Review of: Unveiling Sulfate Aerosol Persistence as the Dominant Control of the Systematic Cooling Bias in CMIP6 Models: Quantification and Corrective Strategies by Jie Zhang et al, for ACP.

Stephen E. Schwartz, Reviewer; 2025-0513

The authors make the point that “Including sophisticated aerosol schemes in the models of the sixth Coupled Model Inter-comparison Project (CMIP6) has not improved historical climate simulations.” They go on to state that “in particular, the models underestimate the surface air temperature anomaly (SATa) when anthropogenic sulfur emissions increased in 1960~1990, making the reliability of the CMIP6 projections questionable.”

The present paper seems to consist of two components: (1) The differences among models in the sulfate forcing and (2) the relation of sulfate forcing to temperature change among the models.

Before going further in this review, I must state major disagreement with the authors’ definition of what they denote as the “effective retention time” of atmospheric sulfur. This quantity is found in the authors’ analysis of the CMIP model results to be as low as 1 day or so. In my judgment this flawed definition casts a cloud over all the findings in the study. This is elaborated below.

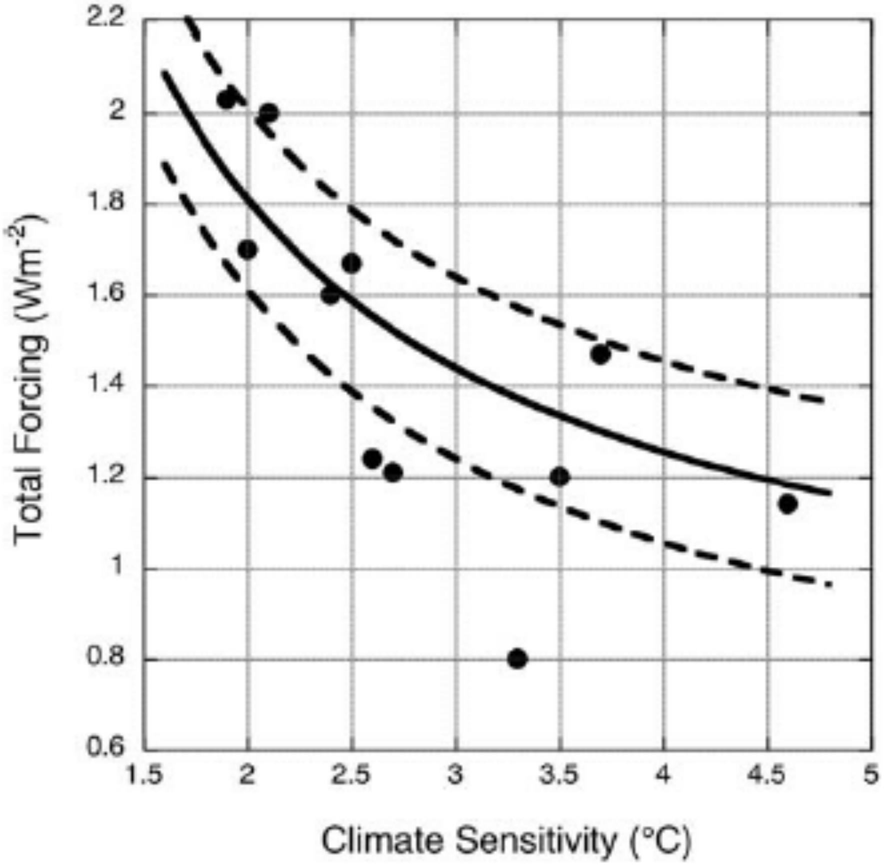
In the present study the authors define and use a quantity that they call the ESRT, effective sulfur retention time scale, equation 1, line 295.

$$\mathrm{ESRT} = \mathrm{loadSO4}/(D_{\mathrm{SO_4}}+D_{\mathrm{SO_2}}),$$

This quantity is central to the author’s analysis so in my opinion should not be hidden back in the appendix and justified. I disagree with the utility of this definition. It is a stock of sulfate upon leaving flux of atmospheric sulfate plus leaving flux of atmospheric SO2. This is contrary to customary definition of residence time (more explicitly, turnover time, to distinguish it from other measures of residence time) which would be stock of the substance of interest in a given compartment upon the leaving flux of that substance from that compartment, not upon multiple fluxes of multiple substances; the definition confounds the utility of the definition and may be responsible for the low residence time found in the study. The definition certainly would seem to preclude the utility of any comparisons with residence time as conventionally evaluated.

Setting aside that concern, Figures1b, 4a and 4b seem to show substantial systematic differences across models in measures of sulfate burden that might provide insight into reasons for differences in sulfate forcing across models.

With respect to the relation, across models, between sulfate burden and change in GMST, the authors seem unaware of the extensive prior literature on this subject that is very pertinent to this study. In early work Kiehl (2007) demonstrated anticorrelation between forcing and sensitivity in the climate model runs reported in the 2007 IPCC report. The relation between sulfate forcing and temperature change in models continues through Stevens (2015) and beyond. Certainly, it would seem that any revision of the current paper should take cognizance of this literature.



From Kiehl (2007)

The role of sulfate lifetime as a key influence on forcing, which is a major thesis of this paper, was highlighted by Charlson et al, 1992, who delineated the factors controlling direct sulfate forcing, as given in the following table:

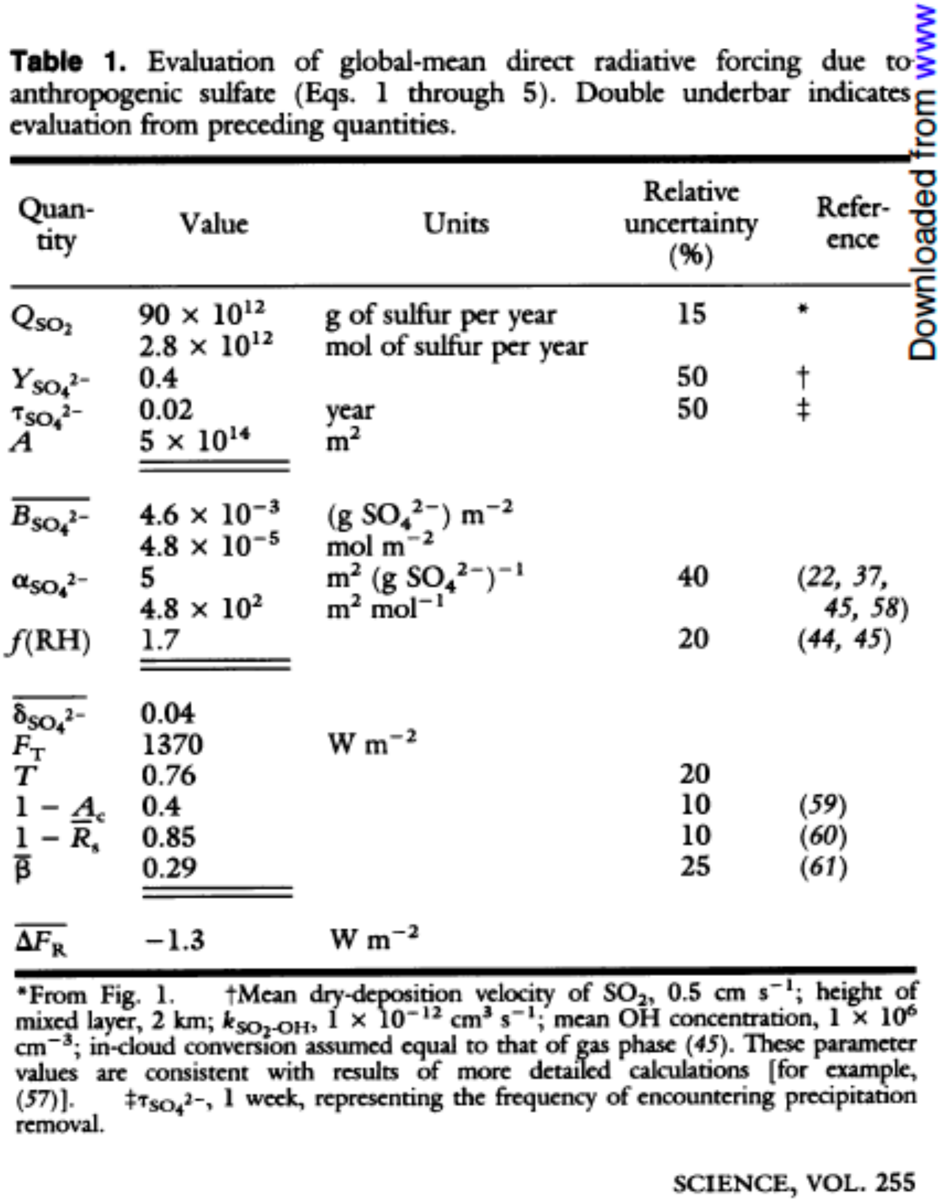


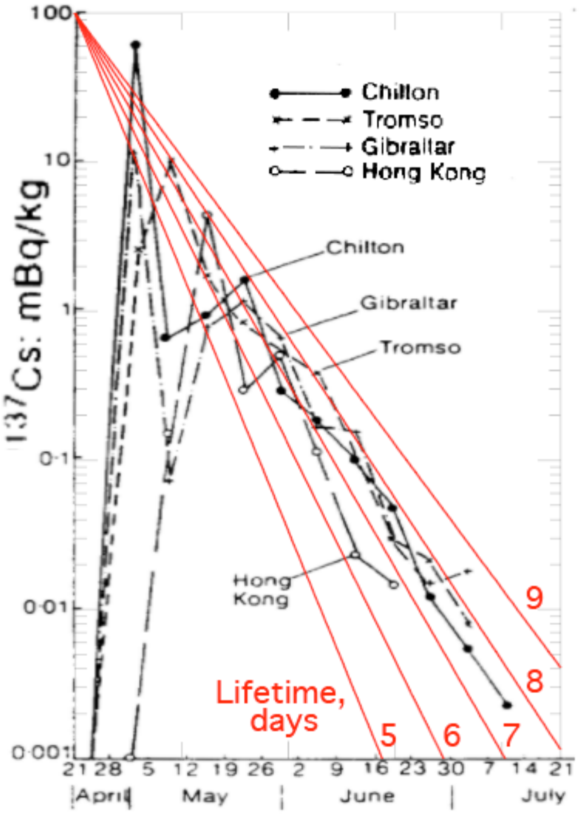
Table 1. Evaluation of global-mean direct radiative forcing due to anthropogenic sulfate (Eqs. 1 through 5). Double underbar indicates evaluation from preceding quantities.

Quantity	Value	Units	Relative uncertainty (%)	Reference
Q_{SO_2}	90×10^{12} 2.8×10^{12}	g of sulfur per year mol of sulfur per year	15	*
$Y_{SO_4^{2-}}$	0.4		50	†
$\tau_{SO_4^{2-}}$	0.02	year	50	‡
A	5×10^{14}	m^2		
$\overline{B_{SO_4^{2-}}}$	4.6×10^{-3} 4.8×10^{-5}	$(g\ SO_4^{2-})\ m^{-2}$ $mol\ m^{-2}$		
$\alpha_{SO_4^{2-}}$	5 4.8×10^2	$m^2\ (g\ SO_4^{2-})^{-1}$ $m^2\ mol^{-1}$	40	(*22, 37, 45, 58*)
f(RH)	1.7		20	(*44, 45*)
$\overline{\delta_{SO_4^{2-}}}$	0.04			
F_T	1370	$W\ m^{-2}$		
T	0.76		20	
$1 - \bar{A}_c$	0.4		10	(*59*)
$1 - \bar{R}_s$	0.85		10	(*60*)
$\bar{\beta}$	0.29		25	(*61*)
$\overline{\Delta F_R}$	−1.3	$W\ m^{-2}$		

*From Fig. 1. †Mean dry-deposition velocity of SO_2, 0.5 cm s^{-1}; height of mixed layer, 2 km; $k_{SO_2\text{-}OH}$, 1×10^{-12} $cm^3\ s^{-1}$; mean OH concentration, 1×10^6 cm^{-3}; in-cloud conversion assumed equal to that of gas phase (*45*). These parameter values are consistent with results of more detailed calculations [for example, (*57*)]. ‡$\tau_{SO_4^{2-}}$, 1 week, representing the frequency of encountering precipitation removal.

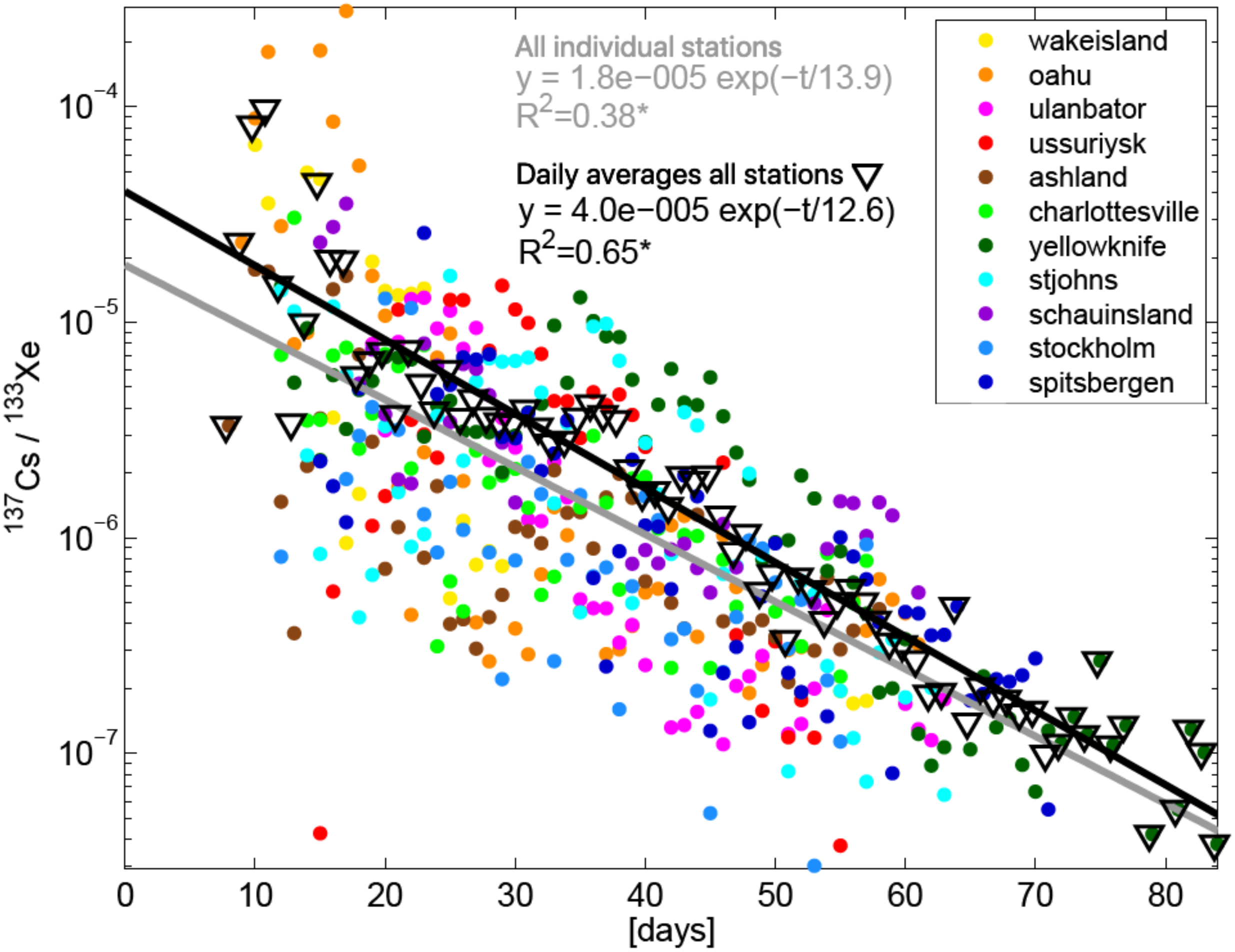
Those investigators also presented an analysis of indirect (Twomey) forcing that rested on mass loading of atmospheric sulfate, which in turn is proportional to sulfate residence time. Unless the authors take exception to this sort of analysis, I would suggest that this might be a good place to start in their assessment; if they do take exception, then it would seem incumbent on them to identify the point of disagreement and to justify their concern. The quantity τ_{SO42-} in the above is

0.02 yr or 7.3 days. This value of $\tau_{\mathrm{SO42-}}$ was based in in that study in large part on the observed rate of decrease of concentration of 137Cs at several midlatitude stations in the Northern Hemisphere subsequent to the release of this isotope from the Chernobyl incident in 1986. The negative of the slope of the log of that concentration vs time gives the decay constant; the inverse of that, the residence time. 137Cs released by the reactor release attaches to accumulation mode particles and thus serves as a good proxy for determination of the lifetime of accumulation-mode particles or sulfate.



Modified from Cambray et al. (1987)

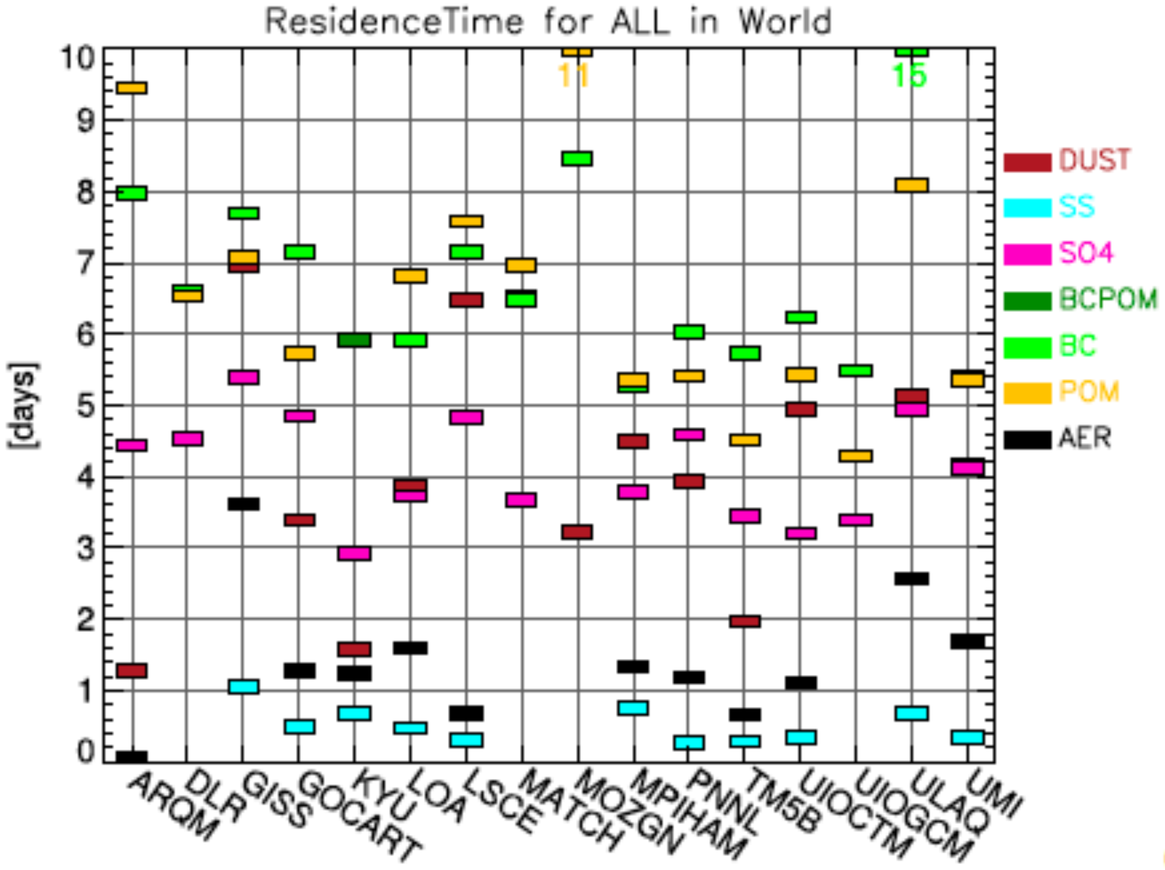
Subsequently, the incident at the Fukushima reactor following the 2011 tsunami provides even better information on lifetime from the ratio of 137Cs to the passive tracer 133Xe (Kristiansen et al, 2012) which removes much of the variability in concentration of a single species arising from variability in transport. That analysis yields a residence time of about 13 days.



Kristiansen et al ACP 2012

In sum, the observational evidence would seem to contradict the short residence time presented by the authors based on their analysis of the CMIP models

Several prior model intercomparisons have been conducted that identify the loading of sulfate (and other aerosol constituents) to try to identify reasons for differences in forcings. I call particular attention to the AeroCom Study (Kinne et al, 2006; Textor et al., 2006). Textor examined residence times of multiple aerosol constituents in a very insightful figure. The model calculations were found to give a much greater residence time for sulfate (3-5.5 days) than reported for the CMIP models as reported in the present manuscript. Stevens (2015) revisited the quantities employed by Charlson et al in their 1992 study, specifically including sulfate lifetime, for which he accepted the results from the model studies taking the value of 3.8 days.



Tropospheric residence times in [days] in the AeroCom models for the species under consideration. Textor et al., 2006.

Whether the short lifetime presented by the authors in the present analysis (about 1 day) is an accurate representation of the lifetime of sulfate in the CMIP models or is erroneous due to their definition cannot be discerned from the manuscript. But it would seem that without further justification of Eq 1, that definition would seem to be fatal to the analysis presented.

Given the concerns raised above I conclude my review here.

References

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