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A Lagrangian view of ozone production tendency in North American outflow in summers 2009 and 2010

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The Pico Mountain Observatory, located at 2,225 ma.s.l. in the Azores Islands, was established in 2001 to observe long-range transport from North America to the central North Atlantic. In previous research conducted at the Observatory, ozone enhancements (> 55 ppbv) were observed in North American outflows containing anthropogenic and biomass burning emissions, and efficient ozone production in these outflows was postulated. One of the major objectives of BORTAS is to better understand chemical composition and evolution during transport of biomass burning outflows. A key to the study of pollution plumes at a ground-based station is identification of emission type and source region(s). Transport pathways of individual plumes are also thought to be critical to plume aging. In this study, by analyzing observations of atmospheric tracer gases at Pico and FLEXPART simulation results, we were able to identify transport events induced by anthropogenic or biomass burning emissions during summers 2009 and 2010. In order to assess ozone production tendency during these long-range transport events, the convolved or "folded" retroplume technique developed by Owen and Honrath (2009) was applied to combine upwind FLEXPART transport pathways with GEOS-Chem chemical fields, providing a semi-lagrangian view of physical properties and production/loss of ozone in polluted North American outflows. Two anthropogenic events from North America were selected for detailed analysis because anthropogenic emissions were considered to be more predictable and consistent over time. Ozone enhancement was observed in both plumes, but due to differing transport mechanisms, ozone production tendency was found to be different between the two. In the first case, ozone production was found during the last two days of transport, when the pollution plume subsided from the free troposphere to the altitude of Pico station in the high pressure system centered over the Azores region at the time. Increase of temperature during the subsidence prompted thermal decomposition of peroxyacetyl nitrate, and consequently, a net ozone production layer ($\sim$2 ppbv/day) was formed at 2 km a.s.l. over the Azores area. In the second case, net ozone production was absent during transport over the North Atlantic, however the plume was transported at low altitude and carried elevated concentrations of ozone to Pico. Modeled non-methane hydrocarbon (NMHC) aging in plumes suggested sufficient performance of the folded GEOS-Chem and FLEXPART technique to diagnose the extent of mixing vs. chemical reaction in determining NMHC ratios. In biomass burning outflows, ozone production tendency may be more complicated due to heterogeneous chemical reactions. Biomass burning outflows will be the subject of future folded retroplume analysis.

Reference

Owen, R. C., and Honrath, R. E. (2009). Technical note: a new method for the Lagrangian tracking of pollution plumes from source to receptor using gridded model output. Atmos. Chem. Phys., 9(7), 2577-2595.