

# Regional Source Apportionment of Summertime Ozone and Its Precursors in the Megacities of Beijing and Shanghai using a Source-Oriented Chemical Transport Model

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The Community Multiscale Air Quality (CMAQ) model with a source-oriented SAPRC-11 photochemical mechanism is developed in this study to quantify the source region contributions to surface O<sub>3</sub> in Beijing and Shanghai in August 2013. Non-background O<sub>3</sub> attributed to NO<sub>x</sub> (O<sub>3</sub>-NO<sub>x</sub>) and VOCs (O<sub>3</sub>-VOC) emitted from different source regions are determined using a three-regime approach that correctly attributes O<sub>3</sub> to NO<sub>x</sub> and VOC precursors throughout the entire range of NO<sub>x</sub>-VOC-O<sub>3</sub> formation sensitivity. Averaged over the entire month and all grid cells, local emissions (51%) and emissions from Hebei (31%) are the two major contributors to non-background daily maximum 8-hour (DM8H) O<sub>3</sub> in Beijing. In Shanghai, local, Zhejiang and Jiangsu emissions account for 53%, 19% and 14% of the non-background DM8H O<sub>3</sub>. Significant variations in local emission contributions are predicted among different model grid cells for both cities (Beijing, 6-80%; Shanghai, 3-76%). On high O<sub>3</sub> days in Beijing, the wind is persistently from the south with high wind speed ( $\sim 5 \text{ m s}^{-1}$ ) in the evening and night. This leads to significant regional contributions of O<sub>3</sub> from Hebei, along with regional transport of VOCs and NO<sub>x</sub>. In Shanghai, high O<sub>3</sub> days are associated with southwesterly/westerly wind in the morning, rotating to southeast in the early afternoon in a counter-clockwise direction. The surface wind then gradually turns back to southwest in the afternoon until the next morning, along with reduced wind speed. In Shanghai, daytime O<sub>3</sub> at the urban center is almost entirely due to local emissions. Low wind speed in the evening and night time allows local NO<sub>x</sub> emissions to efficiently titrate regional O<sub>3</sub>. In both cities, NO<sub>x</sub> emissions are not transported regionally as efficiently as VOCs. Source region contribution analysis of the concentration weighted maximum incremental reactivity (CWMIR) shows that VOCs from other regions are less reactive than locally emitted VOCs. HCHO and acetaldehyde (CCHO) generated from the oxidation of other VOCs are important contributors to regionally transported reactive VOCs. In both regions, the overall CWMIR in both cities is quite similar ( $\sim 4 \text{ moles O}_3 \text{ per mole of VOC}^{-1}$ ).

Keywords: photochemical model, regional transport, nitrogen oxides, volatile organic compounds, emission