The Atmospheric Sulphur Cycle and the Role of Volcanic SO_2

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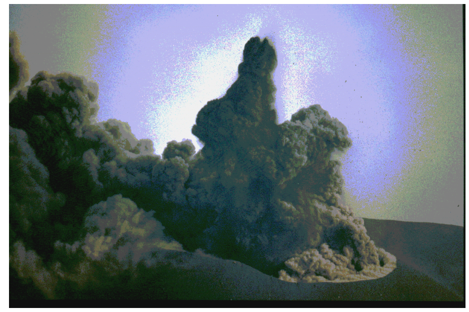

Introduction

Atmospheric sulphur occurs as a variety of compounds, in both gaseous and aerosol forms, and has a range of natural and anthropogenic sources. The life cycles and atmospheric burdens of these compounds are determined by a combination of physical, chemical and biological processes. Many of these processes have been included in the chemistry-climate model used in this study. Results from two model integrations reveal the main features of the S-cycle, and the volcanic and anthropogenic components. A further integration estimates the perturbation caused by the 1783-84 Laki eruption

Model

The chemistry-climate model used here couples the Hadley Centre General Circulation Model (GCM) with an atmospheric chemistry model (STOCHEM). The model has previously been used in several global and regional studies of tropospheric ozone, methane and secondary aerosol formation, from a variety of climate change and air quality perspectives. The chemical code has a detailed description of photo-oxidants as well as a sulphur scheme that includes dimethyl sulphide (DMS) in addition to SO_2 and its gas- and aqueous-phase oxidation to sulphate aerosol. The models have a horizontal resolution of about 5º and a vertical resolution of 1-2 km in the troposphere. The transport and chemistry have time steps of 1 hour and 5 minutes respectively.

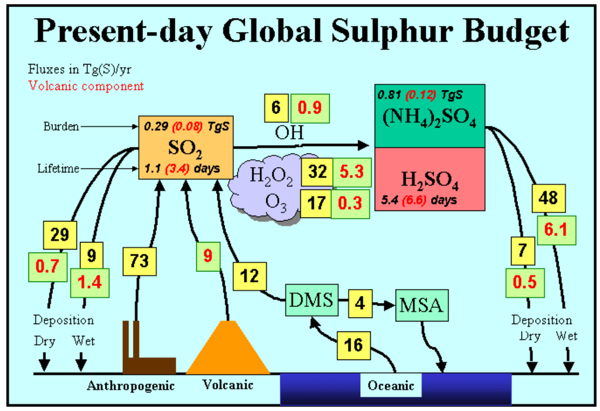

Figure 1. *Model results globally annually averaged.*

Experiments

Each of the runs described here used the same, present-day meteorology, and varied only in the prescribed emissions. Each run was 18 months in length with the last year used for analysis. The first run is for present-day (1990) conditions. The second run excludes volcanic SO_2 emissions. Run 3 is for pre-industrial conditions, setting all anthropogenic emissions to zero, and reducing biomass burning emissions to 20% of present day levels, but keeping all natural emissions constant. Run 4 simulates the Laki eruption of 1783-84, with an emission of 61 Tg(S) over the time period June to January to the atmosphere over Iceland, with 75% of emissions to the upper troposphere/lower stratosphere (8-12 km) and 25% in the lowest 3 km of the atmosphere. Background volcanic emissions are added evenly in the vertical over the lowermost 9 km of the atmosphere, in an attempt to simulate the mixture of passive and explosive inputs.

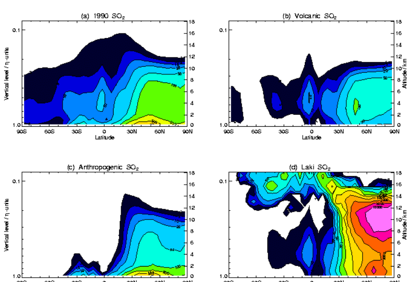

Figure 2. *Zonally annually averaged SO_2 (pptv) for (a) Present-day; (b) Volcanic component; (c) Anthropogenic component; (d) Laki simulation.*

Results

Figure 1 shows the details of the present-day global sulphur budget, as simulated by the model. Fluxes are in Tg(S)/yr. Note that anthropogenic emissions are currently over 3 times larger than natural sources of sulphur, which are roughly equally divided between volcanoes and DMS from oceanic plankton. The sources generate a total atmospheric SO_2 burden of 0.29 Tg(S), with a latitude height distribution shown in Figure 2a. Most of the SO_2 is near the surface in Northern mid-latitudes, close to its industrial source (Figure 2c). The volcanic component (0.08 Tg(S), or 28 % of the global burden) has a larger fraction in the free troposphere (Figure 2b), but is also mainly in the Northern Hemisphere (NH). Volcanic SO_2 is relatively more abundant than other SO_2 because it is emitted into regions of the atmosphere where it has a relatively long lifetime (Figure 4a) – the lifetime for volcanic SO_2 is 3.4 days compared to 1.1 days on average.

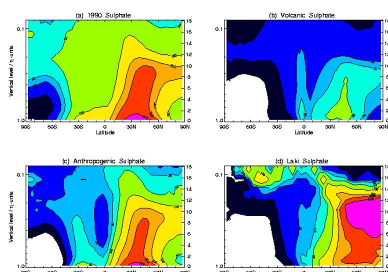

Figure 3. *As Figure 2 but for sulphate aerosol (pptv)*

SO_2 is removed by dry and wet deposition to the surface, and through oxidation to sulphate aerosol, predominantly in the aqueous phase. Deposition accounts for ~40% of present-day SO_2. Hydrogen peroxide (H_2O_2) is the main oxidant, although O_3 is also important, with gas-phase oxidation by OH a relatively minor process. All these oxidants have changed as a result of human activities.

Sulphate aerosol (Figure 3) is formed from the oxidation of SO_2. The aerosol is partitioned roughly equally between sulphuric acid and ammonium sulphate, although this has also changed since pre-industrial times when there was much less available NH3. Aerosol is removed by wet and dry deposition, and has an average lifetime of 5.4 days. Volcanic sulphate has a slightly longer lifetime (6.6 days) as it occurs a little higher up than average sulphate, and so experiences less deposition.

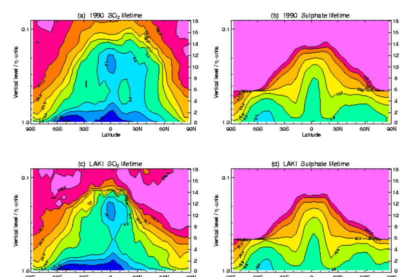

Figure 4. *Lifetimes (days) for present-day (a) SO_2 and (b) Sulphate; (c) are the same for the Laki simulation.*

Laki

Results for the Laki simulation indicate that it introduced a large sulphate perturbation to the NH upper troposphere, and possibly a significant perturbation to the lower stratosphere. The eruption had quite a major impact on oxidants in the NH, as seen in Figure 4c, shown as much longer lifetimes compared to Figure 4a. The sulphate lifetime (Figures 4b and 4d) is largely unaffected as this is only determined by physical processes which are fixed in this simulation.

Conclusions

A chemistry-climate model has been used to simulate the global sulphur cycle. The results are largely in line with other studies, and suggest that volcanic SO2, although making up only about 10 % of current S emissions, contributes closer to 30 % of the global SO2 burden, and 15 % of the global sulphate burden, mainly because it is emitted in regions of the atmosphere where its lifetime is relatively long. Major eruptions of the past, such as the eruption of Laki in 1873-84, are shown to introduce a major perturbation to the background sulphur cycle.