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Atmospheric Submicron Particulate Matter (PM): An Emerging Global Environmental Concern

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Particulate matter (PM) is important for its human health concern, its role in atmospheric physico-chemistry and that for aesthetics. Past few decades have seen raised understanding, awareness and concern on the impacts of PM on human health, air quality and climate globally. However, knowledge gaps related to its nature, formation, transport and impact continue to exist; particularly for those related to smaller PM (mainly 'submicron PM', also referred to as 'PM_1') and their contributions to various processes and impacts.

Adverse impact of particulates on human health has been long established, however, some of the recent findings paint a much grimmer picture. Specific PM sources like traffic, coal, oil and biomass combustion, soil or road dust have shown positive correlation with mortality or adverse health effects.[1,2,3,4,&5] Fine PM can penetrate deep into the lungs[5] and cause increased risk of respiratory morbidity, emergency hospitalization and death risk.[5&6] $PM_{2.5}$ is associated with acute lower respiratory infections and mortality,[7] and submicron PM can influence blood and different organs[8] Evidence even support that ultrafin PM can quickly enter the circulatory system,[9] and can cause major structural damage of epithelial cells when localized in cell mitochondria.[10] Further, it has been reported that iron-bearing nanoparticles can get transported directly into the brain posing serious health hazard.[11] PM, therefore, pose unprecedented human health concerns; particularly with respect to submicron PM.

In atmosphere, PM is intricately linked to various atmospheric phenomenon including climate change. Lately, several works have reported important aspects of atmospheric processes and climate change, where role of submicron PM is considered to be increasingly significant. Particles scatter and absorb light, and they serve condensation surfaces for water vapour, affecting cloud formation, precipitation rates and indirect

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climatic effects due to clouds.[12] Some of the particles are semi-volatile, and they can even modify trace-gas composition, and impact gas phase reaction pathways through heterogeneous reaction in the atmosphere.[5] PM are responsible for a range of striking visual and aesthetic features of our skylines, including stunning sunsets, morning haze, blue haze over forests, and complex cloud formations.[13] These features arise from their direct interaction with solar radiation and their roles in cloud formation.[13] As a consequence, they have both direct and indirect effects on the global radiation budget.[13] The predominantly cooling effect on PM, at least temporarily, acts to mask warming by greenhouse gases. The submicron PM have been identified to play influential role in many of these atmospheric processes.

Particulate size, composition and source decides the impacts from both air-pollution as well as climate perspectives. A thorough understanding of particle concentration, size distribution, chemical composition, state of mixing and morphology are therefore essential to address its radiative, ecological and human health aspects. A lot has been known on these aspects for PM_{10} and $PM_{2.5}$. However, lately, submicron PM has been dominating the environmental concern related to atmospheric PM worldwide. Understanding the share and contribution of submicron PM in the overall PM mass-loading and its physico-chemistry has become highly important. Some of the recent work on size- segregated aerosol characteristics shows that knowledge of patterns and features of the submicron PM can be very helpful for impact studies.

Mass concentration of particulates represent its atmospheric concentration in terms of mass of particulates present per unit vol of air (e.g. $\mu g/m^3$). Numerous studies from different parts of the world representing size-segregated mass concentrations of particulates, mainly as PM_{10}, $PM_{2.5}$ and PM_1, reflect the rising importance of PM_1. Studies from different environmental setting in China[14,15&16] have shown that PM_1 dominates the share and variation patterns of $PM_{2.5}$ and PM_{10}. High share of PM_1 (in total dust) have been found from different forms of combustion in Germany.[17] Studies from India (New Delhi),[18] Greece (Athens),[19] Switzerland (High Alpine)[20] and Italy (Po Valley)[21] show that share and/or trend of PM_1 mass significantly influence/dominate the mass of larger PM. Similarly, non-refractive PM_1 mass have dominated $PM_{2.5}$ in Finland (Helsinki)[5] and smaller biomass burning particles have been found to dominate summer aerosol in USA (Colorado).[22] Many other such studies show that the mass concentration of PM is significantly influenced by that of submicron PM throughout the world. Therefore, submicron PM mass is now turning out to be a relatively bigger concern worldwide.

The chemical nature of particulates have been found to vary widely with source, age, size, atmospheric precessing and environmental condition globally. The chemical characteristics of PM_1, $PM_{2.5}$ and PM_{10} are interesting to analyze, and very commonly the chemistry of PM_1 are found to influence/dominate that of the overall PM. Numerous studies have shown that the spectrum of chemical species found are more often distinct between submicron and larger PM. For example, different studies from Switzerland,[20,23] South Africa,[24] Hong Kong,[25] Finland (Helsinki),[5] USA (Sacramento Valley),[23,26] Africa (Senegal),[23] China,[23] France,[23] Italy,[23] the Netherlands,[23] Spain[23] and 'Greater European Region[27] support dominance of one or more of such species like organics, sulfate, black carbon, nitrate, ammonium, chloride and iron in submicron PM, whereas, of other species like calcium in larger PM. Further, some of the major anions and cations have been majorly found to be part of submicron PM in Greece (Athens).[19] Similarly, certain species were found to be more abundant in fine particles at an urban air pollution site in China (Beijing), where the contributions of pollution sources was found to decrease with increasing PM size.[28] Another global study[29] showed that 'dust' is primarily found in the coarse mode whereas carbonaceous components in fine and ultrafine mode PM, close to emission sources. Thus submicron PM is expected to significantly influence/dominate the overall chemistry of atmospheric PM.

PM size largely govern its physical behavior from various impact perspectives. There has been large amount of data on size distribution of PM, however, in the past few decades there has been significant advancement in measurement facilities and there is now appreciable spectrum of data also on submicron PM and its physical properties from many regions of the world. PM physical properties like those related to light scattering and absorption, hygroscopicity, cloud formation capabilities, etc are being widely studied, also giving striking features of submicron PM; thereby increasing its significance in understanding overall PM impacts. Apart from mass share, other physical properties of PM also show significant contribution from PM_1 in different parts of the world. In Central Himalayas, Nainital, India the scattering and absorption coefficients, scattering and absorption angstrom exponents, backscatter fraction and single scattering albedo share and trends of PM_{10} were seen to be highly influenced by those of PM_1.[30] In Sacaramento, California, USA similar measurements showed significant influence of PM_1 on similar optical properties of PM_{10}.[31,32] In Granada, Spain PM_1 was found to influence the scattering and absorption efficiencies in visible wavelengths.[33] Similar observations were also recorded in Southwestern[34] and Arizona,[35] USA. Several other workers have also observed that the scattering process is mainly due to particles in the fine mode. An important finding in this respect was reported at a rural site in northeast Spain in the Western Mediterranean Basin, which showed that polluted air masses were linked with an increase in PM_1 concentration and consequent increase in the values of scattering, backscattering and absorption coefficients and angstrom exponents. Thus, it is important to note that physical behavior of PM can be significantly influenced by presence of submicron PM.

Thus, several data-set and studies under varying environmental settings from different parts of the world point towards raised significance of submicron PM in larger PM (like $PM_{2.5}$ and PM_{10}). The contribution of submicron PM in terms of share and variation pattern is often dominating mass of larger PM in different regions of the world. The chemistry of larger PM are also in many cases influenced by the chemical nature of submicron PM; as some chemical groups/species largely remain concentrated in the latter. The physical nature and behavior of larger PM, like its optical properties, also in many instances remain highly influenced by those of submicron PM. In this context, it is important to note that rain may not be able to wash out all of submicron PM, as observed in Wuhan, China.[16] Therefore, the significance of submicron PM highlights the need for global efforts to include submicron PM as a major pollutant in all atmospheric PM pollution initiatives and impact studies, and work towards early development and proper utilization of air quality standards for submicron PM.

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