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Evidence for widespread tropospheric Cl chemistry in free tropospheric air masses from the South China Sea

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While the primary global atmospheric oxidant is the hydroxyl radical (OH), under certain circumstances chlorine radicals (Cl) can compete with OH and perturb the oxidative cycles of the troposphere. During flights between Bangkok, Thailand and Kuala Lumpur, Malaysia conducted over two fall/winter seasons (November 2012 – March 2013 and November 2013 – January 2014) the IAGOS-CARIBIC (www.caribic-atmospheric.com) observatory consistently encountered free tropospheric air masses (9-11 km) originating over the South China Sea which had non-methane hydrocarbon (NMHC) signatures characteristic of processing by Cl. These signatures were observed in November and December of both years, but were not seen in other months, suggesting that oxidation by Cl is a persistent seasonal feature in this region. These Cl signatures were observed over a range of $\sim$1500 km indicating a large-scale phenomenon.

In this region, where transport patterns facilitate global redistribution of pollutants and persistent deep convection creates a fast-track for cross-tropopause transport, there exists the potential for regional chemistry to have impacts further afield. Here we use observed relationships between NMHCs to estimate the significance and magnitude of Cl oxidation in this region. From the relative depletions of NMHCs in these air masses we infer OH to Cl ratios of 83$\pm$28 to 139$\pm$40 [OH]/[Cl], which we believe represents an upper limit, based on the technique employed. At a predicted average [OH] of 1.5×10^6 OH cm^{-3} this corresponds to an average (minimum) [Cl] exposure of 1-2$\times10^4$ Cl cm^{-3} during air mass transport. Lastly, in addition to estimating Cl abundances we have used IAGOS-CARIBIC observations to elucidate whether the origin of this Cl is predominantly natural or anthropogenic.