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Interannual variability and long-term trends in global tropospheric ozone and related chemistry during recent decades

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Global distributions and abundances of tropospheric constituents (O_3, CH_4, NO_y, CO, VOCs, NHx, SOx and aerosols) inter-annually change under the influences of meteorology (transport, temperature, water vapor, clouds, rain, etc.) and emissions from anthropogenic/natural sources and biomass burning. Given the importance of climate effects of these species as short-lived climate forcers/pollutants (SLCFs/SLCPs), there have been an increasing number of studies to project future changes in individual constituents and assess impacts of emissions reduction in future. Since chemistry climate models are used for such purposes, model validation against the observations and precise interpretation/understanding of changing processes in a model are essentially needed. In this study, we investigate the inter-annual variability and long-term trend of tropospheric constituents during the years 1980 to 2014 in a chemistry-aerosol coupled climate model. The base chemical model used in this study is CHASER coupled with the aerosol model SPRINTARS. CHASER, also developed in the framework of the MIROC Earth system model (MIROC-ESM), simulates detailed chemistry in the troposphere and stratosphere with an on-line aerosol simulation including production of particulate nitrate and SOA. For the simulations, anthropogenic and biomass burning emissions are specified using the MACCity/MACC inventories. Our results show that temporal variability (anomaly) in surface and lower tropospheric ozone very clearly correlates with that in CO, especially in NH (r >0.8), indicating the principal importance of biomass burning emissions in determining near-surface O_3 variability; surface PM (PM2.5) in NH also well coincides with CO. Changes in middle to upper tropospheric O_3, on the other hand, respond to variability/trends in water vapor, transport from the stratosphere (STE), and lightning NO_x production associated with climate change/variability (inc. ENSO) and/or stratospheric O_3 change. It is also demonstrated that the inter-annual variability and long-term trend in tropospheric mean OH concentration (and hence global mean CH_4 concentration) is largely controlled by tropospheric abundances of O_3, NO_x, and water vapor. In simulations, the emission-driven long-term increase in global mean CH_4 ($\sim$350 ppbv for 1980-2010) is nearly halved ($\sim$170 ppbv) by OH increases due to changes in climate and NO_x emission.