

THE ROLE OF AEROSOLS IN THE EARTH'S ATMOSPHERE

By

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Table of Contents

List of Tables	vii
List of Figures	viii
Abstract	ix
Acknowledgements	x
1 INTRODUCTION	1
1.1 Objective the Study	2
1.1.1 General objective of the study	2
1.1.2 Specific objective of the study	2
2 PHYSICAL PROPERTIES OF ATMOSPHERIC AEROSOLS	4
2.1 Introduction	4
2.2 Types of Aerosols	5
2.2.1 Mineral dust	5
2.2.2 Sea spray	5
2.2.3 Volcanic aerosols	6
2.2.4 Smoke	6
2.2.5 Soot	7
2.3 Organic and Inorganic Aerosols	7
2.3.1 Organic aerosol	7
2.3.2 Inorganic aerosols	8
2.4 Chemical Composition of Aerosols	10
2.5 Sources of Aerosols	10
2.5.1 Natural source of aerosol	11
2.5.2 Anthropogenic aerosol	11
2.6 Size and Shape of Aerosols	11

2.6.1	The area, volume, moments and mass distribution of aerosols	13
3	CHEMICAL PROPERTIES OF AEROSOLS	17
3.1	Formation and Processes of Aerosol	17
3.2	Removal and Transport of Aerosols in the Atmosphere	18
3.2.1	Wet deposition	18
3.2.2	Dry deposition	20
3.2.3	Sedimentation	21
3.2.4	Diffusion	21
3.3	Atmospheric Aerosol Chemistry	22
3.3.1	Nucleation	23
3.3.2	Coagulation	24
3.3.3	Lifetime of aerosol	24
3.3.4	Aerosol chemical reaction	25
4	OPTICAL PROPERTIES OF AEROSOL	30
4.1	Introduction	30
4.2	Aerosol Optical Depth (AOD)	32
4.3	Absorbtion of Radiation by Aerosol	33
4.4	Scattering of Radiation by Aerosols	34
4.5	Reflection of Radiation by Aerosols	38
4.6	Long-Wave Aerosol Radiative Properties	38
4.7	Global Distribution of Aerosol Optical Properties	39
4.8	Aerosol Radiative Forcing	40
4.9	Role Aerosols in Radiative Transfer	46
5	ROLE OF AEROSOLS ON CLIMATE	50
5.1	Aerosol Cloud-Interaction	50
5.2	Role of Aerosols in Rain-Drop Formation (Nucleation Center)	51
5.3	Water Cycle	52
5.4	The Effects Aerosol on Cloud Properties	53
5.4.1	Effects of aerosol on cloud optical thickness and albedo	53
5.5	Effects of Aerosols in Precipitation	56
5.6	Global Aerosol Forcing	57
5.7	Climatology of Stratospheric Aerosols	58
5.8	Climatology of Tropospheric Aerosols	59
5.9	Observation and Measurement of Aerosol	60
5.9.1	Aerosol chemical composition and size distribution	60
5.9.2	Aerosol scattering, absorption, and extinction coefficients	61

5.9.3	Aerosol optical depth(AOD)	62
5.10	Role of Aerosol in Climate Change	63
5.11	Role of Aerosol in the Global Climate System	64
5.12	How do Aerosols Affect Climate	65
5.12.1	Soot and direct radiative effect of aerosol	66
5.12.2	Acidification of precipitation	67
5.13	Aerosol and their Relation to Global Climate and Climate Sensitivity	68
6	SUMMARY AND CONCLUSION	70
	Bibliography	71

List of Tables

4.1	Global-mean radiative forcing due to aerosol radiation interactions [5].	42
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List of Figures

4.1	Scattering of radiation by an aerosol layer [7].	37
4.2	Illustrates the fact that as the solar zenith angle increases, a larger fraction of the forward scattering (scattering angles ranging between 0 and 90°) contributes to scattering in the upward direction. Combining these effects, we show that the radiative effect of a thin aerosol layer increases with the solar zenith angle up to the point and the phase function is represented by an ellipsoid which intercepts the horizontal plane [5].	43
4.3	A schematic representation of interaction between aerosols over the Earth's surface having albedo S with incoming solar radiation, T denotes the transmission (diffuse + direct) due to aerosols in the layer and R is the diffuse reflectance of the aerosols in the layer [4].	47
5.1	schematic illustration of aerosol-cloud interaction. (a) cloud albedo and lifetime effect, and (b) semi-direct effect [4].	51
5.2	The change in the albedo of a cloud per unit change in the droplet concentration ($\Delta A/\Delta N$) as a function of the cloud albedo (A) and the droplet concentration (N), for a cloud with a constant liquid-water content [14].	56
5.3	The effect of aerosols on the climate [4].	65

Abstract

In this project we have review the role of aerosols in the Earth's atmosphere. We considered an important source of aerosol particles in the atmosphere are the processes of physical and chemical interaction of substances, in particular the interaction of the main gas, and water va-pour. In our work aerosol particles interact with sun radiation by reflection, scattering, or absorbing incoming radiation and the amount of reflection, scattering or absorbing we can explain the effect of aerosol in radiative transfer and also determine whether aerosol particles have a net warming or cooling effect on climate. This effect described by three related optical properties of aerosols. Aerosols play a crucial role in Earth's atmosphere, influencing climate, weather patterns, and air quality. These tiny particulate matter, which can be of natural or anthropogenic origin, act as cloud condensation nuclei, facilitating cloud formation and impacting precipitation processes.

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Chapter 1

INTRODUCTION

An aerosol is technical definition as a suspension of a fine liquid and solid particles in gas. Aerosol is derived from the Greek word "aero" air and "sol" solution and is a term applied to solid or liquid materials suspended in the air [1]. In physics, aerosols are a mixture of air and particles (solid, liquid), that are in equilibrium. In meteorology and atmospheric physics, the aerosols is represented by a particles suspended in air, of greatly differing composition, shape dimension, and properties [2]. Aerosol particle have varied physio chemical properties and span several orders of magnitude in size, but are limited to particles with atmospheric residence time above some hours with this limitation aerosol are considered to range approximately from $0.001 - 100\mu m$ in diameter (more see section, 2.6). Atmospheric aerosols are generated from either primary or secondary aerosols and both types of aerosols have natural and anthropogenic origin. Atmospheric aerosols are important in many ways. Aerosol content affect the Earth's albedo and play a major role in the global radiation balance and climate. Various aerosol acts as a cloud condensation nuclei (CCN) and are important in the formation cloud and precipitation (indirect effect of aerosols). Aerosol particles also play a significant role in the transfer of the solar and thermal

radiation affecting the radiation regime of the atmosphere Earth's surface system, and consequently, the weather and climate on the Earth. Aerosol play an important role in the absorption and scattering of solar radiation (direct effect of aerosols) [3].

This project is organized in to six chapters. Chapter one the introduction part of overview the basic definition of aerosol and the role of aerosol in the Earth's atmosphere simply, in chapter two physical properties of atmospheric aerosols that including, types of aerosol, source of aerosol, chemical composition of aerosol, and size and shapes of aerosols. In chapter three, chemical properties of aerosols that includes formation and removal of aerosol, migration and transport of aerosol in the atmosphere, and atmospheric aerosol chemistry, chapter four optical properties of atmospheric aerosol, chapter five the role of aerosol in climate and finally in chapter six summary and conclusion of the study are included.

1.1 Objective the Study

1.1.1 General objective of the study

The general objective of this project is to review the role of atmospheric aerosols in the Earth's atmosphere.

1.1.2 Specific objective of the study

To study the sources, properties, and important of aerosols.

To review the optical characteristics of atmospheric aerosols.

Describe the role of aerosol in radiative transfer through scattering, absorbing, and reflection.

To analyzing and identify the role of aerosol on climate balance systems particularly their roles in radiative forcing, cloud formation, and precipitation patterns.

Chapter 2

PHYSICAL PROPERTIES OF ATMOSPHERIC AEROSOLS

2.1 Introduction

The most important physical characteristics of an aerosol population are the size distribution, chemical composition, and shape of the particles. In this chapter we describe about the properties of atmospheric aerosols. Including their physical properties, sources, different types of aerosol, chemical composition of aerosols, and size and shape of aerosols [4, 5].

2.2 Types of Aerosols

2.2.1 Mineral dust

Mineral dust is emitted when wind blows over deserts otherwise dry soils, lifting the particles get carried off into the atmosphere. Mineral dust is one of the most abundant aerosol types and dust particles are also large compared to other aerosol often size of several micro meters in diameter. Mineral dust is originates mainly from deserts, dry lake beds, arid and semiarid regions, in addition to drier region where vegetation has been reduced or from soil surfaces that are distributed by man made activities. The average lifetime of dust particles in the atmosphere is about 2 weeks, during which dust can be transported thousands of kilometers [4, 17].

2.2.2 Sea spray

Sea spray, the largest component of sea salt, also includes organic materials such as dissolved organic carbon, bacteria, phytoplankton, and micro-algae. It primarily forms when air bubbles burst at the ocean's surface. The characteristics of sea spray depend on factors like wind speed, humidity, and sea surface temperature. The ocean is the main source of sea-salt aerosols, which are produced directly from the ocean and released into the atmosphere. SSA (sea salt aerosols) particles are efficient cloud condensation nuclei (CCN) that can influence cloud properties, reflectivity, lifetime, and precipitation processes [4, 17].

2.2.3 Volcanic aerosols

Volcanoes can emit fragments of pulverized rocks and minerals, usually called volcanic ash, during explosive eruptions. Volcanoes produce two main aerosols ash and sulfate. Ash is a dark, large aerosols by the pulverization of crystallized magma and contains mineral such as silicon and feldspar. Volcanic emit SO_2 which is oxidized in the atmosphere to sulfur aerosols. This aerosols is occur in stratosphere. Volcanoes also emit sulfur-rich gases (in the form of sulfur dioxide (SO_2), and hydrogen sulfide (H_2S) that get oxidized in the atmosphere to form submicronic sulfate aerosols. Volcanic aerosols are a mixture of absorbing and scattering particles which the absorbs usually being large enough to fallout quickly so that the smaller ones dominate. If these sulfur-containing gases are emitted in the troposphere, the residence time of the subsequent aerosols will be short, a few weeks at most. However, if the eruption is powerful enough to inject the sulfur gases in the stratosphere, then the volcanic aerosols have a much longer residence time, of the order of a few months to more than a year, depending on the region and altitude of injection [5, 17].

2.2.4 Smoke

Smoke is a solid or liquid aerosols that result of incomplete composition or condensation of super saturation va-pour. Most smoke particles are sub micrometer in size. It emitted from fires both natural wild fire and human caused (agricultural particles such as land cleaning and waste incineration). There are often also referred to as biomass aerosols and are composed of organic (brown) and black carbon (soot). The composition of smoke is strongly depend on the fuel surface burning and atmospheric condition. Organic and black carbon light to varying extents, which warms,

the atmosphere. Locally while simultaneously shading and cooling the surface below when black carbon is deposited on snow and ice, it decreases their reflectivity of those surface, which can warm them, and speed up melting [6, 11].

2.2.5 Soot

Soot is a black carbon (BC) that is the dominant form of light absorbing particles matter in the atmosphere. Black carbon (soot) is emitted by incomplete combustion processes, both human (diesel engines) and natural (wild fire). Its ability to absorb visible and infrared radiation means black carbon (soot) can heat the atmosphere and darken surface specifically snow and ice. These effects have important consequences of earth climate and climate change. Soot as an air borne contaminant in the environment has many different sources, all of which are results of some form of pyrolysis. They includes soot from burning, internal combustion on engines power plants boilers, hog-fuel boilers, central steam, heat boilers waste incineration, burning, house fires, forest fires and furnaces. Soot is often associated with combustion-generated primary carbonaceous aerosol [6, 17, 21].

2.3 Organic and Inorganic Aerosols

2.3.1 Organic aerosol

Aerosols can be classified broadly as inorganic or organic based on their chemical composition. The organic fraction refers to carbon-containing compounds. One can distinguish primary from secondary organic aerosols. Secondary organic aerosols originate from volatile organic compounds that are oxidized in the atmosphere, some of

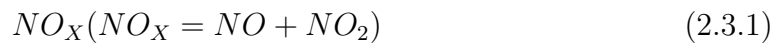
which can actually evaporate from primary organic aerosols. The chemical composition of organic aerosols originating from the combustion of fossil hydrocarbons (hydrocarbon-like organic aerosols) differs from that of organic aerosols originating from the combustion of biomass (biomass burning organic aerosols). Organic aerosols containing oxygen-rich compounds are called oxygenated organic aerosols. Primary organic aerosols particle were regarded as relatively non-volatile composites of organic compounds emitted by individual sources, such as biomass burning, gasoline and diesel vehicles, food preparation, smoking, and numerous other small sources. Secondary organic aerosol is regarded as an additional coating of secondary organic compounds formed via gas-phase oxidation of volatile organic carbon (VOC) precursors [5, 7].

2.3.2 Inorganic aerosols

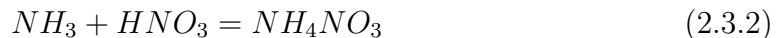
The presence of inorganic material is very usual in aerosols. This is the case of sea spray composed mainly of sodium chloride ($NaCl$), but also of sulphate aerosols whose chemical composition depends on the degree of neutralization of the sulphuric acid. The chemical composition of sulphate therefore varies between pure sulphuric acid (H_2SO_4), ammonium bisulphate (NH_4HSO_4) and ammonium sulphate ($(NH_4)_2SO_4$). Inorganic aerosols are generally very hygroscopic, and do not absorb light much, if at all, in the solar spectrum. There are different types of secondary inorganic aerosol: that including sulphate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+). The secondary inorganic aerosol are formed from gas-phase precursor, their concentration are dependent on emission of NO_x , SO_2 , and ammonia (NH_3) precursors gases. The emission of NO_3 , NH_3 , and SO_2 form both natural and

anthropogenic sources [5].

Sulphate; as the formation of sulphate, in a first stage sulfuric acid aerosol is formed. These include their effect on the radiative properties of the atmosphere (by acting as CCN and affecting cloud albedo and lifetime), their contribution to visible degradation and acid deposition and the impact of aerosol on climate. Ammonium; down wind of sources of gaseous ammonia, NH_4^+ is the principle cation associated with fine sulphate and fine nitrate in the continental aerosol. Ammonia is released by natural soils, coal composition, biomass burning, fertilizer application and production or is emitted as a result of the decay of waste products from domestic animals, wild animals, sea and oceans. Nitrate; in fine particles comes primarily from the reaction of gas-phase nitric acid with gas-phase ammonia forming particulate ammonium nitrate. The gas-phase the nitric-acid (HNO_3) is produced by the oxidation of:



gas, which itself is emitted by the composition of fossil fuels and biomass, but also by natural sources, such as soils. In the presence of ammonia, a fraction of the nitric acid will partition in to the particle phase to form NH_4NO_3 aerosols, as show equation:



Thus, as sources of NH_4NO_3 , both NH_3 and NO_3 sources have been taken into account [8].

2.4 Chemical Composition of Aerosols

Atmospheric aerosols are generally composed of variable amounts of sulphate, ammonium, nitrate, sodium, chloride, trace metals, crustal elements, water and carbonaceous material. The sulphate component is derived predominantly from the atmospheric oxidation of anthropogenic and natural sulphur-containing compounds such as sulphur dioxide (SO_2) and dimethyl sulphide (DMS), respectively. Nitrate is formed mainly from the oxidation of atmospheric nitrogen dioxide (NO_2). Sulphate and nitrate are initially formed as sulphuric and nitric acids, but are progressively neutralized by atmospheric ammonia forming the corresponding ammonium salts. Crustal materials include soil dust and windblown minerals. The carbonaceous fraction of the aerosols consists of both elemental and organic carbon. Elemental carbon (EC), also called black carbon, graphitic carbon, or soot, is emitted directly into the atmosphere, predominantly from combustion processes. While particles containing organic carbon (OC) can be directly emitted into the atmosphere (i.e. from primary sources such as biomass burning and combustion processes), they can also be introduced by secondary organic aerosol (SOA) formation. The latter process occurs when volatile organic compounds undergo atmospheric oxidation reactions, forming products that have low enough volatilities to form aerosol via either nucleation or gas-to-particle partitioning to pre-existing particles [7, 9].

2.5 Sources of Aerosols

In order to distinguished particles from the point view of their source there are two main classification of atmosphere aerosols. The first processes to classify the particles

according to their origin that is anthropogenic and natural aerosols. Where as the second processes to differentiate the particles from their mode of emission in the atmosphere: primary and secondary aerosols. It is finally possible to combine the two classifications for a more complete description, the primary and secondary particles being able to be of natural (biogenic) or anthropogenic origin.

2.5.1 Natural source of aerosol

Natural source of aerosols are originated from the nature. Natural aerosols source including; mineral dust, sea-salt, biomass burning aerosol, biogenic aerosol (bacteria, algae, spores, and debris), dimethyl sulphide(*DMS*), volcanic, organic materials smoke, mist, fog, and forest exudates.

2.5.2 Anthropogenic aerosol

The sources are largely dominated by emission from the combustion of fossil fuels (coal and oil), bio fuels (plant biomass including: food, vegetable, oil, animal wastes), other fuels (peat) or vegetation fire caused by human. Anthropogenic aerosols also including industrial dust, biomass burning, black carbon (from fossil fuel), organic carbon (from fossil), SO_2 , NO_x , NH_3 , and volatile organic compounds [7, 10].

2.6 Size and Shape of Aerosols

Aerosol particles of different sizes, shape and composition play a vital role in many atmospheric processes such as visibility, radiation balance, air pollution, and cloud formation. The sizes of aerosols extend over several orders of magnitude from 0.001

μm to $100 \mu\text{m}$, sweeping from a cluster of molecules at one end to hailstones at the other end. Based on their size ranges, aerosols are divided into three categories: Aitken nuclei ($0.001 \mu\text{m} < r < 0.1 \mu\text{m}$), large particles ($0.1 \mu\text{m} < r < 1.0 \mu\text{m}$), and giant particles ($r > 1.0 \mu\text{m}$).

The aerosol size distribution is represented by a function $n(r)$ which is the number of particles per unit volume whose radius lies between r and $r + dr$ and the radius number density distribution $N(r)$. The problem is avoided by using the differential radius number density distribution $n(r)$ defined as [11, 16].

$$n(r) = \frac{dN(r)}{dr}. \quad (2.6.1)$$

The total number of particles per unit volume, N_0 (the total aerosol concentration) can be obtained by integrating over the size distribution:

$$N_0 = \int_0^{\infty} n(r)dr, \quad (2.6.2)$$

The particle distribution to consider adopting is the normal distribution

$$n(r) = \frac{N_0}{\sqrt{2\pi}\sigma} \frac{1}{\sigma} \exp\left[-\frac{(r - \mu)^2}{2\sigma^2}\right]. \quad (2.6.3)$$

where μ is the mean and σ is the standard deviation of the distribution. The size of particles in an aerosol generally covers several orders of magnitude. The moments of a size distribution are simple metrics to express the shape of the distribution. The i^{th} moment m_i of $n(r)$ is defined

$$m_i = \frac{1}{N_0} \int_0^{\infty} (r - c)^i n(r)dr, \quad (2.6.4)$$

where c is some constant. The raw moment is the moment where $c = 0$. The moments about the mean are called central moments. Generally a size distribution

is characterized by its center and by its spread. The center of a distribution can be represented by the mean, μ defined by

$$\mu = \frac{\int_0^{\infty} rn(r)dr}{\int_0^{\infty} n(r)dr} = \frac{1}{N_0} \int_0^{\infty} rn(r)dr. \quad (2.6.5)$$

The mean is the first raw moment of a size distribution. The mode is the peak (maximum value) of a size distribution, and the median is the "meddle" value of a data set. The variance (σ^2) is defined as

$$\sigma^2 = \frac{1}{N_0} \int_0^{\infty} (r - \mu)^2 n(r)dr, \quad (2.6.6)$$

standard deviation (σ), of a distribution is the square root of the variance.

2.6.1 The area, volume, moments and mass distribution of aerosols

The distribution of particle area can be represented by a differential area density distribution, $a(r)$ which represents the area of particles whose radii lie between r and $r + dr$ per unit volume, i.e.

$$A(r) = \int_r^{r+dr} a(r)dr. \quad (2.6.7)$$

Hence,

$$a(r) = \frac{dA}{dr}. \quad (2.6.8)$$

For spherical particle,

$$a(r) = \frac{dA}{dN} \frac{dN}{dr} = 4\pi r^2 n(r). \quad (2.6.9)$$

The total particle area per unit volume, A_0 , is then given by

$$A_0 = \int_0^{\infty} a(r)dr. \quad (2.6.10)$$

The distribution of particle volume can be represented by a differential volume density distribution, $u(r)$ which represents the volume contained in particles whose radii lie between r and $r + dr$ per unit volume, i.e.

$$V(r) = \int_r^{r+dr} u(r)dr. \quad (2.6.11)$$

Hence,

$$u(r) = \frac{dV}{dr}. \quad (2.6.12)$$

For spherical particle,

$$u(r) = \frac{dV}{dN} \frac{dN}{dr} = \frac{4}{3}\pi r^3 n(r). \quad (2.6.13)$$

The total particle area per unit volume, V_0 is given by

$$V_0 = \int_0^{\infty} u(r)dr. \quad (2.6.14)$$

The distribution of mass can be represented by a differential mass density distribution, $m(r)$ which represents the mass contained in particles with radii between r and $r + dr$ per unit volume, i.e.

$$M(r) = \int_r^{r+dr} m(r)dr. \quad (2.6.15)$$

Hence,

$$m(r) = \frac{dM}{dr} \quad (2.6.16)$$

For spherical particle,

$$m(r) = \frac{dM}{dN} \frac{dN}{dr} = \frac{4}{3}\pi r^3 \rho n(r). \quad (2.6.17)$$

Where ρ is the density of the aerosol material. The total particle mass per unit volume, M_0 , is

$$M_0 = \int_0^{\infty} m(r)dr. \quad (2.6.18)$$

The i^{th} order of moment of the size distribution (raw moment of a log normal distribution) is given by

$$m_i = N_0 \exp\left(i\mu + \frac{i^2\sigma^2}{2}\right), \quad (2.6.19)$$

The first three moments are

$$\begin{aligned} m_1 &= \int_0^\infty rn(r)dr = N_0 \exp\left(\mu + \frac{1}{2}\sigma^2\right) = N_0 r_m \exp\left(\frac{1}{2}\sigma^2\right) \equiv N_0 r_m \exp\left(\frac{1}{2}\ln^2 S\right) \\ m_2 &= \int_0^\infty r^2 n(r)dr = N_0 \exp\left(2\mu + 2\sigma^2\right) = N_0 r_m^2 \exp\left(2\sigma^2\right) \equiv N_0 r_m^2 \exp\left(2\ln^2 S\right), \\ m_3 &= \int_0^\infty r^3 n(r)dr = N_0 \exp\left(3\mu + \frac{9}{2}\sigma^2\right) \equiv N_0 r_m^3 \exp\left(\frac{9}{2}\ln^2 S\right) \end{aligned}$$

The mean radius, the surface area density and the volume density of a log normal distribution are given by

$$Mean = \frac{1}{N_0} \int_0^\infty rn(r)dr = \frac{1}{N_0} m_1 = \frac{1}{N_0} N_0 \exp\left(\mu + \frac{1}{2}\sigma^2\right) \quad (2.6.20)$$

$$= r_m \exp\left(\frac{1}{2}\sigma^2\right) \equiv r_m \exp\left(\frac{1}{2}\ln^2 S\right) \quad (2.6.21)$$

$$Area = \int_0^\infty 4\pi r^2 n(r)dr = 4\pi m_2 = 4\pi N_0 \exp\left(2\mu + 2\sigma^2\right) \quad (2.6.22)$$

$$= 4\pi N_0 r_m^2 \exp\left(2\sigma^2\right) \equiv 4\pi N_0 r_m^2 \exp\left(2\ln^2 S\right) \quad (2.6.23)$$

and

$$Volume = \int_0^\infty \frac{4}{3}\pi r^3 n(r)dr = \frac{4}{3} m_3 = \frac{4}{3} N_0 \exp\left(3\mu + \frac{9}{2}\sigma^2\right) \quad (2.6.24)$$

$$= \frac{4}{3}\pi N_0 r_m^3 \exp\left(\frac{9}{2}\sigma^2\right) \equiv \frac{4}{3}\pi N_0 r_m^3 \exp\left(\frac{9}{2}\ln^2 S\right) \quad (2.6.25)$$

For a log normal distribution the effective radius is

$$r_e = \frac{m_3}{m_2} = \frac{\exp\left(3\mu + \frac{9}{2}\sigma^2\right)}{\exp\left(2\mu + \frac{4}{3}\sigma^2\right)} = \exp\left(\mu + \frac{5}{2}\sigma^2\right) \quad (2.6.26)$$

$$= r_m \exp\left(\frac{5}{2}\sigma^2\right) \equiv r_m \exp\left(\frac{5}{2}\ln^2 S\right) \quad (2.6.27)$$

The effective variance for a distribution can be define:

$$u_e = \frac{\int_0^\infty (r - r_e)^2 \pi r n(r) dr}{r_e^2 \int_0^\infty n(r) dr} = \frac{m m_4}{m_3^2} - 1 \quad (2.6.28)$$

The forth moment of size distribution

$$m_4 = N_0 r_m^4 \exp(8 \ln^2 S) \equiv N_0 \exp\left(4\mu + \frac{16}{2}\sigma^2\right) \quad (2.6.29)$$

$$u_e = \frac{m_2 m_4}{m_3^2} - 1 = \frac{(N_0 \exp(2\mu + 2\sigma^2)) (N_0 \exp(4\mu + \frac{16}{2}\sigma^2))}{(N_0 \exp(3\mu + \frac{9}{2}\sigma^2))^2} - 1 \quad (2.6.30)$$

$$= \exp(\sigma^2) - 1 \equiv \exp(\ln^2 S) - 1 \quad (2.6.31)$$

It is common for the log normal distribution to be expressed in terms of the radius [5, 16]. Nothing that

$$\frac{dl}{dr} = \frac{1}{r} \quad (2.6.32)$$

Then in terms of radius rather than log radius we have

$$n(r) = \frac{dN(r)}{dr} = \frac{dN(l)}{d(l)} \frac{dl}{dr} = n_l(l) \frac{dl}{dr} = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[-\frac{(\ln(r) - \mu)^2}{2\sigma^2}\right]. \quad (2.6.33)$$

Chapter 3

CHEMICAL PROPERTIES OF AEROSOLS

3.1 Formation and Processes of Aerosol

An important source of aerosol particles in the atmosphere are the processes of chemical interaction of substances, found in the gas phase, in particular the interaction on the main atmospheric gases (nitrogen, oxygen, sulfur), and also of water vapour with some gases, such as, sulfur dioxide, chlorine, ammonia, and ozone (this part is more discussed in section 3.3). An important role in the formation of aerosols is played by volcanic emissions, forest fires, biological processes, and production activities of mankind. In actuality, the formation of new particles from the gas phase is only possible through homogeneous nucleation, or through nucleation initiated by molecular ion clusters too small to be classified as aerosol particles. Heterogeneous nucleation on insoluble particles initiates changes in particle size and composition distributions, but does not increase particle number concentration.

3.2 Removal and Transport of Aerosols in the Atmosphere

Near the surface aerosol may attach direct the surface and thus be removed from the atmosphere by aerosol caused wet deposition and dry deposition. Most atmospheric aerosol particles are advocated by wind and primarily removed from the atmosphere by precipitation. The process of wet deposition describes how aerosol particles are removed by precipitation. Dry deposition is when particles are deposited on the surface by turbulence or gravity. Secondary anthropogenic aerosols are transported by the airflows they encounter during their life in the atmosphere, until covering the global scale. In fact, aerosols formed through g-to-p conversion processes can be transported over long distances, since the time required for g-to-p conversion and the relatively small sizes of the particles formed during such a process lead to long atmospheric residence times for such aerosols. This is, for instance, the case of acidic aerosols such as sulfates and nitrates, which contribute to the so called acid rains [7]. For different aerosols injected near the surface, the lifetime and the transports are typically several hours, days, months, and years in the troposphere and stratosphere [12, 13].

3.2.1 Wet deposition

The main sink of aerosols in the atmosphere, at least for soluble species, is wet deposition, also called wet scavenging. It includes both scavenging that occurs within the clouds during the formation of precipitation (i.e. in-cloud scavenging), and scavenging that occurs under the cloud because of the downward flux of precipitating water

(i.e. below-cloud scavenging). If cloud droplets grow to a size where their sedimentation velocity is large enough, the cloud starts to produce drizzle or precipitation, and aerosol matter that is incorporated in the falling droplets is scavenged. If the falling droplets (or drops) reach the surface, then the aerosol mass is removed from the atmosphere [5].

In-cloud nucleation scavenging refers to the incorporation of CCN in to cloud droplets by activation. The total scavenging coefficient (F_s) of aerosol by a cloud can be defined as:

$$F_s = \frac{\text{aerosol}(ca) - \text{aerosol}(ci)}{\text{aerosol}(ci)}, \quad (3.2.1)$$

where "aerosol (ca)" is the concentration of aerosol in the clear air that enter cloud based and "aerosol (ci)" is the concentration of aerosol that is present interstitially between the cloud droplets. The removal of aerosols by precipitation is one of the major processes by which the atmosphere is cleansed and a balance maintained between the source and sink of atmospheric aerosol. The concentration of aerosol $X(D_p)$ with diameter between D_p and $D_p + dD_p$ after removal that has been active for a time period "t" can be represented by:

$$X(D_p) = X_0(D_p) \exp[-A(D_p)t], \quad (3.2.2)$$

where $X_0(D_p)$ is the aerosol concentration at $t = 0$ and $A(D_p)$ is a parameter called scavenging rate. The scavenging collection efficiency $E(D_p, D)$ of aerosol diameter D_p by precipitation particles of size D can be derived from

$$A(D_p) = \int_0^\infty A(D)E(D_p, D)V(D)N(D)dD, \quad (3.2.3)$$

Where $A(D)$ is effective cross-sectional area of a precipitation particle $V(D)$ is the fall speed of the particle, and $N(D)dD$ the concentration of precipitation particle with diameters between D and $D + D_p$. It is assumed that E is a function only of D_p then

$$E \simeq A(D_p) \left[\sum_i A_i(D) V_i(D) N_i(D) \Delta D_i \right]^{-1}, \quad (3.2.4)$$

where i indicates a size interval for the precipitation particles [14].

3.2.2 Dry deposition

Dry deposition is one of the most important processes occurring in the atmosphere, by which trace gases and particles are removed from the atmosphere in the absence of precipitations. The relative importance of the removal effects by dry deposition depends on various factors, such as: the atmospheric turbulence level, the chemical and water solubility characteristics of the particles, and the nature of the surface and terrain characteristics. In fact, the level of turbulence governs the rate at which aerosols are delivered down to the surface, especially within the layer nearest to the ground, such a rate varying as a function of aerosol size, density, and morphological characteristics. Turbulence in the atmosphere controls the dry deposition apart from the shape, size, and density of particles. The dry deposition flux (F) is expressed as

$$F = u_d C, \quad (3.2.5)$$

where u_d is the deposition velocity and C corresponds to the concentration of aerosols that are depositing from a reference height [5, 7, 14].

3.2.3 Sedimentation

Sedimentation or gravitational settling of the aerosols is an important process, not only because it creates a sink of aerosols at the surface, but also because it redistributes aerosols on the vertical through a (relatively slow) downward motion. Ignoring other processes, sedimentation acts in the following way on the continuity equation for aerosols:

$$\frac{\partial \rho c}{\partial t} \text{div}_z (\rho c V_{sed}) = 0, \quad (3.2.6)$$

where C is the aerosol mixing ratio and V_{sed} the sedimentation velocity. Sedimentation is a slow process for accumulation mode particles. In the troposphere, large-scale transport, convective transport, and wet scavenging are the dominant processes. However in the stratosphere, where vertical transport is slow because of a larger stratification and aerosol residence time is long, sedimentation is an important process to transport aerosols into the troposphere. Sedimentation is also important for aerosol particles of the coarse mode in the troposphere. An aerosol particle is subject to the gravity force and to a drag (air resistance) force. The drag force acts on the particle in the opposite direction to its velocity, and its magnitude is proportional to the size of the particle [5].

3.2.4 Diffusion

Particle diffusion results from its Brownian motion, which is the random motion of the particle in the fluid as a result of its continuous bombardment by gaseous molecules. Diffusion of particles is the net transport under the influence of a concentration gradient. The particles move from regions of high concentration to regions of low concentration [11]. The dynamical or transport (removal) mechanisms are

diffusion (horizontal dilution, i.e., spreading of aerosols to other locations and vertical diffusion) and gravitational sedimentation. The rate of diffusion of particles by atmospheric mixing can be derived based [7] from the following relation.

$$\Phi D = -D_{nM} \left(\frac{\partial n}{\partial z} \right)_{diffusion}, \quad (3.2.7)$$

where ΦD is the diffusive flux ($\text{particles cm}^{-2} \text{sec}^{-1}$), D is the eddy diffusion coefficient ($\text{cm}^2 \text{sec}^{-1}$), nM is the air density (molecules cm^{-3}), and n is the diffusing species (number cm^{-3}). In the tropics the eddy diffusion coefficient is about $1 \times 10^5 \text{ cm}^2 \text{sec}^{-1}$ in the stratosphere [7]. The rate of change of particle concentration due to diffusion is written as:

$$\frac{\partial n}{\partial t} = \frac{-\partial \Phi D}{\partial z} \quad (3.2.8)$$

3.3 Atmospheric Aerosol Chemistry

Aerosol chemistry is significant involved in several technological fields as well as a very important one is the technological of the production of new materials by means of aerosol chemical reactions and processes. A further field in which aerosol chemistry is involved in the technology of combustion processes. Heterogeneous and particle surface chemical reactions are very important parts of the formation and modification processes by aerosol production in burning gases, liquids, and powders and also deal with the formation, processes, characteristics, effects, and chemical composition of secondary anthropogenic aerosols (SAA).

The atmospheric aerosol chemistry deal with the emitted gas and vapor molecule can react chemically in the gas-phase can be absorbed into a particle or droplet where they might chemical react can be transported into stratosphere, or can be removed by

interaction with the earth surface. A significant fraction of secondary anthropogenic aerosols; such as sulfate, nitrate, ammonium, carbonaceous, organic and inorganic is produced in the atmosphere by composition of several physio-chemical processes, gas-phase chemistry, aqueous-phase chemistry, condensation, and adsorption [5, 8].

3.3.1 Nucleation

Nucleation is the transformation of matter from one phase to another phase through the formation of nuclei. For atmospheric aerosols, nucleation refers to the transformation of gas-phase molecules into a cluster of molecules called an aerosol embryo or an aerosol nucleus. Nucleation refers to the formation of a new stable solid or liquid aerosol particle from a gas phase consisting of either gaseous species only (homogeneous, homo-molecular nucleation) or involving two or more gaseous species, one of which most commonly is water (homogeneous, heteromolecular nucleation) or when two or more gaseous species condense onto pre-existing particles (heterogeneous, heteromolecular nucleation). The equation to predict homogeneous and heterogeneous nucleation rates is based on classical nucleation theory [5].

$$\Delta G = 4\pi r_n^2 \delta_n - \frac{4\pi r_n^3 \rho_n R^* T}{3 mg} \ln S_g, \quad (3.3.1)$$

which states that the change in Gibbs free energy (ΔG) during nucleation of a cluster of molecules is equal to a surface-tension term minus a saturation ratio term. r_n stands for the radius of the nucleating cluster (cm), ρ is the mass density of the condensed cluster (gcm^{-3}), R^* is the gas constant ($8.314Jmole^{-1}K^{-1}$), mg is the molecular weight of the condensing gas ($gmole^{-1}$), σ_n is the surface tension of the condensed cluster (Jcm^{-2}), and S_g is the saturation ratio of the gas [10, 17].

3.3.2 Coagulation

Coagulation occurs when two particles collide and stick together. This process reduces the number concentration of particles in the atmosphere but conserves the volume concentration of particles. Coagulation is an important microphysical process as neglecting this process would result in an unusually large aerosol number concentration which is erroneous. The derivation of numerical methods for solving coagulation begins with the integro-differential coagulation equation:

$$\frac{\partial n_u}{\partial t} = \frac{1}{2} \int_0^u \beta_{u-w} n_{u-w} n_w dw - n_u \int_0^\infty \beta_{u,w} n_w dw, \quad (3.3.2)$$

where $u-w$ and w are the volumes of coagulating aerosol particles, u is the volume of the new, coagulated particle, n is the time-dependent number concentration (cm^{-3}) of particles of volume u , $u-w$, or w , and β is the coagulation kernel or rate coefficient of the two colliding particles ($cm^3 particle^{-1} s^{-1}$) [17]. The equation indicates that the change in the number concentration of particles of volume u is equal to the rate at which particles of volume $u-w$ coagulate with particles of volume w minus the rate at which particles of volume u are lost due to coagulation with particles of all sizes [4, 5, 10].

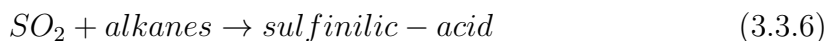
3.3.3 Lifetime of aerosol

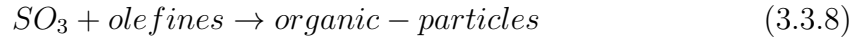
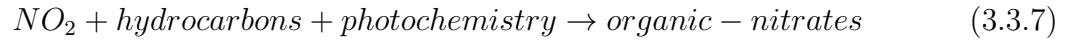
Aerosol lifetime range from hour to years, dependent primarily on the size of the particles and the height at which they are injected into the atmosphere for different aerosol particles injected near the surface the lifetime and transport are typically several hours, days, month's, and years in the troposphere and stratosphere. For fine aerosols injected near the surface the lifetime is typically several day. This increases

to weeks or month's for particle injected into or transported to the upper troposphere, and fine aerosols in the stratosphere (volcanic sulfate) can remain three four years. Because of these short life times, many therefor are not usually transported far from sources [7]. The lifetime of atmospheric aerosol particles depends on their chemical nature, size and on the altitude range. Aerosol particles in the lower stratosphere have a much longer lifetime of up to 1 year before they penetrate the tropopause and undergo tropospheric loss processes. During its atmospheric lifetime, aerosol can easily be transported on intra-hemispheric scales during its residence time of more than one week in the free troposphere [10].

3.3.4 Aerosol chemical reaction

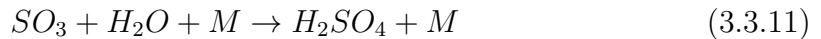
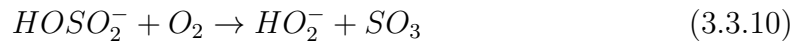
Secondary aerosol (SA) originates in the atmosphere due to gas-to-particle conversion. Many anthropogenic aerosols show irregular shapes, since they originate and grow through two basic processes which both regulate the mass transfer from the gas phase to the particulate phase (g-to-p conversion) where pre-existing particles may grow through material condensation from the gas phase and new particles may form through homogeneous nucleation. In these g-to-p conversion processes, three major families of chemical species are involved, containing sulfur, nitrogen, and organic substances. The most important role in these processes is played by the following different reactions:



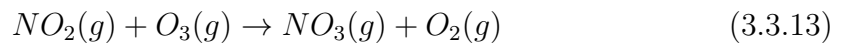


Forming from the above reaction molecules of H_2SO_4 and HNO_3 [4].

Secondary particle reaction from SO_2 : about 35% of the number density concentration of airborne aerosols consists of sulfates forming through oxidation of anthropogenic SO_2 emissions, in large part originated from fossil fuel combustion (primarily coal and oil). The following set of reactions of the transformation of sulfur dioxide to give sulfuric acid and oxidized of SO_2 to give H_2SO_4 is considered.



Secondary particle reaction from NO_X : in situ chemical reactions involving and generating many important atmospheric trace constituents lead to the formation of nitrate aerosols. Most of such gaseous reactions are initiated by photolysis involving radicals and occur with one to three different molecules. These reactions can be classified as homogeneous or heterogeneous, according to the aforementioned definitions. One of the most important homogeneous reactions is the following:



The concentration of ozone (O_3) is calculated as:

$$[O_3] = J[NO_2]/K_2[NO] \quad (3.3.14)$$

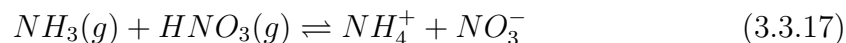
Which constitutes in practice the major source of the nitrate radical (NO_3) in the atmosphere. Mixing an inorganic aerosol, like nitric acid (HNO_3), with organic compounds (aldehydes) which can appreciably increase the rate of aerosol growth is an example of a heterogeneous reaction OH concentrations. Even with rigorous pollution controls, increasing numbers of cars in urban centers can produce high concentrations of NO, which can then be converted to NO_2 through the following reaction:



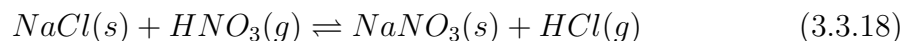
The NO_2 can then be involved in the production of O_3 . Because the rate coefficient of the reaction represented in Eq. (3.3.14) increases as temperature decreases, the production of NO_2 can become significant in urban areas during cold winters. In the ammonianitric acidwater system, NH_4NO_3 is found in the aqueous state for air RH higher than the deliquescence point. In fact, ammonia and nitric acid react in the atmosphere to form ammonium nitrate (NH_4NO_3):



The ammonia nitrate is formed in areas characterized by high ammonia and nitric acid concentrations in the presence of low sulfate concentrations. In ammonium nitrate solutions for RH higher than that of deliquescence, NH_4NO_3 is found in the aqueous state. The corresponding dissociation reaction is:

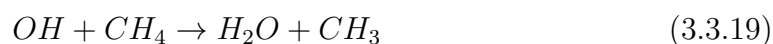


The addition of NaCl to an urban aerosol can have a series of interesting effects, including the following reaction of NaCl with HNO_3 :



As a result of this reaction more nitrate is transferred to the aerosol phase and is associated with the coarse sea-salt particles [7, 10].

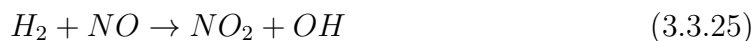
Secondary organic aerosol reaction: in the formation photochemical smog, the OH radical can initiate a chain reaction, which attacks the hydrocarbon pollutants in urban air, through one of the following reaction:



or



The resulting radical CH_3 in Eq. (3.3.19), H in Eq. (3.3.20), and CH_3CO in Eq. (3.3.21) are then involved in the reaction that oxidize NO to NO_2 and regenerate OH. For example, CH_3 formed through the reaction given in Eq. (3.3.19) can initiate the following sequence of reaction.



To form OH and oxidized NO to NO_2 . The net result of these four reactions together with the reaction described in Eq. (3.3.19) is given by :



Where the oxidation of CH_4 result in the oxidation of NO to NO_2 without consuming O_3 . The reaction defined in Eq. (3.3.26) produces form-aldehyde ($HCHO$), which is an eye irritant and constitutes an efficient source of HO_x , as can be seen in the sequence of the two following reactions producing HO_2 [10]:



and



Chapter 4

OPTICAL PROPERTIES OF AEROSOL

4.1 Introduction

Aerosols optical properties are describe the ability of aerosol to scatter, reflect and absorb radiation at different wave length. Since aerosol particles interact with sun's radiation by reflection, scattering or absorbing incoming radiation, and the amount of reflection, scattering or absorbtion determine whether aerosol particles have a net warming or cooling effect on a climate. The optical properties of aerosol in turn depend on the number and size (physical), and their scattering and absorbtion (chemical) nature. The three aerosol related optical properties namely: aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry parameter that are crucial to determine aerosol radiative effect depend on the physical and chemical characteristics of aerosol. Single scattering albedo (SSA) is an aerosol optical depth equation, and depends on the chemical composition of aerosol types, and also it determined by the contribution of scattering due to aerosols vis-'a-vis their extinction (scattering + absorbtion). In combination with surface reflectance, SSA determines the warming

or cooling contribution from aerosol. It is represented in the following equation:

$$SSA(\lambda) = \frac{\beta^{sca}(\lambda)}{\beta^{sca}(\lambda) + \beta^{abs}(\lambda)} = \frac{\beta^{sca}(\lambda)}{\beta^{ext}(\lambda)}, \quad (4.1.1)$$

where $\beta^{sca}(\lambda)$ is the scattering coefficient, $\beta^{abs}(\lambda)$ is absorbing coefficient and $\beta^{ext}(\lambda)$ is the extinction coefficient. The extinction coefficient can be describe in terms of volume coefficient. The volume extinction coefficient $\beta^{ext}(\lambda, r)$ represent the energy removed from a beam per unit distance by extinction. For mono-disperse aerosol they are calculated from

$$\beta^{ext} = \sigma^{ext}(\lambda, r)N(r) = \pi r^2 Q^{ext}(\lambda, r)N(r), \quad (4.1.2)$$

where $N(r)$ is the number of particles per unit volume at some radius, r , $\beta^{ext}(\lambda, r)$ is volume extinction coefficient, $\sigma^{ext}(\lambda, r)$ is extinction cross section, and $Q^{ext}(\lambda, r)$ is efficiency factor. For a collection of extinction aerosol particles the extinction volume coefficient given by

$$\beta^{ext} = \int_0^\infty \sigma^{ext}(\lambda, r)n(r)dr = \int_0^\infty \pi r^2 Q^{ext}(\lambda, r)n(r)dr, \quad (4.1.3)$$

where $n(r)$ represents the number of particle with radii between r , and $r + dr$ per unit volume. It is also useful to define the quantities per particles i.e.

$$\bar{\sigma}^{ext}(\lambda) = \frac{\int_0^\infty \sigma^{ext}n(r)dr}{\int_0^\infty n(r)dr} = \frac{\beta^{ext}(\lambda)}{No}, \quad (4.1.4)$$

where $\bar{\sigma}^{ext}(\lambda)$ is the mean extinction cross section. Asymmetry parameter (g) describes the shape of aerosols, and depends on both size distribution and chemical composition [4, 16].

4.2 Aerosol Optical Depth (AOD)

Aerosol optical depth is directly proportional aerosol loading, and the size distribution of aerosol mass burden in atmospheric column typically, in an aerosol size distribution sub-micron aerosols are order of magnitude than super micro particles. This is defined as the column integrated value of the light coefficient δ_e and is represented mathematically:

$$AOD = \int_Z^{\infty} \delta_e dz, \quad (4.2.1)$$

AOD is strongly dependent on wavelength and it is a common parameter to identify both sources of aerosol generation and their evolution as it represents the aerosol loading in the atmospheric column. AOD tells us how much direct sun light is preventing from reaching the ground by these aerosol particles. The integral of the extinction coefficient over the vertical is called the aerosol optical thickness (AOT) or aerosol optical depth (AOD):

$$\tau = \int_{surface}^{topofatmosphere} \sigma^{ext}(z) dz, \quad (4.2.2)$$

The total integrated columnar optical depth of the atmosphere is given as:

$$\tau = \tau_{rs} + \tau_{aerosol} + \tau_{ma} \quad (4.2.3)$$

where τ_{rs} is the Rayleigh scattering optical depth (scattering due to air molecules), $\tau_{aerosol}$ is the aerosol optical depth (AOD), and τ_{ma} is the optical depth due to molecular absorptions such as ozone, water vapor, or nitrogen dioxide [4, 7, 18]. The aerosol optical depth $\tau_{aerosol}$ can be obtained by subtracting τ_{rs} and τ_{ma} from τ . This is;

$$\tau_{aerosol} = \tau - (\tau_{rs} + \tau_{ma}). \quad (4.2.4)$$

4.3 Absorbtion of Radiation by Aerosol

Absorption is a process that removes energy from the electromagnetic radiative field, and converts it to another form. The absorption factor Q^{abs} and define as the ratio of absorption cross-section and geometric cross-section S_g of the article. $S_g = \pi r^2$ for:

$$Q^{abs} = \frac{S^{abs}}{S_g}, \quad (4.3.1)$$

The volume absorbtion coefficient, $\beta^{abs}(\lambda, r)$ represent the energy removed from a beam per unit distance by absorption. For a mono-disperse aerosols they are calculated from

$$\beta^{abs} = \sigma^{abs}(\lambda, r)N(r) = \pi r^2 Q^{abs}(\lambda, r)N(r), \quad (4.3.2)$$

where $N(r)$ is the number of particles per unit volume at some radius, r , $\beta^{abs}(\lambda, r)$ is volume absorption coefficient, $\sigma^{abs}(\lambda, r)$ is absorption cross section, and $Q^{abs}(\lambda, r)$ is absorption efficiency factor. For a collection of absorption aerosol particles the absorption volume coefficient given by

$$\beta^{abs} = \int_0^\infty \sigma^{abs}(\lambda, r)n(r)dr = \int_0^\infty \pi r^2 Q^{abs}(\lambda, r)n(r)dr, \quad (4.3.3)$$

Where $n(r)$ represents the number of particle with radii between r , and $r + dr$ per unit volume. It is also useful to define the quantities per particles i.e.

$$\bar{\sigma}^{abs}(\lambda) = \frac{\int_0^\infty \sigma^{abs} n(r)dr}{\int_0^\infty n(r)dr} = \frac{\beta^{abs}(\lambda)}{N_0}, \quad (4.3.4)$$

Where $\bar{\sigma}^{abs}(\lambda)$ is the mean absorption cross section. For a population of aerosol particles optical properties needed to be integrated over the size distribution $n(r)$ such as an intensive aerosol properties, we provide the expression for the mass absorption efficiencies ($unitm^2g^{-1}$),

$$\sigma_m^{abs} = \frac{\int_0^\infty \pi r^2 Q^{abs}(r)n(r)dr}{\int_0^\infty \frac{4}{3}\pi r^3 \rho n(r)dr}, \quad (4.3.5)$$

Absorbing aerosols refer to those aerosols that absorb light, it reduce the amount of sunlight reaching the surface (direct effect) and heat their surroundings. By doing so, they modify the vertical distribution of heat in the atmosphere and affect atmospheric thermodynamics and stability, possibly hastening cloud drop evaporation, and thereby affecting cloud amount, formation, dissipation and, ultimately, precipitation. Deposition of aerosol absorption on snow and ice reduces surface albedo leading to accelerated melt. The most abundant aerosol absorption type is black carbon (BC), brown carbon (BrC), and dust. From the absorption coefficients black carbon mass concentrations can be determined as

$$M_{ass}BC(\lambda) = \frac{\beta_{abs}(\lambda)}{\sigma_{abs}(\lambda)} = \frac{\beta_{abs}(\lambda).C}{\sigma_{atn}(\lambda)}, \quad (4.3.6)$$

Where $\sigma_{abs}(\lambda)$ and $\sigma_{atn}(\lambda) = \sigma_{abs}(\lambda).C$ are the mass specific absorption and attenuation cross-sections, respectively. Each of these sources may result from, and be strongly influenced by, anthropogenic activities, such as industrial combustion, as well as by natural activities. The properties and amounts of aerosol absorption depend upon various factors, such as: primarily fuel source and burn conditions, vegetation type, and in the case of dust, soil type and ground cover. The net radiative forcing by aerosol absorption is generally positive (warming), results from a variety of specific effects, and is impacted by different atmospheric processes [4, 16,19].

4.4 Scattering of Radiation by Aerosols

Scattering is the process by which a particle in the path of an electromagnetic wave continuously extracts energy from the incident wave and reradiates that energy in all the 4π directions. The scattering factors Q^{sca} are define as the ratio of the scattering of the geometric cross-section S_g of the particle ($S_g = \pi r^2$) for spherical particle with

radius r

$$Q^{sca} = \frac{S^{sca}}{S^g}, \quad (4.4.1)$$

The volume scattering coefficient, $\beta^{sca}(\lambda, r)$ represent the energy removed from a beam per unit distance by scattering. For a mono-disperse aerosols they are calculated from

$$\beta^{sca} = \sigma^{sca}(\lambda, r)N(r) = \pi r^2 Q^{sca}(\lambda, r)N(r), \quad (4.4.2)$$

where $N(r)$ is the number of particles per unit volume at some radius, r , $\beta^{sca}(\lambda, r)$ is volume scattering coefficient, $\sigma^{sca}(\lambda, r)$ is scattering cross section, and $Q^{sca}(\lambda, r)$ is scattering efficiency factor. For a collection of scattering aerosol particles the scattering volume coefficient given by

$$\beta^{sca} = \int_0^\infty \sigma^{sca}(\lambda, r)n(r)dr = \int_0^\infty \pi r^2 Q^{sca}(\lambda, r)n(r)dr, \quad (4.4.3)$$

where $n(r)$ represents the number of particle with radii between r , and $r + dr$ per unit volume. It is also useful to define the quantities per particles i.e.

$$\bar{\sigma}^{sca}(\lambda) = \frac{\int_0^\infty \sigma^{sca}n(r)dr}{\int_0^\infty n(r)dr} = \frac{\beta^{sca}(\lambda)}{No}, \quad (4.4.4)$$

where $\bar{\sigma}^{sca}(\lambda)$ is the mean scattering cross section. The phase function represents the redistribution of the scattered energy. For a collection of particles, the phase function is given by

$$P(\lambda, \theta) = \frac{1}{\beta^{sca}} \int_0^\infty \pi r^2 Q^{sca}(\lambda, r)P(\lambda, r, \theta)n(r)dr, \quad (4.4.5)$$

where $P(\lambda, \theta)$ is scattering energy and $P(\lambda, r, \theta)$ is angular distribution of phase function. The asymmetry parameter is the average cosine of the scattering angle, weighted by the intensity of the scattered light at a function of angle [16]. It has the value 1 for perfect forward scattering, 0 for isotropic scattering and -1 for perfect backscatter.

$$g = \frac{1}{\beta^{sca}} \int_0^\infty \pi r^2 Q^{sca}(\lambda, r)g(\lambda, r)n(r)dr, \quad (4.4.6)$$

For a population of aerosol particles optical properties needed to be integrated over the size distribution $n(r)$ such as an intensive aerosol properties, we provide the expression for the mass scattering efficiencies ($unitm^2g^{-1}$),

$$\sigma_m^{sca} = \frac{\int_0^\infty \pi r^2 Q^{sca}(r) n(r) dr}{\int_0^\infty \frac{4}{3} \pi r^3 \rho n(r) dr}, \quad (4.4.7)$$

The primary effect of aerosols in the climate system is the scattering of solar radiation, which means that only the direction of the radiation changes. Sulfate, nitrate and sea-salt aerosols can be considered purely scattering at visible wavelengths. Sulfate aerosols scatter primarily solar radiation and cause cooling of the Earth atmosphere system. The increase in the reflected solar radiation at the top of the atmosphere due to such non absorbing aerosols is nearly identical to the reduction in the solar radiation at the surface. The scattering increases the fraction of solar radiation reflected back to space, cooling the climate system. However, high surface albedo and the presence of clouds tend to reduce the net effect. Multiple scattering of radiation by atmospheric aerosols reduces the incoming solar radiation that reaches the Earth's surface and increases the radiation that escapes from the top of the atmosphere, thus cooling the Earth/atmosphere system. The resulting cooling of the Earth's surface is manifest large volcanic eruptions, such as Mt. Pinatubo in 1991, which inject large amounts of aerosol into the stratosphere. The Pinatubo eruption was followed by a noticeable decrease in mean surface temperatures for the following 2 years because of the long residence time of aerosols in the stratosphere [7, 20, 21].

It is estimated that the global average scattering optical depth of aerosols is about 0.1 and that 25% of this optical depth is contributed by anthropogenic aerosol. The radiative forcing from the anthropogenic aerosol (AA) layer is

$$\Delta F = \frac{F_s \Delta A}{4}. \quad (4.4.8)$$

where ΔA is the associated increase in the Earth's albedo (note that ΔF is negative; the effect is one of cooling). In figure below, we decompose the solar radiation flux

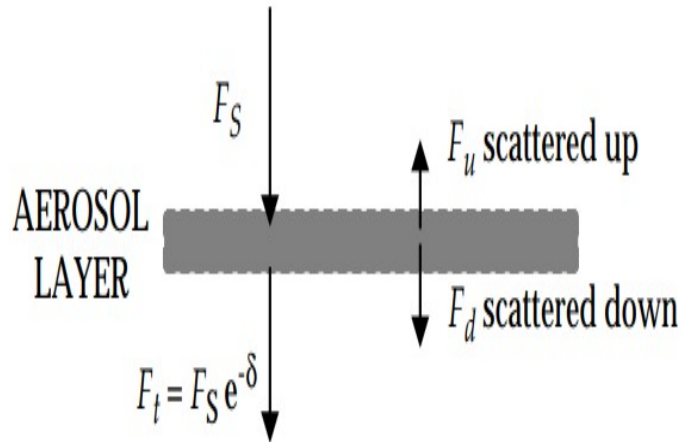


Figure 4.1: Scattering of radiation by an aerosol layer [7].

incident on the aerosol layer (F_s) into components transmitted through the layer ($F_t = F_s e^{-\delta}$), scattered forward (F_d), and scattered backward (F_u). We need to relate δ to ΔA . Because $\delta \ll 1$, we can make the approximation $e^{-\delta} \approx 1 - \delta$. The scattered radiation flux $F_d + F_u$ is given by;

$$F_d + F_u = F_s - F_t = F_s - F_s(1 - \delta) = F_s \delta. \quad (4.4.9)$$

Bright (scattering) aerosols increasing the local albedo when over dark surfaces but have less of an impact when over brighter surfaces [4, 11].

4.5 Reflection of Radiation by Aerosols

The reflective of aerosol particles in the upper layers of the atmosphere (the stratosphere) to reflect short-wave solar radiation back into space. Less solar radiation then reaches the Earth's surface, thus reducing global warming. The increase in the reflected solar radiation at the top of atmosphere due to such none absorbing aerosols is nearly identical to the reduction in the solar radiation at the surface. Most reflective aerosols centers on sulphate aerosols. Volcanic eruptions naturally emit large quantities of sulfate, a compound of sulfur, into the stratosphere. It has long been known that these ash particles reflect short-wave sunlight. For large volcanic eruptions, this can lead to global cooling. An aerosols effect on sunlight (radiation) depends primarily on the composition and color of the particles. Broadly speaking, bright colored or translucent particles tend to reflect radiation in all direction and back to wards space. Pure sulfate and nitrates reflect nearly all directions they encounter, cooling the atmosphere [4, 21].

4.6 Long-Wave Aerosol Radiative Properties

Aerosol radiative forcing exhibits a strong RH dependence. As RH increases SSA increases, resulting in an increase (-ve) in the top of atmosphere and surface forcing. TOA (top of atmosphere) forcing is positive for continental polluted aerosols when the atmosphere is dry (0% RH); because of the increasing SSA TOA forcing becomes negative. TOA and surface forcing in the shortwave region increase as RH (relative humidity) increases, resulting in nearly similar atmosphere warming. The solar (shortwave) radiation always acts to warm the atmosphere. In comparison, the absorption and emission processes in the atmosphere when integrated over the long-wave

region results in either a net loss of long-wave radiative energy (radiative cooling) or a net gain (radiative warming). The fluxes in the long wave region depend significantly on temperature and relative humidity. Interaction of aerosols with long-wave aerosol radiative (thermal radiation) significant only for particles with diameter similar in magnitude to wavelengths in the thermal region of the electromagnetic spectrum; generally only naturally occurring aerosols such as dust and sea-salt have sufficiently large diameter. Also, these large diameter aerosols can have a more significant effect on the thermal energy budget of Earth when at high altitudes vs low altitudes. This is because high altitude aerosols are cold. Thus, the thermal radiation absorbed by the high altitude aerosol is re-emitted at cold temperatures [4, 21].

4.7 Global Distribution of Aerosol Optical Properties

Global three-dimensional models have to account for the atmospheric processes that affect aerosols in the atmosphere, namely emissions, nucleation, condensation, evaporation, coagulation, cloud processing, atmospheric transport, dry and wet deposition, and chemistry/climate feedback mechanisms. Aerosol removal processes and climate impacts are greatly influenced by the size distribution. Moreover, anthropogenic emissions favor the transformation of insoluble to mixed particles by coating them with secondary products formed in the atmosphere. This increases the number of CNs that can grow to critical sizes to become CCNs, affecting the microphysical properties of aerosol, with impacts on aerosol lifetimes, global distributions, and radiative properties [11]. The global distribution of aerosol optical properties described by the principal observation of AERONET (Aerosol Rbotic NETwork), which is ground-based

network of measuring aerosol optical properties. AOD is large over desert such as the Sahara and the Gobi and their down stream areas. AOD large over biomass burning and fossil-fuel combustion areas such as East Asia, South Africa, and Amazon. Based on AERONET observed global aerosol properties the fine-dominated types are separated mainly by their single scattering albedo (SSA) ranging from non-absorbing aerosol such as sulphate and sea-salt ($SSA \approx 0.95$) in-developed or industrial region to moderately absorbing aerosol ($SSA \approx 0.90$) in forest fire burning and developing industrial regions to absorb aerosols ($SSA \approx 0.85$) in region of Savanna or grass land burning. Because this areas emit large amount of black carbon, brown carbon, and relatively small amount of scattering such as sulphate [24, 35].

4.8 Aerosol Radiative Forcing

Aerosol radiative forcing is defined as the effect of anthropogenic aerosols on the radiative fluxes at the top of the atmosphere (TOA) and at the surface and on the absorption of radiation within the atmosphere. The effect of the total (anthropogenic + natural) aerosols is called aerosol radiative effect or total aerosol forcing. Radiative forcing (RF) is often used to quantify and compare the potential climate impact of the various aerosol effects. Atmospheric aerosols influence the earth's radiative balance directly through scattering and absorbing solar radiation, and indirectly through affecting cloud properties. Aerosol radiative effects are considered as either positive or negative perturbations to the radiation balance, and they include direct, indirect (albedo effect and cloud lifetime effect), and semi-direct effects. Both the albedo effect and cloud lifetime effect cool the atmosphere by increasing cloud optical depth and cloud cover, respectively. The radiative impact of aerosols depends on the aerosol

properties but also the characteristics of the incident solar radiation. The difference between the radiative forcing at TOA and surface is defined as the atmospheric forcing and can be written as:

$$ARF_{ATM} = ARF_{TOA} - ARF_{SFC}. \quad (4.8.1)$$

where ARF_{ATM} is aerosol radiative force on the atmosphere, ARF_{TOA} is aerosol radiative forcing the top of the atmosphere, and ARF_{SFC} is atmospheric aerosol in the surface. ARF_{ATM} represents the amount of energy trapped by the aerosols present in the atmosphere. A positive ARF_{ATM} indicates a net gain of radiative flux in the atmosphere and hence warming, while a negative ARF_{ATM} indicates a net loss and thereby cooling. More see table (4.1) for the different anthropogenic aerosol species; such as sulphate, nitrate, and secondary organic aerosols contribute a negative RF_{ari} , while black carbon aerosols contribute a positive RF_{ari} . Biomass burning aerosols are partially absorbing so that their RF_{ari} can be either positive or negative.

The changes arising from the aerosol scattering and absorption of solar radiation are referred to as their direct radiative forcing [5, 21]. The amount of absorbed energy is termed the atmospheric heating rate (Kelvin per day [K/day]) and can be calculated as

$$\frac{\partial T}{\partial t} = \frac{g}{C_p} \left[\frac{ARF_{ATM}}{\Delta P} \right], \quad (4.8.2)$$

where $\frac{\partial T}{\partial t}$ is the heating rate (K/day), g is the acceleration due to gravity, C_p is the specific heat capacity of air at constant pressure, and P is the atmospheric pressure. The heating rates are calculated separately for shortwave and long-wave regions, by inserting the appropriate ATM forcing for the respective wavelength regions [4, 23].

Let us consider a layer of scattering (i.e. non-absorbing) aerosols, with (vertical) optical depth τ_a and up-scatter fraction $\beta_a(\theta_0)$. We neglect for now the presence of

Aerosol type	Net radiative forcing (W m^{-2})	Uncertainty range (W m^{-2})
Fossil-fuel sulphates	-0.40	-0.6 to -0.2
Fossil-fuel and biofuel black carbon	+0.40	+0.05 to +0.6
Fossil-fuel and biofuel organic aerosols	-0.12	-0.4 to +0.1
Nitrates	-0.11	-0.3 to -0.03
Biomass burning	+0.0	-0.2 to +0.2
Desert dust	-0.10	-0.3 to +0.1
All aerosols	-0.35	-0.85 to +0.15

Table 4.1: Global-mean radiative forcing due to aerosol radiation interactions [5].

a gaseous atmosphere. Several effects need to be taken into account to understand the dependence with the solar zenith angle, θ_0 , of the interactions between aerosols and solar radiation (figure 4.2). First of all, the amount of available solar radiation reaching a horizontal surface is equal to $S \cos \theta_0$ where $S \simeq 1362 \text{W m}^{-2}$ is the solar constant. It decreases therefore with $\cos \theta_0$ as the solar zenith angle increases from 0 (the Sun is at the zenith) to 90° (the Sun is on the horizon). Second, the optical depth along the direction of propagation of solar radiation is $\tau / \cos \theta_0$ and increases therefore with $1 / \cos \theta_0$ as the solar zenith angle increases. We can consider that, at least under the single scattering assumption, these two effects compensate each other. Of course, when 0° tends towards 90° , the single scattering assumption is no longer valid and the aerosol radiative effect has to tend towards 0 because there is no energy available to be lost. Third, we need to consider the dependence of the up-scatter

fraction with the solar zenith angle.

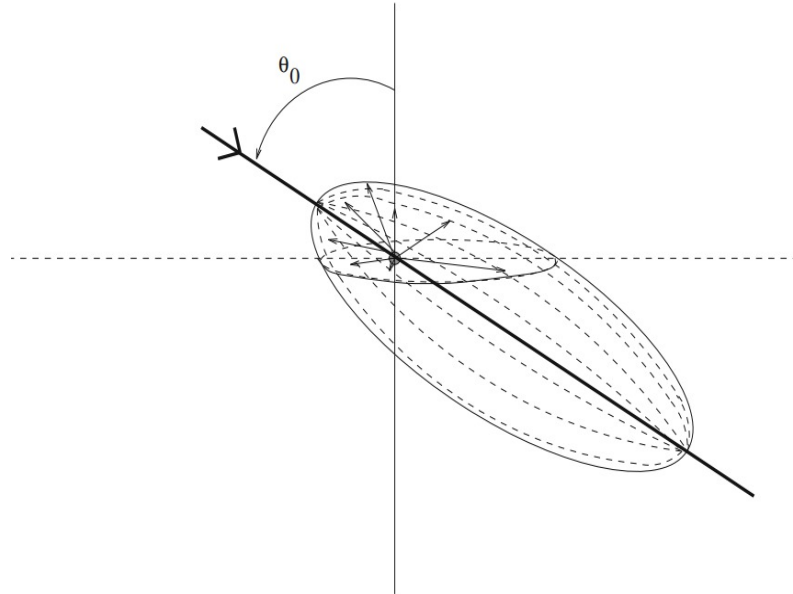


Figure 4.2: Illustrates the fact that as the solar zenith angle increases, a larger fraction of the forward scattering (scattering angles ranging between 0 and 90°) contributes to scattering in the upward direction. Combining these effects, we show that the radiative effect of a thin aerosol layer increases with the solar zenith angle up to the point and the phase function is represented by an ellipsoid which intercepts the horizontal plane [5].

Let us now consider a layer of scattering aerosols in a cloud-free atmosphere and denote β_a the up-scatter fraction, while deliberately ignoring for now its dependence with the solar zenith angle. This aerosol layer is placed between the surface with albedo R_s and an atmospheric layer with transmittance T (defined here as the fraction of solar radiation that goes through the atmosphere without being absorbed or back scattered). The solar zenith angle is noted θ_0 as before. The reflectance of the aerosol layer alone can be approximated by:

$$R_a \simeq \beta_a \tau_a / \cos \theta_0 \quad (4.8.3)$$

and its transmittance

$$T_a = 1 - R_a \simeq 1 - \beta_a \tau_a / \cos \theta_0 \quad (4.8.4)$$

The reflectance of the coupled surface aerosol system can be obtained by adding the successive reflections that come out of the system:

$$\begin{aligned} R_{as} &= R_a + T_a R_s T_a + T_a R_s R_a R_s T_a + T_a (R_s R_a)^2 R_s T_a + \dots \quad (4.8.5) \\ &= R_a + T_a^2 R_s \sum_{i=0}^{\infty} (R_s R_a)^i \\ &= R_a + \frac{T_a^2 R_s}{1 - R_s R_a} \\ &= R_a + \frac{(1 - R_a)^2 R_s}{1 - R_s R_a} \end{aligned}$$

where it has been assumed that the reflectance of the aerosol layer is the same for the upward and downward solar radiation. The change in reflectance due to the aerosol layer is equal to:

$$\Delta R_{as} = R_{as} - R_s = R_a + \frac{(1 - R_a)^2}{1 - R_s R_a} - R_s \approx R_a (1 - R_s)^2. \quad (4.8.6)$$

Under the assumption that $R_a \ll 1$ The change in reflected solar radiation caused by the aerosol layer at a given place and time can therefore be approximated as:

$$\Delta F = -S \cos \theta_0 T^2 \Delta R_{as} \approx -S (1 - R_s)^2 T^2 \tau_a \beta_a. \quad (4.8.7)$$

It can be seen that the radiative effect increases linearly with the aerosol optical depth and with the up-scatter fraction, and tends to zero when the surface albedo tends to 1 (i.e. a fully reflective surface). If we consider that in first approximation the radiative forcing due to aerosol radiation interactions is nonexistent in cloudy conditions and that the various parameters involved in its calculations are not correlated, Eq. (4.8.9) can be integrated in space and time to obtain an estimate of the global-mean radiative forcing by aerosols:

$$\overline{\Delta F} = -\frac{1}{2} S \overline{T}^2 (1 - \overline{A_c}) (1 - \overline{R_s})^2 \overline{\beta_a} \overline{\tau_a}, \quad (4.8.8)$$

where A_c is the cloud cover. Bars indicate an average over the range of atmospheric conditions that exist over the globe, except for $\overline{\beta_a}$ that represents the isotropic up-scatter fraction. The factor $\frac{1}{2}$ appears because every single point of the globe is in the night on average half of the year[5].

The simple Eqs. (4,8,9) and (4.4.10) are only valid for a non-absorbing aerosol. We know however that the atmospheric aerosol can be more or less absorbing, in particular because of the presence of black carbon in combustion aerosols. The previous formulation can be generalized so as to include the effect of aerosol absorption. In this case, the reflectance and transmittance can be written as:

$$R_a \approx \beta_a \varpi_a \tau_a / \cos \theta_0 \quad (4.8.9)$$

$$T_a = 1 - R_a - A_a \approx 1 - \beta_a \varpi_a \tau_a / \cos \theta_0 - (1 - \varpi_a) \tau_a / \cos \theta_0, \quad (4.8.10)$$

where A_a is the absorptivity in the aerosol layer and ϖ_a is the single scattering albedo of the aerosol. Equation (4.8.8) becomes:

$$\Delta R_{as} = R_a + \frac{T_a^2 R_s}{1 - R_s R_a} - R_s \approx R_a \left((1 - R_s)^2 - 2R_s \frac{A_a}{R_a} \right) \quad (4.8.11)$$

The instantaneous radiative forcing for a thin aerosol layer in clear-sky conditions can then be approximated in the following way:

$$\Delta F \approx -ST^2 \varpi_a \beta_a \tau_a \left((1 - R_s)^2 - \frac{2R_s(1 - \varpi_a)}{\beta_a \varpi_a} \right) \quad (4.8.12)$$

It can be seen that this expression reduces to Eq. (4.8.9) when $\varpi_a = 1$

$$\Delta F = -ST^2(1 - R_s)^2 \beta_a \tau_a. \quad (4.8.13)$$

It now appears that it is the triplet of parameters (β_a, ϖ_a, R_s) that determines the sign of the radiative effect due to aerosol radiation interactions. The radiative effect of

aerosols is more likely to be positive which induces a warming of the climate system if the surface albedo R_s is large, the up-scatter fraction β_a is small or the single scattering albedo a is small [5].

4.9 Role Aerosols in Radiative Transfer

The aerosol particles play a significant role in the transfer of solar and thermal radiation, affecting the radiation regime of the atmosphere Earth's surface system, and consequently, the weather and climate on the Earth's. Aerosols play an especially important role in the absorption and scattering of solar radiation. Atmospheric aerosols interact with shortwave radiation from the sun and long-wave thermal radiation from the Earth. These interactions profoundly influence the energy budget and climate of the planet [15, 21]. To estimate the influence aerosols have on solar energy, consider incident solar radiation (flux = I_0) that is incident on a surface of albedo S . The surface will absorb $(1 - S)I_0$ and reflect back SI_0 . This means that an energy flux $(1 - S)I_0$ is available for heating the Earth and indirectly the atmosphere, while the remaining SI_0 is lost to space. This is because air is almost transparent to solar radiation for large wavelengths and not true for longer infrared wavelengths where gaseous absorption is large. A layer containing aerosols which can scatter (both forward and backward) and absorb radiation is now introduced (figure 4.3). In the case of radiative flux, forward scattering by aerosols is generally disregarded as forward scattering only redistributes intensities; however, backward scattering can be considered equivalent to reflection. Let us define optical depth (τ), such that a fraction $e^{-\tau}$ of normally incident radiation emerges unaffected, while the fraction $(1 - e^{-\tau})$ is scattered or absorbed. This scattered or the absorbed fraction is referred to as single scattering

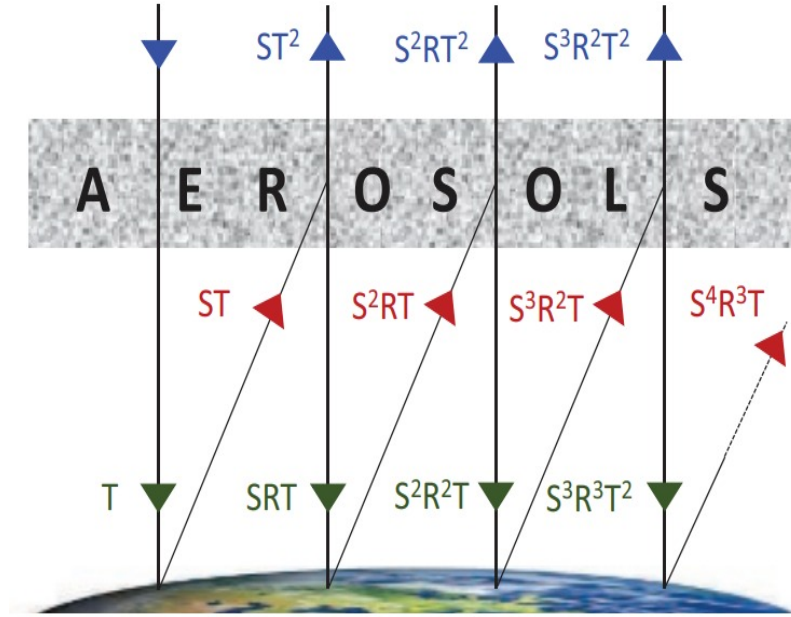


Figure 4.3: A schematic representation of interaction between aerosols over the Earth's surface having albedo S with incoming solar radiation, T denotes the transmission (diffuse + direct) due to aerosols in the layer and R is the diffuse reflectance of the aerosols in the layer [4].

albedo, which is denoted by S here for the purpose of derivation. The aerosol layer interacts with solar radiation as follows: the incident flux I_0 at its first encounter with the aerosol layer has part reflected back, this flux being $(1 - e^{-\tau})(1 - g')sI_0$ the flux $(1 - e^{-\tau})g'sI_0$, though scattered remains in the forward direction and the flux $(1 - e^{-\tau})(1 - s)I_0$ is absorbed and g' is only the incident radiation that is scattered forward is taken into account in the present case. The un-scattered or the flux that is scattered forward is written as

$$(1 - e^{-\tau})sg'I_0 + e^{-\tau}I_0 = ((1 - e^{-\tau})sg' + e^{-\tau})I_0 \quad (4.9.1)$$

The reflected and absorbed fluxes can be given respectively as

$$(1 - e^{-\tau})(1 - g')sI_0 \quad (4.9.2)$$

and

$$(1 - e^{-\tau})(1 - s)I_0, \quad (4.9.3)$$

yields a total of I_0 . The downward flux $((1 - e^{-\tau})sg' + e^{-\tau})I_0$ reach the surface, when $(1 - S)[e^{-\tau} + sg'(1 - e^{-\tau})]I_0$ is absorbed and $S[e^{-\tau} + sg'(1 - e^{-\tau})]I_0$ is reflected. This reflected flux again passes through the aerosol layer, experiencing absorption, reflection (downward), and transmission and forward scattering upward (figure 4.2). In a concise manner the transmission (T) and reflection (R) can be represent:

$$T = e^{-\tau} + (1 - e^{-\tau})g's \quad (4.9.4)$$

$$R = (1 - e^{-\tau})(1 - g')s. \quad (4.9.5)$$

Then the multiple reflections occurring between the surface and the aerosol layer can be followed by writing equations to obtain outward flux that never reaches the ground

$$RI_0 \quad (4.9.6)$$

flux reflected by ground

$$(TS)TI_0 = T^2SI_0, \quad (4.9.7)$$

flux twice reflected by ground

$$TS(RST)I_0 = T^2S^2RI_0 \quad (4.9.8)$$

and the flux that is thrice reflected by ground as

$$TSR(TS^2R^2)I_0 = T^2S^3R^2I_0. \quad (4.9.9)$$

and so on. It is seen that except for the first outward reflected flux all the other flux components contain T^2 ; no higher powers of T can occur as no radiation can pass right through the layer more than twice. The total reflected flux can be obtained by the sum of individual contributions:

$$I_R = RI_0 + T^2S(1 + SR + S^2R^2 + \dots)I_0 \quad (4.9.10)$$

$$I_R = \left(R + \frac{T^2S}{1 - SR} \right) I_0 \quad (4.9.11)$$

The flux which is not reflected back to space is either absorbed by the ground or by the atmosphere so that the absorbed flux is obtained as the difference between incident and reflected fluxes:

$$I_A = \left(1 - R - \frac{T^2S}{1 - SR} \right) I_0 \quad (4.9.12)$$

In comparison, the absorbed flux in the absence of an aerosol layer is only $(1 - S)I_0$ [4].

Chapter 5

ROLE OF AEROSOLS ON CLIMATE

5.1 Aerosol Cloud-Interaction

Cloud droplets are produced in the lower atmosphere by condensation of water on existing aerosol particles. Aerosols can act as cloud condensation nuclei (CCN) and ice nuclei resulting in the formation of cloud droplets and ice crystals. Sulfate, sea salt, and organic aerosols being water soluble serve as good CCN and ice nuclei. The aerosol-cloud interactions which can lead to changes in cloud characteristics and precipitation can occur in many different ways cloud albedo effect also known as Twomey effect (where in the number of cloud particles become more but smaller in size for the same cloud water or ice content), cloud lifetime effect also referred to as Albrecht effect (aerosols can increase the lifetime of clouds and reflectivity (albedo), decrease the precipitation efficiency, reduce the surface reaching solar radiation which would contribute to a cooling of the Earth's surface), semi-direct effect (aerosols can absorb the solar radiation and influence the static stability, re-emit the absorbed radiation as thermal radiation and by heating the air mass may cause evaporation of cloud droplets) [4, 25]. From the below figure small cloud droplets which are polluted

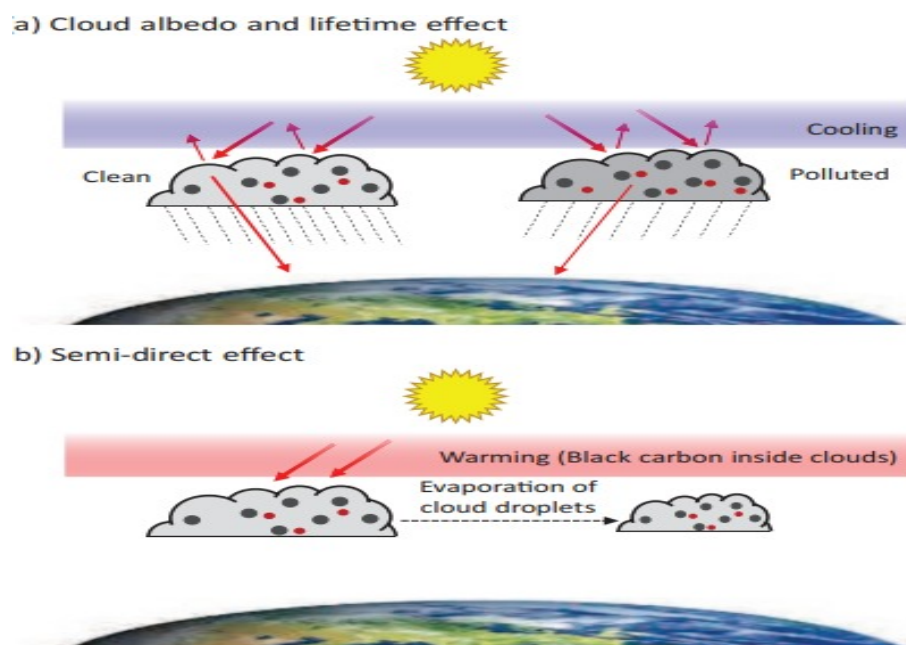


Figure 5.1: schematic illustration of aerosol-cloud interaction. (a) cloud albedo and lifetime effect, and (b) semi-direct effect [4].

by aerosols indirect and semi direct effects.

5.2 Role of Aerosols in Rain-Drop Formation (Nucleation Center)

The significant impact of aerosols on clouds microphysics and dynamics, maritime aerosols can lead to the formation of rain-drops that falls to the ground when the upward motion is increasing. Their simulations also show the increase of aerosols does increase the CCN which causes the formation of a large small number of droplets which can decrease the collision rate so it results in a time delay in the formation of rain-drops. Then this delay prevents the decreases of vertical motion and increase the duration of the diffusion droplet growth stage. Atmospheric aerosols are scavenged by

rainfall, and their size distributions are largely affected by variations in the dispersed raindrop size. The size distribution of aerosols and raindrops are often explained in terms of cloud and precipitation [14]. If the atmosphere had no aerosol particles, it would never have had rain. In the real atmosphere, water vapor must generally find a suitable surface, upon which it can condense to form cloud droplets. This suitable surface is the so called condensation nuclei, which is usually a hygroscopic aerosol particle. Once they are initially formed, cloud droplets will grow by further condensation of water vapor and coagulation with other water droplets and/or dry aerosol particles. This process of removal of aerosol particles is known as rainout. When the cloud produces rain, the rain drops will further collect aerosol particles below the cloud base during falling. This process of removal of aerosol particles is known as washout [27].

5.3 Water Cycle

Human activities influence the regional water cycle directly through modifying and exploiting stores and flows from rivers, lakes and ground water and by altering land cover characteristics. These actions alter surface energy and water balances through changes in permeability, surface albedo, evapotranspiration, surface roughness and leaf area [14]. Tiny air-borne particles including those released by human activities and the secondary organic aerosols have potential impacts on the hydrologic cycle. The land atmosphere interaction through exchanges of energy, water, momentum between the land surface and atmosphere. The land surface characteristics such as the albedo, roughness, and land cover/land use can have an impact on the water cycle. Aerosols play an important role in many areas including human health, atmospheric reactions,

the radiation and precipitation. Increases in aerosol concentration and changes in their composition may affect the Earth's climate and water supply. Aerosols have direct and indirect effects on the Earth's climate system by reducing the amount of solar radiation that reaches the surface and changing the properties of clouds [26].

5.4 The Effects Aerosol on Cloud Properties

Aerosols are largely responsible for the creation of clouds by acting as cloud condensation nuclei, or a sort of foundation for clouds to accumulate water on. Increasing the amount of aerosol in the atmosphere can influence the frequency of cloud occurrence, cloud thickness, cloud droplet, and rainfall amounts. This are indirect radiative effect of aerosols. There is also semi-indirect effect where the local heating of the atmosphere by absorbing aerosol like black carbon can influence whether cloud form at all, or lead to evaporations of existing clouds. Clouds in clean air are composed of a relatively small number of large droplet (left). Higher concentrations of aerosols can lead to large number of small droplet (right). These clouds are typically more reflective (brighter). When absorbing aerosols such as soot or dust are deposited on snow or ice, they decrease its reflectivity (albedo) and cause the surface to absorb more light which has a warming effect (more seen section 5.1) [14].

5.4.1 Effects of aerosol on cloud optical thickness and albedo

Photographs of the earth from satellites show that the brightest objects, and therefore the major reflectors of the sun's radiation, are clouds and snow-covered surfaces. Therefore, if atmospheric aerosol modify the radiative properties of clouds, they have the potential for large effects on the Earth's radiation balance. The potential for

increasing atmospheric aerosol concentrations to affect the radiative properties of clouds and therefore global climate. The optical thickness (τ) of a cloud of depth (h) containing number concentration $n(r)$ of droplets of radius r is given by:

$$\tau = \pi h \int_0^{\infty} Q_e r^2 n(r) dr, \quad (5.4.1)$$

Where Q_e is the extinction efficiency. At visible wavelengths, and when $\lambda \ll r$ Q_e has a value close to 2. Therefore, if we assume a narrow droplet size spectrum with a mean radius \bar{r} , Eq. (5.4.1) simplifies to

$$\tau = 2\pi h(\bar{r})^2 N \quad (5.4.2)$$

Where N is the total number concentrations of droplets

$$N = \int_0^{\infty} n(r) dr \quad (5.4.3)$$

The total liquid water content (w) of the cloud is

$$W = \frac{4}{3}\pi\rho_L \int_0^{\infty} r^3 n(r) dr \quad (5.4.4)$$

where ρ_L is the density of liquid water, or

$$W = \frac{4}{3}\pi\rho_L(\bar{r})^3 N \quad (5.4.5)$$

From equation (5.4.2) and (5.4.5)

$$\tau = 2.4 \left(\frac{W}{\rho_L} \right)^{2/3} h N^{1/3} \quad (5.4.6)$$

It follows from equation (5.4.6) that if the liquid water content and depth of a cloud are constant,

$$\frac{\Delta\tau}{\tau} = \frac{1}{3} \frac{\Delta N}{N} \quad (5.4.7)$$

The albedo (or reflectance) of a cloud is the fraction of the incident radiation that is reflected by the cloud integrated over the hemisphere of backscattering. The albedo A of a cloud is given to a good approximation by (Meador and Weaver, 1988)

$$A = \frac{(1 - g)\tau}{1 + (1 - g)\tau} \quad (5.4.8)$$

Where g is the scattering asymmetry factor, which is the average value of the cosine of the scattering angle. For the scattering of solar radiation by clouds, $g \simeq 0.85$. Hence, Eq. (5.4.8) becomes

$$A \simeq \frac{\tau}{\tau + 0.67} \quad (5.4.9)$$

Provided the liquid water content and cloud depth are held constant, Eq. (5.4.6) and (5.4.9) yield

$$\frac{\Delta A}{\Delta N} = \frac{A(1 - A)}{3N} \quad (5.4.10)$$

It follows from Eq. (5.4.10) that for a given N , $\Delta A/\Delta N$ has a maximum value when $A = 0.5$, although the curve is rather flat for a range of values on either side of $A = 0.5$. For a fixed value of A , $\Delta A/\Delta N$ is inversely proportional to N . Hence, as can be seen from Figure. (5.2), the albedo A of a cloud is most sensitive to changes in N when A has values from about 0.25 to 0.75 and N is small (i.e., in air with low CCN concentrations). Clouds that form in air containing high concentrations of CCN tend to have high droplet concentrations, and this enhances the shortwave (i.e., solar) albedo of the clouds. Because clouds are already optically thick at longer wavelengths, changes in the absorption of terrestrial radiation by clouds due to increases in droplet concentration are negligible. Hence, from this point of view, anthropogenic pollution should produce a cooling of the Earth's surface [14].

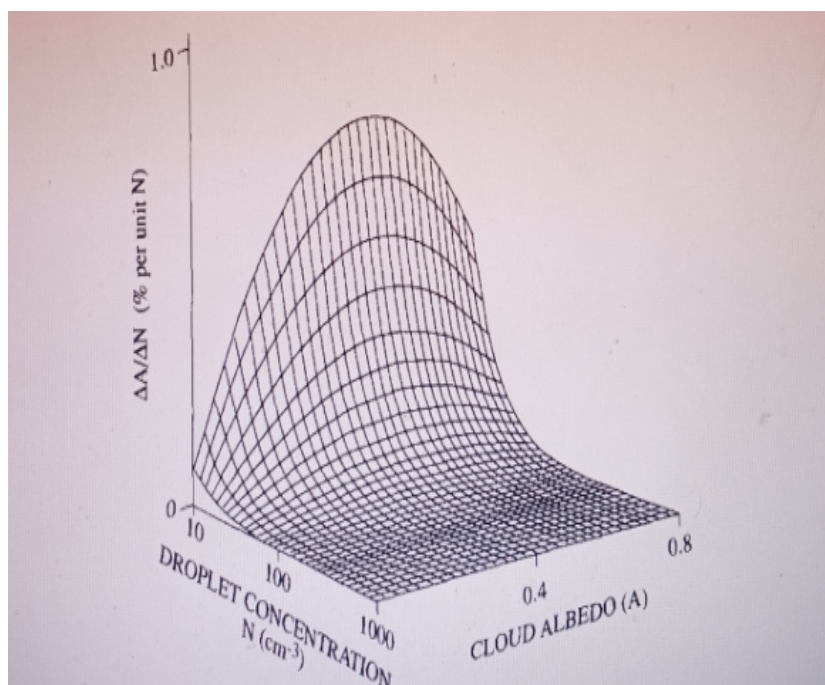


Figure 5.2: The change in the albedo of a cloud per unit change in the droplet concentration ($\Delta A/\Delta N$) as a function of the cloud albedo (A) and the droplet concentration (N), for a cloud with a constant liquid-water content [14].

5.5 Effects of Aerosols in Precipitation

Precipitation is the central mechanism for transport of water from the atmosphere to the Earth's surface. Aerosols are the particles in the atmosphere which act as embryos for cloud droplets and so have a direct impact on cloud microphysics and hence precipitation. Aerosols impact the reflective properties of clouds indirectly through their effect on cloud drop formation. The first indirect effect of aerosols on cloud's albedo is due to the reduction in size of cloud droplets with increasing aerosol concentrations and the second indirect effect concerns the suppression of precipitation due to the collision-coalescence process of rain formation becoming inefficient because

of the smaller droplet size. The result is that the cloud becomes colloidally stable, lives longer and reflects the incoming solar radiation back into space for longer times and also decreases the precipitation [14, 29]. Aerosols exert the control over the formation over cloud and precipitation through their direct effects on the nuclei which is the carrier of cloud droplet. Each cloud droplets requires an aerosol particle to condense on. So the biogenic and anthropogenic aerosols can become the cloud condensation on nuclei to affect the cloud droplet formation and precipitations. Therefore the number of cloud condensation nuclei is critical linked between aerosol and precipitation [26].

5.6 Global Aerosol Forcing

The effect of aerosols on climate is normally quantified in terms of aerosol radiative forcing (this section is more describe section 4.8). Aerosol radiative forcing is defined as the effect of anthropogenic aerosols on the radiative fluxes at the top of the atmosphere (TOA) and at the surface and on the absorption of radiation within the atmosphere. Global aerosol forcing shows the total (natural + anthropogenic) aerosol forcing over the globe. The forcing is mostly negative, and large negative values tend to be associated with high AOD, i.e., large aerosol burden in the atmosphere. Some areas have significantly positive forcing instead. Such as, the aerosols over the eastern tropical Atlantic (between Equatorial and $20^{\circ}S$) have huge positive forcing. This positive forcing is aided by low level cloud. The cancelation between the surface forcing and the atmosphere forcing occurs makes the zero forcing at the TOA. This cancelation occurs when the aerosol SSA is within a certain range associated with certain surface albedo and the presence of low cloud. The globally average aerosol

radiative forcing ΔF_R was calculated by the following equation.

$$\Delta F_R = \frac{S_0}{4} T_{atm} (1 - N) (1 - a)^2 2\beta\tau_{sc} \quad (5.6.1)$$

Where S_0 is solar constant, T_{atm} is the transmittance of the atmosphere above aerosol layer, N is fraction of sky cover clouds, a is the albedo of underling surface, β is the fraction of radiative scattering by aerosol into upper hemisphere and τ_{sc} is the aerosol layer scattering optical thickness. The above equation is the radiative forcing due to the change of reflectance of the earth aerosol system. $\frac{S_0}{4}$ is the globally average incident solar flux at the top of the atmosphere [14, 34].

5.7 Climatology of Stratospheric Aerosols

Stratospheric aerosols play an important role in the chemistry and radiative influence of the middle atmosphere. Stratospheric aerosol plays a significant role in stratospheric chemistry by providing the surface for a class of processes known as heterogeneous chemical reactions. These reactions play a critical role in ozone chemistry and, as a result, the total surface area density (SAD) provided by stratospheric aerosol is a crucial parameter in understanding changes to ozone. In addition, aerosol can transport of material from the troposphere into the stratosphere. Some of these materials, such as water vapor, are active greenhouse gases and thus changes to stratospheric aerosol may play an indirect role in modulating climate. Stratospheric aerosols have much longer residence times, on the order of about 1 year, and therefore have a more uniform distribution. As a result, volcanic explosions in which the debris reaches into the stratosphere can perturb global climate for several years [4]. Once the SO_2 is injected it undergoes a photochemical processing that eventually produces gaseous

sulfuric acid. Since sulfuric acid has a low saturation vapor pressure, it readily condenses onto existing aerosol or nucleates new aerosol using homogeneous (without a condensation nuclei) or using ambient solid particles as nuclei including associated volcanic ash, meteoric dust, and organic particles [21].

5.8 Climatology of Tropospheric Aerosols

Aerosols remain in the troposphere for up to 2 weeks, where they experience chemical transformation and long-range transport. Climatological of aerosols in the troposphere have been developed over time, tailored to the diverse interests in aerosol properties. Aerosols are removed from the atmosphere primarily by spatially inhomogeneous precipitation processes and, to a smaller extent, turbulence. Thus, aerosols have a relatively short residence time in the troposphere of typically up to 1 or 2 weeks. Consequently, aerosol concentrations vary greatly in time and space, both horizontally and vertically, in contrast to well-mixed greenhouse gases like carbon dioxide. Long-range transport depends on the amount of aerosol emitted, weather systems, and aerosol chemistry, especially aging processes. Transport occurs predominantly within the troposphere, either at low level in the boundary layer or aloft in the free troposphere. The potential climatic effects of tropospheric aerosols differ in some important ways from those of stratospheric aerosols. Since tropospheric aerosols are concentrated towards the surface and hence emit thermal radiation at a temperature only slightly lower than that of the surface, their interaction with infrared radiation is less important than for stratospheric aerosols. Tropospheric aerosols can also affect the Earth's radiation budget by modifying the properties of water clouds. Such modifications can be brought about by both the very small tropospheric aerosols that

can act as cloud condensation nuclei and by the larger sized aerosols [21] .

5.9 Observation and Measurement of Aerosol

Aerosols, as a group of atmospheric constituents, exhibit wide variability in their physical, chemical, and optical nature. Aerosol mass and number (bulk as well as size fractionated/segregated) describe the physical nature of aerosols. Depending on the property that is to be measured, observe and the measurement techniques employed to study aerosol characteristics can vary from in situ, to ground-based to air-, balloon-, and space-borne.

5.9.1 Aerosol chemical composition and size distribution

Characterization of aerosols can be done by obtaining representative samples of mass and number as a function of space, time, and size. The key atmospheric parameters such as temperature and relative humidity can influence sampling of aerosols. Filters are used to collect bulk aerosols based on the mechanisms of diffusion, impaction, and sedimentation. Aerosol mass spectrometer used to for determining the composition of individual aerosol particles. This instrument also used to measure and studies the chemical, physical, and optical characteristics of aerosols. Another instrument that used to measurements the size distribution of aerosol is Optical particle counter (OPC). An in situ OPC having a light source, a photomultiplier tube, and a pulse height analyzer was developed for quantitative measurements of aerosol number concentrations. The optical depth of an aerosol population with homogeneous properties on the vertical filling a layer with thickness Δz is:

$$\tau_a = \Delta z \int \pi r^2 Q^{ext}(r, \lambda, m) n(r) dr \quad (5.9.1)$$

Where $Q^{ext}(r, \lambda, m)$ is the extinction factor function of the aerosol radius, r , the wavelength, λ , and the complex refractive index, m , itself a function of λ and $n(r)$ is the size distribution that we seek to inverse. If τ_a is measured at several wavelengths in the visible and near-infrared domain, an approximative aerosol size distribution can be inverted [5, 14].

5.9.2 Aerosol scattering, absorption, and extinction coefficients

An integrating nephelometer measures the light scattered by aerosols in the $0 - 180^\circ$ scattering angle range, and integrates this light to yield scattering coefficient. An integrating nephelometer can measure aerosol scattering (β_{sca}) and the hemispheric backscattering coefficients at one or several wavelengths. One of the instruments that uses this technique and measures aerosol absorption is an aethalometer. Aethalometer measures aerosol absorption from the attenuation of a beam of light transmitted through the sample collected on a filter, which is proportional to the amount of BC (black carbon) mass loading in the filter deposit. The extinction measure one of the new instruments, namely, extinctionmeter combines photoacoustic cell and nephelometer to measure simultaneously aerosol absorption and scattering, respectively, thereby yielding aerosol extinction. The scattering coefficient of aerosols is derived from the total light extinction:

$$I = I_0 \exp(\sigma x) \quad (5.9.2)$$

where I is the intensity of the light after going through the atmospheric path, I_0 is the intensity of light source (reference), x is the thickness of the medium through which the light passes, and σ is the extinction coefficient (scattering + absorption coefficients). Aerosol Absorption coefficients of aerosols as a function of wavelength

are calculated following as

$$\beta_{abs}(\lambda) = \frac{-1}{C.R} \frac{\ln(i_1/i_2)}{Q.\Delta t}, \quad (5.9.3)$$

Where i_1 and i_2 are intensities of the sample and the reference beams, respectively, after a during the time interval Δt , and A is the area of the exposed spot on the filter where aerosols are collected. C is the correction factor applied to account for any change in the absorption occurring due to aerosols on the filter over that of the airborne particles. R is an empirical correction factor and describes the change in the aethalometer response with increased particle loading on the filter.

5.9.3 Aerosol optical depth(AOD)

Radio-meter instruments that quantity the amount of electromagnetic radiation (light) are among the most important tools optical depth, a measure of the amount of light that aerosols scatter and absorb in the atmosphere. Sun photometry is one of the most widely used techniques for measuring aerosol properties by which the aerosol optical depths can be directly obtained, unlike other remote sensing techniques, where the data have to be analyzed using complex inversion algorithms to evaluate the optical depths. Direct Sun photometry at multiple wavelengths is a useful method to characterize aerosols in the Earths atmosphere. The governing equation for Sun photometry measurement is

$$\tau = \frac{-1}{m} \left[\ln\left(\frac{I}{I_o}\right) - 2 \ln\left(\frac{r_o}{r}\right) \right], \quad (5.9.4)$$

where τ is the total integrated columnar optical depth of the atmosphere, I is the instantaneous solar radiation intensity measured by the photo-meter, and I_o is the solar radiation intensity obtained from Langley plot extrapolated for zero air-mass,

m is the atmospheric air-mass, r is the instantaneous value of the Sun-Earth distance, and r_o is the Sun-Earth distance when I_o values are obtained [5].

5.10 Role of Aerosol in Climate Change

By interacting with and by modifying solar and terrestrial radiative fluxes, aerosols act as part of the climate system. Due to variations with time of the aerosol optical depth, as occurs in the stratosphere after volcanic explosions and in the troposphere due to air pollution and dust storms, aerosols act as a driver for climate change. Aerosols may also indirectly affect climate by modifying cloudiness and the radiative properties of cloud non-absorbing aerosols will increase the albedo of the atmosphere thus reducing the amount of solar radiation which reaches the surface. As the aerosol absorbs in the shortwave range of the spectrum, energy is directly transferred to the atmosphere. The effect is a heating of the atmosphere and a cooling of the underlying surface. The net effect depends on the ratio of the absorption coefficients in the visible and infrared but also on the albedo of the surface.

The short-wave solar radiation received at the top of the atmosphere by the ground gas system must be equal to the long-wave or infrared radiation emitted by the ground gas system itself. For whatever reason, if this balance is disturbed or disrupted, it will cause climate change. In other words, this disturbance either reduce or increase the received solar radiation and the emitted long-wave radiation from the earth, which cloud lead to the energy budget of the earth-gas system deviates from its equilibrium state. It is to reduce the solar radiation reaching the ground by scattering and absorbing sunlight then cool the surface of the earth, which could offset part of the warming effect caused by greenhouse gases, and used as condensed nodules in clouds

to change cloud microphysical processes and precipitation properties, aerosols could change the water cycle of the atmosphere [28]. Many important atmospheric heterogeneous chemical reactions occur over the surface of aerosol particles or are impacted by them. Since the industrial revolution, a large amount of pollutant aerosols and gases have been directly emitted into the atmosphere [31].

5.11 Role of Aerosol in the Global Climate System

Aerosol particles in the atmosphere scatter sunlight back into space, they reduce the amount of energy that the planet absorbs, keeping it cooler. But this simplistic view ignored the fact that aerosol particles in the real world may also lead to heating of the lower atmosphere if they contain light absorbers such as elemental carbon and mineral dust. In fact, it was proposed that the warming effect from black carbon in aerosols may balance the cooling effect of the sulphate component, the largest single contributor to aerosol cooling [17]. Scattering and absorbing the solar radiation is known as the direct effect of aerosols on the global climate, which can lead to either cooling or warming of the atmosphere depending on the proportion of light scattered to that absorbed. Aerosols also have an indirect effect on climate by altering the properties of clouds, resulting in a change of their scattering properties and longevity. The latter effect has been supported by certain observations indicating that aerosols from forest fires and urban pollution can suppress rain and snow fall. In this way, changing aerosol in the atmosphere can change the frequency of cloud occurrence, cloud thickness, and rainfall amounts. These changes in cloud droplets number concentration and size and their longevity are, respectively, known as the first and second indirect effects of aerosol on climate and they are both believed to increase the amount of

sunlight that is reflected into space without reaching the Earth's surface, resulting in a cooling effect [9].

5.12 How do Aerosols Affect Climate

They scatter and absorb solar radiation and cause cooling or warming effect in the atmosphere through the direct process whereas indirect effect includes modification of cloud microphysical properties including droplets size, lifetime, cloud height and see the following figure). Eventually, they serve as cloud condensation nuclei for cloud

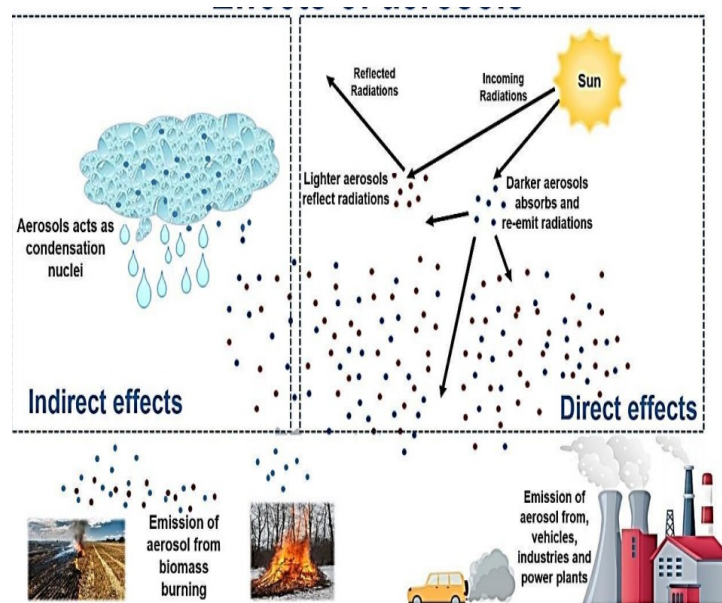


Figure 5.3: The effect of aerosols on the climate [4].

formation. Atmospheric aerosols are produced through natural and anthropogenic processes. Anthropogenic activities include fossil fuel and biomass burning, mining activities, modification of the natural land-use and cover, changes in industrial and

agricultural practices. In chapter three we have seen that, one emitted to the atmosphere SO_2 and NO_x become oxidized to sulfate and nitrate through gas and aqueous phase processes this types of aerosol contains acid deposition and also climate effects [4, 36].

5.12.1 Soot and direct radiative effect of aerosol

Soot incorporated within an aerosol particle will increase the particles absorption in the visible part of solar spectrum and thus it will decrease the particles single scattering albedo. The direct top of the atmosphere radiative forcing, ΔF , of an optically thin aerosol layer is given by:

$$\Delta F = -\frac{S_0}{4} T_{atm}^2 (1 - N) [(1 - \alpha)^2 2\beta\tau_{sc} - 4\alpha\tau_{abs}] \quad (5.12.1)$$

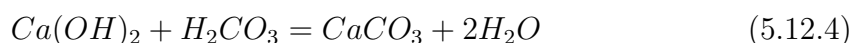
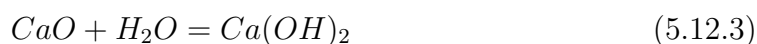
Where S_0 is the solar constant, N the fraction of sky covered by clouds, T_{atm} the transmittance of the atmosphere above the aerosol layer, α the surface albedo, β the fraction of the scattered radiation that is scattered into the upper hemisphere, and τ_{sc} and τ_{sc} the scattering and the absorption optical thickness of an aerosol layer. The negative value of radiative forcing implies cooling of the system, while a positive value implies heating. For non absorbing aerosol $\tau_{abs} = 0$, and Eqn. (5.12.1) implies always a cooling effect. When soot is present within an aerosol, aerosol absorption increases and the direct aerosol effect will be either cooling or heating, depending on the relative magnitudes of the terms inside the bracket on the right-hand side of Eqn. (5.12.1). For an optically thin aerosol layer, $\omega = \frac{\tau_{sc}}{\tau_{sc} + \tau_{abs}}$. The critical single scattering albedo, ω_{sc} , which determines whether an aerosol will heat or cool the system, is derived from Eqn. (5.12.1) in the form

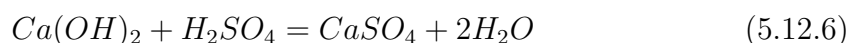
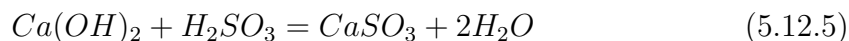
$$\omega_{cr} = \frac{2\alpha}{\beta(1 - \alpha)^2 + 2\alpha} \quad (5.12.2)$$

For given surface albedo, α , and backscattering fraction, β , an aerosol with single scattering albedo $\omega > \omega_{cr}$ will cool the system, while aerosols with $\omega < \omega_{cr}$ will cause heating. Soot within cloud droplets will again increase the droplet's absorption of electromagnetic radiation and decrease their single scattering albedo. This leads to an increased absorption of solar radiation within a cloud layer, to heating, and to a possible increased rate of evaporation of cloud droplets. However, soot in highly polluted regions, produced by industrial activities or biomass burning, can affect cloud absorption [21].

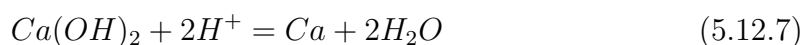
5.12.2 Acidification of precipitation

Raindrops contain a number of trace gases, some of which are acidic, some of which are alkaline, which come from aerosols scavenged by cloud- and rainwater. Aerosols therefore contribute to determine the pH of rainwater. It can be considered that in the absence of pollution the rain pH is generally about 5 to 6, although lower or larger pH can be found in some places. Rainwater becomes acidic in the presence of sulphuric acid and nitric acid, both of which are strong acids. Organic acids can also contribute to decrease the rainwater pH. On the contrary ammonia and calcium carbonate ($CaCO_3$) are weak bases and can increase the pH of precipitations. The chemical properties of the elements calcium (Ca) in the aerosol may have the most important effect on the acidity of precipitation water. Ca in aerosol is in the form of CaO and salt. If the aerosol Ca is in the form of CaO , the following reactions may occur when the aerosol is collected by raindrops containing acids:





For the PH value calculation the above reaction are equivalent to:



The pH of rainwater can indeed be in the alkaline range in semiarid regions such as North Africa and India because of the alkaline property of soil aerosols that are scavenged by the rain. Overall, there are fairly large spatial variations in rainwater pH across the globe with both natural and anthropogenic variations. Anthropogenic emissions of SO_2 have led to rain acidification with typical pH values of 4 over North America and Europe, and values as low as 2 over the most polluted regions. Acid rain and, more generally acid deposition under all its forms, contribute to the acidification of soils and freshwater bodies such as lakes and streams [5, 27].

5.13 Aerosol and their Relation to Global Climate and Climate Sensitivity

Aerosols can directly effect the Earth's climate through their interactions with incoming solar radiation and up-welling terrestrial thermal radiation. By absorbing solar and terrestrial radiation, aerosols tend to warm the regions of the atmosphere within which they ore located and, by changing solar and thermal net fluxes at the top of the atmosphere, they can alter the overall heat balance of the Earth. These radiative perturbations can be significant in that they may result in important changes in atmospheric and surface temperatures and in that they may alter atmospheric circulation patterns and hence indirectly modify precipitation patterns. The change

in global mean surface temperature (T) as a result of R_F can be expressed by the following simple heat balance equation

$$\frac{C_d(\Delta T)}{dt} = R_F - \frac{\Delta T}{\lambda}, \quad (5.13.1)$$

where C is the heat capacity of the land-ocean atmosphere system and λ is the climate sensitivity. At radiative equilibrium ($\frac{d(\Delta T)}{dt} = 0$) Eq. (5.13.1) reduced to $\Delta T = \lambda R_F$. However the earth is not in radiative equilibrium since less thermal radiation is currently emitted to space compared to what is absorption radiation. This radiation imbalance causes the earth to gradually warm with global warming as a result [30].

Chapter 6

SUMMARY AND CONCLUSION

In this study we have seen the role of aerosol in the Earth's atmosphere. Aerosol particles are a solid, like dust and liquids like water, suspended in the atmosphere. Atmospheric aerosols can be either emitted directly into the atmosphere like primary aerosols, or emitted gases under complex chemical reactions and condense as particles like secondary aerosols. Aerosol particles vary greatly in size, shape, source, amount and distribution in space and time, and they survive in the atmosphere. An important source of aerosol particles in the atmosphere are the processes of chemical interaction of substances found in the gas-phase particular interaction processes.

Aerosol particles interact with Sun's radiation by reflection, scattering or absorbing incoming radiation, and determine whether aerosol particles have a net warming or cooling effect on a climate. Radiative forcing (RF) is often used to quantify and compare the potential climate impact of the various aerosol effects. Aerosols are largely responsible for the creation of clouds by acting as cloud condensation nuclei or a sort of foundation for clouds to accumulate water on. Atmospheric aerosols influence the Earth's radiative balance directly through scattering and absorbing solar radiation, and indirectly through affecting cloud properties.

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