Long-range transport of black carbon and its dependence on aging timescale

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Improving the ability of global models to predict concentrations of black carbon (BC) is essential to evaluating the impact of BC on climate. We find that BC burdens around the globe, as well as source-receptor relationships, are remarkably sensitive to the assumed aging timescale for BC in the model. (The aging timescale describes the e-folding time for BC to be converted from hydrophobic to hydrophilic.) This motivates our use of HIPPO observations to optimize BC aging timescale by minimizing model-measurement differences.

In this study, we tag BC tracers from 13 source regions around the globe in a global chemical transport model, MOZART-4. Numerous sensitivity simulations are carried out varying the aging timescale of BC emitted from each source region. The aging timescale for each source region is optimized by minimizing errors in vertical profiles of BC mass mixing ratios between simulations and HIAPER Pole-to-Pole Observations (HIPPO). For most HIPPO deployments, in the Northern Hemisphere, optimized aging timescales are less than half a day for BC emitted from tropical and mid-latitude source regions, and about 1 week for BC emitted from high latitude regions in all seasons except summer. We find that East Asian emissions contribute most to the BC loading over the Northern Pacific Ocean, while South American, African and Australian emissions dominate BC loadings over the Southern Pacific Ocean. Dominant source regions contributing to BC loadings in other parts of the globe are also assessed. The lifetime of BC originating from East Asia (i.e., the world's largest BC emitter) is found to be only 2.2 days, shorter than the global average lifetime of 4.9 days. East Asia's contribution to global BC burden is found to be 64% lower than from the second largest emitter, Africa. Thus, evaluating only relative emission rates without accounting for differences in aging timescales and deposition rates is not predictive of the contribution of a given source region to climate impacts. Our simulations indicate that lifetime of BC increases nearly linearly with aging timescale for all source regions. When aging rate is fast, the lifetime of BC is largely determined by factors that control local deposition rates (e.g. precipitation). The sensitivity of lifetime to aging timescale depends strongly on the initial hygroscopicity of freshly emitted BC. Our findings suggest that the aging timescale of BC varies significantly by region and season, and can strongly influence the contribution of source regions to BC burdens around the globe. Improving parameterizations of the aging process for BC is important for enhancing the predictive skill of air quality and climate models. Future observations that investigate the evolution of hygroscopicity of BC as it ages from different source regions to the remote atmosphere are urgently needed.