Measurement of Sulphur in the Stratosphere from SAGE II

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Introduction

Sulphur containing compounds exert a significant influence on the chemistry of the Earth's atmosphere and hence its climate. Due to its low reactivity in the troposphere the most abundant form of sulphur found in the stratosphere is carbonyl sulphide (OCS) with an expected global mass of approximately 0.57 Tg. This gas is mainly formed from CS_2 oxidation but is also produced by the Earth's oceans and biomass burning.

The predominant anthropogenic sulphur containing species in the stratosphere is sulphur dioxide (SO_2). This gas reacts with H_2O in the atmosphere to form H_2SO_4 droplets.

By applying retrieval techniques to satellite spectral extinction measurements the size distribution described by its number density (N), mode radius (R) and spread (S) of these aerosol droplets is obtained and a value for global stratospheric mass of sulphur in H_2SO_4 form can be measured.

Method

Parameters which describe the aerosol size distribution, N, R and S for a monomodal aerosol distribution have been retrieved using the Optimal Estimation[1] (OE) method. This inversion technique, based on Baysian Statistics, uses a nonlinear least squares fitting algorithm to identify all possible solutions within a measurement uncertainty.

From the size distribution effective radius, surface density and volume density can be calculated. The volume density values are gridded by longitude, latitude and altitude and the mass of H_2SO_4 in each cell is found by:

$$\text{Mass} = \text{Vol Dens} \times \text{Volume} \times \rho$$

Where ρ, the density of H_2SO_4, is a function of temperature and weight fraction *wf*. *wf* itself depends on temperature and the partial pressure of H_2O. The mass of sulphur is then easily obtained from its atomic mass and the molecular mass of H_2SO_4.

Results

A time series from 1984 until 2001 of the total mass of retrieved stratospheric sulphur in droplet form is given in Figure 1 and shows values of approximately 0.25 Tg during background conditions. A maximum stratospheric loading of nearly 10 Tg is reported during the Pinatubo volcanic episode however the assumption of a monomodal distribution is no longer valid during such events and the values obtained should be considered with care. Figure 2 shows one height profile and demonstrates the decrease in sulphur with height.

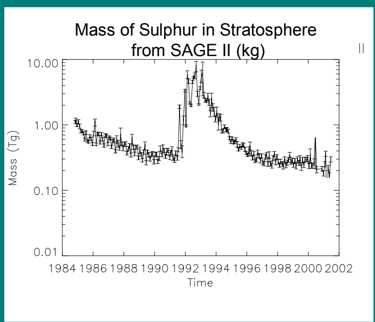

Figure 1

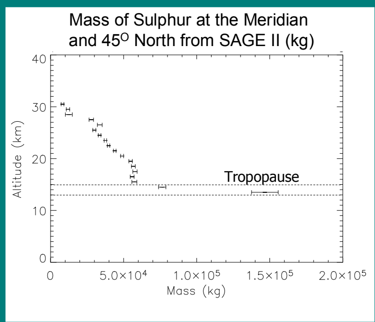

Figure 2

Discussion

From the above plots it is apparent that sulphur droplets are prevalent in the lower stratosphere with a background loading of approximately 0.25±0.03 Tg. During volcanic events a much higher value has been reported but these values have been obtained during conditions not suited to the retrieval's forward model.

A height profile of sulphur mass for 45° North on the meridian has been selected demonstrating a decrease with height. A sudden increase in the sulphur mass is seen to occur at heights which are in the tropopause region.

Conclusion

The mass of stratospheric sulphur in droplet form has been derived from SAGE II data using retrieved values of aerosol volume density.

During background conditions total sulphur mass is typically 0.25±0.03 Tg. The error on the retrieved values is 15% or less during background conditions but can be much larger if the aerosol distribution is not monomodal.

The expected height dependence of sulphur mass is observed. A sudden large increase is consistent with measurements in the tropopause region.

Reference

1. Rodgers, C. D., Inverse methods for atmospheric sounding.

Acknowledgements

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