made on clear days for the period of record.

Monthly averages of $\bar{q}$ at Mauna Loa for 1958 through 1977 are shown in Fig. 1. The scatter (standard deviation) is 1 percent for the morning values and slightly higher for the afternoon values. Secular changes in transmission are evident beginning in 1963 with the eruption of Mount Agung.

The afternoon data are fewer and less

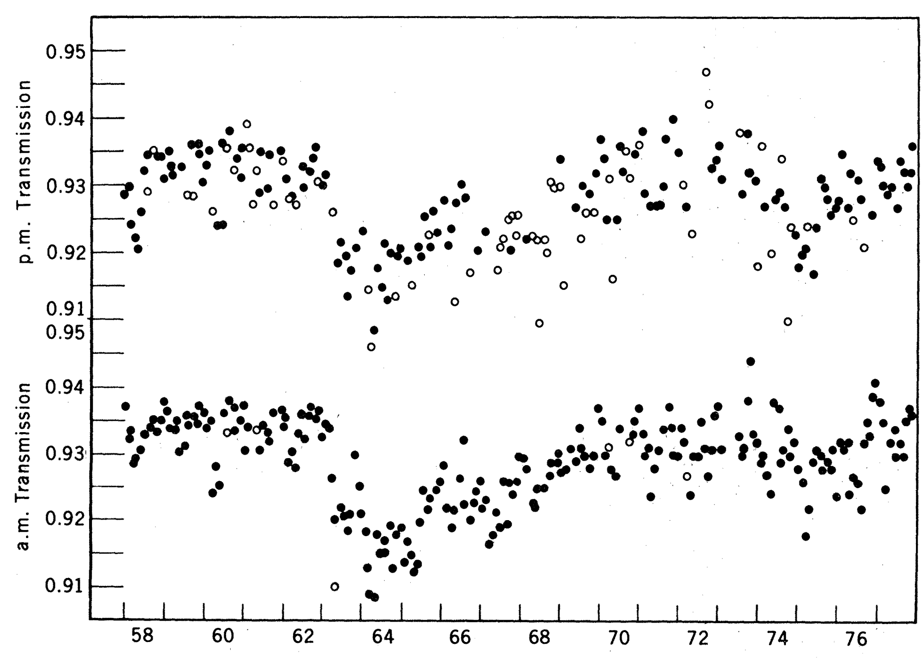

Fig. 1. Plot of monthly averages of transmission of normal-incidence solar irradiation measured at Mauna Loa Observatory. Secular trends are evident and are larger than the normal background scatter of the data. Open circles indicate monthly avarages obtained from five or fewer transmission values.

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reliable because periodic influxes of moisture and more turbid air from levels below the observatory result in maximum cloudiness in the afternoon. In the morning, however, it is common to have uniform conditions and air with a low moisture content (less than 2.0 mm of precipitable H_2O on the average) (*13*), hence we used only the morning observations in the analysis.

To minimize short-term variations in transmission, we computed 6- and 12-month running averages of the data. These are shown in Fig. 2, where the secular trends are discernible and a regular annual period is seen in the 6-month average. Figure 2 also shows all documented explosive volcanic eruptions (from 1958 to the present) where the volcanic dust cloud reached the stratosphere ($\geq$10 km above mean sea level). The mean lifetime of tropospheric aerosols is on the order of only a few days to a few weeks (*14*); therefore, stratospheric penetration was considered requisite for global transport of aerosols with atmospheric lifetimes in excess of several months (*10*).

The record of explosive eruptions since 1958 shows that volcanoes whose effluent reached the stratosphere are located in three major latitude bands: 14°N to 14°S, 50° to 70°N, and 50° to 70°S. In addition, there were four periods of intense activity: the Agung period, 1962 to 1964; the Awu period, 1965 to 1966; the Hekla period, 1970 to 1971; and the De Fuego period, 1974 to 1976. Transient decreases in atmospheric transmission at Mauna Loa occurred near all periods with the exception of the Hekla period, following the eruptions with time delays on the order of 1 week to several months. The time delays seem to depend on the magnitudes and locations of the eruptions and are doubtless related to the zonal separation between the volcanic eruption and Mauna Loa and to atmospheric transport mechanisms (*3*).

Table 1. Changes in atmospheric transparency, Mauna Loa Observatory.

Period	Transmission (%)	Optical depth
Transient changes		
Pre-Agung (1958–1962) to:		
Agung-Awu (1963–1968)	−2.0	−0.032
Recovery period (1970–1973)	−0.20	−0.004
De Fuego (1974–1976)	−0.75	−0.012
*Annual variations: maximum to minimum**		
Total record (1958–1977)	−0.73	−0.012
Undisturbed period, pre-Agung (1958–1962)	−0.60	−0.010
Disturbed period, Agung and Awu (1963–1968)	−0.91	−0.015
Recovery period, Hekla (1970–1973)	−1.10	−0.018

*Maximum, December and January; minimum, April and May.

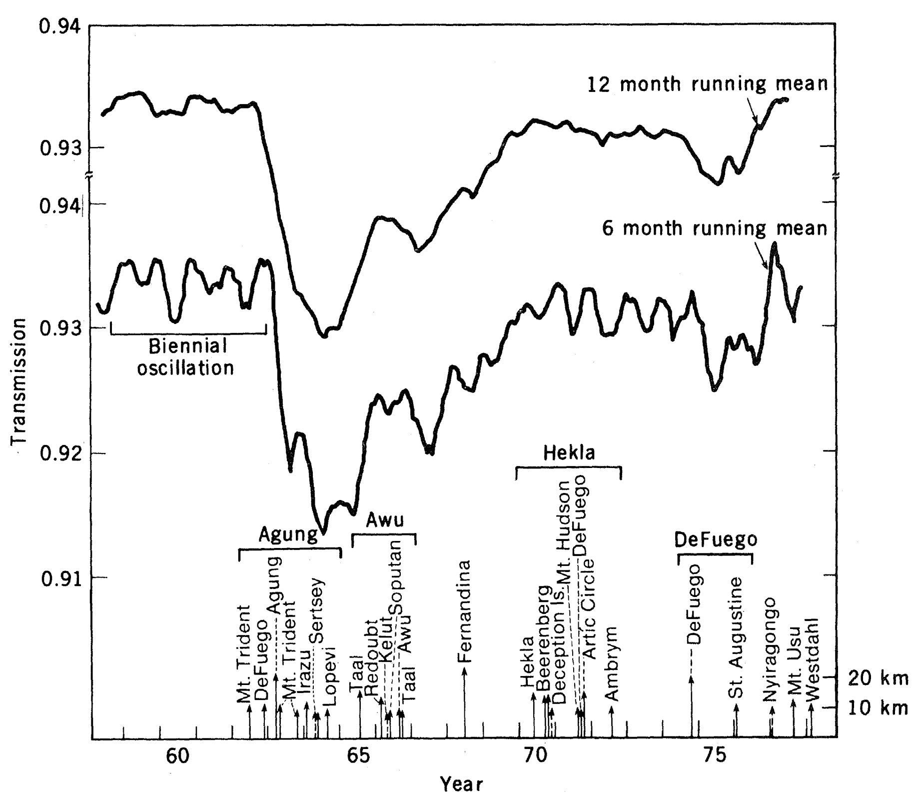

Fig. 2. Running means of morning transmission values at Mauna Loa. Volcanic eruptions with dust clouds reaching stratospheric heights are indicated on the abscissa. The annual cycles in transmission as well as secular trends can be seen. Note that transmissions measured in 1976 to 1977 are as high as those in the pre-Agung period. Three episodic decreases occur in 1963 to 1964, 1966 to 1967, and 1975 to 1976. A gradual decrease from 1970 to 1974 is also evident.

The largest decrease in transmission in Figs. 1 and 2 is associated with the Agung event in 1963. It is closely followed in 1966 to 1967 by another, smaller decrease in transmission. Recovery from these two major decreases is nearly complete 10 years after the initial event when a third major decrease occurs that is associated with the De Fuego volcanic eruption in 1974.

During the Hekla period only De Fuego was active in the equatorial latitude band; all other eruptions in this period were in the high latitude bands. (In the other three periods of activity only the Mount Trident, Sertsey, and Redoubt eruptions were outside the equatorial latitude band.) During the Hekla period only a general monotonic decrease in transmission is seen. However, the annual variations for this period are the largest in the record (see Table 1).

The magnitudes of the atmospheric transmission changes measured at Mauna Loa are summarized in Table 1. The transient decrease associated with the Agung eruption is at least twice any annual variation shown. Note that the De Fuego decrease is of the same magnitude as the annual variation for the undisturbed period 1958 to 1962. Since Agung and the subsequent eruptions, the transmission values at Mauna Loa did not return to the pre-Agung (1958 to 1962) level until 1977 (Fig. 2).

Over the entire range of the record, there is a regular annual cycle in transmission at Mauna Loa with the minimum during April to June and the maximum during November to February (Fig. 3). The average magnitude of the annual cycle for 1958 to 1962 is 0.60 percent, the smallest observed (Table 1). The amplitude of the annual cycle increases during periods of aerosol increase in the stratosphere. Similar annual cycles in atmospheric transmission have been reported in the literature and have been attributed to the natural variation of background tropospheric aerosols (*2*); vertical transport and advection of natural and anthropogenic aerosols with greater instability and convection during summer months, particularly over continents (*15*); and annual migration of trajectories of stratospheric aerosol bands (*16*). We note that there is no change in the phase of the annual variation at Mauna Loa before and after a volcanic eruption.

A biennial periodicity in transmission is evident in the record for 1958 to 1962, when no explosive volcanic activity was reported (Fig. 2). Over this 5-year period, this oscillation is in phase with the stratospheric wind direction (*17*), and a westerly flow in the stratosphere over Mauna Loa is coincident with a larger decrease in transmission in summer. After Agung, the biennial variation in transmission is either nonexistent or masked by the larger episodic variations that followed.

Transmission recovery rates after the initial injections of volcanic effluent into the stratosphere are 0.61 percent per year after the Agung period, 0.33 percent per year after the Awu period, and 0.40 percent per year after the De Fuego period. The initial recovery rate after an episode seems to be more linear than exponential, whereas an exponential rate might be expected if only gravitational settling were acting on an originally Junge aerosol distribution (*18*). If additional aerosols are created sometime later from the gaseous effluent that accompanies the initial influx of aerosols into the stratosphere, they will undoubtedly affect the recovery times, as will the dynamics of natural cleansing processes between the troposphere and the stratosphere (*19, 20*). Complete recovery of atmospheric transmissions after volcanic eruptions, in times ranging from months to a few years, have been reported by others (*7, 21*).

Comparison of the transmission at the beginning (1958) and at the end (1977) of the Mauna Loa record shows no obvious long-term trend. Recovery from all volcanic eruptions in the 20-year period appears to be complete. However, a long-term linear decrease of 0.2 percent can be inferred from 1958 to 1976, excluding the transient decreases in the interim. Although such a small decrease in transmission is within the noise of the data, we attempted to account for it by computing the decrease in transmission that would be caused by a gradual increase in total atmospheric ozone from 270 to 280 m-atm-cm over Mauna Loa (*15, 22*). The calculated decrease amounted to only 0.03 percent and would not be enough to account for the total decrease, if any.

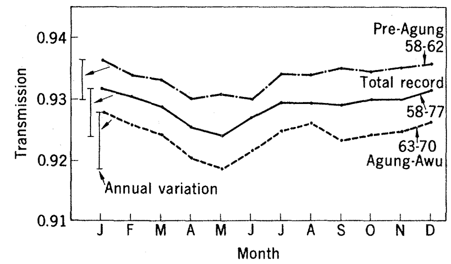

Fig. 3. Annual cycles in transmission for three periods at Mauna Loa. All the annual cycles are in phase. The magnitude of the annual cycle is largest for the period 1963 to 1970 and smallest for the pre-Agung period.

Annual, biennial, and transient variations in transmission have been observed in the continuous Mauna Loa record since 1958. The transient changes appear to be linked to episodic stratospheric aerosol increases due to injections of volcanic effluent into the stratosphere by sufficiently strong eruptions. The effect is greatest with eruptions originating near the equator, where the vertical motion and the subtropical jet and strong zonal stratospheric winds are conducive to rapid transport of aerosols over the equatorial belt. Volcanic injections that occurred north or south of 50° latitude can be detected at Mauna Loa, but are seen as gradual rather than sharp decreases in the transmission and as increases in the magnitude of the annual cycle.

A biennial cycle in the first 5 years of the record and an annual cycle in the entire record strongly suggest natural variations in atmospheric transmission, independent of the transient variations due to volcanic episodes. The cause or causes of these cycles observed at a remote site such as Mauna Loa Observatory are still not fully understood.

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Atmospheric Reactions of Polycyclic Aromatic Hydrocarbons: Facile Formation of Mutagenic Nitro Derivatives

Abstract. *Directly active mutagens are formed on exposure of the promutagen benzo*[a]*pyrene to gaseous pollutants in smog. In simulated atmospheres containing 1 part per million nitrogen dioxide and traces of nitric acid, directly mutagenic nitro derivatives are readily formed from both benzo*[a]*pyrene and perylene, a nonmutagen in the Ames reversion assay. Possible formation of direct mutagens by such reactions on sample collection filters, in exhaust effluents, and in the atmosphere should be recognized.*

The carcinogenicity of atmospheric organic particulate matter in experimental animals has been known for more than 30 years (*1, 2*). It has generally been attributed to the presence in polluted air of benzo[*a*]pyrene (BP) and other polycyclic aromatic hydrocarbons (PAH) as well as their azaheterocyclic analogs (*3, 4*). Several investigators, however, have noted that the carcinogenicity of both urban air and exhaust particulates from spark-ignition engines could not be ac-

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