

Umesh Kulshrestha · Pallavi Saxena *Editors*

Plant Responses to Air Pollution

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 Springer

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About the Editors

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Environment Centre, Lancaster University, UK (2011). She is also an expert of TOAR International Meeting, which focuses on tropospheric ozone and its impact on vegetation, from 2015 onwards. In addition to that, she has been a recipient of various awards like the Young Scientist Award from the Indian Society of Plant Physiology (2013) and DBT Bio-CARe Award (2014) and several travel grant awards from World Meteorological Organization (WMO), International Global Atmospheric Chemistry (IGAC) and National Oceanic and Atmospheric Administration (NOAA) to participate as an expert in the Ozone Pollution and Its Impact on Vegetation in Trpospheric Ozone Assessment Report (TOAR) Meeting (2015–2016). She has also reviewed research articles for reputed journals, like *Atmospheric Environment* (Elsevier) and *Mitigation and Adaptation of Strategies for Global Change* (Springer), *Environmental Technology* (Taylor and Francis), *Polish Journal of Environmental Studies*, *Environmental Monitoring and Assessment* (Springer) and *International Journal of Physical Sciences* (Academic Journal). She has participated in various national (10) and international conferences (8) and presented several papers. Dr. Saxena has also participated in various workshops and trainings (15) in the area of air pollution and plant physiology.

Umesh Kulshrestha and Pallavi Saxena

Being a living community on the planet earth, utilization of natural resources is quite obvious by humans. However, the overexploitation of these resources during past two decades has resulted in major environmental problems. Increasing human population and energy demand has contributed different kinds of harmful chemical and biological species in the atmosphere. Recently, air pollution has become a burning issue of common concern. The process of air pollution can be defined as the atmospheric conditions having high levels of pollutants which may produce undesirable effects on materials, plants, and human health. The major air pollutants include sulfur dioxide, nitrogen oxides, carbon monoxides, hydrocarbons and particulate matters, etc. which play a very significant role in affecting the biochemical and physiological processes of the plants and ultimately lead to yield losses (Heck et al. 1988). In developing countries like India, a very drastic change in the quality of air has been observed during past two decades. Many cities in Asia, Africa, Latin America are facing major challenges of air pollution (Ashmore 2005). Delhi (the capital city of India) is also facing the problem of severe air pollution in spite of the implementation of CNG-driven public trans-

port (Saxena et al. 2012). The air quality in Delhi has been the worst among 1,600 cities of the world (WHO 2014–2015). According to the estimates, around 1.5 million people are killed every year. India has the world's highest death rate from chronic respiratory diseases and asthma.

1.1 Air Pollution and Plant Health

Generally, atmospheric pollutants have a negative effect on the plants; they can have direct toxic effects or indirectly by changing soil pH followed by solubilization of toxic salts of metals like aluminum. The deposition of particulate matter covers the leaf blade reducing light penetration and blocking the opening of stomata. These impediments influence the process of photosynthesis due to which rate of photosynthesis and growth declines sharply. On the other hand, leaves of the trees have role in retaining particulate matter but they are much more affected when the dry depositions are increased (Jyothi and Jaya 2010). Polluting gases such as SO₂ and NO_x entered into the leaves through the stomata following the same diffusion pathway as CO₂. NO_x dissolved into the cells gives rise to nitrite ions (NO₂⁻, which are toxic at high concentrations) and nitrate ions (NO₃⁻) that enter into nitrogen metabolism as if they had been absorbed through the roots. In some cases, exposure to pollutant

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gases particularly SO₂ causes stomatal closure, which protects the leaf against further entry of the pollutant but also curtails photosynthesis. In the cells, SO₂ is dissolved to give rise to bisulfite and sulfite ions, the sulfite is toxic, but, at low concentrations, it is metabolized by chloroplasts to sulfate, which is not toxic. At sufficiently low concentrations, bisulfite and sulfite are effectively detoxified by plants. In such cases, the SO₂ air pollution is a source of sulfur nutrient for the plant. In urban areas these polluting gases may be present in such high concentrations that these cannot be detoxified rapidly enough to avoid injury. For example, ozone is presently considered to be the most damaging phytotoxic air pollutant in North America (Heagle 1989; Krupa et al. 1995). It has been estimated that whenever the mean daily O₃ concentration reaches 40, 50, or 60 ppb (parts per billion or per 10⁹), the combined yields of soybean, maize, winter wheat, and cotton would be decreased by 5 %, 10 %, and 16 %, respectively. Ozone is a highly reactive gas. It combines with plasma membranes and alters the metabolism. As a result, stomatal apertures are poorly regulated, chloroplast thylakoid membranes are damaged, RuBisCO is degraded, and photosynthesis is inhibited. Ozone reacts with O₂ and produces reactive oxygen species, including hydrogen peroxide (H₂O₂), superoxide (O₂⁻), singlet oxygen (¹O₂*), and hydroxyl radical (OH⁻). These denature proteins and damage nucleic acids (thereby giving rise to mutations) cause lipid peroxidation, which breaks down lipids in membranes. Reactive oxygen species (ROS) formed in the absence of O₃ too, particularly in during electron transport process in the mitochondria and chloroplasts, when electrons can be donated to O₂. Cells are protected from reactive oxygen species by enzymatic and nonenzymatic defense mechanisms (Heterich and Heterich 1990; Cape 2003). Defense against ROS is provided by the scavenging properties of molecules, such as ascorbic acid, α-tocopherol, phenolic compounds, and glutathione. Superoxide dismutases (SODs) catalyze the reduction of superoxide to hydrogen peroxide. Hydrogen peroxide is then converted to H₂O by the action of catalases and peroxidases. Of particular impor-

tance is the ascorbate-specific peroxidase localized in the chloroplast. Acting in concert, ascorbate peroxidase, dehydroascorbate reductase, and glutathione reductase remove H₂O₂ in a series of reactions called the Halliwell–Asada pathway, named after its discoverers. Glutathione is a sulfur-containing tripeptide that, in its reduced form, reacts rapidly with dehydroascorbate and becomes oxidized in the process. Glutathione reductase catalyzes the regeneration of reduced glutathione (GSH) from its oxidized form (GSSG) in the following reaction:



Exposure of plants to reactive oxygen species stimulates the transcription and translation of genes that encode enzymes involved in protection mechanisms. In *Arabidopsis*, exposure for 6 h per day to low levels of O₃ induces the expression of several genes that encode enzymes associated with protection from reactive oxygen species, including SOD, glutathione S-transferase (which catalyzes detoxification reactions involving glutathione), and phenylalanine ammonia lyase (an important enzyme at the start of the phenylpropanoid pathway that leads to the synthesis of flavonoids and other phenolics) (Weiss et al. 1997).

Therefore, in relation to air pollution phenomena and physiological processes of plants, the present book covers all the important topics in order to justify the title “Air Pollution and Plant Health: Climate Change Perspectives.” Chapter 1 deals with the general introduction. It basically provides general information about air pollution and its effect on plants. Chapter 2 summarizes the air quality trends along with global and regional emissions of particulate matter, SO_x, and NO_x. Also, this chapter gives an overview of global sources of air pollutants and their future projections. Chapter 3 deals with the effect of these urban air pollutants on biota. It describes about the effects of air pollutants such as particulate matter, SO_x, and NO_x on plants as well as on human health. This chapter highlights the need for imperative shift toward renewable energy sources for sustainable and environmental friendly solution for new technologies and

industrial producers. Chapter 4 describes the characteristics and mechanisms of plant which help in the uptake of air pollutants giving insights about the distribution of pollutants inside the plant machinery. It also describes how the pine needles and mosses accumulate air pollutants demonstrating the role of plants as biomonitors. Chapter 5 opens up the discussion about biogenic VOC emissions and their role in atmospheric sciences. In this chapter, plant chemistry and the conditions under which it emits BVOCs are detailed. Besides, this chapter also provides information about BVOC linkages with global warming and plant signaling processes. Chapter 6 deals with the biochemical effects of air pollutants on plants. This chapter summarizes about the significant effects posed by air pollutants on plant health. Biochemical parameters such as chlorophyll, proline content, and other enzymatic activities act as bioindicators for determining the health of the plant. Chapter 7 deals with air pollutants and photosynthetic machinery of plants. It describes how high concentrations of air pollutants damage leaves and reduce leaf area, thereby affecting the photosynthetic efficiency of plants to a significant extent. The damage to the photosynthetic apparatus accounts for the decline in photosynthetic efficiency, but each pollutant differs in its mode of action for damaging the photosynthetic apparatus of leaves. Photosynthesis and respiration are interrelated processes involved in the basic processes of plants. Therefore, Chap. 8 deals with the effect of air pollutants on plant gaseous exchange process: effect on stomata and respiration. This chapter describes about how respiration acts as an indicator for environmental stress. The study also shows that leaf characters including cuticle, stomata, epidermal cells, and guard cells get affected due to stress induced by the air pollutants. This further affects the gaseous exchange as well as respiration in plants. Chapter 9 deals with the effect of air pollutants on biomass and yield. It also linkages how respiration acts as an indicator in the physiological mechanisms. It is to mention that any book of this theme is incomplete without the provided dedicated chapter on environmental stresses and plant health. Therefore, Chap. 10 deals with air pollu-

tion stress and plant response. This chapter describes about how the structural and physiological changes occur due to stress imposed by air pollutants. It is very important to control the effect so as to the quality of air. Hence, Chap. 11 deals with the biomonitoring and remediation by plants. This chapter describes about how respiration acts as an indicator for environmental stress. The study also shows that leaf characters including cuticle, stomata, epidermal cells and guard cells get affected due to stress induced by air pollutants. Chapter 12 deals with air pollution control by policies and laws. It deals with different laws or amendments made by government to curb air pollution and other mitigation measures. Chapter 13 deals with the pollution and plants in relation to changing policies. This chapter determines the current pollution scenario in India and various interventions made at policy levels in particular for plants. Chapter 14 deals with tropospheric ozone and effects on plants. Ozone is important to study because it is the only phytotoxic pollutant having high oxidative capacity. It can damage plant to a larger extent as compared to other air pollutants. This chapter summarizes the information available on plant responses to O₃ at physiological, cellular, and biochemical levels, crop yield, forest, and grassland communities at present concentrations and also under projected future concentrations.

1.2 Conclusion

The collated information based on several studies concludes that air pollutants not only affect the vegetation near the point sources and urban centers but also affect the crops depending on the environmental conditions in suburban and rural areas. The physiology and metabolism of plants are altered due to the oxidizing potential of the pollutants. In order to survive, the responses of plants vary between different species and their cultivars. Responses of plants to air pollutants also depend on type of pollutants, concentrations' duration, and its magnitude. There is a need to screen out sensitive and tolerant cultivars especially in developing countries and establish

the exposure indices of all the important crops to reduce the crop loss. The detailed description of air pollutant effect on plants, sources of air pollution, and control policies is given in this book through different chapters.

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Air Quality: Global and Regional Emissions of Particulate Matter, SO_x, and NO_x

2

Darpa Saurav Jyethi

Abstract

Poor air quality is known to have deleterious effects on the environment and human health. A variety of air pollutants are identified at unprecedented levels. Particulate matter and oxides of sulfur and nitrogen are common pollutants of the atmosphere. The emissions of these criteria pollutants have been studied widely. An overview of their global and regional emissions is helpful in assessing the status of contributions from various sectors and efficacy of control strategies over the years in both developed and developing nations of the world. Particulate matter levels in developed regions have been observed to have decreased substantially toward the end of the twentieth century, contrary to the trend of emissions in developing countries. Transportation and power generation sectors are key sources of PM emissions. Oxides of sulfur have been observed to have peaked in the 1970s and subsequently decreased thereafter on a global scale; however, the developing economies have registered a rise in emissions. Similar trend has been observed for oxides of nitrogen with decline of emissions from developed regions of the world and subsequent increase from developing countries in the earlier part of the twenty-first century. Timely intervention of suitable strategies to combat emissions in developing regions is crucial to nullify the increasing trend of emissions. The present chapter provides an overview of three criteria air pollutants – particulate matter, oxides of sulfur, and oxides of nitrogen with respect to their changing global and regional emission scenario in the past decades and sector-wise contributions.

Keywords

Air quality • Air pollutants • Particulate matter • Oxides of nitrogen • Sulphur • Nitrogen

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2.1 Introduction

Air quality has gained immense concern because of its wide-ranging implications. Poor air quality has been generally attributed to population explosion and subsequent overexploitation of natural resources, urbanization, and industrialization. Much of the burden in the present times is markedly high in the developing nations of the world. A thorough understanding of sources, global and regional emission scenarios, and spatiotemporal variations is crucial in order to develop and implement strategies suitable to curb regional and local sources of air pollution. Air pollution sources are basically classified as natural (events that are not the result of any human activities, e.g., forest fires, dust, volcanoes) and anthropogenic (resulting from human activities). Stationary sources (e.g., power plants, refineries, and industries) and mobile sources (e.g., cars, trucks, buses) are two broad categories of prevalent sources in urban areas. Outdoor or ambient air pollution is mostly attributed to combustion of fossil fuels; however, fuel evaporation has also been known to contribute to pollution load (Black et al. 1998). Indoor sources (e.g., building materials and activities such as cleaning, cooking, etc.) are also other sources of outdoor air pollution. The United States Clean Air Act has categorized “major” and “area” stationary sources, the former being sources that emit 10 tons per year of any of the listed toxic air pollutants or 25 tons per year of a mixture of air toxics. Equipment leaks when materials are transferred from one location to another or during discharge through emission stacks or vents are considered in the category. On the other hand, “area” sources are those that emit less than the aforementioned limit. Emissions from single area sources are apparently often relatively less; however, their emissions are alarming when they are considered in a collective scenario, for instance, in a densely populated urban area. Apart from sources of air pollution, meteorology and topography are two important factors that determine the prevalence of pollution (Seinfeld and Pandis 2006) and extent of exposure and subsequent effects (Goncalves et al. 2005). Meteorology governs the dispersion,

chemical transformations, residence time, transport, and fate of pollutants in the atmosphere. The following section deals with the implications of air quality deterioration from various perspectives.

2.1.1 Air Quality and Its Implications

Air quality is recognized as an important indicator of environmental sustainability, health status, economic condition, and social well-being. The effects of air pollution are not restricted in the source region but are trans-boundary. Environmental implications include damages of life-sustaining systems such as atmosphere, hydrosphere, and pedosphere, primarily caused by introduction of toxic air pollutants and through formation of secondary pollutants such as acid rain and tropospheric ozone. These harm plants, forests, food crops, wildlife, aquatic bodies, and biota (DEFRA 2013). Air quality has been linked to ocean acidification, eutrophication, long-term changes in water quality, visibility degradation and haze, and alteration of Earth’s radiation budget (Seinfeld and Pandis 2006); bioaccumulation, behavioral, neurological, and reproductive effects in fish, birds, and wildlife; species extinction (Welch 1998); altered soil chemistry; and consequently plant species composition, agricultural yield, and damage to historical built environment (Thomas 1961; Brimblecombe 2003).

Health ramifications due to poor air quality are generally complex. It includes a host of outcomes that have been linked to short- and/or long-term exposure to air pollutants. Short-term or acute exposure to pollutants results in mild outcomes such as eye and throat irritation, headaches, nausea, intense effects such as respiratory tract infections – bronchitis and pneumonia – allergic reactions, and worsening of asthmatics and other sensitive populations such as infants, children, and elderly. Incidences of lung cancer, cardiovascular disorders, and detrimental effects on the brain, nerves, liver, and kidneys are some of the chronic effects of long-term exposure to specific air pollutants (Kim et al. 2014 and refer-

ences therein). The Organisation for Economic Co-operation and Development (OECD) has declared that exposure to outdoor air pollution is expected to become the top environmental cause of premature mortality globally by 2050 (OECD 2012). Cardiovascular, pulmonary, and respiratory systems are predominantly affected. It is estimated that the global burden due to outdoor air pollution manifested in seven million deaths in 2012 (WHO 2014). On a regional scale, the World Health Organization (WHO) Western Pacific and Southeast Asian regions bear most of the burden (WHO 2014). The International Agency for Research on Cancer (IARC), WHO's specialized cancer agency, has declared outdoor air pollution as carcinogenic (Group 1) to human beings. Among various air pollutants studied, particulate matter is most closely related with increased cancer cases, especially lung cancer. Additionally air pollution has been linked to increase in cancer of the urinary tract/bladder (IARC 2013). Air quality index (AQI) is commonly used as a tool to assess the local air quality on a daily basis. It is expressed for individual pollutant (particulate matter, sulfur dioxide, tropospheric ozone, and carbon monoxide) and uses numerical values and color codes to alert general population regarding the status of air quality and its health effects upon exposure (0 to 5 (good, green); 51 to 100 (moderate, yellow); 101 to 150 (unhealthy for sensitive groups, orange); 151 to 200 (unhealthy, red); 201 to 300 (very unhealthy, purple); 301 to 500 (hazardous, maroon) (USEPA 2014).

Air quality degradation has economic consequences. The estimated cost of the deaths and illness attributable to air pollution in OECD member countries was about USD 1.7 trillion in 2010 (OECD 2014). Significant amount of the burden has been found to be arising from vehicular tailpipe emissions. The costs of deteriorating air quality are generally higher for the weaker (in terms of socioeconomic status) sections of the society (Wong et al. 2008; ALA 2013). Studies have concluded that sensitive population (Bateson and Schwartz 2004; Sunyer et al. 2000) and socioeconomically weaker sections of society (Forastiere et al. 2006; Jerrett et al. 2004; Neidell

2004) are most strongly affected by air pollution. The reasons attributed to such an observation are cheaper costs of production and less stringent environmental regulations for the shift of polluting industries into poorer areas from wealthier areas (Pulido 2000).

2.1.2 Air Pollutants

There are a gamut of chemical substances in the atmosphere that are known to be harmful for the environment and human health. The Clean Air Act of the United States Environmental Protection Agency (USEPA) defines air pollutant as "any air pollution agent or combination of such agents, including any physical, chemical, biological, radioactive (including source material, special nuclear material, and byproduct material) substance or matter which is emitted into or otherwise enters the ambient air." Certain air pollutants are ubiquitous. The USEPA classifies air pollutants as criteria and non-criteria pollutants for monitoring and regulation purposes. Commonly found air pollutants that are known to harm environment and health are classified as criteria pollutants. The term "criteria" emphasizes the need for regulation and routine monitoring by developing human health-based and/or environmentally based permissible levels. Presently, six pollutants – particulate matter, tropospheric ozone, carbon monoxide, sulfur oxides, nitrogen oxides, and lead – are designated as criteria pollutants by the USEPA. These are used as key indicators of air quality all over the world. The USEPA has set guideline values for these criteria pollutants referred to as National Ambient Air Quality Standards (NAAQS). Two standards – primary and secondary – are specified. Primary standard values of each of these pollutants provide public health protection, including protecting the health of "sensitive" populations such as asthmatics, children, and the elderly. Secondary standards provide public welfare protection, including protection against decreased visibility and damage to animals, crops, vegetation, and buildings (USEPA 2005). Non-criteria pollutants, unlike criteria pollutants, do not have

assigned guideline value. In addition to criteria and non-criteria pollutants, certain air pollutants are classified as “air toxics” or hazardous air pollutants (HAPs). These include those pollutants that are known to cause cancer and other serious health outcomes, such as reproductive, immune, neurological, and congenital defects. Adverse environmental and ecological effects are also considered in classifying pollutants as HAPs. Acetaldehyde, acetonitrile, asbestos, benzene, carbon tetrachloride, chloroform, dibenzofurans, phenol, polycyclic aromatic, nickel, arsenic, antimony, lead, mercury, and cadmium compounds, radionuclides, and pesticides are among 187 compounds that are listed as HAPs (USEPA 2015a). The USEPA regulates HAPs in two phases – firstly, based on emissions from best available technology and, secondly, on the basis of health risks.

Air pollutants are either primary or secondary in origin. Pollutants that are directly released into the air from the pollutant source are termed as primary pollutants. Once emitted in the atmosphere, some of the primary pollutants are chemically and/or photochemically transformed into secondary pollutants. Gaseous pollutants and particulate matter are two broad categories with marked differences in measurement techniques and control strategies. Tropospheric ozone, carbon monoxide, sulfur oxides, nitrogen oxides, ammonia, and benzene are some of the gaseous pollutants commonly found in ambient atmosphere. Particulate matter is a relatively complex entity suspended in the atmosphere with size-dependent atmospheric residence time. Size distribution of particulate matter in atmosphere along with number, mass concentrations, and chemical constituents is highly variable on a spatial as well as temporal scale.

2.2 Particulate Matter

Particulate matter (PM), otherwise known as atmospheric aerosol, is a criteria air pollutant. The term “aerosol” denotes a system of particles (solid or liquid) suspended in a gas (air). PM underscores the particle component of atmo-

spheric aerosols. It is highly complex in composition. Size is an important factor of particulates and governs the transport and fate in the atmosphere. “Fine” and “coarse” are two broad categories of particulates. Fine particulates are those having an aerodynamic diameter of 2.5 micrometers (μm) or less, whereas particulates having aerodynamic diameter larger than 2.5 μm and smaller than 10 μm are termed as coarse. Coarse particulates are generally mechanically generated, whereas fine particulates are either directly emitted from sources or formed in the atmosphere from gas to particle conversion reactions. Fine particulates are of particular concern due to association with adverse health concerns as they are inhalable and are able to penetrate deep into the lungs. Fine particulates generally comprise the accumulation mode ($0.1 \mu\text{m} < \text{particle size} < 2.5 \mu\text{m}$) and the nuclei mode (particle size $< 0.1 \mu\text{m}$). Nuclei mode particles have relatively short lifetimes in the atmosphere and end up in the accumulation mode. Gas-phase condensations and secondary formations on the existing particles in atmosphere also play a crucial role on the chemistry of the particles. Concentrations of ultrafine particles (UFPs, particulate matter of less than 100 nm in aerodynamic diameter) are also known to be of particular relevance especially from inhalation exposure perspective. It is noteworthy that not only size of particulates but size distribution is also an important attribute in the study of PM. It has been reported that the methods used for measuring PM concentrations are affected by the size distribution. Capareda et al. (2004) found that size distribution of PM emitted in rural areas is significantly larger than that of PM present in urban areas. The authors reported that mass median diameter (MMD) of urban PM is generally less than 10 μm aerodynamic diameter, whereas agricultural PM will have an MMD larger than 10 μm . The size of PM also governs the process of removal from atmosphere. Fine particulates are generally removed through rainout and washout, whereas coarse particulates are mostly removed through sedimentation. Apart from size of particulates and size distribution, the composition of PM is highly variable and complex. PM is mostly composed of

carbonaceous matter and includes inorganic and organic components. These components drive health outcomes upon inhalation and consequent deposition in the respiratory system. Certain chemical components trigger the production of reactive oxygen species and induce oxidative stress in lung cells (Tao et al. 2003). Other attributes that influence the environmental and health effects are mass concentrations and morphology. Size and chemical composition have been often debated to be the competing attributes of PM in relation to health effects. The USEPA, however, reports that the health outcomes due to PM exposure vary by size and composition with both bioavailable metals and organic chemicals playing a crucial role in the health outcomes. Studies have concluded that no known chemical substance is of sufficient toxicity given the current levels of exposure to particulate matter to explain the observed magnitude of health effects (USEPA 2002). Fine and coarse PM has been routinely monitored in various cities across the world. Anthropogenic exploitation of fossil fuels is known to generate significant amount of PM. Naturally occurring PM originates from volcanoes, dust storms, wild fires, and biogenic and sea spray. Long-term exposure to PM has been linked with morbidity and mortality (Dockery et al. 1993; Pope et al. 2004). PM exposure for a short term results inflammatory response in the upper respiratory tract and decreased lung functions in general population. Sensitive populations such as children and elderly, asthmatics, and people suffering from heart and lung ailments are extremely susceptible to initiation of cough, phlegm, wheezing, shortness of breath, bronchitis, increased asthma attacks, and aggravation of lung or heart disease (Cohen and Pope 1995). Ambient and occupational exposure to elevated levels of PM has been associated to chronic pulmonary obstructive diseases (COPD) such as bronchitis and emphysema.

Environmental concerns of PM include absorption or scattering of solar radiation, consequent alteration of the radiation budget of Earth, and haze formation in the lower troposphere. Composition and size of PM are important in the manifestation of these effects. For instance, black

carbon or soot-dominated aerosol is absorbing and sulfate aerosol is scattering in nature. Exposure in plants is either through deposition on vegetative surfaces or soil–root pathway. Effects manifest in alteration of key physiological processes such as photosynthesis and respiration; foliar surface damage by acidic and alkaline component; nutrient uptake from soil; foliar nutrient leaching; reduced vigor, productivity, reproductive success, and resilience; and disturbed nutrient cycling, soil stabilization, water cycling, and even energy flux (Grantz et al. 2003). Other environmental effects of PM include acidification of aquatic and terrestrial ecosystems upon deposition and degradation of heritage monuments and built structures.

2.2.1 Global and Regional Emissions of Particulate Matter

PM is one of the most widely studied criteria air pollutants. Quantification of ambient PM load has been achieved by gravimetric and continuous or real-time measurements. Satellite-derived aerosol optical depth (AOD) has also been used to derive PM concentrations. Sampling and measurement techniques are subjected to significant uncertainty because of sampling artifacts, handling procedures, and by the fact that a significant portion of PM is semi-volatile and partition between gas and particle phase is temperature dependent (Van Dingenen et al. 2004). Satellite-derived PM also suffers from uncertainty due to aerosol vertical profile variability on a seasonal and regional scale and cloud fraction (Li et al. 2015).

van Donkelaar et al. (2015) used satellite observations as a surrogate to report an increased population-weighted fine particulate concentrations (2.1%/year globally) from 1998 to 2012. The authors also found that higher emission trends in some developing countries influenced the trend on a global scale. Figure 2.1 depicts a world map exhibiting a measure of extinction of radiation or total aerosol optical depth at 550 nm due to aerosol scattering and absorption. It is

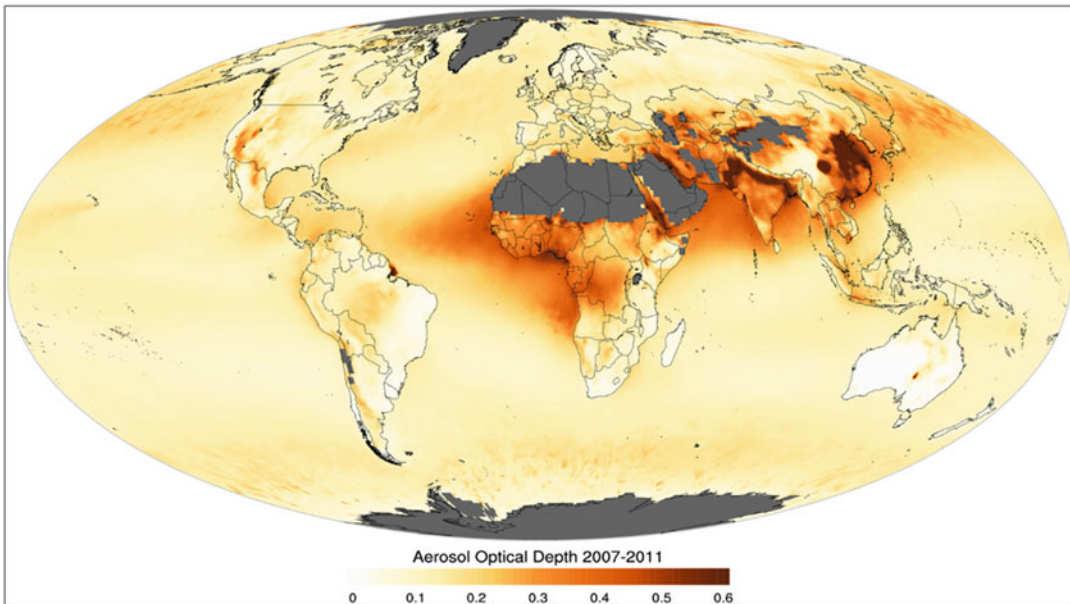


Fig. 2.1 World map showing aerosol optical thickness from Moderate Resolution Imaging Spectroradiometer (MODIS) on NASA's Terra satellite. *Dark brown* pixels show high aerosol concentrations, while tan pixels show

lower concentrations, and *light yellow* areas show little or no aerosols. *Gray* denotes that the sensor could not make its measurement (Source: <https://en.wikipedia.org/wiki/Particulates>)

indicative of the PM pollution status of the regions in the world. The USEPA has reported 35 % and 30 % reduction in the national average of $PM_{2.5}$ and PM_{10} levels, respectively, from 2000 to 2014 in the USA (USEPA 2016). Upadhyay et al. (2011) studied PM levels in Southern Phoenix, Arizona, and found that soil and combustion processes explained the observed variance in PM_{10} levels, whereas combustion sources explained the observed $PM_{2.5}$ concentrations. The difference in urban and rural $PM_{2.5}$ levels during 2009–2012 was studied in the Midwestern region of the USA, and it was concluded that the observed levels in rural areas were generally lower ($8.4\text{--}10.4 \mu\text{g m}^{-3}$) compared to the urban sites ($9.5\text{--}11.6 \mu\text{g m}^{-3}$). Crustal materials dominated in rural areas, whereas secondary aerosols and combustion products dominated in urban areas (Kundu and Stone 2014). Karagulian et al. (2015) reported that 24 % and 30 % of the observed $PM_{2.5}$ and PM_{10} , respectively, in the USA can be attributed to traffic sources. It was also reported by the authors that secondary particle formation contributed as high as 46 % and

44 % to the $PM_{2.5}$ and PM_{10} levels, respectively. Annual mean levels based on daily averages of PM data monitored by the European Environmental Agency (EEA) in various countries of the European Union were found to be relatively higher in nations such as Turkey, Bulgaria, Slovakia, the Czech Republic, Poland, and parts of Italy as compared with the other member nations during the year 2012 (EEA 2014). Primary PM_{10} , $PM_{2.5}$, and $PM_{2.5-10}$ emissions during 1990–2010 have been reported to be decreased by 26 %, 28 %, and 21 %, respectively, across regions in Europe. Increase in emissions from road transport and agriculture since 1990 has been attributed as the reason for the resulting difference in the trends in $PM_{2.5}$ and $PM_{2.5-10}$. The transition of source (from coal to natural gas) for power generation and better pollution control strategies have been the key reasons for the decrease in PM_{10} levels (EEA 2012). Considerable variations with respect to sites and seasons in the contribution of non-combustion fraction to the observed PM were observed in the cities of Athens, Madrid, and London. PM_{10} levels in

Athens and Madrid were found to be higher in hot and dry season indicating toward secondary formations and crustal particles (Kassomenos et al. 2014). On a global scale, the highest contribution of natural dust to observed levels of PM_{2.5} and PM₁₀ has been found in the Middle East region (52% and 44%, respectively) (Karagulian et al. 2015). PM pollution is particularly worse in Asian cities. Mean PM₁₀ concentrations across Northern China was found to be an order higher than those recorded in European Union and the USA. Coal and biomass fuel combustion for heating in winter contributed the highest to the observed levels in China (Luo et al. 2014). PM emissions in the Indian subcontinent have been considerably high in megacities. PM_{2.5} levels in rural areas in the Indo-Gangetic plains are reported to be higher than some urban centers (Dey et al. 2012). Delhi, the capital of India, is one of the worst in the world with respect to particulate pollution. Marrapu et al. (2014) estimated the contribution of transportation, power generation, industrial activities, and domestic sources as 86.8%, 7.9%, 4.6%, and 0.8% and 52.6%, 9.9%, 15.3%, and 22.2% for PM₁₀ and PM_{2.5}, respectively, for the city during 2010. On a global level, Karagulian et al (2015) found 25% of urban ambient air pollution from PM_{2.5} is contributed by traffic, 15% by industrial activities, 20% by domestic fuel burning, 22% from unspecified sources of human origin, and 18% from natural dust and salt. Takeshita (2011) examined the role of transport sector using REDGEM70 global energy system model and concluded that in a business as usual (BaU) situation (wherein an assumption is made that developing countries will adopt identical stringent regulations as in developed countries but with a lag of 20 years), global PM emissions from road vehicles will decrease by 93% from 2000 to 2050. It is estimated that an early implementation of strict regulatory measures in developing countries can potentially alleviate the PM pollution scenario from road vehicles.

2.3 Oxides of Sulfur

Oxides of sulfur (SO_x) is a generic term for a group of compounds containing sulfur and oxygen molecules. SO_x emissions in the atmosphere predominantly result from anthropogenic sources, i.e., combustion of sulfur-containing fuels such as coal and oil used in electricity generation. Petroleum refineries and smelting of metal sulfide ores for metal manufacturing are other sources. Apart from anthropogenic sources, natural sources such as volcanoes and wildfires are also known to emit considerable amounts of SO_x into the atmosphere. Sulfur dioxide (SO₂) is the predominant form emitted from usual high-temperature combustion processes (Bowman 1991) and is considered a criteria air pollutant. Harmful effects of SO₂ are well evidenced on plant and human health (WHO 2000; ATSDR 1998; Chen et al. 2007). Once emitted in the atmosphere, SO₂ is oxidized to sulfate aerosol (Seinfeld and Pandis 2006). Saxena and Seigneur (1987) suggested that in addition to the gas-phase oxidation by hydroxyl (OH) radicals, SO₂ oxidation in aqueous aerosols may also contribute significantly to sulfate formation. Wet and dry deposition are the removal modes of SO₂ and its oxidation products. The most severe environmental effect associated with SO₂ is that of acid deposition with consequent damage to terrestrial and aquatic ecosystems. SO₂ dissolves readily in water present in the atmosphere to form sulfurous acid (H₂SO₃). Sulfur trioxide (SO₃), another oxide of sulfur, is either emitted directly into the atmosphere or produced from sulfur dioxide and is rapidly converted to sulfuric acid (H₂SO₄). Sulfate particles are responsible for scattering visible light, global cooling, and haze formation (IPCC 2007). Human health effects of SO₂ exposure have been associated with respiratory illness and exacerbation of existing cardiovascular and pulmonary diseases in children, the elderly, and asthmatics (Bremmer et al. 1999; Mar et al. 2000; Maynard and Ayres 2014). Plant exposure to ele-