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Title: Source Apportionment of Black Carbon and PM 10 Aerosol Using Optical Properties, Chemical Tracers and Statistical Tools

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Abstract: Tropospheric aerosols are ubiquitous in the natural environment and play an important role in the hydrological cycle of Earth, while also affecting human health, local and global climate, and air quality. Aerosols have the ability to enter the human body and cause disorder in the normal functioning of the cardio-vascular and pulmonary system of the body. As a consequence, the concentration of aerosol particles in the air is a major determinant of its quality, and as a consequence significant efforts are made to mitigate their number and mass concentration density in the atmosphere. Thus, research on various aspects of aerosol particles, including their sources, is an integral and important part of atmospheric sciences. For development of targeted mitigation strategies, it is crucial that the potential sources and their relative strengths are determined and reported. In my thesis, I test the validity and effectiveness of the application of various source-apportionment techniques, in particular that of a new technique based on the optical properties of black carbon. Angstrom exponent measurements of equivalent black carbon (BC_{eq}) have recently been introduced as a novel tool to apportion the contribution of biomass burning sources to the BC_{eq} mass. The BC_{eq} is the mass of ideal BC with defined optical properties that, upon deposition on the aethalometer filter tape, would cause equal optical attenuation of light to the actual PM_{2.5} aerosol deposited. The BC_{eq} mass hence is identical to the mass of the total light-absorbing carbon deposited on the filter tape. Here, I use simultaneously collected data from a seven-wavelength aethalometer and a high-sensitivity proton-transfer reaction mass spectrometer installed at a suburban site in Mohali (Punjab), India, to identify a number of biomass combustion plumes. The identified types of biomass combustion include paddy- and wheat-residue burning, leaf litter, and garbage burning. Traffic plumes were selected for comparison. We find that the combustion efficiency, rather than the fuel used, determines α_{abs} , and consequently, the α_{abs} can be ~ 1 for flaming biomass combustion and >1 for older vehicles that operate with poorly optimized engines. Thus, the absorption angstrom exponent is not representative of the fuel used and, therefore, cannot be used as a generic tracer to constrain source contributions. Subsequently I developed a novel method for quantifying the contribution of long-range transport, and regional and local sources of PM₁₀ to the aerosol loading at receptor sites. Subsequently I apply the same to quantify their contribution at multiple receptor locations in Hessen, Germany. This study uses two newly developed statistical source apportionment models, MuSAM and MuReSAM, to perform quantitative statistical source apportionment of PM₁₀ at multiple receptor sites in South Hessen. MuSAM uses multi-site back trajectory data to quantify the contribution of long-range transport, while MuReSAM uses wind speed and direction as proxy for regional transport and quantifies the contribution of regional source areas. On average, between 7.8 and 9.1 $\mu\text{g}/\text{m}^3$ of PM₁₀ ($\sim 50\%$) at receptor sites in South Hessen is contributed by long-range transport. The dominant source regions are Eastern, South Eastern, and Southern Europe. 32% of the PM₁₀ at receptor sites in South Hessen is contributed by regional source areas (2.8- 9.41 $\mu\text{g}/\text{m}^3$). This fraction varies from $<20\%$ at remote sites to $>40\%$ for urban stations. Sources located within a 2 km radius around the receptor site are responsible for 7% to 20% of the total PM₁₀ mass (0.7- 4.4 $\mu\text{g}/\text{m}^3$). The perturbation study of the traffic flow due to the closing and reopening of the Schiersteiner Brücke revealed that the contribution of the bridge to PM₁₀ mass loadings at two nearby receptor sites increased by approximately 120% after it reopened and became a bottleneck, although in absolute terms, the increase is small. In the final chapter, I present the major conclusions from the studies compiled in this thesis. I highlight the importance of source-apportionment for identification and quantification of source contributions to aerosol mass loadings in the atmosphere and the importance of using source-apportionment techniques customised for the environment of the study region. Giving due consideration to the underlying assumptions of the technique in view of the area of application, is an important pre-requisite for effective source-apportionment. Extrapolation of the application area without due consideration of potential artefacts due to local conditions must be avoided for all source-apportionment methods, else erroneous results are likely to be produced. This is crucial to provide accurate information to policy makers who need to know the role of transported versus local pollution sources for smart and focused practical mitigation efforts.


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