

Response to comments from Referee #1

Thank you for your comments. We certainly agree that there are several assumptions in the model which limit it and it is a good idea to discuss these further. The effect of cosmic rays on Forbush Decreases is an area that has received a lot of attention very recently (e.g. Sloan and Wolfendale 2008, Svensmark et al 2009, Beer et al 2009, Laken et al 2009) and we feel that it is important to start to understand the issue from a modeling perspective. While it would be most interesting to repeat this study with an established global model we think that a simple box model study such as this has merit as well and is a good starting point for the modeling effort.

We agree that the assumptions you have listed could be discussed in more detail and we will attempt to do so.

Below we answer your specific comments, point by point.

1) The hiding of different sink processes in "k" is problematic as many of the processes depend on each other (e.g. condensation loss of vapours vs. effect of coagulation on total particle surface) and many of the processes are also strongly size-dependent.

We agree that it would have been better to include the sea salt explicitly in the model. However we did estimate the loss from the sea salt by making a single run with the addition of a starting population corresponding to the sea salt distribution. By disabling condensation we then estimated the loss rate for the nucleating particles to the sea salt. Since this is not an ideal way to do it we included the loss parameter as one of those tested in the sensitivity study. This is already discussed on p. 22841 l. 23ff and we will extend this discussion.

2) The model simulations cover several days. It is nowhere said what is the behaviour of sulphuric acid production term: in the atmosphere, sulphuric acid is produced by OH-reactions which are solar radiation dependent. As many of the aerosol processes are non-linear, the potential assumption of constant sulphuric acid production is clearly wrong => if the sulphuric acid formation rate is sinusoidal, the new particle formation rates should also follow the similar pattern.

The previous paper by Enghoff et al., 2008 does not answer this question.

The sulphuric acid production term is constant within each run. In the development of the model we have tried running with a sinusoidal production term. However since this is supposed to be a box model of an "average" marine environment we decided it was better to keep the production term constant since when it is night in one place it is day in another. We will add this clarification to p. 22839 where the equation for sulphuric acid is introduced.

Nucleation mechanisms:

- The authors assume that the formation rate depend only on "Q", and "s" is constant is problematic. Clearly, sulphuric acid concentration (or ion-induced cluster formation

rate) is not constant, and due to nonlinear processes, the resulting particle formation can not represent the real situation.

We agree. This is one of the assumptions we have had to make in order to be able to run this box model study, which depicts a situation that is less noisy than the real world would be. This will be discussed on p. 22839 where “s” is introduced.

3) Due to highly non-linear processes, coagulation and condensation can not be treated separately. A half-time approach does not take into account normal short term competition between the growth and loss processes.

We are not sure what you mean here? Condensation and coagulation is solved simultaneously within each time step. We will include the GDE used directly in the paper to make it more clear what the model is doing.

3) What are e.g. the nucleation mode particle growth rates resulting from the initial assumptions? The authors should also present an example of development of aerosol size distribution during the modelling period.

We will include an example of a resulting particle distribution and discuss this. With regards to growth rates the model has been checked to comply roughly with the rule of thumb of 1 nm/h of growth pr. 10^7 cm⁻³ of sulphuric acid (Boy et al 2005).

4) what is the effect of the particle formation size? Typically the modelling results are very sensitive to this parameter, and a sensitivity study over e.g. 2-6 molecules should be made.

This is one thing we have not tested the sensitivity of. Since the model assumes that clusters are stable upon formation we do not imagine that it will make a big difference if the clusters have to start growing from a size of a few molecules more or less.

5) The comparison with AERONET data can not really be made, as the AERONET sites are above the continents and in the approach of this paper, aerosol consists only of sulphuric acid and sea-salt: above land, main fraction of aerosol is typically organic, and depending on season, location and meteorology, soil compounds can have a significant influence as well. How is the comparison with AERONET sites exactly done? Can it really be done?

As we also point out in the paper (p.22850 l. 25ff) there is certainly a difference between the (mostly) land based AERONET data and our model. However this is the only data available at the small wavelength pair and we felt it was important to try to make the comparison nevertheless. Since there actually seems to be a signal in the AERONET data this also makes the comparison more valid – even if the main fraction is organic there is still some contribution from sulphuric acid. We will add additional text to the manuscript to elaborate on this.

6) Above oceans, cloud processing is one of the main factors modifying aerosol size distribution, not only the number, but also the size. What is the effect of cloud processing on results?

Cloud processing could not be included directly into the model, as mentioned on p 22842 l. 11ff. We've tried to accommodate a bit for this by including loss to rain out in the loss rate but we realize this is not a perfect approach.

References:

Sloan and Wolfendale, Testing the proposed causal link between cosmic rays and cloud cover, *Environmental Research Letters* 3, 2008

Svensmark et al, Cosmic ray decreases affect atmospheric aerosols and clouds, *GRL* 36, 2009

Beer et al, Sudden Cosmic Ray Decreases: No Change of Global Cloud Cover, *GRL*, in press

Laken et al, Cosmic ray decreases and changes in the liquid water cloud fraction over the oceans, *GRL* 36, 2009

Boy et al, Sulphuric acid closure and contribution to nucleation mode particle Growth, *Atmos. Chem. Phys.*, 5, 863–878, 2005