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Source-receptor relationships for airborne measurements of CO_2, CO and O_3 above Siberia: a cluster-based approach

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Abstract. We analysed results of three intensive aircraft campaigns above Siberia (April and September 2006, August 2007) with a total of ~70 h of continuous CO_2, CO and O_3 measurements. The flight route consists of consecutive ascents and descents between Novosibirsk (55° N, 82° E) and Yakutsk (62° N, 129° E). We performed retroplume calculations with the Lagrangian particle dispersion model FLEXPART for many short segments along the flight tracks. To reduce the extremely rich information on source regions provided by the model calculation into a small number of distinct cases, we performed a statistical clustering – to our knowledge for the first time – into potential source regions of the footprint emission sensitivities obtained from the model calculations. This technique not only worked well to separate source region influences but also resulted in clearly distinct tracer concentrations for the various clusters obtained. High CO and O_3 concentrations were found associated with agricultural fire plumes originating from Kazakhstan in September 2006. A statistical analysis indicates that summer uptake of CO_2 is largely explained (~50% of variance) by air mass exposure to uptake by Siberian and sub-arctic ecosystems. This resulted in an average 5 to 10 ppm difference with overlaying air masses. Stratosphere-troposphere exchange is found to strongly influence the observed O_3 mixing ratios in spring, but not in summer. European emissions contributed to high O_3 concentrations above Siberia in April 2006 and August 2007, while emissions from North Eastern China also contributed to higher O_3 mixing ratios in summer, but tend to lower mixing ratios in spring, when the airmass aerosol burden is important. In the lower troposphere, large-scale deposition processes in the boreal and sub-arctic boundary layer is a large O_3 sink, resulting in a ~20 ppb difference with overlaying air masses. Lagrangian footprint clustering is very promising and could also be advantageously applied to the interpretation of ground based measurements including calculation of tracers' sources and sinks.

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