

Response to comments from Referee #2

General remarks

The manuscript explores the idea of quantifying the response of tropospheric aerosol optical properties to short-term reductions in atmospheric ionization during Forbush decreases, with interesting results. The authors are commended for relating their results to observations, and for writing in a clear and organized way. The research is done in a reasonably sound manner given the limitations of the modeling approach, which are not concealed from the reader, with some exceptions listed below. These should be discussed in the manuscript before publication. In the light of the uncertainties and limitations in the modeling approach, the manuscript should be considered a waypoint in the understanding of atmospheric ionization for tropospheric aerosol and motivation for research that validates the authors' findings, rather than a closure.

Thank you for your comments. We agree completely that the manuscript is to be viewed as a waypoint and definitely not as a final word on the topic. Below we will answer your specific comments point by point.

Page 22834/line 10

"... in the cases with atmospherically realistic output parameters ..."

"output parameters" is a bit confusing here, would "... input parameters to our model ..." be appropriate?

This is meant to refer to the cases where the output from the model look mostly like what is to be expected in the real atmosphere. We will try to make this clearer.

Page 22839/line 25

"Assuming that the concentration of sulphuric acid particles is exponentially decaying with height (h) in the troposphere (extending from 0–10 km)..."

This assumption introduces a large amount of uncertainty and constitutes a limitation on the manuscript's conclusions: It is equivalent to assuming that aerosol nucleation events produce the same mixing ratio of new particles throughout the troposphere, or that vertical mixing in the troposphere takes place on the same time scale as the nucleation process. Both alternatives are somewhat unrealistic: Marine nucleation events are often triggered by removal of pre-existing aerosol by precipitation, e.g. in the boundary layer by stratiform clouds, or by the introduction of aerosol precursor molecules into a region that is favorable for nucleation, such as by lifting of boundary layer DMS and SO₂ by convection into the upper troposphere. In the first case the nucleation event would be confined to the boundary layer, in the second to the upper troposphere, and mixing throughout the tropospheric column can be expected to take place on time scales much longer than that of the nucleation event and perhaps even the lifetime of the particles. Nucleation events are therefore unlikely to produce a vertical distribution of ultrafine aerosol that is consistent with the assumption. This is supported by observations which do not show vertically well-mixed ultrafine

aerosol (Clarke and Kapustin, 2002). A lesser actual response of aerosol optical properties to Forbush decreases should be expected from aerosol nucleation taking place in confined tropospheric layers than based on the assumption. On the other hand, precipitation events that trigger aerosol nucleation by removing pre-existing aerosol also remove pre-existing sea-salt, which may result in a greater response of the aerosol optical properties than presented in the manuscript (until the sea-salt aerosol population is replenished). The limitations imposed by the assumption on the conclusions of the manuscript are a motivation for further research and not a reason to discount it in toto, but they should be discussed.

This is a good point. We will discuss the implications of this assumption in the final paper as you've suggested.

Page 22841/line 10

"To establish a steady state of background sulphuric acid particles, initially, the sulphuric acid model is run for a month for various constant cluster formation rates (s), constant sulphuric acid production rates (PH_2SO_4), and half lives (τ) of nucleated particles against primary particles."

In addition to the processes used in the model to produce a steady-state marine background aerosol size distribution, other processes shape the aerosol size distribution in the actual atmosphere, such as transport, mixing, dry deposition, and cloud processing.

In addition, sulfuric acid production has a diurnal cycle and nucleation occurs mostly during day-time and often within limited periods of time in the marine atmosphere.

It would therefore be a great asset for the manuscript to demonstrate that the simulated aerosol size distributions are realistic, e.g. by plotting them against observed size distributions, or to give their integrated number concentration and geometric mean diameters, and compare these to observed values of marine Aitken mode particles, such as given by Heintzenberg et al. (2000). A fair agreement would countervail to a large extent the limitations of the chosen modeling approach.

We will show a plot of the resulting size distribution from our "single run" case and compare it with the suggested reference. Our "single run" case has a mean diameter of 246 nm and a total particle concentration of $\sim 90 \text{ cm}^{-3}$ (excluding sea salt). The size is somewhat higher than the accumulation mode cases shown in table 3 of the Heintzenberg paper. On the other hand it fits well with the 266 nm given in Seinfeld and Pandis 2. ed (p.371, table 8.3) for the second mode of a marine aerosol distribution. Also this particular run lies in the high end of the parameter space with regards to size (see our fig. 3).

Page 22842/line 15

"... which fits well with our choice of loss rates"

Full stop missing.

Page 22842/line 23

"... dotted ..."

Dashed?

Page 22842/line 28

Equation (5): $dQ(t)(t) ! dQ(t) ?$

Page 22849/line 14

"... high wind speed of 18 m2."

Units should probably be m/s.

Page 22850/line 24, and Figures 2, 4, 5, and 6

"The dashed line is the average of the FD signal over the events listed above."

It is not clear how the FD signal is defined; in addition the dashed line in Figures 2, 4, 5, and 6 showing the response of the Climax neutron monitor count to the Forbush decrease is not sufficiently well described in the figure labels and captions, and the units are missing. Please add a sentence in the figure caption(s) that explains the dashed line, declaring what it represents precisely, or modify the above sentence so that the reader can easily understand what the dashed lines stand for.

Figure 4

Caption: dotted ! dashed ?

Figure 6

- MODIS label: Should it read "AE(550-865) MODIS"?*
- Please use a more descriptive label than "CLMX 5 events"*

Figure 2-6

A larger font for the figure labels/tick marks would be an asset.

We will correct the typos and try to make the figure captions better.