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Long-term trends and periodical variations of surface ozone and their influencing factors at the Mt. Waliguan GAW station, China

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Tropospheric ozone is an important atmospheric oxidant, greenhouse gas and atmospheric pollutant at the same time. Long-term trends of baseline ozone is highly needed information for environmental and climate change assessment. The variation characteristics, long-term trends and periodicities of surface ozone measured at a global baseline Global Atmospheric Watch (GAW) station in the north-eastern Tibetan Plateau region (Mt. Waliguan, 36˚17' N, 100˚54' E, 3816m a.s.l.) for the period of 1994 to 2013 were uncovered using the various trend tests and the Hilbert-Huang Transform (HHT) analysis. Significant positive trends were detected for both daytime (0.24±0.16 ppbv yr^{-1}) and nighttime (0.28±0.17 ppbv yr^{-1}) ozone. The largest nighttime increasing rate occurred in autumn (0.29±0.11 ppbv yr^{-1}), followed by spring (0.24±0.12 ppbv yr^{-1}), summer (0.22±0.20 ppbv yr^{-1}) and winter (0.13±0.10 ppbv yr^{-1}), respectively. The HHT spectral analysis identified 2-4, 7 and 11 year periodicities. The factors driving the observed ozone changes at WLG were explored using backward trajectory analysis, chemistry-climate model hindcast simulations (GFDL-AM3), a trajectory-mapped ozonesonde dataset and several climate indices. A stratospheric ozone tracer implemented in GFDL-AM3 indicates that stratosphere-to-troposphere transport (STT) can explain ~60% of the simulated springtime ozone increase at WLG, consistent with an increase in the NW air mass frequency inferred from the trajectory analysis. Enhanced STT associated with the strengthening of the mid-latitude jet stream contributes to the observed high-ozone anomalies at WLG during the springs of 1999 and 2012. During autumn, observations at WLG are more heavily influenced by polluted air masses originated from Southeast Asia than in the other seasons. Rising Asian anthropogenic emissions of ozone precursors is the key driver of increasing autumnal ozone observed at WLG, as supported by the GFDL-AM3 model with time-varying emissions, which captures the observed ozone increase (0.26±0.11 ppbv year^{-1}). AM3 simulates a greater ozone increase of 0.38±0.11 ppbv year^{-1} at WLG in autumn under conditions with strong transport from Southeast Asia and shows no significant ozone trend in autumn when anthropogenic emissions are held constant in time. During summer, WLG is mostly influenced by easterly air masses but these trajectories do not extend to the polluted regions of eastern China and have decreased significantly over the last two decades, which likely explains why summertime ozone measured at WLG shows no significant trend despite ozone increases in Eastern China. Analysis of the Trajectory-mapped Ozonesonde dataset for the Stratosphere and Troposphere (TOST) and trajectory residence time reveals increases in direct ozone transport from the eastern sector during autumn, which adds to the autumnal ozone increase. We further examine the links of ozone variability at WLG to the QBO, the East Asian summer monsoon (EASM) and the sunspot cycle and found that they are respectively linked to the 2-4, 7 and 11 year periodicities. A multivariate regression analysis is performed to quantify the relative contributions of various factors to surface ozone concentrations at WLG. Through an observational and modelling analysis, this study demonstrates the complex relationships between surface ozone at remote locations and its dynamical and chemical influencing factors.