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Title:	Quantifying the contribution of distant, regional, and local sources to particulate matter (PM) loadings in north-west India using a combination of statistical tools and low-cost PM sensors
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Abstract:	<p>Particulate matter (PM) emitted from natural and anthropogenic sources is ubiquitous in the atmosphere. It impacts the atmospheric composition, air quality, climate, human health, and the biosphere. Rising industrialization and urbanization in the Indo-Gangetic Plains (IGP), also referred to as India's food basket, have led to severe air quality deterioration. Several sites in this densely populated region frequently exceed the national ambient air quality standards (NAAQS) of PM₁₀ and PM_{2.5}. The PM levels in IGP are modulated by a complex interplay of unique and diverse sources and varying meteorological conditions that transport the emissions from the source to the receptor. However, the lack of spatially and temporally robust PM measurements and inadequate understanding of the source apportionment limits the effective mitigation of PM pollution. This thesis attempts to apportion the PM over a regionally representative site in the IGP into distant, regional, and local sources using a combination of statistical techniques and low-cost PM sensors. The first part of the thesis quantifies the contribution of long-range transport to elevated PM levels and the number of exceedance events compared to the regional sources of PM present in the NW-IGP using long-term ground-based measurements and back-trajectory analysis. I find that long-range transport from the distant regions in the west (Arabia, Thar Desert, Middle East, and Afghanistan) leads to significant enhancements in the average coarse-mode PM mass loadings during all seasons. However, local sources (wind speed < 1 m s⁻¹) contributed significantly to the enhanced PM_{2.5} and coarse-mode PM during the winter season. Regional sources in the NW-IGP were responsible for the exceedance of PM₁₀ and PM_{2.5} NAAQS on more than 36% and 48% of the days in the study period. This study revealed that inefficient regional combustion sources (municipal waste burning, biomass burning) in the NW-IGP should be targeted for ensuring compliance with the existing air quality standards. Less than 300 government-owned air quality monitoring stations in India publicly share real-time PM measurements. This degree of coverage is inadequate for locating pollution point sources, gauging spatio-temporal variations in PM mass loadings, and devising efficient mitigation strategies. Several low-cost sensors have permeated the consumer market with little to no field validation to meet the rising demand for open-source air quality data. Although these sensors are cost-effective and compact compared to the US-EPA approved Federal Reference Methods (FRMs) and Federal Equivalent Methods (FEMs), conventionally used by regulatory bodies, very few studies have assessed their long-term performance in complex Indian environments. The second part of my thesis evaluates the efficacy of Laser Egg, a low-cost optical sensor, for monitoring ambient PM in Mohali during the summer and monsoon seasons in the year 2016. The Laser Egg's PM measurements were contrasted with two separate US-EPA approved β-attenuation monitors (5014i Beta Continuous Ambient Particulate Monitor) installed at the IISER Mohali Atmospheric Chemistry Facility. The Laser Egg sensors reported precise measurements of PM₁₀ and PM_{2.5} (coefficient of variance < 12 % and correlation coefficient > 0.9). However, hygroscopic growth, aerosol density, aspiration losses of particles at high wind speeds affect the accuracy of sensor PM measurements. I address each of the issues using site-specific empirical corrections, which increase the correlation coefficient of the two Laser Egg sensors from 0.41 - 0.53 to 0.51 - 0.67. This study demonstrates that the successful integration of low-cost sensors in monitoring networks needs rigorous multi-season site-specific calibrations. The government-owned air quality monitoring stations are disproportionately located in urban areas. As a result, the emission strength of activities occurring primarily in rural regions, like harvest, large-scale paddy residue burning, the use of solid biofuels for cooking and heating, remains poorly constrained. In the third part of my thesis, I use the Airveda monitor, a low-cost PM sensor, to contrast the contribution of paddy harvest, local and regional paddy residue burning, and solid biofuel burning to the PM mass loading at rural (Nadampur) and urban (Mohali) sites in Punjab. The raw sensor measurements were corrected using the Random Forest (RF) machine learning algorithm based on several multi-season field calibrations against a FEM. The RF corrected sensor measurements showed Pearson's R > 0.9 and a slope of 0.8 to 1.0 with respect to the reference analyzers and hence provided sufficient accuracy and precision to substitute the bulkier and costlier FRMs and FEMs for field deployments. In the descending order, paddy harvest enhanced the PM₁₀ (PM_{2.5}) mass concentration in Nadampur and Kalal Majra by 44.8 \pm 1.7 μg m⁻³ (20.1 \pm 5.2 μg m⁻³), 41.1 \pm 7.0 μg m⁻³ (12.0 \pm 4.4 μg m⁻³), respectively. In comparison, local and regional paddy residue fires enhanced the PM₁₀ (PM_{2.5}) mass concentration in Patiala, Nadampur, Mohali, and Kalal Majra by 103.1 \pm 13.7 μg m⁻³ (50.1 \pm 7.6 μg m⁻³), 97.0 \pm 36.6 μg m⁻³ (53.4 \pm 16.8 μg m⁻³), and 58.1 \pm 6.6 μg m⁻³ (34.1 \pm 8.7 μg m⁻³), 55.4 \pm 12.8 μg m⁻³ (22.1 \pm 5.6 μg m⁻³) respectively, from 20 October to 19 November 2019. The dip in the daily average temperature from 17.0 oC to 9.9 oC increased the heating demand, enhancing the residential burning of solid biofuels for space and water heating. In the descending order, the increased heating demand enhanced the PM₁₀ (PM_{2.5}) mass concentrations in Nadampur, Kalal Majra, Patiala, and Mohali by 151.2 \pm 47.2 μg m⁻³ (120.1 \pm 8.8 μg m⁻³), 84.2 \pm 24.6 μg m⁻³ (34.1 \pm 7.3 μg m⁻³), 38.3 \pm 7.6 μg m⁻³ (22.5 \pm 0.3 μg m⁻³), and 30.7 \pm 0.4 μg m⁻³ (22.3 \pm 17.3 μg m⁻³), respectively, from 20 November to 19 December 2019. My thesis evaluates the contribution of distant, regional, and local PM sources over the NW-IGP to air quality deterioration and finds that mitigation of local sources has the greatest potential to improve the air quality at all receptor sites studied. I find that different mitigation policies are needed to tackle urban and rural PM pollution. An appropriately calibrated network of low-cost sensors can validate existing emission inventories and can help to locate point sources of PM. A dense low-cost PM sensor monitoring network employing spatial interpolation techniques that account for dispersion could potentially be used to locate PM sources with a disproportionate impact on air quality in urban environments and industrial areas in the future.</p>
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