

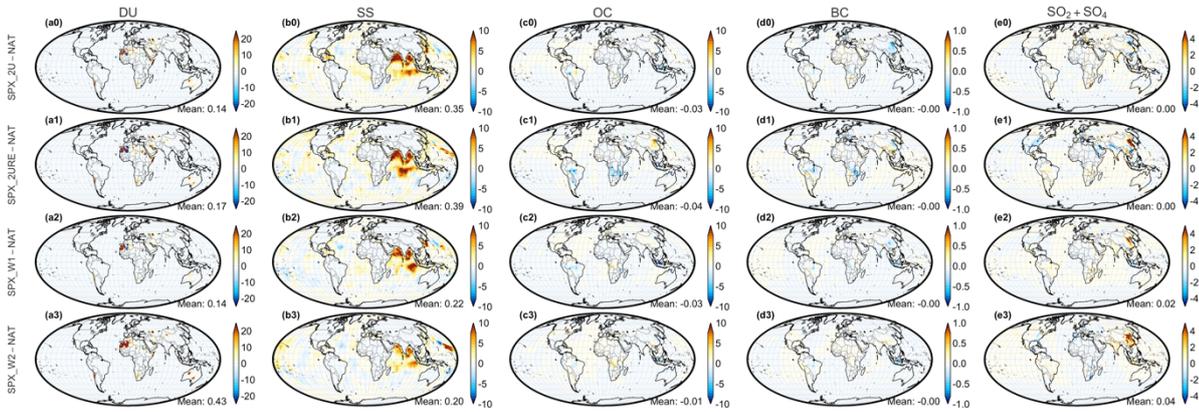
Supplement for the manuscript: Estimating aerosol emission from SPEXone on the NASA PACE mission using an ensemble Kalman Smoother: Observing System Simulation Experiments (OSSEs)

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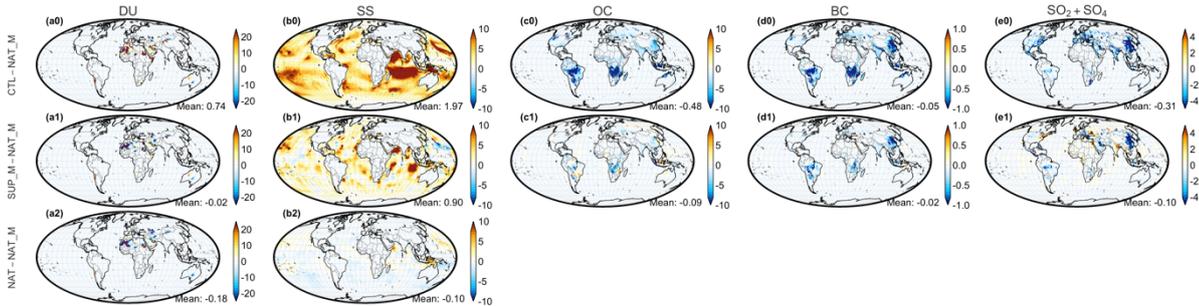
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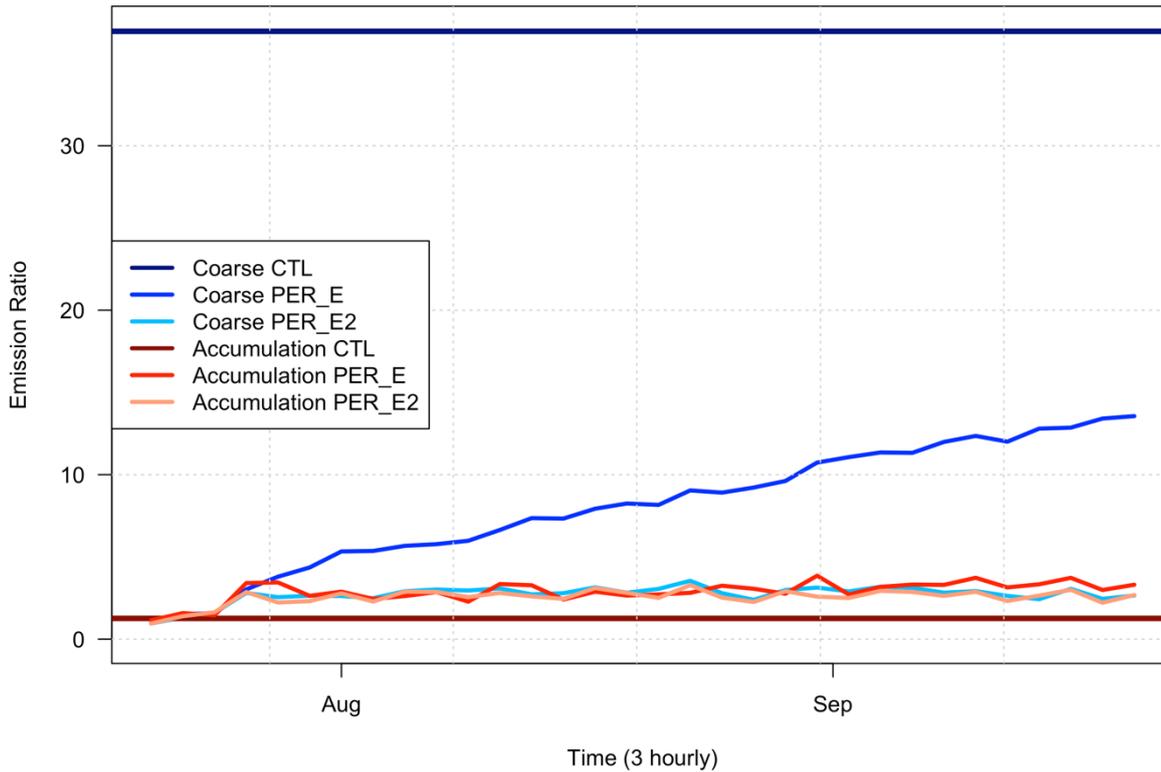
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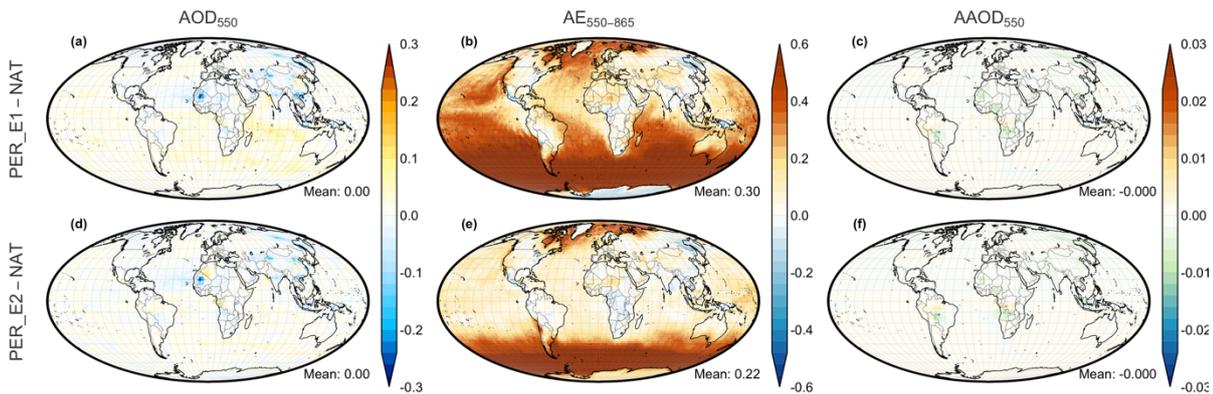
10 **Figure 1.** Aerosol emission ($\text{kg km}^{-1} \text{day}^{-1}$) differences of three data assimilation experiments from NAT for various species (a) DU, (b) SS, (c) OC, (d) BC, (e) SO_2+SO_4 . The subplots of the first, second and third row depict the differences between SPX_2U – NAT, SPX_2URE – NAT, SPX_W1 – NAT and SPX_W2 – NAT respectively.



15 **FigureS 2.** Aerosol emission ($\text{kg km}^{-1} \text{day}^{-1}$) differences by species (a) DU, (b) SS, (c) OC, (d) BC, (e) SO_2+SO_4 . The subplots of the first, second and third row depict the differences between CTL – NAT_M, SUP – NAT_M and NAT – NAT_M (ERA-5 – ERA-I) respectively. Note that the differences of the last row indicates changes in aerosol emissions only due to different meteorology, hence only the emissions of DU and SS are different, since the emissions of other species are not dependent on meteorological parameters.

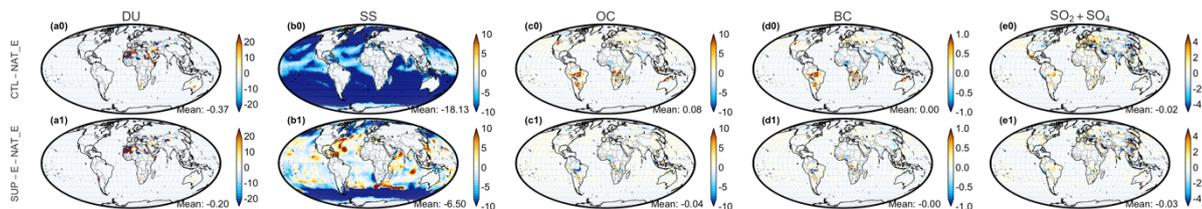


20 **FigureS 3.** Timeseries of sea salt emission ratio NAT_E to CTL over high latitudes in the southern hemisphere ($<-55^\circ$). Coarse and
 accumulation mode are shown with the dark-blue and the dark-red bold horizontal lines. The perturbations used for the coarse and
 25 the accumulation mode assimilation experiment SUP_E and SUP_E2 are shown with the fluctuating (in time) lines. SUP_E2 is
 identical to SUP_E with the difference that it doesn't use the prior correction option. Coarse mode perturbations for SUP_E are
 slowly rising during the course of the experiment, although it needs more time to reach the very high SS coarse emission of NAT_E
 which are 36 times greater than CTL. At the same time accumulation mode perturbations remain low and prior correction option
 does not affect negatively their estimation.



30 **FigureS 4.** Aerosol optical properties differences of SUP_E1 - NAT_M (a,b,c) and SUP_E2 - NAT_M (d,e,f). Left column depict
 AOD (a,d), middle column AE (b,e) and right column AAOD (c,f). PER_E1 emissions are perturbed by species, while PER_E2

emissions are perturbed by species and by size mode for DU and SS. PER_E (discussed in the main paper) is the succeeding experiment where emission perturbations were perturbed by species and by size mode for DU and SS, while prior correction was also enabled.



35 **FigureS 5.** Aerosol emission ($\text{kg km}^{-1} \text{day}^{-1}$) differences of two data assimilation experiments from NAT_E for various species (a) DU, (b) SS, (c) OC, (d) BC, (e) $\text{SO}_2 + \text{SO}_4$. The subplots of the first and the second row depict the differences between CTL – NAT_E and SUP_E – NAT_E respectively.