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Tropospheric halocarbons in Southeast Asia – levels, sources and delineation of regional and local-scale emissions

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Tropospheric halocarbons are important from both scientific and policy perspectives owing to their roles in stratospheric ozone depletion, global warming and radical-induced atmospheric chemistry. Following the implementation of the Montreal Protocol in 1989, long-term ground-based measurements at remote locations have reported gradually decreasing or stabilizing trends for tropospheric CFCs, CCl4 and CH3CCl3, showing the general effectiveness of the global phase-out. However, these trends insufficiently reflect the effectiveness of MP regulations at regional/national scales. Measurement evidence is therefore urgently needed at such scales, especially from Southeast (SE) Asia where the present understanding of halocarbon levels and sources is rudimentary at best.

To address this issue, we report here the first ground-based atmospheric measurements of 26 halocarbons in Singapore, an urban-industrial city-state in SE Asia, and discuss their levels, potential sources, and effects of policy interventions. To this end, we collected a total of 166 whole air canister samples during two intensive 7 Southeast Asian Studies (7SEAS) campaigns (August-October, 2011 and 2012) and analyzed C1-C2 halocarbons using gas chromatography-electron capture/mass spectrometric detection. We supplemented the halocarbon dataset with measurements of selected non-methane hydrocarbons (NMHCs), C1-C5 alkyl nitrates, sulfur gases and carbon monoxide to better understand sources and atmospheric processes. We utilized clustered air-mass backward trajectories to identify source sectors, and employed the receptor model positive matrix factorization (PMF) to quantitatively apportion halocarbon sources.

We report three major observations. First, we find that median atmospheric levels of CFCs, halons, CCl4 and CH3CCl3 in Singapore are close to global tropospheric backgrounds, with enhancements in the 1-17% range. This provides the first measurement evidence from SE Asia of the effectiveness of MP and related national-scale regulations instituted in the 1990s. We also find that first- and second-generation CFC replacements (HCFCs and HFCs) have subsequently emerged as the dominant halocarbon species with enhancements of 39-67%. In view of their high global warming potentials, the recent MP-mandated implementation of HCFC phase-out management plans (HPMPs) in SE Asian countries is both timely and justified. Second, by combining near-source measurements in Indonesia with receptor data in Singapore and clustered air trajectories, we conclude that regionally transported peat-forest burning smoke affects atmospheric levels of several NMHCs (ethane, ethyne, benzene, and propane) and short-lived halocarbons (CH3I, CH3Cl, and CH3Br) in a subset of the receptor samples. We note that the strong signatures of these species near peat-forest fires are diluted during atmospheric transport and are often masked by mixing with substantial urban/regional backgrounds from industrial, marine, and terrestrial biogenic sources at the receptor. Third, we conclude using PMF that industrial emissions related to refrigeration, foam blowing, and solvent use in chemical, pharmaceutical and electronics industries is the major source of halocarbons (34%) in Singapore, followed by regional marine and terrestrial biogenic activity (28%), residual levels from pre-MP operations (16%), seasonal incidences of peat-forest smoke (13%), and fumigation related to quarantine and pre-shipment (QPS) applications (7%). These findings provide a much-needed baseline for tropospheric halocarbon data in SE Asia and identify potential research directions for the future.