Title:	Non-methane hydrocarbon (NMHC) source fingerprints, emissions, and ambient variability over north india quantified using thermal desorption-gas chromatography-flame ionization detection (TD-GC-FID)
Authors:	Kumar, Ashish
Keywords:	hydrocarbon desorption-gas
Issue Date:	Oct-2021
Publisher:	IISER Mohali
Abstract:	Air pollution is a major contemporary environmental threat deleterious effects on on human health and the economy. Poor air quality over north India is a priority area for mitigation by State and National regulatory authorities. As significant constituents of gaseous air pollution, the class of trace gases termed volatile organic compounds (VOCs) are key contributors to both primary and secondary air pollution through their atmospheric chemistry. They act as precursors for surface ozone pollution, peroxyacetylnitrate (PAN) and secondary organic aerosol (SOA), which are the key constituents of gaseous and particulate smog, which affect human health, visibility and crop yields. Reducing the emissions of VOCs therefore can significantly help in pollution mitigation and air quality improvement. However over India, there is still no national monitoring program for VOCs and hence knowledge of their sources and sinks are woefully inadequate. Fossil fuel usage and industries are known major emission sources all over the world but unregulated large-scale anthropogenic activities like crop residue burning and garbage burning which are majorly active in agrarian economies and the developing countries can be significant emission sources too. For example, every year during the post-monsoon season (October and November), the densely populated northwest Indo-Gangetic Plain (NW-IGP) experiences massive burning of paddy stubbles over an area of excess of 12600 km2 which not only aggravates the regional air pollution but results in poor air quality across the entire IGP. Non-methane hydrocarbons (NMHCs) comprise the largest fraction of VOCs. Till date, the measurement of NMHCs in ambient air and 2 from emission sources over India has been limited only to 10-15 speciated VOCs spanning C2-C5 aliphatics and C6-C8 aromatic compounds. This thesis work presents the laboratory method development and customisation of a thermal desorption gas chromatograph equipped with a flame ionization detection (TD-GC-FID) for quantification of 52 NMHCs, field sampling and measurements of NMHC emission profiles from emission sources, and ambient air measurements over two contrasting sites of the Indo-Gangetic Plain. In the first part of my thesis work, I present the method development for quantification of NMHCsin air samples using TD-GC-FID, in line with recommendations of the World Meteorological Organization (WMO). Using this TD-GC-FID method, I then determined the NMHC chemical fingerprints of major urban and agricultural emission sources active in South Asia, such as paddy stubble burning, garbage burning, vehicular exhausts from petrol, diesel, LPG and CNG vehicles, and evaporative emissions from petrol, diesel and LPG fuels. Source specific molecular tracers and molar ratios were derived to distinguish the sources and petrol vehicular exhaust, paddy stubble fires and garbage fires were identified as the most polluting sources based on their ozone formation potentials and BTEX (benzene, toluene, ethylbenzene and xylenes) toxicity fractions. In the second part of the thesis work, I compiled a novel “hybrid” gridded emission inventory of 77 VOCs and all criteria air pollutants emitted from paddy stubble burning over Punjab and Haryana at 1 km x 1 km spatial resolution for 2017. For this I measured /co-measured the emission factors of 77 VOCs present the paddy smoke samples during both flaming and smouldering stage of the fires. The samples were collected from on-field paddy fires. These were then combined with 1 km × 1 km stubble burning activity data constrained by both district-level annual crop production yields and satellite-detected fires. The results revealed that paddy stubble burning was a significant source of oxygenated VOCs like acetaldehyde (37.5±9.6 Ggy-1), 2-furaldehyde (37.1±12.5 Ggy-1), acetone (34.7±13.6 Ggy-1), and toxic VOCs like benzene (9.9±2.8 Ggy-1) and isocyanic acid (0.4±0.2 Ggy-1) many of which are either unaccounted by, lumped together or significantly underestimated (by upto 25 times) in existing global emission inventories. It was also worth noting that the emissions from this source alone were more than 20 times larger than the emissions from traffic and municipal waste burning over Punjab and Haryana during the same period. This work 3 containing the first speciated VOC emission inventory for the paddy stubble burning activity at 1 km2 spatial resolution sheds new light on the air quality impact due to this source. In the third part of the thesis work, I present the wintertime ambient measurements and source analyses of 52 NMHCs over Delhi and Mohali. The ambient abundances of NMHCs were approximately twice as high in Delhi relative to Mohali, and 2-12 times higher as compared to most other megacities in the world, except Lahore and Karachi. Using the source specific molar emission ratios, vehicular emissions, particularly from petrol vehicles, were identified as the major wintertime NMHC emission sources, with minor contributions from biomass burning, garbage burning and fuel evaporative emissions. The results also suggested that a large fraction of the vehicular fleet at Mohali and Delhi was comprised of vehicles with older emission control technologies. Additionally, comparisons with the global emission inventory EDGARv4.3.2 showed that it poorly constrains the emissions of reactive NMHCs like propene, ethene and trimethylbenzenes over north India and requires substantial improvements for faithful representation in atmospheric chemistry models. Overall the results of my thesis provide a new foundational framework for quantitative source apportionment studies in understudied complex emission environments of India and South Asia and equip the atmospheric chemistry and modelling community with a new region-specific high resolution gridded emission inventory for paddy stubble burning in north-west India. Peer-reviewed publications arising from thesis work: 1. Kumar, A., Sinha, V., Shabin, M., Hakkim, H., Bonsang, B., Gros, V., Non-methane hydrocarbon (NMHC) fingerprints of major urban and agricultural emission sources for use in source apportionment studies, Atmospheric Chemistry and Physics, 20, 12133-12152. doi: 10.5194/acp-20-12133-2020, 2020. 2. Kumar, A., Hakkim, H., Sinha, B. and Sinha, V., Gridded 1 km× 1 km emission inventory for paddy stubble burning emissions over north-west India constrained by measured emission factors of 77 VOCs and district-wise crop yield data, Science of The Total Environment, 789 (1), 148064. doi:10.1016/j.scitotenv.2021.148064, 2021. 3. Kumar, A., Hakkim, H., Ghude, S.D., Sinha, V., Probing wintertime air pollution sources over the Indo-Gangetic Plain through 52 rarely measured hydrocarbons at Delhi & Mohali, Science of The Total Environment, 801 (1), 149711. doi:10.1016/j.scitotenv.2021.149711, 2021.
URI:	http://hdl.handle.net/123456789/4072
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