sea: absolute difference surface concentration of SO4 – high–so4 upwelling longwave flux at TOA – high–so4 surface flux surface flux surface concentration surface concentration of BC - high-so4 of SO2 - high-so4 of BC - high-so4 of SO2 - high-so4 6.3e-21 0.0e + 00 $\Delta$  emiso2 (kg m<sup>-2</sup> s<sup>-1</sup>)  $\Delta\,\mathrm{emibc}\,(\mathrm{kg}\,\mathrm{m}^{-2}\,\mathrm{s}^{-1})$ ∆ mmrbc (kg kg-1)  $\Delta \cos (kg kg - 1)$  $\Delta rlut (Wm-2)$ 2.7e-21 ∆ mmrso4 (kg kg -1.0e-01 2.5e-1 1e-13 -8.2e-22 5.0e-02 2.0e-1 -9.0e-12 -8 0e-14 \_4 4e\_2 -1.2e-1 2003 2004 2000 2001 2003 2004 2001 2002 2003 2004 2002 2003 2002 2003 2004 2002 2003 2004 2002 2000 2000 2001 2004 2000 2001 2000 2001 2000 2001 2002 Year Year Year upwelling shortwave flux at TOA – high–so4 incident shortwave flux at TOA – high–so4 net radiative flux at TOA – high–so4 upwelling clear–sky longwave flux at TOA – high–so4 upwelling clear-sky shortwave flux at TOA - high-so4 clear-sky net radiative flux at TOA - high-so4 0.0e+00  $rsut (W m^{-2})$ 0e+00  $\Delta$  rsutcs (W m – 2) 0e+00  $\Delta \operatorname{rsut}(\operatorname{Wm}-2)$  $\Delta rsdt (Wm-2)$ ≥ ∆ rlutcs (W m-00+00 5.0e-08 -1.0e-01 0.0e+00 -1.0e-0 -5.0e-08 ∆ rlut -2.0e-0 -4e-01 -1.0e-07 2000 2001 2002 2003 2002 2003 2002 2003 2002 2003 2002 2003 2001 2002 2003 2004 2000 Year Year Year Year Year Year dry deposition rate of BC – high–so4 wet deposition rate of BC – high–so4 total deposition rate of BC – high–so4 implied cloud response dry deposition rate wet deposition rate  $rlutcs-rsutcs\left(W\,m^{-2}\right)$ of SO2 - high-so4 at TOA – high-so4 of SO2 – high-so4 wetbc (kg  $m^{-2} s^{-1}$ ) 2.4e-15 8.6e-16 1.6e-15 1e-01 0e+00 wetbc (kg  $m^{-2}$  s<sup>-1</sup>)  $dryso2 (kg m^{-2} s^{-1})$ drybc (kg  $m^{-2} s^{-1}$ ) 0e+00 -6e-14 vetso2 ( -3e-14 -6.6e-10 ∆ drybc ∆ rlut + 2002 2003 2004 2003 2004 2003 2004 2002 2003 2004 2001 2002 2003 2000 2002 2000 2001 2000 2001 2000 2004 2001 2000 2001 2002 2002 2003 2004 2000 2001 Year Year Year Year Year Year dry deposition rate of SO4 – high–so4 total deposition rate of S – high–so4 wet deposition rate ambient aerosol optical total cloud cover - high-so4 convective cloud cover - high-so4 of SO4 – high–so4 thickness at 550nm - high-so4 8e-02 dryso2 + wetso2)/2 + (dryso4 +0e+00  $\Delta$  dryso4 (kg m<sup>-2</sup> s<sup>-1</sup>) wetso4  $(kg m^{-2} s^{-1})$ (percent) ∆ clt (percent) 1.2e-13  $(kg m^{-2} s^{-1})$ -2e-02 ∆ od550aer 0e+00 4e-14 -4e-02 8.0e-14 ∆ cltc ( 2e-02 -6e-02 4.0e-14 -2e-14 0e+00 0e+00 2000 2001 2002 2003 2004 2000 2001 2002 2003 2000 2001 2002 2003 2004 2000 2001 2002 2003 2000 2001 2002 2003 2000 2001 2002 2003 Year Year Year Year Year Year surface cloud cover - high-so4 ice water path - high-so4 surface concentration column mass burden column mass burden column mass burden of DMS - high-so4 of BC - high-so4 of SO2 - high-so4 of SO4 - high-so4 4e-02  $\log \log (kg m^{-2})$ 4e-07  $\Delta$  dms (kg kg – 1)  $\Delta$  loadbc (kg m<sup>-2</sup>) clivi (kg m<sup>-2</sup>) ∆ cl (percent) 2e-02 1.5e-12 1.0e-12 2e-07 5.0e-13 1e-07 -4e-08 -2e-02 0.0e+00 -2e-04 -8e-10 2000 2001 2002 2003 2004 2000 2001 2002 2003 2004 2002 2003 2004 2000 2001 2002 2003 2002 2003 2000 2001 2002 2003 2004 2000 2001 2004 2000 2001 2004 Year Year Year Year Year ∆ loadso4/(dryso4 + wetso4) (days) SO4 lifetime - high-so4 SO2 timescale - high-so4 ∆ loadso2/emiso2 (days) 2e+01 4e+00 1e+01 2e+00 2000 2002 2003 2000 2001 2002 2003 Year Year CAM5 E3SM GISS OsloCTM3 - UKESM - CESM1 -GEOS - MIROC

CESM2

-GFDL

NorESM2