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Introduction

1.1 BRIEF HISTORY OF METEOROLOGICAL SCIENCES

The history of atmospheric sciences begins with weather forecasting. Forecasting originally grew out of three needs – for farmers to produce crops, sailors to survive at sea, and populations to avoid weather-related disasters such as floods. Every society has forecast wind, rain, and other weather events. Some forecasts are embodied in platitudes and lore. Virgil stated, "Rain and wind increase after a thunderclap." The Zuni Indians had a saying, "If the first thunder is from the east, winter is over." Human experiences with the weather have led to more recent forecast rhymes, such as, "Rainbow in morning, sailors take warning. Rainbow at night, a sailor's delight."

Primitive forecasts have also been made based on animal and insect behavior or the presence of a human ailment. Bird migration was thought to predict oncoming winds. This correlation has since proved unreliable. Rheumatism, arthritis, and gout have been associated with the onset of rain, but such ailments are usually unrelated to the weather. The presence of locusts has correctly been associated with rainfall in that locusts fly downwind until they reach an area of converging winds, where rain is likely to occur.

In the 1870s, forecasting based on observations and experience became a profession. Many felt that early professional forecasting was more of an art than a science, since it was not based on scientific theory. Although the amount of data available to forecasters was large and increasing, the data were not always used. Data were gathered by observers who used instruments that measured winds, pressure, temperature, humidity, and rainfall. Many of these instruments had been developed over the previous two centuries, although ideas and crude technologies existed prior to that time.

The Greeks, around 430 BC, may have been the first to measure winds. Yet, reliable instruments to measure wind force and direction were not developed until nearly two millennia later. In 1450, the Italian mathematician Leone Battista Alberti (1404–72) developed the first known anemometer, a **swinging-plate anemometer** that consisted of a disc placed perpendicular to the wind. It was used to measure wind speed based on the angle between the disc in its original position and its displaced position. In 1667 Robert Hooke developed a similar device, the **pressure-plate anemometer**, which consisted of a sheet of metal hanging vertically. Wind-mills were used as early as AD 644 in Persia, but the first spinning-cup anemometer,

which applies the principle of the windmill to measure wind speed, was not developed until the nineteenth century. In 1846, the Irish physicist John Thomas Romney Robinson invented a **spinning-cup anemometer** that consisted of four hemispherical cups mounted on a vertical axis. In 1892, William Henry Dines invented the **pressure-tube** (**Dines**) **anemometer**, which is a device that measures wind speed from the pressure difference arising from wind blowing in a tube versus that blowing across the tube. The pressure difference is proportional to the square of the wind speed.

In 1643, Evangelista Torricelli (1608–47) invented the mercury barometer, becoming the first to measure air pressure. He filled a glass tube 1.2 m long with mercury and inverted it onto a dish. He found that only a portion of the mercury flowed from the tube into the dish, and the resulting space above the mercury in the tube was devoid of air (a vacuum). Thus, Torricelli was also the first person to record a sustained vacuum. He suggested that the change in height of the mercury in the tube each day was caused by a change in atmospheric pressure. Air pressure balanced the pressure exerted by the column of mercury in the tube, preventing the mercury from flowing freely from the tube. The **aneroid barometer**, which represented an advance over the mercury barometer, was not developed adequately until 1843. The aneroid barometer contains no fluid. Instead, it measures pressure by gauging the expansion and contraction of a tightly sealed metal cell that contains no air.

A third important invention for meteorologists was the thermometer. Prior to 1600, Galileo Galilei (1564–1642) devised the **thermoscope**, which estimated temperature change by measuring the expansion of air. The instrument did not have a scale and was unreliable. Torricelli's mercury barometer, which contained fluid, led to the invention of the **liquid-in-glass thermometer** in Florence in the mid seventeenth century. In the early eighteenth century, useful thermometer scales were developed by Gabriel Daniel Fahrenheit of Germany (1686–1736) and Anders Celsius of Sweden (1701–1744).

A fourth important invention was the **hygrometer**, which measures humidity. In 1450, the German cardinal, philosopher, and administrator Nicolas of Cusa (Nicolas Cryfts) (1401–64) described the first hygrometer with the following:

If someone should hang a good deal of wool, tied together on one end of a large pair of scales, and should balance it with stones at the other end in a place where the air is temperate it would be found that the weight of the wool would increase when the air became more humid, and decrease when the air tended to dryness.

(Brownawell 2004). In 1481, Leonardo da Vinci (1452–1519) drew Cryfts' hygrometer in his *Codex Atlanticus*, using a sponge instead of wool. The purpose of the hygrometer, according to da Vinci, was

to know the qualities and thickness of the air, and when it is going to rain.

(White 2000). In 1614, Santorio Santorre developed a hygrometer that measured vapor by the contraction and elongation of cord or lyre strings. Later hygrometers

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were made of wood, seaweed, paper, hair, nylon, and acetate. The hair hygrometer is still used today, although another instrument, the **psychrometer**, is more accurate. A psychrometer consists of two liquid-in-glass thermometers mounted together, one with a dry bulb and the other with a bulb covered with a moistened cloth.

Following the inventions above, observations of pressure, temperature, humidity, wind force, wind direction, and rainfall became regular. By the nineteenth century, weather-station networks and meteorological tables were common. Observers gathered data and forecasters used the data to predict the weather, but neither observers nor forecasters applied significant theory in their work. Theoreticians studied physical laws of nature but did not take advantage of the abundance of data available.

One of the first weather theoreticians was Aristotle, who wrote *Meteorologica* about 340 BC. In that text, Aristotle attempted to explain the cause of winds, clouds, rain, mist, dew, frost, snow, hail, thunder, lightning, thunderstorms, halos, rainbows, and mock suns. On the subject of winds, he wrote (Lee 1951),

These, then are the most important different winds and their positions. There are two reasons for there being more winds from the northerly than from the southerly regions. First, our inhabited region lies toward the north; second, far more rain and snow is pushed up into this region because the other lies beneath the sun and its course. These melt and are absorbed by the Earth and when subsequently heated by the sun and the Earth's own heat cause a greater and more extensive exhalation.

On the subject of thunder, he wrote,

Let us now explain lightning and thunder, and then whirlwinds, firewinds and thunderbolts: for the cause of all of them must be assumed to be the same. As we have said, there are two kinds of exhalation, moist and dry; and their combination (air) contains both potentially. It condenses into cloud, as we have explained before, and the condensation of clouds is thicker toward their farther limit. Heat when radiated disperses into the upper region. But any of the dry exhalation that gets trapped when the air is in process of cooling is forcibly ejected as the clouds condense and in its course strikes the surrounding clouds, and the noise caused by the impact is what we call thunder.

Aristotle's monograph established a method of qualitatively explaining meteorological problems. Since Aristotle was incorrect about nearly all his meteorological conclusions, *Meteorologica* was never regarded as a significant work. Aristotle made observations, as evidenced by diagrams and descriptions in *Meteorologica*, but he did not conduct experiments. Lacking experiments, his conclusions, while rational, were not scientifically based.

Aristotle's method of rationalizing observations with little or no experiment governed meteorological theory through the seventeenth century. In 1637, René Descartes (1596–1650) wrote *Les Météores*, a series of essays attached to *Discours de la Méthode*. In some parts of this work, Descartes improved upon Aristotle's treatise by discussing experiments. In other parts, Descartes merely expanded or

reformulated many of Aristotle's explanations. On the subject of northerly winds, Descartes wrote (Olscamp 1965),

We also observe that the north winds blow primarily during the day, that they come from above to below, and that they are very violent, cold and dry. You can see the explanation of this by considering that the Earth EBFD [referring to a diagram] is covered with many clouds and mists near the poles E and F, where it is hardly heated by the sun at all; and that at B, where the sun is immediately overhead, it excites a quantity of vapors which are quite agitated by the action of its light and rise into the air very quickly, until they have risen so high that the resistance of their weight makes it easier for them to swerve, . . .

Like Aristotle, Descartes was incorrect about many explanations. Despite some of the weaknesses of his work, Descartes is credited with being one of the first in meteorological sciences to form hypotheses and then to conduct experiments.

Between the seventeenth and mid nineteenth centuries, knowledge of basic physics increased, but mathematics and physics were still not used rigorously to explain atmospheric behavior. In 1860, William Ferrel published a collection of papers that were the first to apply mathematical theory to fluid motions on a rotating Earth. This work was the impetus behind the modern-day field of **dynamical meteorology**, which uses physics and mathematics to explain atmospheric motion.

Between 1860 and the early 1900s weather forecasting and theory advanced along separate paths. In 1903, Vilhelm Bjerknes of Norway (1862–1951) promulgated the idea that weather forecasting should be based on the laws of physics. This idea was not new, but Bjerknes advanced it further than others (Nebeker 1995). Bjerknes thought that weather could be described by seven primary variables – pressure, temperature, air density, air water content, and the three components of wind velocity. He also realized that many of the equations describing the change in these variables were physical laws already discovered. Such laws included the continuity equation for air, Newton's second law of motion, the ideal-gas law, the hydrostatic equation, and the thermodynamic energy equation.

Bjerknes did not believe that meteorological equations could be solved analytically. He advocated the use of physical principles to operate on graphical observations to analyze the weather. For example, from a map of observed wind barbs, which give horizontal wind speeds and directions, he could draw a map of streamlines (lines of constant direction) and isolines (lines of constant wind speed), then use graphical differentiation and graphical algebra to determine vertical wind speeds, which would be drawn on another map. This technique was called **graphical calculus**.

Between 1913 and 1919, Lewis Fry Richardson (1881–1953) developed a different method of analyzing the analytical equations describing the weather (Richardson 1922). The method involved dividing a region of interest into rectilinear cells (grid cells), then writing a finite-difference form of the analytical meteorological equations for each grid cell and solving the equations by hand over all cells. Whereas Carle Runge and Wilhelm Kutta developed a method of finite-differencing ordinary 1.2 Brief history of air-pollution science

differential equations in the 1890s, Richardson extended finite-differencing (central differencing in his case) to partial differential equations and to multiple grid cells. Richardson was not satisfied with his solution technique, though, because data available to test his method were sparse, and predictions from his method were not accurate. Nevertheless, his was the first attempt to predict the weather numerically in detail.

Until the 1940s, much of Richardson's work was ignored because of the lack of a means to carry out the large number of calculations required to implement his method. In 1946, John von Neumann (1903–57), who was associated with work to build the world's first electronic digital computer ENIAC (Electronic Numerical Integrator and Computer), proposed a project to make weather forecasting its main application. The project was approved, and the first computer model of the atmosphere was planned. Among the workers on von Neumann's project was Jule Charney, who became director of the project in 1948. Charney made the first numerical forecast on ENIAC with a one-dimensional model (Charney 1949, 1951). Since then, numerical models of weather prediction have become more elaborate, and computers have become faster.

1.2 BRIEF HISTORY OF AIR-POLLUTION SCIENCE

Meteorological science is an old and established field; air-pollution science has a shorter history. Natural air pollution has occurred on Earth since the planet's formation. Fires, volcanic eruptions, meteorite impacts, and high winds all cause natural air pollution. Anthropogenic air-pollution problems have existed on urban scales for centuries and have resulted from burning of wood, vegetation, coal, oil, natural gas, waste, and chemicals.

In the nineteenth and early twentieth centuries, most air pollution was due to chimney and smokestack emission of coal and chemical-factory combustion products. In 1905, Harold Antoine Des Voeux described the combination of smoke and fog he observed in cities in Great Britain as **smog**. Smog from coal and chemical combustion resulted in several air pollution episodes that killed thousands of people between 1850 and 1960. The worst of these was in December 1952, when smog resulted in over 4000 deaths in London. Pollution resulting from coal and chemical-factory combustion in the presence of fog is commonly referred to as **London-type smog**.

In the early twentieth century, the widespread use of automobiles and the increase in industrial activity increased the prevalence of another type of air pollution, called **photochemical smog**. This pollution was most noticeable and formed almost daily in Los Angeles, California. It became so serious that an Air Pollution Control District was formed in Los Angeles in 1947 to combat it. The composition of photochemical smog was not elucidated until 1951, when Arie Haagen-Smit produced ozone in a laboratory from oxides of nitrogen and reactive organic gases, in the presence of sunlight and suggested that these gases were the main constituents of Los Angeles air pollution. Photochemical smog has since been observed in most cities of the world.

Before the twentieth century, air pollution was not treated as a science but as a regulatory problem (Boubel *et al.* 1994). In Great Britain, emission from furnaces and steam engines led to the Public Health Act of 1848. Emission of hydrogen chloride from soap making led to the Alkali Act of 1863. In both cases, pollution abatement was controlled by agencies. In the nineteenth century, pollution abatement in the United States was delegated to municipalities. In most cases, regulation did not reduce pollution much, but in some cases it led to pollution control technologies, such as the electrostatic precipitator for reducing particle emission from smokestacks. In one case, the development of a pollutant-control technology, the scrubber for removing hydrochloric acid gas from chemical factory emission, provided incentive for the swift passage of a regulation, the Alkali Act of 1863. Inventions unrelated to air-pollution regulation reduced some pollution problems. For example, in the early twentieth century, the advent of the electric motor centralized sources of combustion at electric utilities, reducing local air pollution caused by the steam engine.

1.3 THE MERGING OF AIR-POLLUTION AND METEOROLOGICAL SCIENCES

In the 1950s, laboratory work was undertaken to understand better the formation of photochemical and London-type smog. Since the computer was already available, box models simulating atmospheric chemical reactions were readily implemented. In the 1960s and 1970s, air-pollution models, termed **air-quality models**, were expanded to two and three dimensions. Such models included treatment of emission, transport, gas chemistry, and gas deposition to the ground. Most models used interpolated fields of meteorological data as inputs. Today, many air quality models use meteorological fields calculated in real time as inputs.

In the 1970s, atmospheric pollution problems, aside from urban air pollution, were increasingly recognized. Such problems included regional acid deposition, global ozone reduction, Antarctic ozone depletion, and global climate change. Initially, ozone reduction and climate change problems were treated separately by dynamical meteorologists and atmospheric chemists. More recently, computer models that incorporate atmospheric chemistry and dynamical meteorology have been used to study these problems.

1.4 WEATHER, CLIMATE, AND AIR POLLUTION

A model is a mathematical representation of a process. An atmospheric computer model is a computer-coded representation of dynamical, physical, chemical, and radiative processes in the atmosphere. In atmospheric models, time-dependent processes are mathematically described by ordinary differential equations. Space- and time-dependent processes are described by partial differential equations. Ordinary and partial differential equations are replaced with finite-difference or other approximations, then computerized and solved.

1.4 Weather, climate, and air pollution

Computer models also solve parameterized and empirical equations. A parameterized equation is an equation in which one parameter is expressed in terms of at least two other parameters. The equation of state, which relates pressure to temperature and air density, is a parameterized equation. An empirical equation is an equation in which one parameter is expressed as an empirical function (e.g., a polynomial fit) of at least one other parameter. Whereas parameterized equations are derived from insight, empirical equations do not always make physical sense. Instead, they reproduce observed results under a variety of conditions. In this text, computer modeling of the atmosphere is discussed. Such modeling requires solutions to ordinary differential equations, partial differential equations, parameterized equations, and empirical equations.

Since the advent of atmospheric computer modeling in 1948, models have been applied to study weather, climate, and air pollution on urban, regional, and global scales. **Weather** is the state of the atmosphere at a given time and place, and **climate** is the average of weather events over a long period. Some basic weather variables include wind speed, wind direction, pressure, temperature, relative humidity, and rainfall. Standard climate variables include mean annual temperatures and mean monthly rainfall at a given location or averaged over a region.

Air pollutants are gases, liquids, or solids suspended in the air in high enough concentration to affect human, animal, or vegetation health, or to erode structures. Standard air pollution problems include urban smog, acid deposition, Antarctic ozone depletion, global ozone reduction, and global climate change. Urban smog is characterized by the local concentration buildup of gases and particles emitted from automobiles, smokestacks, and other human-made sources. Acid deposition occurs following long-range transport of sulfur dioxide gas emitted from coalfired power plants, conversion of the sulfur dioxide to liquid-phase sulfuric acid, and deposition of sulfuric-acid-related species to the ground by rain or another means. Acid deposition also occurs when nitric acid gas, produced chemically from automobile pollutants, dissolves into fog drops, which deposit to the ground or lungs. This form of acid deposition is acid fog. Acids harm soils, lakes, and forests and damage structures.

Antarctic ozone depletion and global ozone reduction are caused, to a large extent, by chlorine and bromine compounds that are emitted anthropogenically into the atmosphere and break down only after they have diffused to the upper atmosphere. Ozone reduction increases the intensity of ultraviolet radiation from the Sun reaching the ground. Some ultraviolet wavelengths destroy microorganisms on the surface of the Earth and cause skin cancer in humans. Global climate change is characterized by changes in global temperature and rainfall patterns due to increases in atmospheric carbon dioxide, methane, nitrous oxide, water vapor, and other gases that absorb infrared radiation. The addition of particles to the atmosphere may in some cases warm, and in other cases cool, climate as well.

Historically, meteorological models have been used to simulate weather, climate, and climate change. Photochemical models have been used to study urban, regional, and global air-pollution emission, chemistry, aerosol processes, and transport of

Table 1.1	l Scales	of atmos	pheric	motion
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Scale name	Scale dimension	Examples
Molecular scale	≪2 mm	Molecular diffusion, molecular viscosity
Microscale	2 mm–2 km	Eddies, small plumes, car exhaust, cumulus clouds
Mesoscale	2–2000 km	Gravity waves, thunderstorms, tornados, cloud clusters, local winds, urban air pollution
Synoptic scale	500–10 000 km	High- and low-pressure systems, weather fronts, tropical storms, hurricanes, Antarctic ozone hole
Planetary scale	>10 000 km	Global wind systems, Rossby (planetary) waves, stratospheric ozone reduction, global warming

pollutants. Only recently have meteorological models merged with photochemical models to tackle these problems together.

One purpose of developing a model is to understand better the physical, chemical, dynamical, and radiative properties of air pollution and meteorology. A second purpose is to improve the model so that it may be used for forecasting. A third purpose is to develop a tool that can be used for policy making. With an accurate model, policy makers can try to mitigate pollution problems.

1.5 SCALES OF MOTION

Atmospheric problems can be simulated over a variety of spatial scales. **Molecular-scale** motions occur over distances much smaller than 2 mm. Molecular diffusion is an example of a molecular-scale motion. **Microscale** motions occur over distances of 2 mm to 2 km. Eddies, or swirling motions of air, are microscale events. **Mesoscale** motions, such as thunderstorms, occur over distances of 2–2000 km. The **synoptic scale** covers motions or events on a scale of 500–10 000 km. High- and low-pressure systems and the Antarctic ozone hole occur over the synoptic scale. **Planetary-scale** events are those larger than synoptic-scale events. Global wind systems are planetary-scale motions. Some phenomena occur on more than one scale. Acid deposition is a mesoscale and synoptic-scale phenomenon. Table 1.1 summarizes atmospheric scales and motions or phenomena occurring on each scale.

1.6 ATMOSPHERIC PROCESSES

Atmospheric models simulate many processes and feedbacks among them. Figure 1.1 shows a diagram of an air pollution–weather–climate model that simulates gas, aerosol, cloud, radiative, dynamical, transport, and surface processes.

A gas is an individual atom or molecule suspended in the air in its own phase state. Gas molecules have diameters on the order of $2-5 \times 10^{-10}$ m.

An **aerosol** is an ensemble of solid, liquid, or mixed-phase particles suspended in air. Each particle consists of an aggregate of atoms and/or molecules bonded together. An **aerosol particle** is a single particle within an aerosol. Aerosol particles

1.6 Atmospheric processes



Figure 1.1 Diagram of processes simulated in an air pollution-weatherclimate model.

have diameters that range in size from a few tens of gas molecules to 10 mm and can contain many components, including liquid water.

A hydrometeor is an ensemble of liquid, solid, or mixed-phase predominantly water-containing particles suspended in or falling through the air. A hydrometeor particle is a single particle within a hydrometeor. Examples of hydrometeor particles are cloud drops, ice crystals, raindrops, snowflakes, and hailstones. The main difference between an aerosol particle and a hydrometeor particle is that the latter contains much more water than the former. Hydrometeor particles generally range in diameter from 10 μ m to 10 mm. In this text, the term particle used alone may refer to an aerosol particle or a hydrometeor particle.

Figure 1.1 lists the major processes affecting gases in the atmosphere. Emission is the addition of a pollutant to the atmosphere. Photochemistry encompasses gas kinetic chemistry and photolysis. Gas kinetic chemistry is the process by which reactant gases collide with each other and transform to product gases. Photolysis is the process by which reactant gases are broken down by sunlight to form products.

Gases are also affected by **gas-to-particle conversion**. Conversion processes include heterogeneous chemistry, nucleation, condensation/evaporation, dissolution/evaporation, and deposition/sublimation. Gases react chemically on the surfaces of particles to form gas, liquid, or solid products during **heterogeneous chemistry**. **Nucleation** occurs when gas molecules aggregate and change phase to a liquid or solid to form a new small aerosol particle or a cluster on an existing particle surface. **Condensation** occurs when a gas diffuses to and sticks to the surface of a particle and changes state to a liquid. **Evaporation** occurs when a liquid molecule on a particle surface changes state to a gas and diffuses away from the surface. **Dissolution** occurs when a gas molecule diffuses to and dissolves into liquid on the surface of a particle. Evaporation, in this case, is the opposite of dissolution.

Gases are physically removed from the atmosphere by dry deposition and washout. Gas dry deposition (different from solid deposition) is a removal process that occurs when a gas (or particle) impinges upon and sticks to a surface, such as the ground or a house. Gas washout is the dissolution of a gas in precipitation (rainfall) that falls to the ground.

Figure 1.1 lists major aerosol processes. Some, such as emission, nucleation, condensation/evaporation, dissolution/evaporation, and heterogeneous chemistry, affect gases as well. Aerosol–aerosol coagulation occurs when two aerosol particles collide and coalesce (stick together) to form a third, larger particle. Aerosol–hydrometeor coagulation occurs when an aerosol particle collides and coalesces with a hydrometeor particle. Equilibrium chemistry is reversible chemistry between or among liquids, ions, and/or solids within aerosols and hydrometeors. Aqueous chemistry is irreversible chemistry important in water-containing aerosols and in hydrometeors.

Aerosol particles are physically removed by dry deposition, sedimentation, rainout, and washout. Sedimentation is the process by which particles fall from one altitude to another or to the surface due to their weight. This differs from aerosol dry deposition, which occurs when particles contact a surface and stick to the surface. Aerosol rainout is the growth of cloud drops on aerosol particles and the eventual removal of the aerosol particle to the surface by precipitation. This differs from aerosol washout, which is the coagulation of aerosol particles with precipitation that subsequently falls to the ground.

Clouds are affected by several of the same processes affected by aerosol processes. In addition, clouds are affected by ice deposition, which is the growth of water vapor onto aerosol particles to form ice crystals. Sublimation is the conversion of the ice crystals back to vapor. During freezing, liquid water within a hydrometeor changes state to ice. Melting is the reverse. Hydrometeor particles may coagulate with themselves or with aerosol particles. Lightning occurs when ice crystals collide with then bounce off of other ice crystals, creating a charge separation that eventually leads to a flash of light.

Gases, aerosol particles, and hydrometeor particles scatter (redirect) and absorb solar radiation (emitted by the Sun) and infrared radiation (emitted by the Earth, atmosphere, and Sun), affecting heating rates (the rates at which the atmosphere heats or cools), actinic fluxes (a parameter used to calculate photolysis rate