



Factors controlling pollutant plume processing in the lower troposphere

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In summer 2004, intense anthropogenic polluted plumes from Boston and New York City were observed during the IGAC Lagrangian 2K4/ European ITOP (Intercontinental Transport of Ozone and Precursors) campaign. All these plumes were highly polluted and some of them showed very high levels of HNO₃ (up to 40 ppbv). One of these high HNO₃ level plumes was tracked as it moved over northeast North America, and was sampled, by the NOAA-P3 aircraft, on 3 subsequent days. The plume was then transported across the North Atlantic at low altitude and sampled again off the west coast of Ireland by the DLR-Falcon aircraft and possibly one day latter over the English channel. Due to these multiple Lagrangian samplings, the plume composition evolution over 6 days can be examined. The plume showed a decrease in HNO₃, CO and O₃ over the 5 first days.

A Lagrangian photochemical model, CiTTyCAT, was used to assess the phenomena influencing the chemical composition in the plume. The model was initialized with upwind measurements made in the plume and compared with downwind measurements. Several factors were found to be important in governing the chemical evolution of this plume. Large HNO₃ decreases during the first 3 days are almost certainly due to wet deposition. Several deposition schemes were tested and the most realistic results obtained using analyzed ECMWF precipitation data. This HNO₃ decrease contributes indirectly to decreasing O₃ levels because the slow production of O₃ through HNO₃ photolysis is important at high

HNO₃ levels. Dry deposition over North America also decreases O₃ levels in the plume. Photochemical processes were also found to be important for CO and O₃ levels. Overall, the plume was in a net O₃ destruction regime (mean net O₃ production rate of -5.7 ppbv/day) mainly due to high water vapor content. CO levels in the plume were also reduced due to high OH levels showing that CO cannot always be used as a tracer of pollution. Mixing processes were slow and did not have much influence on the chemical composition of the plume in this case. Comparison of multiple integrations with O₃/CO and NO_y/CO correlations confirms these results. Finally, all the processes (dry deposition, wet deposition and photochemistry) contributed to decrease O₃ levels in the plume, leading to a small impact over Europe.