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Title: First Estimate of Ambient Hydroxyl Radicals in the NW-IGP Derived Using in-situ Measurements

of Aromatic VOCs and the Toluene/Benzene Photo-chemical Clock

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Abstract:

Aromatic volatile organic compounds, which are emitted primarily from anthropogenic sources are amongst the most reactive precursors of tropospheric ozone and secondary organic aerosol. Till date however, the ambient concentrations and diel variability of compounds such as benzene, toluene, xylenes and trimethylbenzenes remain poorly constrained over the Indian region. The concentration of ambient hydroxyl radicals that are the primary tropospheric oxidants and control the removal rates of climate and air quality relevant gases has also not been previously quantified at any Indian site. Here, we address the aforementioned gaps in our understanding related to ambient aromatic compounds and hydroxyl radicals using a novel in-situ VOC and meteorological dataset measured at a sub-urban site in the NW-IGP during April-May 2012. The observed concentrations (average $\pm 1\sigma$ ambient variability) were as follows: benzene (1.6 \pm 1.7 ppbV), toluene (2.7 ± 3.1 ppbV), xylenes (1.9 ± 2.3 ppbV) and trimethylbenzenes (1.1 ± 1.2 ppbV). These values were found to be comparable in magnitude to average summertime mixing ratios reported in Tokyo, (benzene: 1-4 ppbV, toluene: 3-9 ppbV, C8 aromatics: 1.3 ppbV, C9 aromatics: 0.3 ppbV) but in general higher than those reported in Houston, USA, (benzene: 0.6 ppbV, toluene: 0.8ppbv, xylene: 0.6 ppbV). The strongest sources of the aromatic VOCs at our site were found to be point sources (e.g. congested traffic intersections such as Tribune Chowk) located in the urban clusters of Chandigarh, Panchkula and Mohali. Analogous trends observed in the time series of benzene, toluene, C8 and C9 aromatics and a high-degree of inter-VOC correlation (r2 > 0.6) showed that common sources drive their emission profiles. By applying a chemical kinetics "photochemical clock" method, that exploits the differential OH reactivity of toluene and benzene, their measured emission ratios at the urban fetch region and the measurement site, we derived an average ambient hydroxyl radical concentration of (17 ± 3) × 106 molecules cm-3 or 0.68 ± 0.001 pptV. This suggests that the ambient oxidising capacity is similar to the oxidising capacity observed in sites at higher latitudes influenced by urban emissions such as New York where ambient hydroxyl radical concentrations of 20 × 106 molecules cm-3 were reported. As the ambient chemical mixture has high concentration of both reactive aromatic compounds and oxidants, the potential for chemical transformations and reactions of aromatic compounds to form aerosol and fuel ozone production locally on short time scales (few hours) appears to be particularly high and is important to consider for mitigation strategies.

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