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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric impact of the 1783-1784 Laki eruption: Part I Chemistry modelling" *by* D. S. Stevenson et al.

D. S. Stevenson et al.

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We would like to thank the referee for his thoughtful and useful comments, which will help to clarify the paper. A point-by-point response to the comments is given below.

Consistent use of units Tg(S) will be used in the revised manuscript, as we think this will reduce confusion. The units Tg(H_2SO_4) and Tg($H_2SO_4.2H_2O$) were used in comparisons of total aerosol yield and burden, as several previous studies had discussed the 'total aerosol mass' resulting from Laki, referring to either the total sulphuric acid mass, or the total hydrated sulphuric acid mass. It is straightforward to convert between different units, using the molecular masses of S, SO_2, H_2SO_4, and $H_2SO_4.2H_2O$ (32, 64, 98, and 134).

In the Abstract, to place the Laki values in context, pre-industrial and industrial values will also be quoted, as suggested.

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

The previous studies that estimated the aerosol loading from Laki did not account for feedbacks in atmospheric chemistry; indeed they did not employ atmospheric models, except in the simplest sense. Estimates were made from ice-core measurements of acid deposition (Clausen and Hammer, 1988; Zielinski, 1994; Clausen et al., 1997), or from observed optical depths (Stothers, 1996), or inferred from the emissions (Thordarson et al., 1996; Thordarson and Self, 2003), assuming complete conversion of SO_2 to H_2SO_4 aerosol.

The crucial factor referred to the previous sentence in the text; the crucial factor in determining the impact of an eruption on the planetary radiation budget, and hence climate.

The term 'dry fog' has been widely used in previous papers (e.g., Stothers, 1996; Grattan and Pyatt, 1999), and appears to equate to an aerosol haze, resembling smoke. In the case of Laki, this seems to have been a haze at both ground level (at least intermittently), and also at higher levels, obscuring the Sun. The Laki dry fog was exceptionally long-lived and widespread, persisting for much of the summer and affecting many parts of the Northern Hemisphere. Some descriptions also indicate a sulphurous smell, and a serious impact on vegetation, e.g., shrivelling of leaves, indicating that SO_2 and/or H_2SO_4 was a major component. The term 'dry' is used because unlike normal fog, there appears to be no accompanying humidity. A recent analogy is perhaps the aerosol-rich haze seen over S.E. Asia in 1997 caused by widespread biomass burning, although this example was not sulphur rich.

As mentioned above, the previous studies did not use atmospheric models, but used either inversions of ice-core data, or used emissions estimates and assumed all of the volcanic SO_2 was converted to aerosol.

Highwood and Stevenson (2003; Part 2 of this work) took monthly mean aerosol distributions from this study and inserted them into the Reading IGCM (Intermediate General Circulation Model). Comparing results for an ensemble of runs to a control (clean pre-industrial atmosphere) run, radiative forcings and climate impacts for the simulated Laki cases were calculated. Northern Hemisphere monthly mean forcings reached nearly -6 W m^{-2} in August 1783. The Laki Hi case generated a statistically significant NH cooling of -0.21 K for 1783, with some cooling persisting for over a year after the eruption, in broad agreement with the observed climate anomalies. Many areas of uncertainty remain in modelling the climate impact of aerosols.

The present-day sulphur cycle in STOCHEM is described in more detail in Stevenson et al. (in press) and Derwent et al. (2003). The version of STOCHEM used here shows generally good agreement with other current models summarised in Penner et al. (2001). Derwent et al. (2003) uses a slightly different version of STOCHEM (different meteorological resolution, similar sulphur scheme), and shows more comparisons with surface data for sulphate. These show reasonable agreement over Europe, but the model underestimates N. American sites by about a factor of two. Compared to other state-of-the art global aerosol models, STOCHEM is strong in that it employs detailed and comprehensive S and oxidant chemistry; its main weaknesses are the relatively coarse resolution, and the lack of any aerosol microphysics.

The referee correctly points out that all of the model validation described refers to earlier versions of the model, and the present model will behave somewhat differently. This is true, but most features of the new model version have been tested in similar ways, e.g., simulation of ^{222}Rn transport, surface ozone and ozone profiles, albeit this work has not yet been published.

The random walk component in the advection scheme introduces a turbulent diffusive mixing.

At Icelandic summer tropopause levels (10 km, or 250 hPa), the model vertical resolution of 25 hPa is equivalent to about 600 m.

Clouds, along with all the meteorological variables (winds, temperatures, humidities, etc.) are calculated by the Met Office Hadley Centre GCM (HadAM3). STOCHEM-Ed

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Interactive Discussion

Discussion Paper

runs as a subroutine of this GCM, receiving (Eulerian, or grid-point) meteorological data every 3 hours. This Eulerian data is then interpolated to air parcel centres, where it is used by the Lagrangian chemistry-transport model. Cloud transport does not occur within the Lagrangian framework. Cloud variables are used by STOCHEM-Ed in several ways. Distributions are used in the photolysis code, to modify photo- dissociation rates. Convective cloud amounts and depths, together with convective precipitation rates are used to determine vertical convective mixing. Cloud liquid water amounts are used in the aqueous-phase chemistry scheme. Precipitation rates are also used to calculate wet scavenging of soluble species.

The conversion of N_2O_5 to nitrate aerosol only occurs on sulphuric acid aerosol in this model version. The model aerosol distribution is used, with an assumed aerosol size (and surface area) distribution that is globally constant.

Emissions are specified on a 5° x 5° grid, and are evenly distributed between any air parcels located within the boundary layer above each grid square, after each advection time step. Boundary layer heights are supplied by the GCM, and typically are 100-200 hPa (1-2 km) in thickness, i.e. the lowest 1-2 model layers in STOCHEM-Ed. Occasionally, very shallow boundary layers occur, and as the layer gets smaller in volume, the likelihood of an air parcel being present decreases. In situations when no air parcels are within the boundary layer for a given grid square, the emissions are stored until an air parcel does arrive.

A 'total' column has been added to Table 2, the emissions.

Species-dependent scavenging coefficients (rather than rates) are taken from Penner et al (1994). Coefficients for SO_2 are 0.8 and 1.5 cm^{-1} for dynamic and convective precipitation, respectively. Equivalent values for sulphuric acid aerosol are 5.0 and 1.5 cm^{-1}. These coefficients are then used with dynamic and convective precipitation rates, and scavenging profiles, to calculate loss rates for all the soluble species from each air parcel. The normalised scavenging profile for dynamic precipitation increases linearly with pressure from zero at 400 hPa to one at 800 hPa, remaining at this value down to the surface. A similar profile is used for convective precipitation, but with the profile starting at the cloud top. Wet scavenging is only switched on (for both convective and dynamic precipitation) above a threshold precipitation rate of 10^{-8} kg m^{-2} s^{-1}. This differs from the use of Henry's Law (e.g. Table 1), which is used to determine the partitioning of soluble species between gaseous and aqueous phases in the presence of cloud droplets.

All species (including SO_2) are subject to convective mixing. Convective mixing is achieved by fully mixing a fraction of the air parcels beneath a convective cloud top. The precipitation rate and the fractional cloud cover determine the fraction mixed, and this fraction has been tuned using observations of ^{222}Rn (e.g., Stevenson et al., 1998a).

We have added a sentence noting that the model aerosol fields do not influence the photolysis rates. The effects of large concentrations of SO_2 on radiative transfer are also not included, but have been considered in Part 2 (Highwood and Stevenson, 2003). SO_2 behaves as a weak greenhouse gas, but even for the very high SO_2 concentrations calculated at the peak of the eruption, the radiative forcing effects were relatively small (<10 %) compared to the magnitude of the aerosol forcing.

We agree with the referee that 2 years of simulation is rather short, and that ideally an ensemble of runs, each with differing meteorology would have been preferable. This approach was taken in Part 2 of the study, but this was mainly because the climate simulations were relatively less computationally intensive, and an ensemble of runs was needed to generate statistically significant results. Earlier STOCHEM-Ed simulations (Stevenson et al., 2001) with a different meteorology generated very similar results; these results are not presented in the paper however, as there were some minor inconsistencies and problems with the chemical scheme in these simulations. Nevertheless, they give the firm impression that monthly to seasonal mean fields (as reported here) are not likely to be strongly dependent on the individual year's meteorology used. Similarly, the use of a 1990s meteorology is not thought to introduce major differences

Full Screen / Esc
Print Version
Interactive Discussion
Discussion Paper

compared to a pre-industrial meteorology.

In the Lo scenario, all of the emissions are below tropopause levels. Over Iceland, the tropopause occurs at 250 hPa (10 km). In the Hi scenario, 25 % (15.3 Tg(S)) of the SO_2 emissions are added to the lower troposphere, 43 % (26.4 Tg(S)) are added to the upper troposphere, and 32 % (19.6 Tg(S)) to the lower stratosphere.

In the Hi scenario, the split in emissions of 75:25 (S. Self, pers. comm. 2002) between the upper troposphere/lower stratosphere and lower troposphere is derived from models of eruption columns (e.g., Woods, 1993). A revised split of 80:20 has been reported subsequently (Thordarson and Self, 2003). The main inputs to these models are the magma eruption rate and gas content, together with the morphology of the eruption conduit and vent, all of which can be estimated from eyewitness accounts of the eruption and examination of the eruption products (Thordarson and Self, 2003).

The figure of 61 Tg(S) for the total release of sulphur from the eruption, is from Thordarson and Self (2003), and is also reported in earlier papers (Thordarson et al., 1996). Unfortunately, these authors fail to report an uncertainty associated with this figure. The amount is derived from analysis of the volume of magma erupted, together with the S content of degassed lava and undegassed melt inclusions, trapped within phenocrysts (crystals in the lava). Uncertainties in these S contents are reported in Thordarson et al. (1996), and vary between ± 5 % and ± 17 %. Coupled with uncertainties in the total volume of magma released, we estimate an uncertainty of about ± 20 % on the figure of 61 Tg(S). This method of estimating SO_2 emissions from an eruption has been shown to give a minimum amount (Wallace, 2001), as there can be a significant pre-eruption vapour content in the magma chamber, which would not be recorded in the melt inclusions. This perhaps indicates that the actual emission is more likely to be larger than 61 Tg(S). However, these problems are usually related to more explosive silicic magmas (e.g., Pinatubo, El Chichon), rather than basaltic magmas.

Figure 1 shows some transport of SO_2 from the eruption latitude (60°N) into the tropics

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over the first 3 months; this is mainly in the lowermost stratosphere (I have added tropopause heights onto the zonal mean figures). Note that the concentrations have a log scale, so the amounts are quite small (the green colour indicates 0.2-0.5 ppbv, 2 orders of magnitude smaller than the peak zonal mean values of 20-50 ppbv). The vast majority of the emitted SO_2 is constrained to the high-latitude Northern Hemisphere (>40°N), but a small fraction does leak into the tropics, in regions where its lifetime is sufficiently long. These results are consistent with our understanding of the circulation of the atmosphere and inter-hemispheric exchange.

As mentioned above, and in section 4.6, this modelling study represents an improvement over previous estimates of the aerosol loading from Laki because it has employed a detailed atmospheric model of transport and chemistry. Previous studies (e.g., Thordarson and Self, 2003) have made sweeping assumptions about the fate of the SO_2 in the atmosphere, such as that it was all converted to aerosol (we find only about 30 % is converted to aerosol), and that it remained airborne for around 1 year (we find mean atmospheric lifetimes of about a week). Furthermore, our results generate a reasonable simulation of deposition of H_2SO_4 to Greenland ice, and in Part 2, we show that our results also generate an impact on climate of the correct magnitude.

In the Conclusions, to place the Laki values in context, pre-industrial and industrial values will also be quoted, as suggested.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 551, 2003.