sea: absolute difference upwelling longwave flux at TOA – so2-at-height surface flux surface flux surface concentration surface concentration surface concentration of BC - so2-at-height of SO2 - so2-at-height of BC - so2-at-height of SO4 - so2-at-height of SO2 - so2-at-height 6.9e-21  $\Delta\,\mathrm{emibc}\,(\mathrm{kg}\,\mathrm{m}^{-2}\,\mathrm{s}^{-1})$  $^{-2} \, \mathrm{s}^{-1}$ ∆ mmrbc (kg kg-1)  $\Delta \cos (kg kg - 1)$ ∆ mmrso4 (kg kg- $\Delta$  rlut (W m – 2) 3.7e-21 0e+00 1e-13 5.0e-2 00+00 2e-14 -2.7e-2 2002 2003 2000 2002 2003 2002 2003 2000 2001 2004 2001 2004 2000 2001 2002 2003 2004 2000 2001 2002 2003 2004 2000 2001 2003 2004 2000 2001 2002 2004 Year Year Year Year upwelling shortwave flux at TOA – so2–at–height upwelling clear-sky longwave flux at TOA - so2-at-height upwelling clear-sky shortwave flux at TOA - so2-at-height clear-sky net radiative flux at TOA - so2-at-height net radiative flux incident shortwave flux at TOA - so2-at-height at TOA – so2–at–height 1e-01  $rsut (W m^{-2})$ rsutcs (W m-2) 0e+00 rlutcs (Wm-2) $\Delta \operatorname{rsut} (W m - 2)$  $\Delta \operatorname{rsdt}(\operatorname{Wm}-2)$ ≥ -2e-01 ∆ rlut + -3e-01 2000 2001 2002 2003 2000 2001 2002 2003 2000 2001 2002 2003 2000 2002 2003 2002 2003 2002 2003 2004 Year Year Year Year Year Year dry deposition rate of SO2 – so2–at–height implied cloud response dry deposition rate wet deposition rate total deposition rate rlutcs – rsutcs (W  $\mathrm{m}^{-2}$ ) wet deposition rate at TOA - so2-at-height of BC - so2-at-height of BC - so2-at-height of BC - so2-at-height of SO2 - so2-at-height wetbc (kg  $m^{-2} s^{-1}$ ) 7.7e-16 1.6e-15 wetbc  $(kg m^{-2} s^{-1})$  $\frac{1}{100}$  dryso2 (kg m<sup>-2</sup> s<sup>-1</sup>)  $\mathrm{drybc}\,(\mathrm{kg}\,\mathrm{m}^{-2}\,\mathrm{s}^{-1})$ 0e+00 ķ 1.0e-13 0e+00 -1e-01 5 0e-14 -2.0e-10 ∆ drybc ∆ rlut + 2003 2004 2002 2002 2003 2004 2002 2003 2003 2001 2003 2004 2000 2001 2004 2000 2001 2002 2000 2001 2002 2000 2001 2002 2003 2004 2000 2000 2001 Year Year Year Year Year dry deposition rate of SO4 – so2–at–height wet deposition rate total deposition rate ambient aerosol optical total cloud cover - so2-at-height convective cloud cover - so2-at-hei of SO4 - so2-at-height of S - so2-at-height thickness at 550nm - so2-at-heigh  $\frac{dryso2 + wetso2}{2 + (dryso4 + }$  $\Delta \, dryso4 \, (kg \, m^{-2} \, s^{-1})$ wetso4  $(kg m^{-2} s^{-1})$ 3e+35 (percent) ∆ clt (percent)  $(kg m^{-2} s^{-1})$ 1.2e-13 -2e-02 -4e-02 ∆ cltc ( 5 0e-13 4.0e-1 -5e-022001 2002 2003 2004 2000 2002 2003 2002 2003 2004 2000 2001 2002 2003 2002 2003 2000 2001 2002 2003 Yea Year Year Year Year Year surface cloud cover - so2-at-heigh ice water path - so2-at-height surface concentration column mass burden column mass burden column mass burden of DMS - so2-at-height of BC - so2-at-height of SO2 - so2-at-height of SO4 - so2-at-height 0e+00 8e-07  $\log \log (kg m^{-2})$ 0.0e + 00 $\Delta$  loadbc (kg m<sup>-2</sup>)  $\Delta$  clivi (kg m<sup>-2</sup>) ∆ cl (percent) loadso4 (kg m 6e-07 -1e-02 ∆ dms (kg kg-1e-04 1 0e-12 -4.0e-10 -2e-02 -8.0e-102e-07 1e-07 0.0e+00 2000 2001 2002 2003 2004 2000 2002 2003 2000 2002 2003 2000 2002 2003 2000 2002 2000 2002 2003 2001 2004 2001 2004 2001 2001 2003 2004 2001 2004 2004 Year Year Year Year Year SO4 lifetime - so2-at-height SO2 timescale - so2-at-height 1.2e+0 ∆ loadso2/emiso2 (days) 4e+08 3e + 08A loadso4/(dryso4+ 2001 2002 2003 2001 2002 2003 Year Year CAM5 E3SM GISS OsloCTM3 - UKESM - CESM1 -GEOS - MIROC

CESM2

-GFDL

NorESM2