Sun photometer and lidar measurements of the plume from the Hawaii Kilauea Volcano Pu’u O’o vent: Aerosol flux and SO2 lifetime

John N. Porter, Keith A. Horton, Peter J. Mouginiis-Mark, Barry Lienert, Shiv K. Sharma, and Eric Lau
Hawaii Institute of Geophysics and Planetology, University of Hawaii, USA

A. Jeff Sutton and Tamar Elias
U.S. Geological Survey, Hawaiian Volcano Observatory, Hawai’i National Park, USA

Clive Oppenheimer
Department of Geography, University of Cambridge, UK

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0094-8226/02/2002GL014744.00

Received 16 January 2002; revised 4 March 2002; accepted 7 March 2002; published 23 August 2002.

1. Introduction

[2] Hawaii’s Kilauea Volcano has been in semi-continuous eruption since January 1983, consisting of 55 episodes to date, dominated by tube-fed pahoehoe lava [Hederer et al., 1998]. Since the eruption began, Pu’u O’o vent has been the main source of SO2 gas release, with much lower emission rates from the summit caldera [Sutton et al., 2001]. In the atmosphere, the SO2 is oxidized and converted to sulfuric acid aerosols through reactions with OH radical and H2O in clear sky and cloud reactions [Finlayson-Pitts and Pitts, 1986]. The aerosol and gas mixture comprises a hazy form of air pollution, locally referred to as volcanic smog, or vog [Sutton et al., 1997]. Once formed, in situ measurements suggest the vog aerosols reach an equilibrium accumulation mode size distribution with a peak dry mass distribution from 0.2–0.4 μm diameter [Porter and Clarke, 1997]. Filter samples (Antony Clarke, University of Hawaii, personal communication, 2001) and temperature volatility tests have shown the vog aerosols are typically composed of sulfuric acid with little neutralization.

[3] Measurements of volcanic plume sulfur gas and aerosol emission rates can be based on either in situ [Hobbs et al., 1982; Stith et al., 1987; McGee and Gerlach, 1998] or remote sensing techniques [Andres et al., 1989; Realmuto et al., 1997]. A widely accepted technique to measure SO2 column abundance is by correlation spectrometer (COSPEC), which employs spectral absorption features of SO2 gas [Rose et al., 1986; Sutton et al., 2001]. Lidars have also been used to image plume shape [Casadevall et al., 1984; Hobbs et al., 1982]. Sun photometer measurements have also been used to derive volcanic aerosol size information [Watson and Oppenheimer, 2000, 2001]. Here we combine three remote sensing techniques, aerosol sun photometry, lidar, and COSPEC measurements to estimate the total oxidized sulfur emissions from the Pu’u O’o plume and the SO2 lifetime.

2. Sun Photometer and Lidar Aerosol Measurements

[4] On August 17, 2001, vehicle-based sun photometer and lidar measurements were made under the Pu’u O’o plume along the Chain of Craters Road within Hawai’i Volcanoes National Park. Sun photometer measurements were made while the vehicle was stationary at different points under the plume. Lidar measurements were made continuously. A portable GPS system and computer recorded time and position of the measurements. The sun photometer used here was a five channel (380, 440, 500, 675, 870 nm) handheld Microtops. The average aerosol optical depths in the volcano plume were 0.38, 0.328, 0.312, 0.214, 0.185 at 380, 440, 500, 675, 870 nm. Calibration for this system is maintained using the Langley plot measurements at the Mauna Loa Observatory. The major errors in making sun photometer measurements are due to pointing, calibration and to a lesser extent sensor thermal drift. Based on theoretical and experimental studies, the error in the aerosol optical depth measurements is expected to be less than 0.01 [Porter et al., 2001]. Figure 1 shows the average aerosol optical depth (at 500 nm) measured at each location along the Chain of Craters road during the third pass. Figure 1 also shows the location of the plume on January 7, 2001, when the Landsat 7 image was taken.

[5] Figure 2 shows a range of lognormal aerosol size distributions, which have spectral scattering coefficients (from Mie theory) that fit the measured aerosol optical depths within their uncertainty. Although a more elegant approach is certainly possible [Lienert et al., 2001], here we have simply tested a range of different size distributions with geometric mean diameters (0.02–0.55 μm) and standard deviations (1.45–3.0) which extend well past those that best fit the data. A real index of refraction of 1.35 with no absorption was assumed here. The uncertainty in the aerosol...
optical depth measurements and the limited range of wavelengths prevents us from further defining the size distribution.

During each pass, lidar measurements were obtained (zenith angle of \( \approx 38^\circ \)) through the vehicle open door (see Figure 3). The custom lidar system used for this experiment was a co-axial 12.7 cm telescope system using a 15 mJ pulsed frequency doubled (532 nm) Nd-YAG laser with 20 Hz pulse rate. A photomultiplier tube detector equipped with a custom log-amp yielded signal from 90-m out to 3-km. In order to avoid damage to the detector from bright sunlight, we always pointed the lidar away from the sun. The inversion of the lidar data is based on a forward stepping approach similar to the one used by Porter et al. [2000] but with different constraints. Here the lidar calibration (which is largely unknown) is adjusted so that the largest lidar integrated optical depths are in agreement with the largest sun photometer measured aerosol optical depths. For the lidar inversion, we also assume the aerosols have no absorption and have an aerosol phase function value (at 180° scattering angle) of 0.3 based on Mie calculations from existing aerosol models of the volcanic aerosol (vog) [Porter and Clarke, 1997]. Based on the lidar measurements, it was seen that the majority of the volcano plume was below 500 m height but was irregularly shaped possibly due to atmospheric turbulence.

### 3. Aerosol Flux Rates

In order to estimate the flux of dry sulfuric acid aerosol we follow equation (1),

\[
\text{Flux Rate} = \left( \frac{\text{Wind Speed}}{\gamma} \right) \sum_{i=1}^{n} \left[ w_i \tau_i \right]
\]

where \( \tau_i \) is the aerosol optical depth for interval \( i \) across the plume, \( w_i \) is the effective width for that interval and \( \gamma \) is the aerosol mass scattering efficiency (in \( \text{m}^2/\text{g} \)) (discussed below) at the same wavelength as the aerosol optical depth being used (500 nm in this case). An aerosol mass scattering coefficient of 7.7 \( \text{m}^2/\text{g} \) was used for these calculations. This value was based on the average of Mie theory calculations from the size distributions shown in Figure 2 with a standard deviation of 2.4 \( \text{m}^2/\text{g} \). In obtaining this value we have assumed the hygroscopic aerosols have a water uptake.

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**Figure 1.** Landsat 7 satellite image collected on January 7, 2001 (at approximately 10:30 am local time) combined with a digital elevation map showing the location of the Chain of Craters road and the Pu‘u O‘o vent. Vertical stems show the aerosol optical depths (at 500 nm) measured at each point along the road on August 17, 2001. The points labeled A and B are for reference in Figure 4.

**Figure 2.** Aerosol size distributions inverted from the aerosol optical depths measurements.

**Figure 3.** Aerosol scattering coefficient (at 500 nm) derived from the lidar while passing under the plume during the third pass. The log of the aerosol scattering coefficient is shown on the right. The data are shown at the height from the surface.
Aerosol optical depths (at 500 nm) plotted along a line perpendicular to the volcano plume. The x symbols show measurements made on the upper part of the road while o symbols show the measurements made on the lower part of the road. The plume perpendicular angle was chosen to produce the best overlap. The symbols A and B show the position of the measurements on Figures 1 and 4.

that is slightly less than sulfuric acid [Tang, 1980] due to the presence of small amounts of ammonium [Porter and Clarke, 1997]. An average relative humidity of 65% was assumed for the lowest 500 m based on the HiLO sounding at 2 p.m. Although the HiLO sounding is ~30 km away, the cloud base predicted from parcel theory from the sounding data was in good agreement with cloud bases observed by lidar, suggesting similar conditions existed at both sites.

The aerosol optical depth across (perpendicular to) the plume is needed to use equation 1. Unfortunately, the road does not cut straight across the plume (Figure 1) so it was necessary to convert the position of the aerosol optical depth measurements to their position along a line perpendicular to the plume. Figure 4 shows the aerosol optical depths along this perpendicular line. All the measurements at each location are shown in Figure 4 while only the average values at each location are shown in Figure 1. The measurements labeled A and B are indicated in Figures 1 and 4. A curve fit to the measurements was carried out and equation 1 was then used to calculate dry aerosol mass flux rates of 47, 60, and 52 (±40%) Mg d⁻¹ for passes 2, 3 and 4. Background aerosol optical depths (on either side of the plume) were averaged and subtracted from the in-plume optical depths prior to using equation 1. Aerosol flux rates calculated from the lidar data gave very similar values, which is expected since the lidar is calibrated with the sun photometer measurements. The first pass could not be used because part of the plume was beyond the road. Accounting for the difference in molecular weight between SO₂ and H₂SO₄, the formation of 53 Mg d⁻¹ of vog aerosol (the average of the three passes) corresponds to 35 Mg d⁻³ of SO₂ loss between the vent and the measurement point. We have assumed that all the S(VI) was derived from S(IV).


