Title:	Detection and quantification of trace gases in ambient air and vehicular exhaust: Decoding the urban atmosphere
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Keywords:	quantification atmosphere
Issue Date:	Oct-2021
Publisher:	IISER Mohali
Abstract:	Air pollution is now recognized by governments, international institutions and civil society as a major global public health risk factor, accounting for 8.7% of global mortality in 2017. Besides endangering health and shortening lifespan, air pollution adversely affects economic productivity as well as welfare. Lost output from premature deaths and morbidity attributable to air pollution accounted for financial losses of ~US$36 billion in India in 2019, which is equivalent to 1.4% of India's gross domestic product (GDP). These assessments are based on direct exposure to two of the most potent pollutants, particulate matter (PM2.5) and ozone (O3), for both of which volatile organic compounds (VOC) act as precursors. Yet, the abundance, identity, spatial-temporal variability and relative source strength of VOCs remain unknown, especially in developing countries, due to the lack of in-situ ambient data and large emission uncertainties. This thesis work aims to improve the existing spatial and temporal VOC data coverage and develop a high-resolution inventory with an improved spatial distribution. In the first part of my thesis work, I reported the fog and non-fog wintertime ambient VOC speciation and diurnal variability using the first time measurements of thirteen key VOCs over two successive winter seasons (2015–16 and 2016–17) at Delhi, in addition to examining changes in emission activity that could be triggered due to fog using VOC tracers. Remarkably, ambient VOC composition changes during fog were not governed by solubility. Acetaldehyde, toluene, isomers of xylenes and trimethyl benzenes decreased by ≥30% (>95% confidence interval), whereas acetonitrile and benzene showed significant increases by 20% (>70% confidence interval), even after accounting for boundary layer dilution. During fog, the lower temperatures appeared to induce an emissions feedback from enhanced open biomass burning within Delhi for warming, releasing gaseous and aerosol pollutants with consequences for fog chemistry, sustenance and intensity. In the second part of my work, I compiled a new regionally accurate and highly resolved emission inventory for India's road transport sector constrained by measured emission factors for 74 non-methane VOCs and regional vehicular activity data, including calculations for all criteria air pollutants. Petrol-2-wheelers and LPG-3-wheelers emitted the highest VOCs (EFs> 50 gVOC L-1) and had the highest secondary pollutant formation potential, so their replacement with electric vehicles would improve air quality. EDGARv4.3.2 and REASv.2.1 global emission inventories overestimated total road sector emitted VOCs due to obsolete EFs and activity data, in particular 2 overestimating ethene, propene, ethyl benzene, 2,2- dimethyl butane, CO, NOx while significantly underestimating acetaldehyde. Nitromethane emissions were missing from previous inventories and with isocyanic acid and benzene contributed significantly to toxic emissions (sumtotal~41±4 Ggyr-1). In the third part of my work, I evaluated the air pollution impact of three potential fleet substitution strategies relative to existing annual emission fluxes of 74 VOCs and criteria air pollutants from the road transport sector to allow well-informed policy formulation through hypothesis testing of whether significant emission reductions can be accomplished by changes in vehicle fleet composition. The results demonstrated that changes in the fleet and fuel portfolio by vehicles that emit lower on-road air pollution could achieve a significant reduction in direct emission of particulate matter (PM2.5, -82%; BC, -99%), gaseous pollutants (Non-methane VOCs, -84%; isocyanic acid, -73%; BTEX, -95%; CO, -73%), and secondary pollutant formation (SOA, -89%; O3, -78%). Overall, this thesis work has addressed the long-standing issue of inaccurate and poorly speciated traffic emission inventory over the Indian region. The comprehensive representation of VOCs from the road transport sector that has not been available before will aid source apportionment studies, improve air quality forecasts, and better understand the global sources of VOCs such as nitromethane and acetaldehyde. First author peer-reviewed publications from thesis work: 1. Hakkim, H., Sinha, V., Chandra, B.P., Kumar, A., Mishra, A.K., Sinha, B., Sharma, G., Pawar, H., Sohpaul, B., Ghude, S.D. and Pithani, P.: Volatile organic compound measurements point to fog-induced biomass burning feedback to air quality in the megacity of Delhi, Sci. Total Environ., 689, 295-304, https://doi.org/10.1016/j.scitotenv.2019.06.438, 2019. 2. Hakkim, H., Kumar, A., Sinha B., Sinha, V.: RTEII: A new high-resolution (0.1° × 0.1°) road transport emission inventory for India of 74 speciated NMVOCs, CO, NOx, NH3, CH4, CO2, PM2.5 reveals massive overestimation of NOx and CO and missing nitromethane emissions by existing inventories, Atmos. Environ.:X, 11, 100-118, https://doi.org/10.1016/j.aeaoa.2021.100118, 2021.
URI:	http://hdl.handle.net/123456789/4075
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