Text S1

**The Effect of Volcanic sulfate aerosols in PI Control**

The PI control simulation is designed to establish the baseline climate for historical transient simulations (Schmidt et al., 2012). In this study, V2-IVA-NPI ensemble experiments are conducted to explore the effect of volcanic sulfate aerosols in the PI-control configuration on simulated historical climate (Table 3). The hypothesis is that without including historical averaged volcanic sulfate aerosols in the PI control, the additional sulfate aerosol emissions from historical volcanic eruptions would contribute to enhancing aerosol-cloud interactions and cooling of the climate. Additionally, without the inclusion of volcanic sulfate aerosols in the PI control, the volcanic quiescent warming and surplus cooling effects cannot be represented. The V2-IVA-NPI experiment replicates the setup of the V2-IVA experiment, but it omits the historical averaged explosive volcanic sulfate aerosols in the V2-IVA-NPI's PI control run, which serves as the initial condition for the V2-IVA-NPI's historical run. Although both V2-IVA and V2-IVA-NPI experiments have the identical emissions in the historical run, we anticipate that the anomaly in V2-IVA-NPI, relatives to its 1850-1899 climatology, result in more low clouds, an enhancement of cooling via aerosol-cloud interactions, and a cooler climate compared to its V2-IVA counterpart.

Table S1 presents a comparison between the V2-IVA and V2-IVA-NPI experiments. During the 1940-1959 period, both V2-IVA and V2-IVA-NPI show an increase in low cloud anomaly compared to their 1850-1899 climatology. Notably, the increase in low cloud fraction in V2-IVA-NPI has been significantly higher than that in V2-IVA. Additionally, the V2-IVA-NPI simulation has simulated a larger liquid water path anomaly, compared to that in V2-IVA. Consequently, the net cloud forcing anomaly in V2-IVA-NPI is 0.10 W/m² cooler than that in V2-IVA, indicating a cooling from aerosol-cloud interactions. Furthermore, the V2-IVA-NPI simulation exhibits a temperature anomaly that is 0.09 K cooler than that in V2-IVA, which is statistically significant. A similar, albeit less pronounced, pattern is observed during the 1960-1979 period, with V2-IVA-NPI simulating a cooler climate compared to that in V2-IVA. In general, the results are qualitatively agree with previous study about the importance of the volcanic sulfate aerosols in PI control simulation (Chim et al., 2023; Schmidt et al., 2012).

Table S1. Same as Table 4 but for comparison between V2-IVA and V2-IVA-NPI.

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| V2-IVA - V2-IVA-NPI | 1940-1959 | 1960-1979 |
| Net Cloud Forcing (W/m2) | 0.099 (-0.611, -0.710) | 0.016 (-0.985, -1.001) |
| Low Cloud Fraction (%) | -0.133\* (0.131, 0.265) | -0.032 (0.369, 0.401) |
| Cloud Liquid Water Path (kg/m2) | -9.65 E-05 (9.73E-4, 1.07E-3) | -1.03E-4 (1.48E-3, 1.58E-3) |
| Surface T (oK) | 0.089\* (0.004, -0.084) | 0.052 (-0.190, -0.242) |

**Reference**

Chim, M. M., Aubry, T. J., Abraham, N. L., Marshall, L., Mulcahy, J., Walton, J., and Schmidt, A.: Climate Projections Very Likely Underestimate Future Volcanic Forcing and Its Climatic Effects, Geophysical Research Letters, 50, e2023GL103743, https://doi.org/10.1029/2023GL103743, 2023.

Schmidt, A., Carslaw, K. S., Mann, G. W., Rap, A., Pringle, K. J., Spracklen, D. V., Wilson, M., and Forster, P. M.: Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate, Atmospheric Chemistry and Physics, 12, 7321–7339, https://doi.org/10.5194/acp-12-7321-2012, 2012.