

## ***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 her thoughtful and useful comments, which raise several important issues. A point-by-point response to the comments is given below.

### 1. Use of monthly mean emission fields

In the original submission, we used monthly mean emissions for the Laki eruption, and no attempt was made to introduce episodicity. In order to assess the importance of short-term variability in the emissions, we have repeated the Laki Hi experiment, using daily varying emissions. We estimated daily emissions using the values attributed to individual episodes in Thordarson and Self (2003, Table 1). For each episode, we made the assumption that emissions fell to  $1/e$  of the initial value by the last day of each episode. Between episodes, emissions are assumed to be zero. Results from this new experiment have been incorporated into the figures and tables of the revised

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manuscript. Overall, results from this new experiment are quite similar to the one using monthly mean emissions, supporting our original suggestion that the use of monthly mean emissions would not have a large effect on the results. Over the JJA period, slightly less of the  $\text{SO}_2$  is oxidised, and less sulphate is produced. The impact on oxidants, averaged over JJA, is slightly less, although the impact in June is larger, as in the daily emissions case, more  $\text{SO}_2$  is released in June. The impact on oxidants is not exactly linear with increasing  $\text{SO}_2$  emission, but is not strongly non-linear. As the referee suggested, using episodic emission data has the effect of slightly reducing the efficiency of conversion to sulphate, and over the course of the whole eruption, the total aerosol yield is reduced by 17 %, although its lifetime is increased by 8 %, and the peak aerosol loading (in July) is very similar.

## 2. Aerosol microphysics

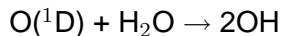
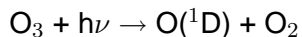
We use a bulk approach in our treatment of sulphate aerosol, and the model calculates no information about aerosol size distributions or microphysical processes. Aerosol nucleation, coagulation and condensation processes are assumed to occur relatively rapidly. Gas-phase sulphuric acid is therefore assumed to instantaneously convert to the equivalent mass of accumulation mode aerosol. This lack of detailed treatment of microphysical processes is a further source of uncertainty in our results, but we feel the assumptions made are probably reasonable.

## 3. $\text{H}_2\text{O}$ emission

The referee correctly points out that the eruption also emitted a large quantity of magmatic water (235 Tg), and that this may play a role in the atmospheric chemistry, particularly at the plume scale. It is important to note that  $\text{SO}_2$  does not react directly with water to produce sulphuric acid, as has been implied in some earlier studies (Thorndarson and Self, 2003). Water is a precursor of two major  $\text{SO}_2$  oxidants, the hydroxyl radical (OH) and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ), and levels of these oxidants will increase as water vapour levels increase. However, there is a large amount of water readily

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available in the atmosphere, and ambient temperatures will limit the increase in water vapour, such that humidity cannot exceed 100 %. Assuming that the plume rapidly comes into thermal equilibrium with the environment, and a conservative estimate of 50 % humidity in the background air above Iceland, then we could expect at maximum a doubling of water vapour associated with the eruption plume, with perhaps similar increases in oxidant levels, and hence sulphate production. However, to generate OH, UV radiation and ozone are also required, in order to generate excited oxygen atoms ( $O(^1D)$ ):



As the OH concentration rises, so will concentrations of the hydroperoxyl radical ( $HO_2$ ). At high radical concentrations, radical loss rates are proportional to  $[HO_x]^2$  ( $HO_x = OH + HO_2$ ), and this acts as a strong buffer to radical concentrations rising further. Any rise in OH concentrations will therefore be limited by the availability of ozone, sunlight, and by radical self-destruction. The importance of magmatic water will also decrease as the plume mixes with the background atmosphere, which is likely to occur on timescales of hours to days. These timescales are short compared to the chemical lifetime of  $SO_2$  in the upper troposphere/lower stratosphere ( $>20$  days). Overall, we suspect that the additional magmatic water vapour can be expected to locally increase oxidant levels (and hence sulphate production), but only to have a limited impact at larger scales. To simulate these processes in detail, a plume or cloud-scale model is required (e.g., the ATHAM model, Trentmann et al., 2002). The global model employed here is not an appropriate tool to model such processes. We do explicitly state that there may be important plume-scale processes that will modify our results.

#### 4. Wet and dry deposition

Some clarification of the wet deposition scheme has already been discussed in our response to Referee 1. Dry deposition rates are calculated using a resistance analogy

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scheme (e.g., Seinfeld and Pandis, 1998). Dry deposition velocities used for SO<sub>2</sub> are 6.0, 8.0 and 0.5 mm s<sup>-1</sup> over land, sea, and snow/ice surfaces respectively. These values are quite uncertain, but are consistent with the review of Erisman and Baldocchi (1994). Equivalent values for sulphate aerosol are 2.0, 1.0 and 0.05 mm s<sup>-1</sup> (Wesely et al., 1985). An aerodynamic resistance term is also calculated using the surface stresses and heat fluxes from the GCM. There is no dependence of deposition on vegetation type. Both the wet and dry deposition schemes are relatively simple, but are typical of the current generation of global scale sulphur cycle models (e.g., Koch et al., 1999). As discussed in Stevenson et al. (in press) and Derwent et al. (2003), the global sulphur cycle in STOCHEM is generally in good agreement with a range of other models, summarised by Penner et al. (2001).

#### 5. Sink processes for sulphate

Sedimentation processes for aerosol are neglected here, but may well be important. Because the model lacks any description of the aerosol size distribution, sedimentation cannot be modelled. It is worth noting however, that if this process were included, it would tend to increase the removal rate of aerosol, and hence reduce the atmospheric aerosol loading.

#### 6. Figure 6

We failed to indicate that Figure 6 compares surface observations with modelled values from the lowest model layer. Simulated deposition fluxes at individual sites show similar levels of inter-annual variability.

#### 7. Comparison of results with ice core data

We think that there are unlikely to have been major changes in circulation patterns since 1783, so we feel justified in using a 1990s meteorology to represent the time period of the eruption. However, as mentioned in our response to Referee 1, there will be inter-annual meteorological variability, and this will influence the dispersion of

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the volcanic emissions and products, nevertheless, the seasonal-global zonal mean distributions are unlikely to be significantly affected. However, the referee is correct to point out that simulated deposition fluxes at an individual site (central Greenland) will reflect much more strongly any inter-annual variability. The simulated ice-core deposition fluxes in Figure 9 and Table 4 therefore have quite large error bars, and should only be treated as an indication of the magnitude of acid deposition to Greenland. This is partially evident in the results from the new episodic simulation, which shows a 45 % reduction in total acid deposition to Greenland. The episodic emissions are effectively seeing a different meteorology to the monthly mean experiment.

#### Additional references

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