SEASONAL AND REGIONAL VARIATIONS OF PRIMARY AND SECONDARY ORGANIC AEROSOLS OVER THE CONTINENTAL UNITED STATES: OBSERVATION-BASED ESTIMATES AND MODEL EVALUATION

Shaocai Yu[+$], Robin L. Dennis[+], Prakash V. Bhave[+] and Rohit Mathur[+]
Atmospheric Sciences Modeling Division, Air Resources Laboratory,
National Oceanic and Atmospheric Administration, RTP, NC 27711
[+]On assignment to National Exposure Research Laboratory,
U.S. Environmental Protection Agency, Research Triangle Park, NC 27711
[$] On assignment from Science and Technology Corporation,
10 Basil Sawyer Drive, Hampton, VA 23666-1393
e-mail: yu.shaocai@epa.gov
Voice (919) 541-0362 Fax (919) 541-1379

Due to the lack of an analytical technique for directly quantifying the atmospheric concentrations of primary (OC_{pri}) and secondary (OC_{sec}) organic carbon aerosols, different indirect methods have been developed to estimate their concentrations. In this study, seasonal and regional variations of OC_{pri} and OC_{sec} over the continental U.S. for the year 2001 were estimated by using observed OC and elemental carbon (EC) data from Interagency Monitoring of Protected Visual Environments (IMPROVE) and Southeastern Aerosol Research and Characterization (SEARCH) networks, coupled with the primary OC/EC ratios, $(OC/EC)_{pri}$, obtained from an emission/transport-model (US EPA Models-3/Community Multiscale Air Quality (CMAQ) model) (i.e., emission/transport of primary OC/EC ratio method). These observation-based estimates show that the yearly mean OC_{pri} concentrations vary greatly by location from 0.27 μg C m^{-3} to 3.72 μg C m^{-3} (0.71±0.82 μg C m^{-3}, mean±standard deviation), while the yearly mean OC_{sec} concentrations vary from 0.16 μg C m^{-3} to 1.76 μg C m^{-3} (0.61±1.04 μg C m^{-3}). Seasonal analysis shows that OC_{sec} concentrations are 0.34±0.62, 0.47±0.86, 0.80±1.03, and 0.72±1.30 μg C m^{-3} in winter, spring, summer, and fall, respectively, making 30, 41, 51, and 42% contributions to OC respectively. Regional analysis indicates that both OC_{pri} and OC_{sec} concentrations are the highest over the southeast area (yearly mean OC_{pri}: 1.00±1.23 μg C m^{-3} and OC_{sec}: 1.25±0.96 μg C m^{-3}) in all seasons. It is found that OC_{pri} accounts for a large fraction of OC over the Northeast, Southeast, Midwest, West and Midwest (>56%) in all seasons except summer, during which OC_{pri} and OC_{sec} concentrations make approximately equal contributions to OC. On the basis of comparison of OC_{pri} and OC_{sec} at the nearest paired urban/rural sites from SEARCH, the yearly mean OC_{pri} concentrations at the urban sites contribute 71 to 80% to OC, larger than the OC_{pri} contributions at the corresponding paired rural sites (54 to 74%). In contrast, the yearly mean OC_{sec} concentrations at rural sites frequently exceed those at the nearby urban sites. The higher OC_{sec} concentrations during the summer at the three rural sites result in the higher yearly contributions of OC_{sec} to OC (41 to 46%) than at the corresponding paired urban sites (20 to 29%).

The comparisons of the model and observation-based estimates show that over the continental United States, the modeled OC_{sec} concentrations are 0.49±0.45, 0.67±0.59, 0.96±0.99, 1.06±1.03, and 0.84±0.87 μg C m^{-3} in winter, spring, summer, fall and year, respectively, slightly higher than the observation-based concentration estimates listed above. The modeled OC_{pri} concentrations are 0.74±0.96, 0.58±0.81, 0.86±1.02, 0.69±0.63, and 0.72±0.86 μg C m^{-3} in winter, spring, summer, fall and year, respectively, very close to those of the observations which are 0.70±0.82, 0.60±0.58, 0.73±1.00, 0.78±0.80, and 0.71±0.82 μg C m^{-3}.

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