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Introduction

Air pollution effects on human health and environment are an important reasearch topic. Air pollution is a complex phenomenon whose investigation needs a multidisciplinary approach. Field observations are necessary to measure the variables. Modeling activity gives further insight into the problem allowing one to perform analysis even when field observations are not available. We report a brief description of air pollution problems and a first approach to modeling.

1.1 Air Pollution

Air pollution is the result of anthropogenic gas and particle emissions into our environment. The chemical composition of the current atmosphere considerably differs from the chemical composition of the natural atmosphere, as it was in pre-industrial times. Nowadays nowhere on earth there is *clean* air. Our atmosphere is polluted everywhere, its chemical composition differs from the pre-industrial time. The chemical composition of the atmosphere has shown gradual changes as long as the earth has existed. Life started on earth, in an atmosphere that hardly contained any oxygen, only about 0.015% against the current level of about 21%. The atmosphere at that moment contained nearly 99% CO_2, some N_2, and only traces of H_2O and O_2. Because of the low oxygen level, no stratospheric ozone layer could have been formed. So, the surface of the earth received all the UV-B radiation that is captured these days by the ozone layer. This also explains why life had to start in the oceans, at about 10 m below sea level - a depth where the UV-B radiation was substantially lower [10].

The ecosystem *life* created the chemical composition of the atmosphere in which this ecosystem can exist, i.e., a chemical composition in which life can sustain. The chemical composition with its high oxygen level is not in chemical equilibrium, but this non-equilibrium state can be maintained by life itself. Based on this fact, James Lovelock developed the Gaia-theory [31, 32]. In short, his theory states that the earth, including the atmosphere, is a *living*, homeostatic organism [10].

1.1.1 What is Air Pollution?

There are several approaches to define air pollution [10]. For example, the change in the global, chemical composition of the pre-industrial atmosphere, which is due to human influence, can be called *air pollution*; all man-made, anthropogenic emissions into the air can be considered air pollution. So air pollution did not start until mankind started *to play with fire*. The global increase in the concentrations of CO_2, CH_4 and N_2O, all greenhouse gases, could, and should be called *air pollution* in the broad sense, even though these species are not toxic for human beings and the ecosystem. Another approach is to distinguish between the emissions of safe, non-toxic compounds vs harmful ones and only consider the last as air pollution. This distinction, however, has two clear drawbacks. About 1940 and even much later, man-made emissions of CFCs were considered safe because they are inert in the troposphere. However, the decrease of the stratospheric ozone layer has taught us differently. In the same way, CO_2 emissions are safe in the sense that they are not toxic, but their increase leads most likely to a climate change, which in turn will be harmful to large parts of the ecosystem [10]. The second drawback is that natural emissions can also be harmful, such as emissions of dioxine caused by a forest fire as a result of lightning. Next to anthropogenic emissions, it is possible to distinguish between:

1. **Natural emissions:** emissions caused by the non-living world, such as volcanic emissions, sea-salt emissions, and natural fires.

2. **Biogenic emissions:** emissions resulting from the ecosystem, like Volatile Ortanic Compounds (VOC) emissions from forests, and CH_4-emissions from swamps.

In principle, natural and biogenic emissions lead to the chemical composition of the pre-industrial, natural atmosphere. The philosophical question [*whether man-made emissions should also be considered as biogenic, because man is part of the ecosystem*] can be retorted by the distinction that mankind, by making fires, creates anthropogenic emissions [10]. Although the distinction in these three categories: anthropogenic, natural, and biogenic could be useful, quite a number of intermediate emissions exist. Examples are the NO-emissions by soil bacteria, which is a function of the earlier deposited nitrogen on the soil due to anthropogenic emissions of N-compounds or earlier deposited manure containing nitrogen. There is the question of whether or not VOC-emissions are due to planting or not planting of trees, and whether or not dust-emissions are the consequence of paving or not paving sandy roads. These are such intermediate emissions, biogenic or natural, but with a clear human influence. Although anthropogenic emissions started when man learned to make fire, and the air quality, especially the concentrations of fine particles, surpassed air quality guidelines in and around the cave dwellings of the Neanderthal man, the impact of air pollution has been of a local character for a long time. In Europe, elevation of concentration levels occurred for the first time in the middle ages, resulting in the first laws on air pollution that were often focused on odor nuisance around local factories. Also, burning coal for heating and cooking led to air pollution. It should be emphasized here that air pollution in the strict sense (*toxic*) and global (climate) change are interrelated phenomena. Directly, because they often have the same emission sources, and more indirectly because species like tropospheric ozone and aerosols play a role both in local and regional air quality, as well as in climate change [10].

Primary and secondary pollutants

The main, **primary** i.e., directly emitted *gaseous* pollutants are the following:

- Carbon compounds, e.g. CO_2, CO, CH_4, the VOC's (volatile organic compounds);
- Nitrogen compounds, e.g. N_2O, NO, NH_3;
- Sulfur compounds, e.g. SO_2, H_2S;
- Halogen compounds, e.g. chlorides, fluorides, bromides

The main, primary *particle* pollutants are the following:

- Some particles smaller then 2.5 μm in diameter. Included are the Aitken nuclei, particles smaller than 0.1 μm in diameter, which grow rather fast by coagulation to larger particles. The chemical compound of these primary particles is, to a large extent, carbon but also heavy metals as iron, zinc, copper, etc.

- Some particles with a diameter from 2.5 to 10 μm. These larger particles are often composed of sea salt and dust.

Most air pollutants, except the halogen compounds, will be chemically transformed in the troposphere by the OH-radical. The OH-radical is formed in the troposphere by photo-dissociation of O_3, and subsequent reaction of oxygen with H_2O-vapour to OH [29]. The OH-radical do not react with N_2, O_2, H_2O, CO_2, but with other compounds as CO, CH_4, H_2, NO, NO_2, SO_2, NH_3. The OH-radical can be seen as the cleansing agent of the atmosphere, since it transforms *primary* air pollutants into *secondary* pollutants, which are subsequently removed from the atmosphere by dry and wet deposition. In this way the OH-radical determines the atmospheric residence time of most compounds in the atmosphere.

The main, secondary (i.e., formed in the atmosphere) gaseous pollutants are:

- NO_2 and HNO_3 formed from NO;

- O_3 formed through photochemical reactions,

The main, secondary particles are:

- Sulfate aerosols formed from SO_2, and Nitrate aerosols formed from NO_2 followed by the reaction with NH_3 to form ammonium (bi) sulfate and ammonium nitrate.

- Organic aerosols formed from gaseous organic compounds. These *secondary* particles consist mainly of small particles with a diameter less than 2.5 μm

1.1.2 Air pollution modeling

Air pollution modeling is an attempt to describe the causal relation between emissions, atmospheric concentrations, and deposition. Air pollution measurements give quantitative information about concentrations and deposition, but they apply only specific locations. Air pollution modeling can give a more complete and consistent description, including an analysis of the causes - emissions sources, meteorological processes, physical and chemical transformations - that led to these concentrations/deposition. Air pollution models play an important role in science, because of their capability to assess the importance of the relevant processes. Air pollution models quantify the relationship between emissions and concentrations/depositions, including the consequences of future scenario and the determination of the effectiveness of abatement strategies.

The concentrations of species in the atmosphere are determined by transport and diffusion. This means that in considering the history of air pollution modeling, some remarks should be made concerning transport and diffusion. Transport phenomena, characterized by the mean velocity of the fluid, have been measured and studied for centuries. The study of diffusion (turbulent motion) is more recent. Although turbulent motions have been observed from the moment people looked at rivers and streams, one could mention Reynolds paper in 1895 as the scientific starting point for the formulation of the famous criterion for laminar-to-turbulent flow transition in pipes [10]. One of the first articles in which turbulence in the atmosphere is [51]. In later years, Taylor developed the *Taylor-theory of turbulent diffusion*, (1921). In this theory, it is shown that the diffusion from a point source can be described by an eddy diffusivity constant for travel times which are much larger than the turbulent integral time scale, the so-called *diffusion limit*. For smaller time-scales the effective turbulent diffusivity is proportional to the travel time. Until about 1950, a number of studies were performed on the subject of diffusion in the atmosphere [40, 50, 6, 11, 52, 24, 4]. The paper by Richardson [40] considered long-range aspects; up to over 80 km. Bosanquet [6] is one of the first who published about the impact of chimney plumes.

Modeling of Point Sources

The analysis of the dispersion from low and high level point sources, especially experimental, was a major topic shortly after 1955. Papers on this subject [47, 18, 17, 21, 38], are devoted to the Prairie grass experiment, [49]. Perhaps the first paper on this subject was [41]. Publication by Pasquill, *Atmospheric Diffusion*, in 1962 [34], was a major milestone in summarizing the work performed until that moment. Air pollution modeling around the beginning of the sixties was focused on local dispersion phenomena, mainly from point sources with SO_2 as major component in the application studies. The Gaussian plume model [19] was formulated, in which the horizontal and vertical spread of the plume was experimentally determined. Tables appeared with the famous Pasquill-Gifford sigma-values in the horizontal and vertical direction, as a function of the atmospheric stability ranging from very stable, class F, to very unstable, class A. As functions of distance from the source the experimental sigma values are in reasonable agreement with Taylor theory. Differences are due to the fact that Taylor-theory holds for homogeneous turbulence, which is not the case of atmosphere. During sixties, the studies concerning dispersion from a point source continued and were broadening in scope. Major studies are available in [22, 53, 7, 33, 26]. Use and application of the Gaussian plume model spread over the whole globe, and became a standard technique in every industrial country to calculate the stack height required for permits. See for example [5] who published a standard work in Russian. The Gaussian plume model concept was soon applied also to line and area-sources. Gradually, the importance of the mixing height was realized [23, 13] together with its major influence on the magnitude of ground level concentrations. Air pollution modeling papers published in the sixties and seventies were mainly written by meteorologists, specialized in boundary layer meteorology and atmospheric turbulence. These studies focused on the effect of atmospheric stability on plume spread [10].

Air Pollution Modeling at Urban and Larger Scales

Shortly after 1970, scientists began to realize that air pollution was not only a local phenomenon. It became clear - firstly in Europe - that the SO_2 and NO_X emissions from tall stacks could lead to acidification at large distances from the sources. It also became clear - firstly in the US - that ozone was a problem in urbanized and industrialized areas. These situations could not be tackled by simple Gaussian-plume type modeling. Two different modeling approaches were developed, Lagrangian modeling and Eulerian modeling.

Lagrangian modeling, directed at the description of long-range transport of sulfur [42, 14, 15]. The work by Eliassen [14] was the start for the well-known EMEP-trajectory model which has been used over the years to calculate trans-boundary air pollution of acidifying species and later, photooxidants. Lagrangian modeling is often used to cover longer periods of time, up to years.

Eulerian modeling began with studies for ozone in urbanized areas[39], for SO_2 in urban areas and for regional scale [46]. Modeling studies by Reynolds on the Los Angeles basin, the well-known Urban Airshed Model-UAM originated.

Eulerian modeling, in these years, was used only for specific episodes of a few days. In general, Lagrangian modeling was mostly performed in Europe, over large distances and longer time-periods, and focused primarily on SO_2. Eulerian grid modeling was predominantly applied in the US, over urban areas and restricted to episodic conditions, and focused primarily on O_3. Also hybrid approaches were studied, as well as particle-in-cell methods. Early papers on both Eulerian and Lagrangian modeling are by [16, 30]. A comprehensive overview of long-range transport modeling in the seventies was presented. The next step is global modeling of earth troposphere. The first global models were 2-D models, in which the global troposphere was averaged in the longitudinal direction [25]. The first, 3-D global models were developed in [35]. Since approximately 1980, the basic modeling concepts and tools were available to the scientific community. Developments after 1980 concerned the fine-tuning of these basic concepts.

1.2 Venice

The atmosphere in Venice, Italy, like in other European cities, is influenced by complex PM_{10} and $PM_{2.5}$ multi-emission sources with a net tendency to exceed the limits fixed by the directive 99/30/EC. Venice is located in the northeast part of Italy between the Adriatic Sea and an intensively and industrialized mainland. Venice atmosphere is affected by a variety of emissions from both point and diffused sources, as the city is in the middle of a not, vert, similar 550 km^2-wide lagoon, close to the Adriatic Sea and not far from cultivated fields and a heavily populated and industrialized mainland which encompasses:

- chemical, metallurgical and oil-refinery industrial plants in Porto Marghera (almost 12 km^2);
- Glass Factories in Murano;

- a Coal Power Plant;
- a medium-size urban area with more than 270 000 inhabitants;
- several heavy traffic roads;
- a motorway;
- commercial and industrial harbours.

Past decades have been dedicated principally to study water problems that characterized Venice all along time. In the last years air pollution problems have become a critical subject, specifically due to its complex and sensitive ecosystem. Venice University [37] and Public Administration [20] started to investigate the presence of PM_{10} and its influence on air pollution. ARPAV (Veneto Region Environmental Protection Agency) developed an emission distribution *pie* for an air pollution responsibility in Venice area.

Air pollution dispersion is a very complex phenomena on many aspects and skills are required to develop a complete research program. Hourly measurements of meteo variables and of emissions are required to provide a good model of air pollution dispersion. In order to develop a specific analysis of the problem a multidisciplinary team composed by three University Departments (Scienze Ambientali, Informatica and Chimica Fisica) has grown up with the aim of reaching a new step in Venice air pollution knowledge.

Chapter 2

Atmosphere

Meteorology has practical applications in the area of control and management of air quality. Its significance was first realized when the increasingly heavy use of coal for home heating and industrial powers led to extreme sulfur pollution during certain weather conditions. One famous case occurred in London during foggy December in 1952, when approximately 4000 people died as the direct result of air pollution. Four years later, in January 1956, under similar conditions, 1000 deaths were blamed on an extended fog in London. Since that time, the problem has grown as a result of industrialization. High air pollution concentrations are no longer local and restricted to urban areas, but can be transported for long distances by large-scale weather patterns.

2.1 Air

Earth atmosphere is a very thin layer surrounding the globe. Its height is about 80 km, which is 1.25% of the earth radius. The composition of the atmosphere is almost uniform with height (see table 2.1). The gases listed in the left column of the table are permanent, i.e., their concentrations do not change in time and space. The concentrations in the right column can change with time e/o position.

Nitrogen and oxygen are the most abundant atmospheric gases. Their total amount is about 99.03%. The abundance of the remaining permanent gases is only about 0.9324%. The concentration of water vapor, which is also one of atmospheric gases, changes as a part of the natural hydrologic cycle.

Table 2.1: Atmospheric Gases

Permanent Gas	Symbol	%	Gas	Symbol	%
Nitrogen	N_2	78.08	Water vapor	H_2O	0.0 - 4.0
Oxygen	O_2	20.95	Carbon dioxide	CO_2	0.0351
Argon	A_r	0.93	Methane	CH_4	0.00017
Neon	N_e	0.0018	Carbon Monoxide	CO	0.00002
Helium	H_e	0.00052	Ozone	O_3	0.000004
Hydrogen	H_2	0.00005	Sulfur dioxide	SO_2	0.000001
Xenon	X_e	0.000009	Nitrogen dioxide	NO_2	0.000001

The concentration of the carbon dioxide (CO_2) and methane (CH_4) show cyclic oscillations associated with the annual vegetation cycle. The amount of sulfur dioxide (SO_2) may vary due both to volcanic eruptions into the upper atmosphere and to anthropogenic activities.

Most of the ozone is found in the higher atmosphere, about 30 km above the earth's surface. At this height, ozone is produced naturally and forms the so- called *ozone layer* which absorbs most of the ultraviolet radiation from the sun. Ultraviolet radiation is harmful to life. The ozone layer protects life on earth against radiation. The amount of variable atmospheric gases changes as a result of anthropogenic industrial activities. For example, ozone (O_3), nitrogen dioxide (NO_2), and carbon monoxide (CO) are emitted into the lower atmosphere by motor vehicles, due to high-temperature fuel combustion. Ozone is also the result of photochemical reactions. Carbon dioxide (CO_2) and sulfur dioxide (SO_2) are produced by burning wood and coal. Sulfur dioxide readily oxidizes to sulfur trioxide (SO_3). In moist air, sulfur trioxide reacts with water and produces sulfuric acid (H_2SO_4). Sulfuric acid can be transported within clouds for hundreds of kilometers. When it is removed from the clouds, it can produce acid rain.

2.2 Energy

Earth's atmosphere acts as a giant heat engine transforming available energy into the movement of huge masses of air. Practically all *fuel* for this engine is supplied by the sun. The contribution of other sources (e.g. the earth's interior) is smaller than 0.02%. Since earth atmosphere is semi-transparent to incoming solar radiation, roughly 20% of its energy is strictly obtained by absorption. About 30% of solar radiation is reflected or scattered into space. The remaining part goes through the atmosphere and is absorbed by earth surface.

Earth surface has a considerable influence on air temperature. Differences in temperature near the ground are caused by the variation of thermal properties of the underlying surface. Because water has an enormous specific heat, it takes far more heat to raise the temperature of water than it does to raise the temperature of rocks or soil. Besides, the heating energy is deposited only in a few decimeters of soil, while in the oceans it is mixed through the top few meters of water. Consequently, since water covers 61% of the Northern Hemisphere and 81% of the Southern Hemisphere, there are considerably smaller annual temperature variations in the water-dominated Southern Hemisphere, as compared to the Northern one.

Temperature in the atmosphere changes with height. The troposphere is the lowest layer of the atmosphere, extending up to about 10 km above the ground, the average temperature decreases with height at the rate of about 0.6 C per 100 m (Figure 2.1). Above, in the stratosphere, temperature generally increases with height, due to an absorption of solar radiation in the ozone layer. In the mesosphere the temperature decreases with height while in the thermosphere is the opposite. Departures from the plot in the figure can occur because of seasonal and latitudinal variations.

Within the first few hundred meters, the temperature significantly changes diurnally. At night, the earth surface cools radiatively and causes a decrease in air temperature near the surface. As a result, the so-called *temperature inversion layer* is formed near the earth surface. In the inversion layer, the temperature increases with height. On the other hand, during the day, the earth surface is heated by the sun. The warm surface causes an increase of the air temperature in a thin layer above the ground. As a result, the daytime air temperature near the ground readily decreases with height. Temporal variations of the air temperature in the atmosphere can also be related to various natural, and anthropogenic factors, such as volcanic eruptions and increased levels of air pollutants. During volcanic eruptions, tons of dust and ash are thrown into the atmosphere. Some of this material reaches the levels above the troposphere and is redistributed around the earth. The temporary presence of volcanic debris causes some of the sun's energy to be reflected back into outer space before it reaches the earth surface. Volcanic particles

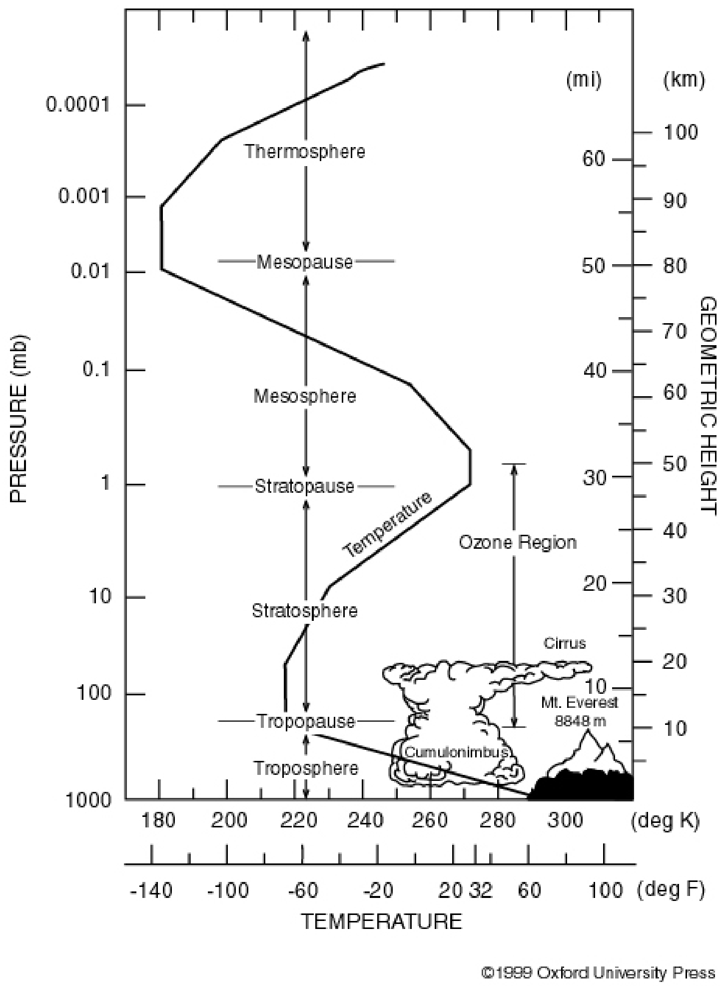

Figure 2.1: Temperature vertical distribution, (after: `http://www.engr.colostate.edu`)

also intensify cloud formation. More clouds reflect more solar energy back into outer space. Even though the volcanic dust and clouds also prevent some of the earth heat from escaping, the resulting effect is a cooling of the soil. There has been concern that further industrial emissions of pollutants into the atmosphere will cause global warming, commonly called the *greenhouse effect*. It is triggered by such pollutants as carbon dioxide, methane, nitrous oxide, and chloro-fluoro-carbons. Greenhouse gases prevent part of the earth radiation from escaping into space, and keep the earth warmer. An average surface temperature increase of 0.5 C has been observed since 1880. As a result of the warming effects, glacier and continental ice could melt, resulting in rising sea levels. More water vapor could be released into the air causing greater precipitation. A weakening of the Gulf Stream current might also occur. The warming trend could shift climate zones around the world and make floods, droughts, storms, cold and heat waves to be more extreme and more frequent. There is some skepticism in the scientific community about such an *apocalyptic* vision of the future. The observed 0.5 C temperature increase could merely be a natural climate fluctuation. Global warming caused by the greenhouse effect could be canceled due to radiative cooling by the increased presence of anthropogenic aerosols and clouds. Reduction of the ozone content, and consequently in the amount of the absorbed solar energy, is expected to cool the stratosphere. Nonetheless, the recent model simulations imply that only about 3.3% of the greenhouse energy is used to warm the atmosphere. Most of this energy (92%) is stored in the oceans. In addition, about 4.1% of the greenhouse energy causes the melting of the ice cover in Antarctica and Greenland, and about 0.5% melts the world glaciers [48].

2.3 Forces

Atmospheric motion results from the action of atmospheric forces. Such forces may be classified into two categories:

- body forces: they act at a distance on the bulk of the air parcel (hence the word *body*)

- surface forces or *stresses* (forces per unit area): they act through direct contact and they are exerted directly on the surface of the air parcel. The surface forces can be divided into:

1. normal stresses
2. tangential (*shear*) stresses.

Surface forces reflect an interaction among air molecules or air parcels. If two layers of air flow at slightly different velocities, the random sidewise intrusions of some slower molecules into the faster stream slows down the faster stream. The intrusion of faster molecules into the slower stream speeds up the slower stream. The wandering of individual molecules produces internal friction in the fluid which is called viscosity. The atmospheric pressure can be regarded as the weight of the atmosphere per unit area.

Pressure differences in space generate the pressure gradient force which acts from higher to lower pressure regions of the atmosphere. As the atmospheric pressure decreases with height, one might expect that the vertical component of the pressure force is able to move the atmospheric air out into space. But this does not occur, because the vertical pressure gradient force is approximately balanced by the gravity force. This balance is expressed by the *hydrostatic equation*, which can be written in the following form:

$$-\frac{1}{\rho}\frac{dp}{dz} = g \tag{2.1}$$

where z is height, ρ is the air density, p is the air pressure, g is the gravity acceleration. The hydrostatic balance may be disturbed by the buoyancy force, due to density differences between parcels of moving air and their vicinity, as well as by varying motions in time and space. Buoyancy forces act on individual parcels of air only when there are differences in density between the parcels and the ambient atmospheric air. For instance, the buoyancy force appears when a number of water vapor molecules is added to a fixed volume of air. In this case the same number of air molecules must leave this volume to keep constant the total number of molecules (*Avogadro's law*), as well as temperature and pressure.

The rotation of the earth introduces the *Coriolis force*. The Coriolis force is perpendicular to an object's relative velocity, and is oriented to the right of the velocity vector in the northern hemisphere, and to the left in the southern one. Its magnitude is proportional to the product of an object's mass, its velocity, earth angular velocity (7.29x10^{-5} radians/s), and sin φ, where φ is the latitude at which wind occurs. The Coriolis effect combines two factors, one that exerts its strongest force on objects traveling on a north-south axis, and another which affects objects moving on an east-west axis. The first factor results from the rotational velocity of the earth surface, which varies with latitude. Air moving north from the Equator begins with a greater rotational speed and outruns slower moving portions of the globe. As a result, it relatively curves eastward and ahead of the earth rotation. Similarly, air traveling southward, toward the Equator, begins with a low initial velocity and curves west, as the faster-moving earth exceeds it. The east-west component of the Coriolis force is a consequence of the tendency of any orbiting object to fly off in a straight line. This tendency, together with the rotation of the earth, produces a force which lies on the plane perpendicular to the earth axis, and thus has a sideways component in relation to the earth surface. Consequently, an object moving east will curve toward the Equator, while an object moving westward will curve toward the pole.

2.4 Static Stability

The buoyancy force can also appear when air parcels travel vertically in the atmosphere. A dry parcel of air moving upward expands adiabatically due to a decrease in the atmospheric pressure with height. The process of expansion decreases the internal energy of parcel molecules, and causes the parcel to cool. When a dry parcel of air moves downwards, it contracts adiabatically due to an increase in the atmospheric pressure with height. The contraction increases the internal energy of parcel molecules, causes the parcel to warm. Changes of the temperature in a dry adiabatic process are described the Poisson equation:

$$\frac{T(z)}{T_0} = \left\{ \frac{p(z)}{p_0} \right\}^k \tag{2.2}$$

where T is the absolute temperature in Kelvins, p is pressure, $k = R/c = 0.286$, R is the gas constant of dry air (R = 287 $\mathrm{m^2s^{-2}K^{-1}}$), c is the specific heat of dry air (c = 1007 $\mathrm{m^2s^{-2}K^{-1}}$), z is the actual height and o indicates the initial level of the moving parcel. Differentiating the above equation, with respect to height, with the help of the hydrostatic equation (1), and the equation of state we obtain:

$$p = \rho R T \tag{2.3}$$

which relates pressure, temperature and density. One finds that the temperature of a vertically moving parcel changes in the atmosphere at the constant lapse rate γ = - dT/dz = g/cp 1 C per 100 m. This value is called the *dry adiabatic lapse rate* (the lapse rate is defined as a negative vertical gradient). When the atmosphere is well mixed by vertical motions of air parcels, its temperature is not constant but decreases with height at the dry adiabatic lapse rate. In a well mixed atmosphere the so-called *potential temperature* is constant. The potential temperature is defined as the temperature of a parcel which is brought adiabatically to the reference level of 1000 hPa.:

$$\Theta = T(z)\left\{\frac{1000}{p(z)}\right\}^k \tag{2.4}$$

The pressure p in (4) is expressed in hPa. By differentiating (4) with respect to height, with the help of (1), one can obtain:

$$d\Theta/dz = dT/dz + \gamma_a \tag{2.5}$$

The potential temperature is indeed conserved (constant) during vertical adiabatic motions. More specific considerations on temperature variation in the atmsphere can be found in [48].

2.5 Scales of Atmospheric Motions

In 1926, L.F. Richardson (1881-1953) noted that atmospheric motion occurs over a broad range of horizontal length scales - from thousands of kilometers to millimeters. The largest scale motion is caused by thermal and pressure contrasts over the globe, modified by the rotation of the earth. Land and oceans introduce additional modifications to this primary flow and help to initiate secondary circulations. Local topography introduces tertiary circulations. A cascade process, in which eddies of the largest (global) size trigger smaller and smaller (local) ones, continues down to molecular motions, which finally cease due to viscosity.

The range of scales of atmospheric motions can be adequately defined by considering fundamental frequencies of the atmospheric motions [2]:

- the Brunt-Våisålå frequency: $N = (g/T_0 d\Theta/dz)^{1/2} \sim 10^{-2}s^{-1}$

- the inertial frequency: $f = 2\Omega sin\phi \sim 10^{-4}s^{-1}$
- the planetary frequency: $P = Ub^{1/2} \sim 10^{-6}s^{-1}$

where f is the Coriolis parameter, Ω the angular velocity of the earth, ϕ is the latitude, U is the horizontal velocity of air, b is the rate at which the Coriolis parameter changes with latitude.

The Brunt-Våisålå frequency N defines gravity oscillations in the stratified atmosphere. The inertial frequency f results from the rotation of the earth. The planetary frequency P is related to oscillations in the westerlies flow in the middle and upper troposphere. Based on these fundamental frequencies N, f, and P listed above, the following scales of atmospheric motions can be defined:

- planetary scale: $F < P$ *or* $F < 10^{-6}s$
- large scale: $P < F < f$ *or* $10^{-6}s < F < 10^{-4}s$
- meso-scale: $f < F < N$ *or* $10^{-4}s < F < 10^{-2}s$
- small scale: $F > N$ *or* $F > 10^{-2}s$

where F is the frequency of the atmospheric circulation under consideration. The corresponding length scale L can be obtained by assuming $L = 2\pi U/F$, where U is the velocity of the air ($U \sim 10$ km/h). Consequently:

- planetary scale: $L > 2\pi U/P$, *or* $L > 1500$ *km*, but limited by the circumference of the Earth
- large scale: $2\pi U/f < L < 2\pi U/P$, *or* 200 *km* $< L < 1500$ *km*
- meso-scale: $2\pi U/N < L < 2\pi U/f$, *or* 2 *km* $< L < 200$ *km*
- small scale: $L < 2\pi U/N$, *or* $L < 2$ *km*

Atmospheric transport and diffusion closely follow this classification. Pollutants from local/urban sources (isolated factories, power plants, waste disposals, e.t.c) are quickly dispersed by small-scale motions near the earth. Pollutants emitted from major industrial areas or forest fires retain high concentrations for greater distances and are dispersed on small and meso-scales. In cases of powerful sources (e.g., nuclear plant explosions or the burning of oil wells), high concentrations of air pollution can remain in the atmosphere for a very long time. The planetary scale circulations transport material injected into higher levels of the atmosphere. Such injections can occur during volcanic eruptions, when tons of dust and ash are thrown into the atmosphere. Some of this material reaches to the levels of the stratosphere and can be slowly redistributed around the earth by planetary-scale circulations. Pollutants which do not reach above the troposphere are dispersed much faster by large, meso- and small-scales motions. A more detailed discussion of atmospheric motions of various scales is introduced in the following sections.

2.5.1 Planetary-scale Circulations

The planetary-scale circulation of the atmosphere is schematically illustrated in Figure 2.2. Understanding of planetary-scale circulations is necessary to achieve an appropriate description of tropospheric and stratospheric transport, and transformations of carbon dioxide, methane, ozone, water vapor, nitrous oxide, chloro-fluoro-carbons, and aerosols. For this reason the analysis of planetary- scale circulations is often performed in conjunction with atmospheric chemistry. This approach was used, for example, to explain the ozone-hole effect. As depicted in Figure 2.2, at the equator (latitude 0) air is thermally forced upward and begins its high-level flow to the north and to the south. At the same time, the air over the north pole begins its low-level journey southward. This simple convective transfer between the equator and the poles is disrupted by the earths rotation, and three separate circulation cells are established. The subtropical cell is called the Hadley cell, the middle one is called the Ferrel cell, and the third one is called the Polar cell.

In the northern hemisphere, the Coriolis force turns the air to the right. As a result, below latitude 30, lower winds become easterly and upper winds westerly. At the same time, the air over the north pole is deflected to the right, and becomes easterly above latitude 60. A similar picture occurs in the southern hemisphere. A semi-permanent high pressure belt (marked by a letter H) is formed near latitude 30, in both the northern and southern hemispheres. Air ascends around latitude 60 in subpolar low pressure zones (marked by a letter L), which are created in both hemispheres. Subpolar low zones are accompanied by a distinct boundary, which separates cold air

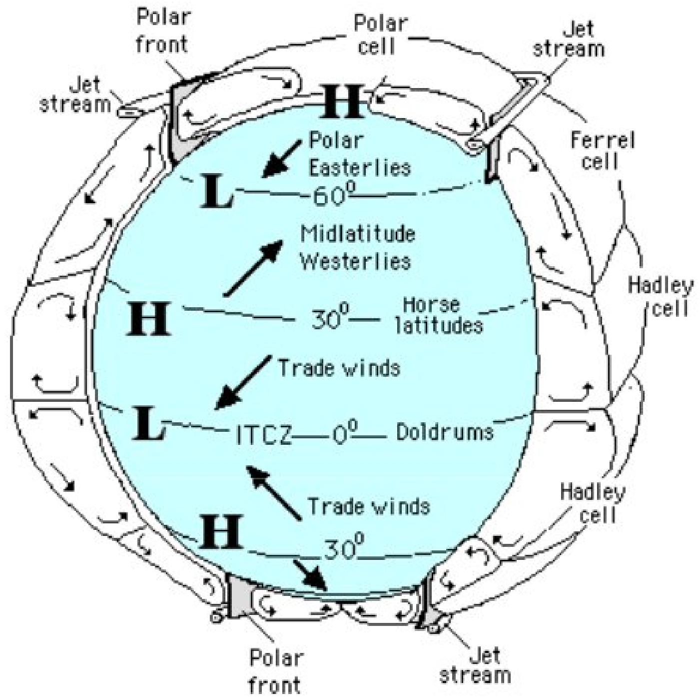

Figure 2.2: The general circulation of the atmosphere after [48]. Idealized zonal belts of high and low pressure systems are marked by letters H and L.

moving south, and mild air traveling poleward. This boundary between the Polar and Ferrel cells is called the *polar front*. In the tropopause, above the Polar front, there is a meandering globe-circling current of westerly winds. The current, called a *jet-stream*, moves air with a typical speed of about 150 km/h. It is hundreds of kilometers wide and only a few kilometers deep.

A zone near the equator is called the *intertropical convergence zone* (ITCZ). This region of very monotonous weather and weak winds is referred to as the *doldrums*. Steady east winds in the zone 0 to 30 are called the *trade winds*. Trade winds provided sailing ships with a route from Europe to America. From the 16th to the 19th centuries, the northeast trades were used to transport goods to Africa, where they were exchanged for slaves. From Africa, sailing boats filled with human cargo voyaged to America, exploiting southeast trades. From America, with the help of prevailing westerlies, they returned to Europe loaded with sugar, rum and cotton. Latitudes of about 30 are called the *horse latitudes*. In this specific region, sailing was frequently very slow [48].

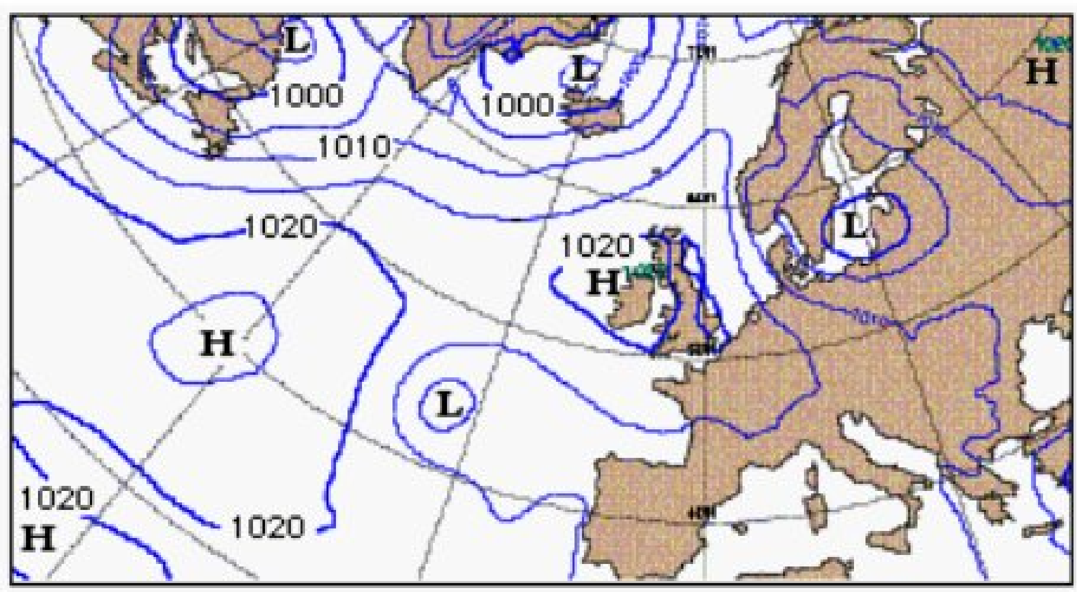

Figure 2.3: A typical surface weather chart [48].

2.5.2 Large-scale Circulations

Large-scale circulations are characterized by horizontal length scales varying from few hundreds to few thousands kilometers. Such circulations affect mostly horizontal dispersion of pollutants in the troposphere on time scales of days to several weeks. Large-scale circulations in the atmosphere depend on pressure distribution patterns. Five distinct pressure systems can be defined surface weather charts (see Figure 2.3):

1. *Lows* (marked by a letter L), also called cyclones, pressure systems surrounded on all sides by higher pressure;

2. *Highs* (marked by a letter H), also called anticyclones, pressure systems surrounded on all sides by lower pressure;

3. *Troughs*, elongated areas of low pressure with the lowest pressure along a line marking the maximum curvature of isobars;

4. *Ridges*, elongated areas of high pressure with the highest pressure along a line marking the maximum curvature;

5. *Cols*, neutral areas between two lows and two highs.

Large-scale circulations transport various air masses. Atmospheric air masses are bodies of air with nearly uniform temperature and moisture. Air masses can be classified depending upon their source regions as polar (P), arctic (A), or tropical (T). Each of those masses can also be specified as continental (c), or marine (m). Moreover, a small letter (w) or (k) was used to indicate that an air mass are warmer or cooler than the underlying surface. Therefore, mTw means hot and humid tropical air initiated over the ocean and warmer than the underlying surface.

A transition zone between two different air masses is called a *front*. Fronts form at outer boundaries of high-pressure systems and extend all the way to the center of the low-pressure system (Figure 2.4). Across the frontal zone, temperature, humidity and wind often change rapidly over short distances. Fronts can be classified into four groups: *cold*, *warm*, *occluded* and *stationary*. The cold front is the leading edge of an advancing cold air mass.

Analogously, the leading edge of an advancing warm air mass is called the warm front. When the cold front catches up with the warm front, the two occlude (close together). The result is an occluded front. When neither the cold nor the warm air masses are advancing, the front is called *stationary*. At the cold front, colder and denser air wedges under the warmer air and forces it upward. The frontal edge has an average slope of 1:50 (1 unit of height: 50 units of length), which is due to friction which slows the flow near the ground. As the moist, unstable air rises, its water vapor condenses into a series of cumulus clouds, cumulonimbus (Cb), and altocumulus (Ac). Strong, upper level winds blow the cirrostratus (Cs) and cirrus (Ci) clouds, far in advance of the approaching front, as shown in Figure 2.5.

The frontal surface of the warm front is less steep (about 1:100). Behind a warm front the *stratus clouds* (St) and fog are observed near the earths surface. Stratus clouds can produce drizzle. As air moves upward along the

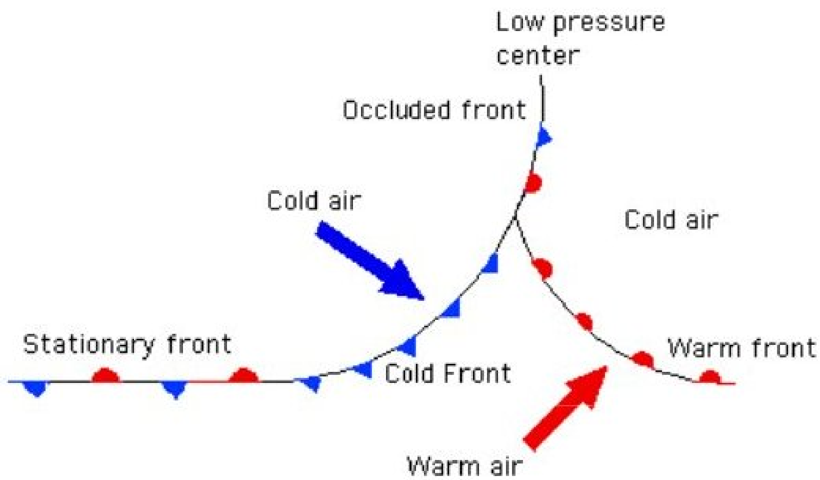

Figure 2.4: Atmospheric fronts on a surface weather chart, [48].

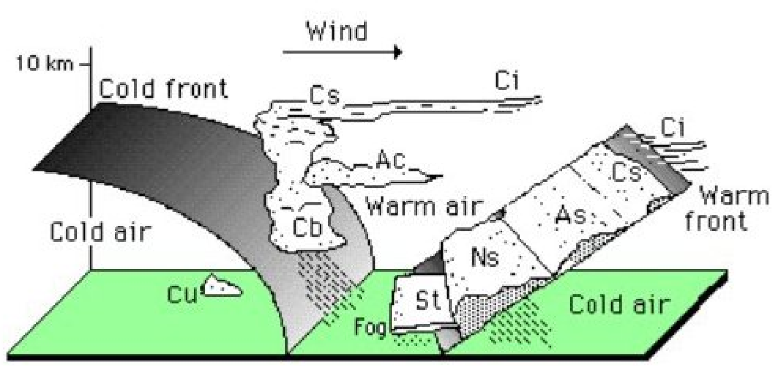

Figure 2.5: Frontal cloud systems. [48]

warm front, *nimbostratus clouds* (Ns) form, producing a broad area of rain or snow. Farther along the front, clouds gradually transform into altostratus (As), and then into a thin, white veil of cirrostratus (Cs). On top of the frontal surface, there are usually cirrus (Ci) clouds. There is a simple empirical rule, known as the Buys-Ballot law, which relates the direction of the wind near the earth's surface to the pressure field. The rule is strictly valid above the near-surface layer of frictional influence. According to the Buys-Ballot law, if you stand in the Northern Hemisphere, with the wind blowing at your back, the low pressure center will be located to the left. In the southern hemisphere, with the wind blowing at your back, the low pressure will be located to the right. The explanation of the rule is shown in Figure 2.6. When the air mass is pushed by the horizontal pressure gradient force, it initially moves toward the low pressure area (vector V1 in the figure). The moving parcel is simultaneously under the influence of the Coriolis force (outlined arrows indicated as C1 and C2), which changes its direction (vectors V2 and V3). The air changes its direction until an equilibrium between the pressure and the Coriolis forces is reached. The wind resulting from this equilibrium blows along isobars (lines in the figure marked 1000 mb, 1004 mb), and is called *geostrophic*.

In cases with strongly curved isobars, there is a balance of the pressure gradient, Coriolis, and the centrifugal forces. In the Northern Hemisphere this balance is associated with a clockwise circulation in anticyclonic (high) pressure systems, and counterclockwise circulation in cyclonic (low pressure) ones (Figure 2.7). Near the earth surface, in the presence of friction, the pressure force is no longer balanced by the Coriolis and centrifugal forces, and the wind is directed from high pressure to low pressure, crossing isobars at an angle of about 30 (Figure 2.8).

The resulting inward motion toward a low pressure center is called horizontal convergence. In just the opposite, outflow in a high pressure center is called horizontal divergence. Horizontal convergence near the ground in the low- pressure system causes the accumulation of air in the center. To remove inward- flowing air, a very slow (a few cm/s) but persistent vertical upward motion is generated. On the other hand, there is a descending flow of air to compensate for the high-pressure divergence near the ground.

Because most fluid phenomena on the earth involve rotation, the concept of vorticity is useful to explain complex atmospheric motions. Vorticity occurs as a result of different portions of fluid being moved by different amounts.

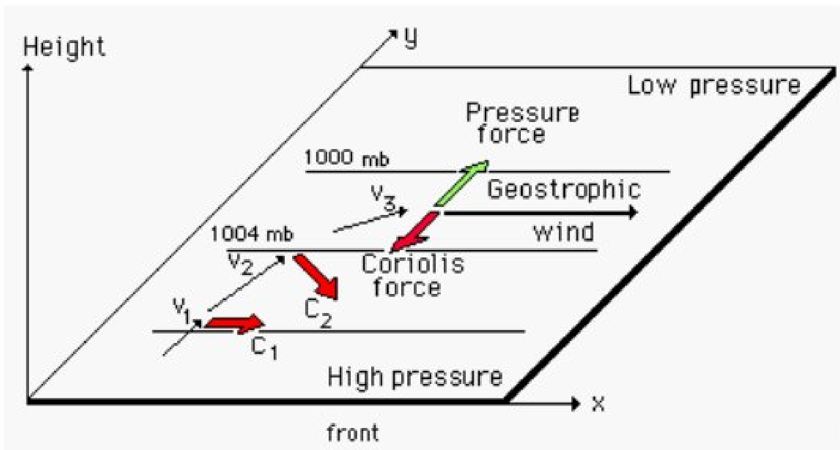

Figure 2.6: The geostrophic wind on the surface weather chart. [48]

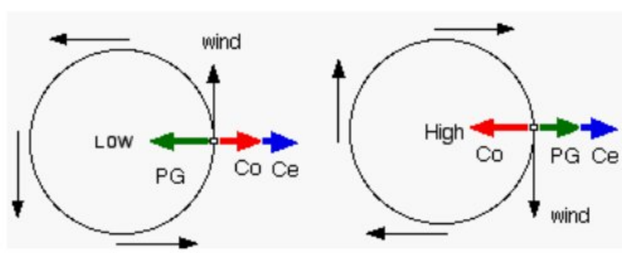

Figure 2.7: Flow in the pressure systems without friction (upper row) near the earth's surface. PG-the pressure gradient force, Co-the Coriolis force, Ce-the centripetal force [48]

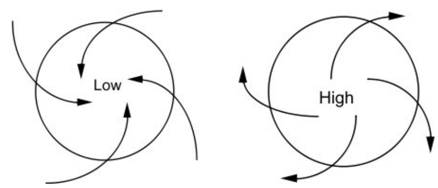

Figure 2.8: Flow in the pressure systems with friction near the earth's surface. PG-the pressure gradient force, Co-the Coriolis force, Ce-the centripetal force. [48]

Vorticity is a vector quantity, since it depends on the orientation of the axis of rotation. In meteorology, rotation about a vertical axis is often considered. Vorticity is defined to be positive (cyclonic) when the fluid spins counterclockwise, and negative (anticyclonic) when the fluid spins clockwise (when viewed from above). Because the earth spins, it also has vorticity. In the northern hemisphere, the earth vorticity is always positive, because the earth spins counter-clockwise about its vertical axis. The amount of the earth vorticity depends on latitude. If the vorticity-meter is placed on the north pole, it will spin about its vertical axis, with the speed of one revolution per day. Thus, according to our definition, earth vorticity is twice the angular velocity of the earth. When the vorticity-meter is placed on the equator, it will not spin about its vertical axis. Its vorticity is nil. The absolute vorticity is defined as a sum of the earth's vorticity and the vorticity of the air relative to the earth. The concept of vorticity is useful for explaining many phenomena in the atmosphere. For instance, it can be used to explain the development of Rossby waves in westerlies flow (Figure 2.9), in the middle and upper troposphere. Rossby waves are wavelike patterns, usually three to five in number, which extend completely around the earth. The wave flow of the westerlies provides an important mechanism for heat and contaminant transfer across mid-latitudes.

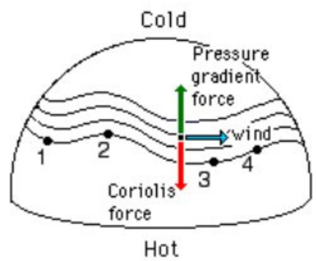

Figure 2.9: Rossby waves: wave-like patterns in westerlies flow. [48]

2.5.3 Meso-scale Circulations

Meso-scale circulations can be characterized by horizontal length scales on the order of a few tens to several hundred kilometers. Meso-scale flows can be mechanically or thermally forced and generated near the earth surface due to the effects of the earth topography, or in the free-atmosphere (figure 2.10). For example, up-slope or down-slope winds are circulations which are mechanically forced by topography. Examples of circulations, which are thermally forced in the free atmosphere, include hurricanes, severe convective storms.and frontal circulations. Gravity waves are circulations which are dynamically or thermally forced by topography.

Meso-scale convection is often induced by the temperature contrasts at the earth surface. Resulting circulations include sea/land breezes, lake breezes, urban heat islands, mountain and valley winds, and monsoons. They are best developed when large-scale winds are weak. One example is the sea/land breeze generated by the diurnal differences of temperature between the sea and the land. During daytime, the coast heats more rapidly than the sea, which causes convection over hot land. Conversely, at night, land cools more quickly than the sea, which causes subsidence of air. As a result, the compensating flow during the day (sea breeze) is directed toward the land, and in the opposite direction at night (land breeze). Studies of Brown [8] that show the angle between the roll direction and the free stream vary from about 3 in the stable case to about 5 for the convective case. Kuettner [27, 28] found the following typical properties of rolls:

- length 20 - 500 km;
- spacing: 2 - 8 km;
- height: 0.8 - 2 km;
- width/height ratio: 2 - 4 : 1.

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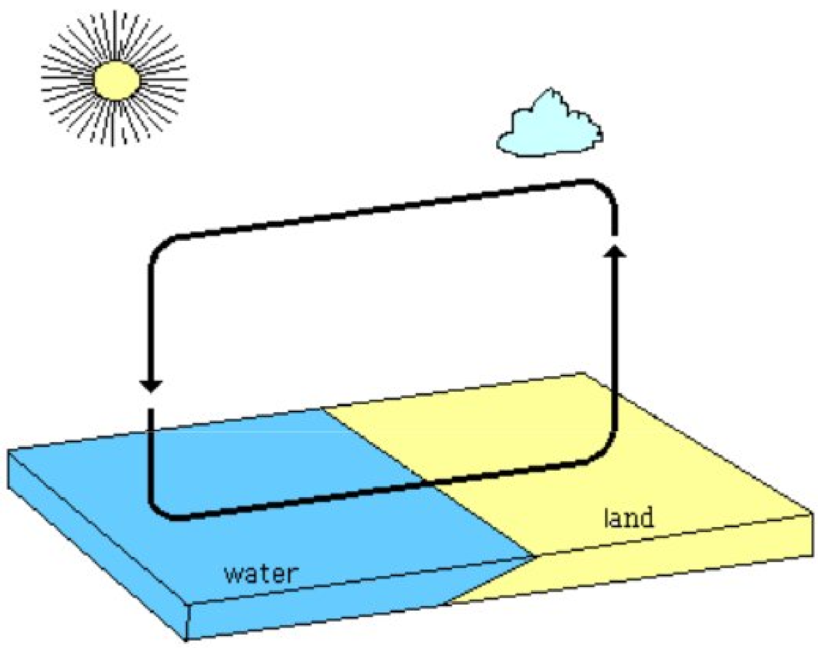

Figure 2.10: A schematic structure of the sea breeze. [48]

2.5.4 Small-scale Circulations. Turbulence

Small-scale motions are characterized by an horizontal length scales from millimeters to a few kilometers. Consequently, they are considered *local* with respect to air pollutant sources. Small-scale motions can be generated by temperature contrasts at the earth surface, wind shear, effects of the earth topography. They can also occur above the earth surface in statically stable flows or on a density discontinuity interfaces, when the destabilizing influence of the wind shear overcomes the stabilizing effect of the buoyancy force. The last effect is called Kelvin-Helmholtz instability. If the static stability of the flow is not sufficient to dampen perturbations excited by the wind shear, they may amplify. Eventually the waves may break and dissipate into smaller scale complex and chaotic motion called turbulence.

Turbulence is an essential part of the mechanism which disperses air pollutants and is crucial for the efficiency of many natural processes, such as the evaporation of water, dissipation of fog, and dispersion of plant seeds. To describe turbulence, it is convenient to adopt the approach of Reynolds (1842-1912) who in 1895 proposed decomposing any flow variable into a mean quantity (marked by overbars) and a fluctuation about the average (letters with primes). For example, for wind components and the potential temperature we have:

$$u_i = \bar{u}_i + u_i' \qquad \Theta_i = \bar{\Theta} + \theta' \tag{2.6}$$

The averaging procedure can be defined in different ways, as time average, space average or ensemble average. In a conventional theoretical procedure the mean quantities are *ensemble averages*. It is assumed that atmospheric flows are members of an ensemble whose individual realizations obey the Navier-Stokes equations. Details can be found in [48, 45].

2.6 Planet Boundary Layer

The *planetary boundary layer* (PBL) is the lowest portion of the atmosphere, about 1 - 2 km deep, which intensively exchanges heat as well as mass (water, gases) with the earth surface. Although the PBL contains only about 2% of the total kinetic energy of the atmosphere, it contributes as much as 25% to its total generation and 35% to its total dissipation. The atmospheric boundary layer is of great practical and scientific importance. Essentially, all human and biological activities take place in this layer. Practically all air pollutants from natural and anthropogenic sources are emitted within the

PBL.

Flow in the boundary layer is controlled by the diurnal cycle of the surface energy budget. The energy balance at the surface is expressed as:

$$R_n + G + H + E = 0 \tag{2.7}$$

where:

- R_n is the flux of net radiation (global solar radiation received by the surface, plus atmospheric radiation, minus terrestrial radiation);
- G is the vertical heat flux into the soil;
- H and E are the sensible (conduction) and latent (resulting from water phase changes) heat fluxes to the atmosphere.

A quantity has a positive value, when energy is transferred away from the interface, and a negative value in the case of transfer towards the interface. Net radiation flux Rn is negative (down) during the day, reaching minimum values at local solar noon. At night, Rn is positive, illustrating the loss of energy by terrestrial radiation. Rn is zero just before sunset, and just after sunrise. At night, the term E can become negative, if dew forms. During the day, energy gained at the surface is transferred to the atmosphere, to the soil, and also is used in the evaporation processes. This transfer of heat (from the ground surface to the air directly above it) can generate vertical motions, called *convection*. Convection redistributes heat throughout the atmospheric boundary layer. The influence of the surface sensible heat flux decreases with height. As a result, the diurnal temperature amplitude also decreases with height.

It is generally assumed that over a flat and homogeneous surface, the planetary boundary layer is *horizontally homogeneous*, but organized vertically into *several layers*.

Within a few millimeters on the surface, there is a viscous sublayer, where the flow is mostly laminar. Above this layer, there is a surface layer, 1-100 m deep, where the turbulent fluxes of momentum, heat, and moisture are approximately constant with height (i.e., they change in magnitude no more than 10% from their surface values). The wind direction in the surface layer is approximately constant with height. In the first few meters of the surface layer the wind velocity, humidity, and temperature are nearly logarithmic in neutral, and also in stable and convective conditions. Above the logarithmic sublayer the profiles of various meteorological parameters differ depending on thermal stratification. The portion of the boundary layer beyond the surface layer is called the *outer layer*. In day-time conditions the potential temperature, humidity and wind velocity in the outer layer are approximately constant with height, and the layer is often called the *mixed layer*. At night, the temperature inversion is usually formed near the surface. In the upper portion of the outer layer, called the residual layer, the potential temperature remains constant with height. At the level of few hundred meters above the surface, the wind velocity reaches a maximum, and exceeds the value of the geostrophic wind. This maximum is often called the *low-level nocturnal jet*.

The diurnal changes of temperature are accompanied by changes of wind velocity and wind direction. Above the surface layer the winds are observed to reach maximum speeds at night and minimum speeds during the day. But near the surface, the opposite behavior of wind velocity is observed. The surface wind speed increases after sunrise, peaks in the early afternoon and decreases near sunset. Above the surface layer, the diurnal wave is nearly 12 hours out of phase. Friction causes the wind velocity in the boundary layer to be zero at the surface.

During the last several decades, the structure of the atmospheric boundary layer has been intensively studied, not only because of the striking beauty of its coherent structures [1], but mostly because understanding its flows and clouds is essential in environmental studies, numerical weather prediction, and climate analyses. As a result, since the 1950s, the fundamental knowledge of boundary layer turbulence has been achieved as a result of extensive experimental effort, and numerical modeling.

2.6.1 Monin-Obukhov theory

A simple analytical formulation of idealized conditions in the atmospheric boundary layer can be obtained based on similarity and dimensional analysis [9]. During the last four decades an elegant similarity framework has allowed classification of knowledge on atmospheric turbulence obtained through extensive experimental effort. The mathematical aproach to describe PBL can be found in [45].

Turbulence in the thermally stratified surface layer can be elegantly characterized in terms of the surface layer similarity theory. The theory was formulated about five decades ago by A.S.Monin and A.M.Obukhov (1954). This approach has found many practical applications in experimental analysis of surface layer turbulence, as well as in the parametrization of mass and energy exchange across the earth surface.

According to the Monin and Obukhov (M-O) theory, similarity scales in the surface layer can be formulated on constant values of the kinematic turbulent fluxes:

$$H_0 = \bar{w'}\theta' \tag{2.8}$$

$$Q_0 = \bar{w'}q' \tag{2.9}$$

$$\tau_0/\rho = \bar{w'}u' \tag{2.10}$$

then:

$$u_*^2 = \tau_0/\rho; \tag{2.11}$$

$$T_* = -H_0/u_*; \tag{2.12}$$

$$q_* = -Q_0/u_*; \tag{2.13}$$

$$L = u_*^2/\beta\kappa T_* \tag{2.14}$$

where u_*, T_*, q_*, L are the velocity, temperature, humidity (or other scalars, such as ozone or carbon dioxide concentration), and height scales respecively, $\beta = g/To$ is the buoyancy parameter, and L is called the Monin-0bukhov length. The sign convention is chosen so that L is negative in unstable and positive in stable stratification. The potential temperature flux H_0 in the definition of T_* and L should be replaced by the virtual potential temperature flux H_V when moisture stratification is included. The virtual potential temperature flux is defined as $H_V = H_0 + 0.61TQ_0$, where Q_0 is the surface humidity flux. The virtual temperature Θ_V is defined as $\Theta_V = \Theta(1+0.61q)$, where q is the specific humidity.

The Monin-0bukhov length L is roughly the height at which the shear production of turbulent kinetic energy u_*/L is equal to the buoyant production βH_0.

One can classify the following two main layers:

- stable boundary layer: $H_0 < 0,\ T_* > 0 \ and\ L > 0$;
- unstable boundary layer: $H_0 > 0,\ T_* < 0 \ and\ L < 0$.

In the latter case, the turbulent energy production is:

- mainly mechanical (produced by shear) for z < -L (close to the surface);
- buoyant (produced by the heat flux) when z > -L.

Similarity theory predicts that any turbulent characteristics of the flow, non- dimensionalized against surface layer scales,is a universal function of the stability parameter $\varsigma = z/L$, e.g.:

$$\frac{\bar{w'}}{u_*^2} = \psi_w(z/L); \tag{2.15}$$

$$\frac{\bar{\theta'}}{T_*^2} = \psi_\theta(z/L); \tag{2.16}$$

$$\frac{\bar{q'2}}{q_*^2} = \psi_q(z/L); \tag{2.17}$$

$$\frac{L}{u_*}\frac{dU}{dz} = \psi_m(z/L); \tag{2.18}$$

$$\frac{L}{T_*}\frac{d\Theta}{dz} = \psi_h(z/L); \tag{2.19}$$

$$\frac{L}{q_*}\frac{dq}{dz} = \psi_q(z/L); \tag{2.20}$$

where U, Θ, and q are mean wind velocity, potential temperature and specific

humidity. Usually, the nondimensional gradients are divided by $\varsigma = z/L$ and substituted by new similarity functions ϕ, defined as:

$$\phi_m = \varsigma\,\psi_m \quad \phi_h = \varsigma\psi_h \quad \phi_q = \varsigma\psi_q \tag{2.21}$$

Consequently:

$$\frac{z}{u_*}\frac{dU}{dz} = \phi_m(z/L) \quad \frac{z}{T_*}\frac{d\Theta}{dz} = \phi_h(z/L) \quad \frac{z}{q_*}\frac{dq}{dz} = \phi_q(z/L) \tag{2.22}$$

where ϕ_m, ϕ_h and ϕ_q are new similarity functions.

Chapter 3

Meteo Models

3.1 Introduction

The atmosphere is an active chemical medium in which chemical reactions are continuously taking place. Their rates are governed by concentrations of the participating species, and external factors (e.g., incident solar energy). Modeling air quality requires an accurate modeling of all the factors that control the concentrations of these chemical species, including the movement of these airborne species from one location to another. Hence, accurate air quality modeling is predicted by accurate meteorological modeling.

The phrase *meteorological modeling* (or *atmospheric modeling* or *numerical weather prediction, NWP*) refers to any numerical representation of the atmosphere and its processes. A numerical representation is based on dynamical, thermodynamical, physical and chemical properties of the atmospheric system. Even though atmospheric modeling started out examining just the atmospheric processes, it is commonly understood that the atmospheric motions depend on the properties of the earth surface as well as the dynamics of the oceans. Therefore, it is appropriate to extend the definition of *meteorological modeling* to include the effects of the surface of the earth and the oceans [43].

Meteorological models are developed and used for two main purposes:

1. to understand and forecast local, regional, or global meteorological phenomena;

2. to provide the meteorological input required to run air pollution models.

Numerical meteorological models can be divided in two groups:

1. diagnostic models, i.e., models that are based on interpolation/extrapolation of available measurements and contain no time-tendency terms;
2. prognostic models,i.e., models that perform space-time integration of the conservation equations of mass, heat, motion, water, and if necessary other substances, such as gases and aerosols.

3.2 Models and Processes

A mathematical representation of the atmospheric system is possible just for some of the processes involved. It is not mathematically feasible to solve all of the equations in an analytical framework. Hence, it is possible to approximate numerical representations of the atmospheric system using computers. At a given time, in-situ as well as remote instruments sense only a very small portion of the atmosphere. Numerical modeling helps to fill the gaps in the sensed data and complete the three-dimensional picture of the atmospheric system in a physically consistent manner. Solar radiation forms the primary energy source for the atmospheric system. The atmospheric gases and the surface of the earth absorb the solar energy. The absorbed energy is emitted back in the infrared wavelengths (long-wave) to be re-absorbed by atmospheric gases or to be emitted to space. This radiation exchange process is modulated by intervening clouds, aerosols, pollutant species, and also by the properties of the surface of the earth. The properties of the earth surface vary from water surfaces (e.g., oceans and lakes), dry and arid surfaces of the deserts, and moisture-laden vegetation (e.g., rain forests) to the reflective surfaces of the snow and ice of the polar and high mountainous regions. These variations impact the thermodynamic structure of the atmosphere, and in turn affect its dynamics. It is helpful to classify the different processes into broad classes:

1. atmospheric dynamics
2. microphysical processes of the water cycle
3. atmospheric radiation
4. atmospheric and aerosol chemistry

5. processes that involve the air-surface interactions

The paragraphs in the sequel describe these processes in detail.

3.2.1 Atmosphere Dynamics

Atmospheric dynamics refers to the processes that directly impact the movement of air and airborne constituents. These processes are related to the fundamental forces acting on a parcel of air.

- The pressure gradient force - the force exerted by air, moving from high-pressure regions to low-pressure regions. This force acts perpendicular to the isobar through a given location and it is pointed toward low pressure zones.
- The gravitational force - acts towards the center of the earth.
- The buoyancy force - the force related to difference in densities between the parcel of air and its surroundings.
- The frictional forces - forces caused as the parcel of air moves with a velocity different than that of the surrounding air. These forces are pointed against the direction of motion.
- The centrifugal the force caused by the rotation of the earth. This force is pointed along a direction perpendicular to the axis of rotation of the earth through the air parcel.

3.2.2 Water Cycle and Cloud Microphysics

Water is a key component of the atmosphere. It makes the earth atmosphere unique in this solar system, and also makes life possible. The term *water cycle* refers to the set of processes that move water from the oceans to the atmosphere, in water phase state vapor, then form clouds, generate rains that lead to streams and rivers, thus taking the water back to its source, the oceans. During this cycle, water is present in all three of its phases vapor, liquid and solid. The processes include evaporation and condensation, as well as growth of ice crystals (and water droplets) on heterogeneous microscopic particle substrates called *ice nuclei*. These processes are collectively termed *microphysics*. The phase change that occurs in several of these processes prodeuces the release of latent heat, which significantly changes the thermodynamics of the environment.

Many of the microphysical processes are not easy to represent in mathematical terms, to be incorporated into numerical models. These processes are represented by parameterizations that have been derived empirically from the results of numerous field and laboratory experiments and measurements. Even though significant progress has been achieved in the understanding of these processes in the last few decades, cloud microphysics still poses considerable challenge to NWP researchers.

3.2.3 Atmospheric Radiation

The sun is the primary source of energy that drives the atmosphere. Solar energy reaches the earth in the form of electromagnetic radiation. Some of this energy is reflected back to space and the rest is absorbed by the various constituents of the atmosphere as well as by various features on the earth surface. Part of this absorbed energy is re-emitted in longer infrared range wavelengths. Gases such as carbon dioxide and methane reabsorb this emitted long wave radiation, thus trapping the energy and giving rise to the term *greenhouse effect*. Clouds also play a major role in reflecting some of the incoming solar radiation and absorbing some of the outgoing long wave radiation. This complex energy cycle along with the rotation of earth about its axis, the tilt of the axis with respect to the plane of earth orbit, and the fact that the equatorial regions receive more direct insolation than the Polar regions, give rise to the large-scale planetary waves and global circulations such as the Hadley cells. As radiation directly impacts the temperature of both the atmosphere and the earth surface, it plays an important role in local circulations, especially thermally driven circulations such as land- and sea-breezes, drainage flows, and Katabatic winds. Atmospheric radiation is modeled at various levels of complexities ranging from simple bulk calculations and two-stream models to more complex models that treat different parts of the electromagnetic spectrum.

3.2.4 Atmospheric Chemistry

The atmosphere is a complex mixture of gases and aerosols. This mixture is augmented with chemical species that result from human activities. This mixture gives rise to numerous chemical reactions of varying strengths and speeds. Some of the chemical reactions are catalyzed by the presence of other chemicals (catalysts) or the presence of ultraviolet radiation. Cloud microphysics and atmospheric radiation, play a major role in atmospheric chemistry. Atmospheric radiation is a key process to several reactions of interest providing the actinic flux for the photochemical reactions. Heterogeneous reactions involve the presence of reactant species and catalysts dissolved in water. Also, water facilitates the transport of material spatially, especially in the vertical direction, due to washout by precipitation.

3.2.5 Atmosphere-surface interactions

Atmospheric conditions are significantly affected by the properties of the earth surface at any location. These interactions occur via several mechanisms:

- dynamical processes due to terrain forcing and friction;
- radiational processes through the absorption and emission of radiation, and
- physical and microphysical processes due to the injection into the atmosphere of water vapor as well as aerosol particles, which act as condensation and depositional nuclei

Atmospheric phenomena that are caused by these interactions can be found almost anywhere. Along the coastal areas, sea and land breezes are driven by the thermal gradient generated by the unequal heating of the land and water surfaces. Mountains block the mean wind. Down-slope winds that accelerate and warm the air are found in many places and are known by names such as Chinook and Fehn. Katabatic winds are found in the Antarctic and are acredit to the rapid cooling at the surface due to the high albedo of the ice surface. Hurricanes are strengthened by the thermal energy and water vapor from the surface of warm oceans. They rapidly decay if either of these sources is cut off. Urban areas impact the local meteorology by a complex interaction between the surface (including buildings and other urban scenery) and the atmosphere, which include the slowing down the mean wind by the increased resistance, and the input of moisture and heat. The urban landscape also provides cloud-forming aerosols and other pollutant species. This phenomenon is called the urban heat island.

3.3 Scales and scale interactions

Atmospherics processes are usually classified into different scales based on their spatial or temporal extent. Dynamical features in the atmosphere range from large-scale planetary waves to small-scale turbulent motions. The atmospheric scales of motion are generally divided in the following wavelength regimes: planetary, synoptic scale or continental scale, mesoscale, and local or cloud scale. These scales have typical wavelengths of 10,000, 1000, 100, and 1 km respectively. The wavelengths represent the spatial size of oscillations that are significant at those scales. Other scales can be easily defined depending on the processes of interest. However, such a classification does not imply that atmospheric dynamical processes do not occur in between wavelengths. The scales have traditionally been defined to make modeling of atmosphere a tractable problem. The definitions of the scales themselves change as faster computers become available. Even though the scales are defined for numerical convenience, it is widely accepted that the processes at one scale significantly affect processes in other scales.

3.4 Modeling Approaches

Numerical models of the atmosphere can be divided into two major categories:

1. diagnostic models
2. prognostic models.

This classification is based solely on the functionality of the model. Diagnostic models are used to describe the state of the atmosphere at a particular time step or during a period of time, based on the known values of the state variables (variables that describe the state of an air parcel pressure, temperature, moisture content and density), as well as the dynamical variables (speed and direction or the velocity components). In general, the equations in this modeling approach do not contain time derivatives. Prognostic models, on the other hand, attempt to describe the atmospheric state at a future time based on the current and past conditions. The prognostic models are described by appropriate differential equations, which represent the time-variability of the state and dynamical variables. The choice among these models depends on the application at hand.

3.4.1 Deterministic modeling and stochastic processes

The atmosphere is stochastic; transport and dispersion exhibit random behavior. Model outputs should in principle be expressed as distributions that display the random character of the variables of interest. A a matter of fact, all models in use are deterministic; they display the average behavior of the spectrum of random outcomes.

3.4.2 Diagnostic models

In diagnostic models the variables of interest are *diagnosed* or *inferred* from the state of the atmosphere, which has been described by other models and/or observational analyses. In most cases, atmospheric states at various times are read into these models. The models make some fundamental assumptions regarding the behavior of the atmosphere in between these states. For example, the variables can be assumed to be constant, or have a linear behaviour. In any case, these models are restricted to cases where the atmospheric states are well known at sufficiently high resolution in space and time. Diagnostic models are used in air quality modeling and plume dispersion modeling arenas, primarily in conjunction with prognostic models, which can provide the four-dimensional state of the atmosphere to provide an input to the diagnostic model.

3.4.3 Prognostic models

Prognostic models use predictive equations to describe model variables. Model variables are quantities that are conserved under spatial motion. A quantity is defined as *conservative* if it does not change in value with time. For example, linear momentum, potential temperature, and mass mixing ratios (ratios between the concentrations of the material to the density of air) of airborne material are conserved quantities in the atmosphere.

3.4.4 Hierarchical modeling

In a numerical model of the atmosphere, the spatial scales of motion that can be resolved are dependent on the grid spacing used to represent the computational domain. Traditional numerical models use a constant grid spacing, hence they suffer from prescribed lower limits for the spatial scale. However, atmospheric motions occur over all spatial scales. Hence, the concept of *hierarchical modeling* was created: a set of numerical models is run, each addressing a specific range of motions. The models are sometimes run in sequence with the lowest grid resolution model first followed by higher grid resolution models, which derive boundary conditions from the solution of a coarser grid resolution model. They can also be run in tandem with information exchanged from lower to higher resolution models during each time step. This is referred to as *nested-grid modeling*. When the information transfer occurs only from lower to higher resolution, the models are called *one-way nesting*. If the solution from the higher resolution model is used to update the lower resolution model, the models are termed *two-way nesting*. Almost all the operational, numerical forecast centers, use this approach of nested-grid modeling. A few examples of nested-grid models include: the ETA model of the National Centers of Environmental Prediction (NCEP), Mesoscale Model 5 (MM5) of the National Center for Atmospheric Research (NCAR), the Regional Atmospheric Modeling System (RAMS) of Colorado State University (CSU), and the Coupled Ocean Atmospheric Mesoscale Prediction System (COAMPS) of the US Fleet Numerical Meteorological and Oceanographic Center (FNMOC). The new Weather Research and Forecast (WRF) model, under development under the leadership of NCAR and NCEP, is also a nested-grid model. From air quality standpoint, any of these models can be used to provide the environmental conditions required by atmospheric chemistry models. For example, the US Environmental Protection Agencys (EPA) Models-3 system encompasses MM5, the Community Multiscale Air Quality (CMAQ) modeling system, Sparse Matrix Operator Kernel Emissions (SMOKE) System and several pre- and post-processors (Byun and Ching, 1999). *Nesting* provides an efficient method for operational models to place higher resolution over areas of interest, especially when uniform grid spacing is exploited over the whole domain. However, when dealing with specific atmospheric phenomena such as hurricanes or the transport and diffusion of a toxic cloud, nested grids have some inherent disadvantages. Nested grid models require an a priori knowledge of the solution so that high-resolution nests can be strategically placed to capture the features of interest. Another problem with nested grids is the existence of internal boundaries in the computational domain, which rise spurious features if not properly treated. Unstructured grid methods provide a paradigm to circumvent these problems.

3.4.5 Multiscale modeling

Even though nested grid models have shown considerable skill in forecasting atmospheric processes, the high number of scales of motion raises considerable challenges to the modelers. Bacon et al. [3] introduced a new paradigm in order to automatically change the grid resolution, based on physical features in the computational domain and in the evolving solution. The Operational Multiscale Environment model with Grid Adaptivity (OMEGA) is built upon an adaptive unstructured grid, where the fundamental computational element is a triangular prism. Such a grid takes advantage of the flexibility of unstructured grids as well as the vertical correlation of the atmosphere.

3.5 Modeling framework

A *model* consists of a set of equations or relationships between variables representing processes,defined on a *computational* domain. The structure of the computational domain is determined by the way in which the equations are solved. In general, models can be divided into two main types based on the frame of reference. If the frame of reference moves with an air parcel, the model is called *Lagrangian*. If the frame of reference is fixed while the air parcel moves relative to it, the model is called *Eulerian*.

3.5.1 Lagrangian models

If the equations are solved following an air parcel, the model is said to be d-Lagrangian as the process solves the Lagrangian (or total) derivative. d-Lagrangian models are also sometimes called *grid-free models* as they do not depend on a specific grid. The Lagrangian framework has certain advantages. Usually, there are no spurious computational modes that can make the calculations unstable. However, the accuracy of the calculations depends on the magnitude of the time steps used for integration, as well as the accuracy of the underlying environment. Long time steps make the calculations less accurate. The magnitude of the integration time step must represent the time-scales of the underlying physics. Lagrangian models are ideally suited to manage localized problems, such as plume dispersion, where the plume itself covers a small volume compared to the dynamic environment. The plume can be represented as a collection of puffs and/or particles whose movement and growth (in the case of puffs) are determined by the dynamic and turbulent nature of the environment they are in. Plume modeling using puffs suffer from the drawbacks of Lagrangian models. As the puffs are followed independently, *it is difficult to compute interaction between the puffs*, while, in nature, the plume is a continuous field.

3.5.2 Eulerian models

∂ Eulerian models are grid based and integrate the partial differential equations. The ∂t prognostic variables are defined in an appropriate geometric grid. The partial differential equations (PDEs) describe the rate of change of a quantity at a grid point.

3.5.3 Semi-Lagrangian techniques

Semi-Lagrangian techniques combine the Lagrangian and the Eulerian methods to provide a method, which is unconditionally stable in a numerical sense. As for Lagrangian schemes, accuracy does depend on the time step used.

3.5.4 Plume models

Plume models describe the plume of material in a background flow. These models infer the background flow either as a constant field or derived from data generated by other models. Plume models need to describe: 1) the source of the airborne material and 2) the transport and dispersion of this material in the ambient flow. In early plume models, the plumes were characterized by a Gaussian distribution of concentration in the transverse directions (across the direction of travel). Such a characterization was based on observations of power plant plumes, fairly close to their source. The plume representation is valid near the source, where the plume is *pristine* or untouched by other effects, and is expanding only by diffusion. However, the planetary boundary layer (PBL), provides a very complex environment in which the plume can be moved around in many ways. Thermals in the PBL can cause entrainment and detrainment to the plume. Once the plume expands enough to touch the ground, some effects (such as ground reflection) have to be taken into account. Other complexities include plume splitting by geographical features such as hills or dynamical features such as eddies. In order to better represent the plume, researchers came up with the *puff model* in which the plume is considered as a collection of discrete puffs, each puff having Gaussian attributes.

3.6 Dynamical and thermodynamical processes

The processes that directly affect the state of the atmosphere at any given time can be broadly classified into: 1) dynamic processes that drive the movement of air, and 2) thermodynamic processes that control the transfer of thermal energy. The equations that make up a model are basically equations that describe these processes. They represent the behavior of the variables describing the state of the atmosphere.

Atmospheric Variables
Atmospheric variables can be classified into three broad classes:

1. variables representing the current state of an air parcel, known as the *state variables*;
2. variables representing the motion of air, or *dynamic variables*;
3. variables describing the airborne constituents.

The state variables include density, temperature and pressure. The dynamic variables include momentum (or velocities) and turbulent kinetic energy (in models that use high-order turbulence parameterizations). Airborne constituents are usually represented by concentration (mass per unit volume) or mixing ratio (mass per unit mass of dry air).

Fundamental Equations
The equations that govern the circulations in the atmosphere are derived from the fundamental physical processes that are involved.

Advection
Advection is the term used to refer to the process of transport of a material in the fluid flow solely due to the point-to-point movement.

Turbulent Diffusion
Turbulent diffusion refers to the seemingly random motions of air that occur at small scales. They come from molecular diffusion (due to Brownian motion) and eddy diffusion (eddies are generated by either velocity shear or buoyancy driven circulations). This is a very complex and challenging process to model. Representing these processes explicitly in a model requires high spatial and temporal resolution, making the model unsuitable for forecasting. Most models resort to parameterizations to describe these processes. Turbulent diffusion plays a very important role in defining PBL, which is defined as the layer of the atmosphere closest to the earths surface that is dominated by the effects of the terrain surface. Outside the PBL, turbulent diffusion is seen in boundaries between high-speed atmospheric layers in which waves are generated that break in a manner similar to ocean waves. Such waves can also be initiated downstream from high mountain ridges as air streams across them. These regions are of great danger ti aircraft due to clear air turbulence (CAT).

3.7 Physics parameterizations

The atmosphere undergoes many physical processes, ranging from large-scale, nearly steady influx of solar radiation, to rapidly changing turbulent eddy processes. In most cases, explicitly representing these processes from first principles is numerically expensive. Hence, it is customary to simplify the equations using empirical relationships derived from field and laboratory experiments. Several parameters were proposed for the same processes. Most prognostic models differ in the type of parameterizations, and the manner they are applied. Some of these processes and their parameterizations are discussed in the following sections.

3.7.1 Cloud microphysics

Cloud microphysics refers to the collection of processes that define the water cycle, with an emphasis on the evolution of clouds and precipitation. Water is unique, in the sense that it can occur in the atmosphere in all phases concurrently. Also, phase changes exchange significant thermal energy with the air. Most models classify the water substance into several categories: water vapor, cloud droplets (non-precipitating), rain, ice crystals (non-precipitating), snow, and sometimes hail. However, models differ in how each of these categories is represented. Some models represent cloud ice (ice crystals) as a single species type, the average size and number changes with time. Other models solve conservation equations for various ice crystal types: needles, plates, stellar crystals as well as hybrid shapes.

Phase changes of water considered are:

- condensation/evaporation;
- deposition/sublimation;
- feezing/melting;
- autoconversion;
- collection;

3.8 Convection

Air moves upward due to the buoyancy forces. This upward motion cools the air adiabatically and can become saturated in the process, resulting in convective/cumulus clouds. The release of latent heat, associated with the condensation and deposition processes, adds extra buoyancy, accelerating updrafts. Convection poses specific challenges to a numerical model. First, convection occur over relatively small scales, O(1-10km). A very small grid spacing is required. Second, the high-speed updrafts impose severe limitations to integration time step. Most mesoscale and regional-scale models use convective parameterizations to grossly represent the effects on the larger scale circulations of the sub-grid scale convective clouds .

3.8.1 Implementation of convective parameterization

Since convective parameterization is scale specific, particular attention has to be paid to its implementation, based on the typical spatial scales of the numerical model. For example, when sufficient resolution is available in order to simulate convection at its coarsest levels, including a convective scheme will double-count the energy and water vapor distributed via the convective scheme. To circumvent this problem, nested-grid models selectively activate or deactivate the convective parameterization based on the grid scale. In the high-resolution grids, only the explicit microphysics is kept active. This poses some interesting questions on implementation on variable-resolution (multi-scale) models. When grid cells are larger than a few kilometers, convection is truly sub-gridscale, when cells are less than a kilometer, the convection bulk may be explicitly captured. One expects a behavior that smoothly varies from full impact of the convective parameterization at coarse-resolution, to no impact at the high- resolution part of the grid.

3.9 Planetary Boundary Layer

The Planetary Boundary Layer (PBL) is the layer closest to the surface of the earth, which is directly impacted by the effects of the terrain. The thickness of this layer is not constant in space and time. It is, in general, shallow over homogeneous and water surfaces, and thicker over rough and land surfaces. The dynamics within the PBL is controlled by convective and turbulent processes, which in turn are controlled by the frictional forces and the thermal (sensible) and water vapor (latent heat) fluxes at the surface. The effects of the PBL can be incorporated into a mesoscale model in two ways. One way is to parameterize the entire PBL as one layer. This involves identifying and relating unresolved processes in the PBL with resolvable ones. The complexity of this single-layer PBL parameterization lies in the variety and interdependence of atmospheric processes acting on different scales. The second approach is to include several computational levels in the PBL in order to resolve the boundary layer structure effectively and explicitly. Such multi-level PBL formulations require near-surface turbulent fluxes of momentum, heat, and moisture within the PBL. Thus, they require some type of closure scheme to relate turbulent fluxes to mean quantities.

3.10 Atmospheric radiation

The fundamental source of energy for the atmospheric engine is the sun. To understand atmospheric dynamics and physics, it is essential to have a good understanding of the energy transformations, starting with the incoming solar radiation. Solar radiation plays a crucial role in atmospheric chemistry by enabling and modulating several key reactions commonly grouped under the term *photochemical reactions.*

3.10.1 Radiation parameterizations

Radiation parameterization raises several major challenges. Though we have classified the radiation broadly into two classes (short wave and long wave), the absorption and the emission parameters changes with the wavelength and the composition of the medium. Scattering by intervening aerosol and gases, as well as the presence of clouds, make modeling intractable. Another problem is modeling the balance between the incoming solar radiation and the outgoing terrestrial radiation. If the balance is not computed accurately, it can result in major errors in circulation, especially in models integrating over long time domains such as climate models.

3.10.2 Incoming solar short wave radiation

The earth-atmosphere system receives a total of 1380 W/m_2 of solar energy at the top of the atmosphere. This quantity is relatively constant and is called as solar constant. There are several processes that redistribute the energy received from the Sun. Of the total 1380 W/m_2 of short wave radiation reaching the top of the atmosphere from the Sun:

- 17% is absorbed by the atmosphere,
- 44% reaches the surface (20% directly and 24% through clouds), 4% being reflected back,
- 20% is absorbed by the surface,
- 20% is reflected by clouds,
- 3% is absorbed by clouds, and
- 6% is scattered by the atmosphere back to space, an equal amount being scattered to the ground.

Aproximately 30% is reflected back to space, the atmosphere absorbs 20% and 50% is absorbed by the earth surface. Of course, this is an aggregated scenario; local situations can be different.

3.10.3 Outgoing Terrestrial Long Wave Radiation

The earth surface and the atmosphere produces radiation primarily in the infrared part of the spectrum. Called *long wave radiation* to be distinguished from *short wave radiation* from the Sun. Some of the radiation emitted by the surface is absorbed by atmospheric components such as carbon dioxide (CO_2) and water vapor. Clouds also play a major role as they reflect back a good portion of the outbound radiation. Other factors that affect the radiant energy budget include aerosols and greenhouse gases (e.g., methane). Long wave radiative flux parameterization of the in atmospheric models is typically treated as a function of the normal optical thickness, which when integrated over all wavelengths is represented by the broadband emissivity. In clear or cloudy air, this emissivity is dominated by the water content of the air. Water vapor and carbon dioxide are considered as sources of long wave radiation.

3.11 Numerics

Model numerics represent the equations in a discretized domain. The discretization process involves the conversion of the continuous conservation equations into piecewise sets of discrete representations. Integration is performed in space (solving and computing gradients) as well as in time (solving the partial differential equations).

3.11.1 Domain discretization

The discretization can be separated functionally into spatial and time discretizations.

1. **Spatial Discretization**: usually a simple rectilinear box is subdivided into a structured three-dimensional grid by sets of uniformly separated planes that are parallel to the sides of the domain.

2. **Temporal Discretization**: marching in time is performed by computing the rate of change of each prognostic variable at each instant in time, then calculating the difference from the last time step to the current time step.

3.11.2 Computational stability considerations

Numerical models solve a set of finite difference equations, which are approximations to the partial differential equations. Truncation errors can accumulate in an unbounded manner depending on the numerical integration scheme chosen. In most cases, the error is directly linked to the local velocity, the spatial resolution and the time step used in the integration. Specific limits on time steps can be derived by performing an eigen-analysis.

3.12 Data ingest

Atmospheric modeling is an initial, boundary value problem. Regarding initial conditions, we depend on measurements of the key state variables. Ideally, initial conditions should define the state of the atmosphere accurately at all points within the computational domain. This is impractical, if not impossible, to achieve measurements and we have to be satisfied with a few finite points in space and time.

3.12.1 Data types

Measured meteorological data can be broadly divided into two types: 1) in situ measurements and 2) remotely sensed measurements. In situ measurements, take place at the location of the measuring instrument. A thermometer, a pressure sensor, a wind-vane, an anemometer are examples of in situ measurements. Remotely sensed measurements come from instruments that look into a region. These include like satellite based instruments, radars, sodars and lidars. In situ measurements are considered ground truth. Remote measurements,even though they have improved in accuracy over the years, are still fraught with errors of various types.

3.12.2 Data quality control and quality assurance

Various types of data are needed to initialize and bound numerical models. The quality of data directly impacts the quality of model output. Errors in data come from various sources. First, from instrument errors. This comes from fundamental physical limitations and from calibration errors. The second type of error is data retrieval error. This is especially applicable for remote sensing platforms. For example, let us consider a radiometer, which measures the radiance of an atmospheric layer. Radiances are converted to temperature. The retrieval algorithms required for this process are only approximations.

3.12.3 Data assimilation techniques

Taking a set of data values and build a complete state of the atmosphere at a time step, or during a time period is called: *data assimilation*. Sparseness of atmospheric data makes it challenging to build a three-dimensional state of the atmosphere using only observations. It is usual to start from a known state of the atmosphere at a time close to model initialization time and modify this first guess state according to the differences between the observations and corresponding first-guess values. This process includes constraints based on fundamental physical principles. If these constraints are not met properly, the inclusion of the observations results in generating noise in the numerical modeling system. Forecasts from a previous operational cycle can be used as the first guess. Data assimilation has received much of attention in the past two decades, resulting in techniques of various complexities. These techniques include schemes such as 1) Optimum Interpolation, 2) Three-dimensional Variational Schemes, and 3) Four- dimensional Variational Schemes. A new class of assimilation techniques receiving much attention by researchers is based on the *ensemble approach*.

3.13 Model verification and validation

Verification and validation are important steps in the development of any model. *Verification* refers to checking whether a model indeed represents the relevant physics. *Validation* is checking model results for accuracy. Verification is usually performed during model development. At times, model validation can be challenging. The greatest challenge in model validation is the disparity between the modeled variables and their observed counterpart. Most models output volume-averaged values (averaged over a grid cell) values while most observations are point observations. However, it is important to note that some observations, as for some chemical species, may be time-averaged values.

Chapter 4

Dispersion Models

4.1 Air Quality Models

Air quality simulation models (AQSM) relates emissions and air quality. Many types have been developed during the past three decades. However, two have emerged as the main types in use: (a) the Gaussian model, used in simulating dynamic plumes, and (b) the grid-based photochemical AQSM, used originally in simulating ambient ozone concentrations. More recently exploited for aerosols, SO_2, its reaction products, other reactive pollutants. The framework of the grid-based model (omitting chemistry), is also used to simulate CO concentration fields. The main premises in adopting models for use are the following:

1. They serve as accurate estimators of air quality for any selected combinations of emissions;
2. The time, cost, and staffing requirements that attend their use will be commensurate with the need, and
3. if the accuracy of estimates falls short, the model deficiencies will be correctable within resources availability.

Assuming that a suitable model is available, it may undergo a number of uses:

- Regulatory planning and analysis, such as the preparation of federal and state implementation plans (FIPs and SIPs).

- Estimation of uncertainties through sensitivity analysis.
- Planning of field studies, and
- identification of research and development needs.

4.2 Dispersion Modeling

The primary focus of dispersion modeling is estimation of primary pollutants concentrations. Strictly speaking, this modeling category applies to pollutants that do not undergo atmospheric chemical transformations. However, it also applies for pollutants for which simple assumptions are incorporated to mirror chemical transformation, such as linear decay terms. Models in use include the following:

- Gaussian formula in one of its many manifestations. This formula represents a first commonly used models. It is applied primarily to both individual and multiple plumes. It may also be applied to groups or aggregations of sources under some hypotheses. Gaussian formula can be written in a form to simulate the dispersion of individual puffs, instead of plumes.
- Another is the approximate solution of the governing equation of mass conservation, which includes a simplifying assumption that relates turbulent fluxes $< u'c' >$, to concentration gradients, $\partial c/\partial x_i$, through the adoption of an eddy diffusivity, K_i,

  $$< u'c' > = -K_i(\partial c/\partial x_i) \tag{4.1}$$

  This equation is commonly applied for more widely or uniformly distributed pollutants such as carbon monoxide (CO), where large individual plumes are not dominant.
- An approximate solution of the governing equations of mass conservation in a coordinate system that moves with the average wind velocity the so called *trajectory model*. Solutions in the fixed and moving coordinate systems are related. Acceptance of the trajectory model implies that parcel integrity is reasonably maintained through the length of time simulation.

- Governing equation of mass solution, usually in parallel with the governing equation of momentum, using more rigorous and complex procedures, hence avoiding K-theory. They are not of common use.

4.2.1 Modeling of Chemical Transformations

By far the most common approach is use of coupled mass balance equations incorporating K-theory, one for each pollutant that is being modeled. Virtually all models now in use for estimating tropospheric ozone concentrations and the concentrations of secondary fine particles are based on these equations. There are differences in the submodels or modules for one or more dynamic processes, such as transport, chemistry, and deposition, and in numerical integration procedure. These models are used for SIP and FIP preparation, regional planning, and other regulatory applications.

4.2.2 Modeling of Pollutant Deposition

Generally models, based on the governing equation of mass conservation, is used to estimate deposition fluxes as a function of location, integrating over time the accumulation of deposited material. Use of the non- reactive form of the model, incorporating simplifying assumptions, allows for calculation over longer simulated times. Less common than calculation of whole concentrations, deposition calculations are of interest for estimation of the following variable:

- Acidic deposition and acid loadings over a seasonal period.
- Ecosystem impacts of air pollutants, such as deposition of nitrogen compounds onto sensitive watersheds.
- Contributions to accumulation of pollutants in lakes and subsequent eutrophication.

4.2.3 Modeling of Adverse Impacts

The objective of modeling *impacts*, in contrast to ambient concentrations, is to examine more directly certain selected effects. Health effects of pollution are a major issue as far as adverse impacts are concerned. Visibility degradation also falls under the heading of *impacts*, as does ecosystem loading.

Again, the same category of models solution of one or more of the governing equations of mass conservation serves as the most common approach for such analyses, incorporating those modifications or additions needed to address the specific effect.

4.3 Estimating Inputs

Three major categories of information are required to formulate inputs to models:

1. air quality,
2. emissions,
3. meteorology.

Boundary and initial conditions are needed to drive models based on conservation of mass. Boundary conditions are generally difficult to estimate. Data are sparse, and often no independent means of estimation exist. Two primary approaches to estimation include acquisition of data at the inflow boundaries, both upwind and overhead, and estimation using a model of much broader spatial scale, but coarser spatial resolution. Emissions are estimated using a wide array of options, from hand-counts and bookkeeping to sophisticated modeling. Where possible, computer-based emissions models and management of emissions data are used to insure uniformity of procedure, reduce error rates, greatly enhance data handling, and increase the rate at which estimation is evalueted. A wide range of approaches to emission estimation, for the different emissions categories, might be adopted. The availability of appropriate data to derive model inputs, to evaluate model performance, and to diagnose and rectify model performance problems is crucial to the successful application of an air quality model.

4.4 Photochemical Grid Models

Photochemical grid models are mostly used for ozone simulations and require several data sets for input preparation and model evaluation: air quality, meteorological, emissions, and geophysical. Such models require a complete specification of the spatial and temporal variations of key atmospheric phenomena. Unfortunately, available data are lacking. A typical air quality data set consists of hourly surface measurements of ozone and oxides of nitrogen (NOx) from monitoring stations operated by air regulatory agencies, usually located in or immediately downwind of urban areas. Those monitoring sites located in rural are often in the general proximity of commercial or industrial sources. Very little routine NO/NOx monitoring is conducted at true rural sites, nor is there routine collection of total or speciated volatile organic compounds (VOC) data. No routine monitoring of ozone or precursors aloft is conducted. Data are rarely available for direct specification of pollutant concentrations on upwind boundaries of the modeling domain.

Photochemical grid models require a complete specification of the temporal and spatial variations of key meteorological variables, such as wind velocity, temperature, and cloud cover. These data supplemented by air monitoring stations constitute the typical meteorological database available for developing meteorological inputs to photochemical grid models. Photochemical grid models also require a complete specification of gridded, temporally resolved emissions estimates for all chemical species. An emission modeling system is required in order to organize, manipulate, and process emissions data for a large modeling domain. Geophysical data are needed for specifying gridded terrain and land use inputs.

4.5 Emission Modeling

In the context of air quality modeling, emission modeling is the process by which emissions estimates are prepared for use in an air quality model. In general terms, the emissions model is the suite of tools that are used to estimate and spatially and temporally allocate emissions for use in deterministic and statistical air quality models. Yet the emissions estimates that result from the emissions modeling process are the critical link in the air quality modeling process. The *emissions estimates model* is a computerized system that utilizes data to estimate emissions from a specific source.

The *emissions modeling system* is a computerized framework under which emissions estimates models operate.

4.6 Categories of Air Quality Models

The primary models (and modeling systems) in use today are those based on the numerical integration of the equations of conservation of mass and those based on the Gaussian formula, the latter for a range of source configurations and extensions of the basic equation.

4.6.1 Numerical Solution of Mass Conservation

The governing equations of mass conservation are:

$$\frac{\partial c_i}{\partial t} + u_x \frac{\partial c_i}{\partial x} + u_y \frac{\partial c_i}{\partial y} + u_z \frac{\partial c_i}{\partial z} = \frac{\partial}{\partial x}\left\{K_x \frac{\partial c_i}{\partial x}\right\} + \frac{\partial}{\partial y}\left\{K_y \frac{\partial c_i}{\partial y}\right\} + \frac{\partial}{\partial z}\left\{K_z \frac{\partial c_i}{\partial z}\right\} + R_i(c_1, c_2, .., c_n) + E_i(x, y, z, t) - S_i(x, y, z, t)$$

where:

- u_x, u_y, u_z = velocity;
- c_i = concentration of i^{th} species
- R_i = chemical generation rate of species i
- E_i = emissions flux
- S_i = removal flux

Emissions, meteorological, and air quality fields are provided as inputs. The equations are integrated numerically in time to produce pollutant concentration fields.

4.6.2 Gaussian Models

The fundamental formula is:

$$c(x, y, z) = \frac{q}{2\pi \bar{u} \sigma_y \sigma_z} exp\left(-\frac{y^2}{2\sigma_y^2}\right) \cdot \left[exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) + exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right)\right] \tag{4.2}$$

where:

- q = source strength
- h = stack height
- σ_y, σ_z = lateral and vertical dispersion coefficients

The formula represents a solution to the equation of mass conservation when steady state ($\partial c/\partial t = 0$), velocity $\bar{u}$ is constant, and diffusion in the x-direction can be neglected. See [45] for a full derivation.

4.7 Air quality modeling methods

Basically two model types are available to numerically simulate air pollution dispersion: Eulerian models and Lagrangian ones. The main difference is that the Eulerian reference system is fixed (with respect to the earth) while the Lagrangian reference system follows the instantaneous fluid velocity.

4.7.1 Eulerian Models

In the Eulerian approach, the observer adopts a fixed frame of reference, usually the surface of the earth. Most Eulerian models use a grid system defined in an orthogonal set of coordinates to describe atmospheric dynamics (advection and diffusion), emissions sources, and chemical production and destruction. Most numerical weather prediction models and comprehensive air quality models rely on this paradigm. Eulerian models generate four- dimensional (space and time) trace species concentration fields for each of the species modeled. Eulerian models generally use fewer simplifying assumptions in the simulation of atmospheric transport compared to other modeling techniques. By the nature of the grid discretization, Eulerian models cannot resolve trace species concentration features at sub-grid scales because emissions are instantly mixed into the grid. Although one can attempt to use very small grid size to resolve the detailed emissions distributions, there is a practical limit at which atmospheric turbulence statistics cannot be described with parameterizations in terms of the mean state variables (such as wind and temperature) as well as the inhibiting high computational cost. To compensate for this deficiency, some Eulerian models include either trajectory submodels or Gaussian dispersion submodels to treat initial transport and chemical transformations of pollutants coming from large point source emissions within the grid. These hybrid (e.g., "plume-in-grid") grid models attempt to minimize the effect of instantaneous dilution of pollutants over the entire grid box assumed by pure Eulerian models. Once the point source plumes reach a certain size, they are added to the existing concentrations in the appropriate grid cells, and subsequently go through transformation and transport processes within the grid model. Numerical diffusion in the advection process and difficulties in representing atmospheric mixing processes are some of the drawbacks of Eulerian models (see section 5).

4.7.2 Lagrangian Models

In a Lagrangian stochastic model (LSM), also called Lagrangian Particle or Random Walk model, the motion of air masses or particles passively following the flow is studied. To simulate the presence of turbulent eddies, particle velocities are subjected to a random forcing. Consequently, these models are of stochastic type. The fictitious particles (computer-particles), which represent pollutant gases or aerosols, are considered small enough to follow the motion of the smallest eddies and, at the same time, big enough to represent a large number of molecules. Each particle is moved, at each time step, by transport, due to the mean wind, and diffusion, related to the turbulent wind velocity fluctuations, without any grid. In the single particle models, the trajectory of each particle represents an individual statistical realization in a turbulent flow characterized by certain initial conditions and physical constraints. Thus the motion of any particle is independent of the other particles, and consequently the concentration field must be interpreted as an ensemble average. The Lagrangian approach is a more natural way of describing the dispersion process. It allows a high resolution, particularly in complex terrain. Moreover, due to present days computer capabilities, these models begin to be used for regulatory applications in some European Countries.

4.8 Modeling Alternatives

While grid models and Gaussian models provide a means for simulating a broad range of atmospheric processes, alternative modeling approaches may prove as or more useful in supporting particular avenues of research and analysis. For example, *box models* play a central role in air chemistry research studies. *Receptor models* provide direct emissions-air quality relationships using basic source information and measured ambient pollutant concentrations. In recognition of the stochastic character of the atmosphere, limited efforts have been devoted to developing suitable statistical models. Although each of these approaches has a limited range of applicability, they provide insight into certain aspects of air pollution phenomena and in some cases may serve to corroborate or validate results obtained from comprehensive simulation models.

- Box Models: a mathematical representation of pollutant dynamics that take place in a well-mixed volume of air.
- Receptor Models: are based on statistical analyses of ambient pollutant measurements and pertinent emissions information.
- Statistical Models: provide estimates of concentration levels as a function of space, time, meteorological, emissions and other pertinent variables.

4.9 Spatial and Temporal Scales

Models are typically applied to study impacts of individual sources, multiple-source industrial facilities, metropolitan areas, or larger regional areas up to subcontinental scale. The spatial scales range from up to few kilometers (for large industrial point sources), to hundred kilometers (for individual urban areas), to few thousand kilometers (for larger regional areas). When applying models to regional-scale domains, the spatial scale of important atmospheric phenomena that ultimately contributes to regional air quality problems must be accurately analyzed. *Nested grid* capabilities, an important feature of contemporary regional models, allow them to resolve important phenomena and concentration gradients in areas of the domain where significant sources are present. The time scales of concern are related to ambient air quality standards, which have averaging times ranging from one hour to one year. Air quality models that include a detailed treatment of chemistry tend to be limited in their applications to a few days of simulation because of the computational costs associated with the numerical integration of the chemical kinetic equations. Models that use a simplified treatment of atmospheric chemistry can be applied to longer time periods (e.g., one year or more) without prohibitive computational costs. The ability to simulate long time periods is generally obtained at the expense of some accuracy (since the treatment of chemistry is less accurate in long-term models). Another approach for estimating annual-average concentrations is to apply an episodic model for several typical meteorological scenarios and to reconstruct a full year by combining these scenarios with appropriate weighting factors. This approach involves making approximations with the representativeness of the meteorology, whereas the use of a long-term model involves making approximations with the chemistry.

4.10 Spatial and Temporal Resolution

Short-term Gaussian plume models are typically applied using hourly meteorological data spanning a period of up to five years. Such models provide hourly concentration estimates at any user-specified point downwind of the source. However, because these models are based on steady-state assumptions, they cannot truly resolve concentration fluctuations. Grid-based models provide concentration estimates that are spatially averaged over the volume of a grid cell, whose size may range from 1 to 40 km or more in the horizontal directions and from ten meters to several hundred meters in the vertical direction. Contemporary grid models exploit nested grids with relatively fine spatial resolution in dense and/or heterogeneous source areas (such as cities) and relatively coarse resolution in rural areas. Use of nested grids is largely motivated by a desire to optimize the computational time required to perform a simulation. The ability to provide variable vertical resolution can also be important. In general, relatively fine vertical resolution is used near the ground where large vertical gradients in the concentration field are likely to occur. Concentration gradients aloft are often much smaller, allowing the use of coarser vertical grid resolution. In establishing the vertical grid structure, careful consideration must be given to the spatial features of elevated stable layers aloft and the possible need to adequately resolve elevated plumes from large point sources. If such plumes are not adequately resolved, they may be subject to significant averaging errors. In addition, the timing and location of plume fumigation to the ground may be in error. For nitrogen oxides (NOx) plumes, this can have a significant effect on VOC/NOx and can also have a profound influence on the relative effectiveness of VOC versus NOx controls on ozone formation.

Chapter 5

Software

Venice encompasses a complex ecosystem where the land/lagoon/sea systems are in relation one with each other. The physical processes that arises in this context are very difficult to understand and to study.

5.1 Introduction

Venice atmosphere is affected by a plethora of emissions from both point and diffuse sources. The city is situated in the middle of a 550 km^2-wide lagoon, close to the Adriatic Sea and to a heavily populated and industrialized mainland with:

- chemical, metallurgical and oil-refinery industrial plants in Porto Marghera (almost 12 km^2);
- Glass Factories in Murano;
- a Coal Power Plant;
- a medium-size urban area with more than 270 000 inhabitants;
- several heavy traffic roads;
- a motorway;
- commercial and industrial harbours.

Venice historical and monumental importance increase the attention on this place that is a particular target for environmental studies. A new project started in 2007 at Ca' Foscari University. Different apporaches has been considered: field survey to collect data for chemical analisys and a support modeling apporach.

A first modeling apporach has been followed to estimate instruments position for field survey with the help of Ente Zona. Ente Zona is an agency whose aim is to control big Marghera's enterprises emissions. In collaboration with different public agencies Ente Zona makes remote meteo data monitoring and every year make a technical report to evaluate enterprises situation ([36]).

5.2 Modeling group work

Our modeling group implemented a modeling system that can simulate meteo profiles, as well as the dispersion of pollutants. Secondary $PM_{2.5}$ formation is the target, so a photochemical model is needed. We have chosen a free code model: CAMx. For meteorological representation we implemented CALMET, a diagnostic model that allow to interpolat data collected from filed survey. Input data collection for model system is a specific and cardinal point because data are difficult to find. This report deals with modeling system implementation.

5.3 CAMx model

To study air pollution dispersion and photochemical reactions CAMx has been implemented [12].

CAMx stands for **C**omprehensive **A**ir quality **M**odel with extensions. It is an Eulerian photochemical dispersion model that allows for an integrated one-atmosphere assessment of gaseous and particulate air pollution (ozone, PM2.5, PM10, air toxics, mercury) over many scales ranging from sub-urban to continental [12]. It is designed to unify all of the technical features required of state-of-the-science air quality models into a single system that is computationally efficient, easy to use, and publicly available.

CAMx simulates the emission, dispersion, chemical reaction, and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species l on a system of nested three-dimensional grids. The Eulerian continuity equation describes the time dependency of the average species concentration (c_i) within each grid cell volume as a sum of all of the physical and chemical processes operating on that volume. This equation is expressed mathematically in terrain-following height (z) coordinates:

$$\frac{\partial c_i}{\partial t} = -\nabla_H V_H c_i + \left[\frac{\partial (c_i \eta)}{\partial z} - c_i \frac{\partial}{\partial z}\left(\frac{\partial h}{\partial t}\right)\right] + \nabla \cdot \rho K \nabla \left(c_i/\rho\right)$$

$$+ \left.\frac{\partial c_i}{\partial t}\right|_{Chemistry} + \left.\frac{\partial c_i}{\partial t}\right|_{Emission} + \left.\frac{\partial c_i}{\partial t}\right|_{Removal}$$

where V_H is the horizontal wind vector, η is the net vertical entrainment rate, h is the layer interface height, ρ is atmospheric density, and K is the turbulent exchange (or diffusion) coefficient. The first term on the right-hand side represents horizontal advection, the second term represents net resolved vertical transport across an arbitrary space- and time-varying height grid, and the third term represents sub-grid scale turbulent diffusion. Chemistry is treated by simultaneously solving a set of reaction equations defined from specific chemical mechanisms. Pollutant removal includes both dry surface uptake (deposition) and wet scavenging by precipitation.

CAMx can perform simulations on three types of cartesian map projections: Universal Transverse Mercator, Rotated Polar Stereographic, and Lambert Conic Conformal. CAMx also offers the option of operating on a curvi-linear geodetic latitude/longitude grid system as well. Furthermore, the vertical grid structure is defined externally, so layer interface heights may be specified as any arbitrary function of space and/or time. This flexibility in defining the horizontal and vertical grid structures allows CAMx to be configured to match the grid of any meteorological model that is used to provide environmental input fields.

5.3.1 CAMx features

In addition to the attributes it shares with most photochemical grid models, some of the most specific features of CAMx are the following:

- Two-Way Nested Grid Structure.
- Flexi-Nesting.
- Multiple Photochemical and Gas Phase Chemistry Mechanism Options.
- Treatment of Particulate Matter.
- Mercury Chemistry.
- User-Defined Chemistry Mechanism.
- Chemical Kinetics Solver Options.
- Plume-in-Grid (PiG) Module.
- Horizontal Advection Solver Options.
- Advanced Photolysis Model.
- Parallel Processing.

CAMx extensions include:

- Ozone Source Apportionment Technology (OSAT).
- Particulate Source Apportionment Technology (PSAT).
- Decoupled Direct Method (DDM) for Source Sensitivity of Ozone and Other Species.
- Process Analysis (PA).
- Reactive Tracer (RTRAC) Source Apportionment.

CAMx version 4.5 includes the major updates and additional capabilities over version 4.4, [12].

Module	Physical Model	Numerical Method
Horizontal advection/diffusion	Eulerian continuity equation closed by K-theory	Bott or PPM for advection, explicit diffusion
Vertical transport/diffusion	Eulerian continuity equation closed by K-theory	Implicit advection and diffusion
Gas-Phase Chemistry	Carbon Bond IV, Carbon Bond 2005, or SAPRC99 mechanisms	ENVIRON CMC solver, IEH solver, or LSODE
Aerosol Chemistry	Dry and aqueous inorganic and organic chemistry/ thermodynamics; static 2-mode or evolving multi-section size models	RADM-AQ, ISORROPIA, SOAP, CMU sectional model
Dry deposition	Separate resistance models for gases and aerosols	Deposition velocity as surface boundary condition for vertical diffusion
Wet deposition	Separate scavenging models for gases and aerosols	Uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size

Table 5.1: Summary of the CAMx modules for key physical processes.

5.3.2 Model Formulation

This section introduces the basic numerical approach exploited in CAMx, and describes the technical formulation of the transport and removal processes.

Numerical Approach The physical representations and the numerical methods used for each term of the pollutant continuity equation are summarized in Table 5.1. CAMx includes peer-accepted algorithms and component formulations, and its modular framework permits easy substitution of additional and/or updated algorithms in the future.

The continuity equation is numerically marched forward in time over a series of time steps. At each step, the continuity equation is replaced by an operator-splitting approach that calculates the separate contribution of each major process (emission, advection, diffusion, chemistry, and removal) to concentration change within each grid cell. The specific equations that are solved individually in the operator-splitting process are shown in order below:

$$\left.\frac{\partial c_i}{\partial t}\right|_{Emission} = m^2 \frac{\partial E_i}{\partial x \partial y \partial z}$$

,

$$\left.\frac{\partial c_i}{\partial t}\right|_{Xadvection} = -\frac{m^2}{A_{yz}} \frac{\partial}{\partial x} \left(\frac{u A_{yz} c_i}{m} \right)$$

,

$$\left.\frac{\partial c_i}{\partial t}\right|_{Yadvection} = -\frac{m^2}{A_{xz}} \frac{\partial}{\partial y} \left(\frac{v A_{xz} c_i}{m} \right)$$

,

$$\left.\frac{\partial c_i}{\partial t}\right|_{Ztransport} = \frac{\partial(c_i\eta)}{\partial z} - c_i\frac{\partial}{\partial z}\left(\frac{\partial h}{\partial t}\right)$$
,

$$\left.\frac{\partial c_i}{\partial t}\right|_{Zdiffusion} = \frac{\partial}{\partial z}\left[\rho K_v \frac{\partial(c_i/\rho)}{\partial z}\right]$$
,

$$\left.\frac{\partial c_i}{\partial t}\right|_{XYdiffusion} = m\left\{\frac{\partial}{\partial x}\left[m\rho K_X \frac{\partial(c_i/\rho)}{\partial x}\right] + \frac{\partial}{\partial y}\left[m\rho K_Y \frac{\partial(c_i/\rho)}{\partial y}\right]\right\}$$
,

$$\left.\frac{\partial c_i}{\partial t}\right|_{Wetscavenging} = -\Lambda_i c_i$$
,

$$\left.\frac{\partial c_i}{\partial t}\right|_{Chemistry} = Mechanism:\ \ specific\ \ Reaction\ \ Equations$$

where c_l is species concentration (μmol/m^3 for gasses, μg/m^3 for aerosols), E_l is the local species emission rate (μmol/s for gasses, μg/s for aerosols), Δt is timestep length (s), u and v are the respective east-west (x) and north-south (y) horizontal wind components (m/s),A_{yz} and A_{xz} are cell cross-sectional areas (m_2) in the y-z and x-z planes, respectively, m is the ratio of the transformed distance on the various map projections to true distance (m=1 for curvi-linear latitude/longitude coordinates), and Δ_l is the wet scavenging rate (s_{-1}). Dry deposition is an important removal mechanism, but it is not explicitly treated as a separate process in the time-splitting approach. Instead, deposition velocities for each species are calculated based on species chemical properties and local meteorological/surface conditions, and used as the lower boundary condition for vertical diffusion. This approach appropriately couples the surface removal of pollutants through each column of cells via the vertical mixing process.

A master driving time step for the model is internally determined during the simulation for the largest and coarsest (master) grid. Time steps typically range from 5-15 minutes for grid cell sizes of 10-50 km, to a minute or less for small cell sizes of 1-2 km. As a result, nested grids require multiple driving time steps per master step depending on their grid sizes relative to the master grid spacing. Furthermore, multiple transport and chemistry time steps per driving step are used as necessary to ensure accurate solutions for these processes on all grids. The first process in each time step for a given grid is the injection of emissions from all sources. CAMx then performs horizontal advection, but alternates the order of advection in the x and y directions each master timestep. This alleviates any potential numerical biases that can develop when the x/y advection order is constant. Vertical advection is performed after horizontal advection, followed by vertical diffusion, horizontal diffusion, wet scavenging, and finally chemistry. Although advection is performed separately in the x (east-west), y (north-south), and z (vertical) directions, the numerical linkage between these components has been developed in a mass consistent fashion to preserve the density field at each time step. This maintains the flexibility to allow many types of meteorological models, and modeling grid resolutions, projections, and layer structures, to characterize transport in CAMx.

5.3.3 CAMx grid configuration

Grid Cell Arrangement

CAMx carries pollutant concentrations at the center of each grid cell volume, representing the average concentration over the entire cell. Meteorological fields are supplied to the model. CAMx internally carries these variables in an arrangement known as an Arakawa C grid configuration (Figure 5.1). State variables such as temperature, pressure, water vapor, and cloud water are located at cell center along with pollutant concentration, and represent grid cell average conditions. Wind components and diffusion coefficients are carried at cell interfaces to describe the transfer of mass in and out of each cell face. Note in Figure 5.1, for example, that horizontal wind components u and v are staggered from each other. This facilitates the solving of the transport equations in *flux form*. Depending upon the source of meteorological data, it is recommended that the user directly provide the gridded horizontal wind fields in the staggered Arakawa C configuration. However, this is not always feasible, and so CAMx offers the option for the user to supply all meteorological variables, including horizontal wind components, at cell center; in this case CAMx internally interpolates the winds to cell interfaces. Note that this leads to a slight smoothing effect on the horizontal

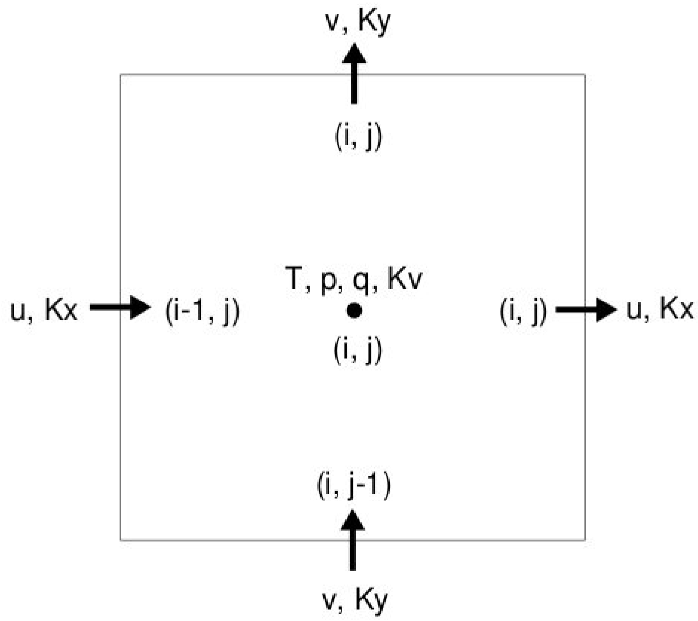

Figure 5.1: Horizontal variable configuration, [12]

wind fields.

Figure 5.1 also describes the horizontal cell indexing convention used in CAMx. Each cell is defined by the index pair (i,j), where:

- i ranges from 1 to nx (the number of cells in the east-west direction);
- j ranges from 1 to ny (the number of cells in the north-south direction).

The eastern and northern faces of the cell are indexed (i,j), while the western and southern faces are indexed (i-1,j) and (i,j-1), respectively. In the vertical, most variables are carried at each layer midpoint (defined as exactly half way between layer interfaces).

Grid Nesting

CAMx incorporates two-way grid nesting, which means that pollutant concentration information propagates into and out of all grid nests during model integration. Any number of grid nests can be specified in a single run, while grid spacings and vertical layer structures can vary from one grid nest to another. The nested grid capability of CAMx allows cost-effective application to large regions in which regional transport occurs, yet at the same time providing fine resolution to address small-scale impacts in selected

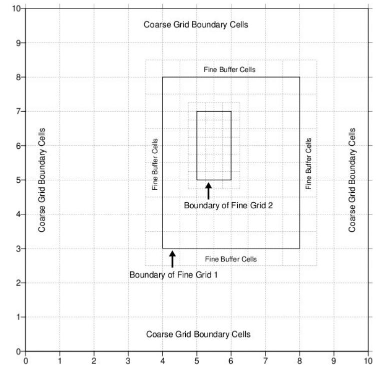

Figure 5.2: An example of horizontal grid nesting, showing two telescoping nested grids within a 10 × 10 cell master grid. The outer nest contains 1012 cells, and the inner nest contains 6 × 10 cells.

areas. Each grid nest is defined over a subset of master (coarsest) grid cells (fig 5.2. For every nested grid must be specified in the *run control file*:

- the range of master grid row and column indices that define the coverage of each nested grid;
- the integer number of nested grid cells that must span one master grid cell

Buffer cells are added around the perimeter of each nested grid to hold boundary conditions. Buffer cells are added automatically within CAMx and should not be specified by the user in the run control file. All nested grid output files contain data for the entire array of computational and buffer cells; however, buffer cell concentrations are considered *invalid* and should be ignored. Additionally, all nested grid input files must provide data for the entire array of computational and buffer cells.

Nesting in the vertical is allowed, but only by sub-dividing parent grid layers into a series of finer layers. To maximize flexibility in the vertical grid structure, each parent grid layer may be individually split into a unique set of fine layers, or not split at all. The vertical layer division is defined by the input height/pressure file for each grid. In figure 5.3 displays an example of how layers may be defined for a master grid and two fine grid nests.

Restrictions on specifying the size and resolution of all grid nests include the following:

1. the ratio of master grid cell size to nested grid cell size must be an integer number;
2. for telescoping grids (a nested grid containing an even finer grid), the cell size of the finest grid must be a common denominator for all parent grids above it;

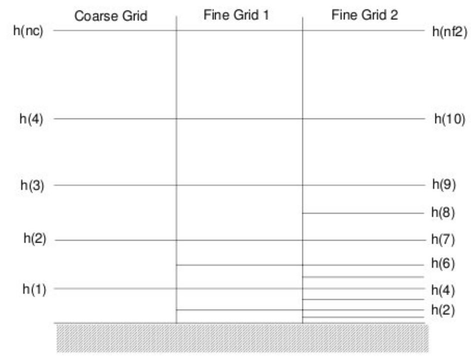

Figure 5.3: A schematic illustration of the CAMx vertical grid indexing convention and vertical nesting.

3. the restriction in (2) above does not apply to parallel nested grids of the same generation;
4. nested grids have hanging nodes;
5. nested grids cannot extend into a boundary, or non-modeled, area of the master grid;
6. CAMx is currently configured to allow four generations of nests (e.g., four levels of telescoping grids); this can be extended in the code easily if more than four levels of nests are required;
7. the total vertical depth of each nested grid must exactly match the depth of the master grid;
8. nested grid vertical layers may be individually split into several finer grid layers.

The following Fortran binary I/O files must be provided for the master grid:

- gridded surface emission;
- fractional landuse distribution;
- height/pressure (defines the layer interface structure);

- horizontal wind components;
- temperature;
- vertical diffusivity;
- water vapor;
- clouds and precipitation.

Any of these input files may be supplied for each nested grid, or none at all. If any of these files are not supplied for a particular nested grid, the *Flexi-Nest* algorithm within CAMx interpolates the missing fields from the parent grid. Clearly it is desirable to provide nested grid data whenever possible. However, the ability to interpolate data is useful for testing sensitivity to grid configurations or for situations when it is not possible to run a meteorological model for all grid nests. The any of these files are not supplied for a particular nested grid, the *Flexi-Nest* option also allows users to redefine the nested grid configuration at any point in a simulation. Nested grids can be introduced or removed only at the time of a model restart since a new CAMx user control file must be used to redefine the grid configuration. CAMx will internally reconcile the differences in grid structure between the nested grid restart files and the new user control file, and then interpolate any data fields not supplied to CAMx for the new nests from the parent grid(s).

5.3.4 Treatment of emissions

Pollutant emissions are treated in two basic ways within CAMx:

1. *low-level (gridded) emissions* that are released into the lowest (surface) layer of the model;
2. and *elevated stack-specific (point) emissions* with buoyant plume rise that can be emitted into any model layer.

During the simulation emissions are injected into each cell of every grid at every time step. Gridded and point emissions are provided to CAMx in separate input files. External emission processing systems are used to develop gridded and point, time- and space-resolved, chemically-speciated input files for CAMx.

Gridded Emissions

Two-dimensional gridded low-level emissions are defined by space- and time-varying rates for each individual gas and PM species to be modeled. Gridded emissions represent sources that emit near the surface and that are not sufficiently buoyant to reach into the upper model layers. Such emission categories include:

1. low-level stack (point) emissions that are too small to result in plume rise above the model surface layer;
2. other non-point industrial sources (fugitive leaks, tanks, etc.);
3. mobile sources (cars, trucks, non-road vehicles, railroad, marine, aircraft, etc.);
4. residential sources (heating, cooking, consumer products);
5. commercial sources (bakeries, refueling stations, dry cleaners);
6. biogenic sources;
7. natural sources (small fires, wind-blown dust).

The spatial distribution of each individual source within these categories is defined by the modeling grid. Information such as population distribution, housing density, roadway networks, vegetative cover, etc. is typically used as a surrogate to distribute regional emission estimates for each source to the grid system. Processing tools are used to combine emissions from all sources into a single input file for each grid.

Elevated Point Emissions

Similarly to gridded emissions, elevated point emissions are defined by space- and time-varying rates for each individual gas and PM species to be modeled. The only difference is that these sources emit from individual stacks with buoyant rise that may take them into upper model layers. These types of sources are almost always associated with large industrial processes, such as electric generators, smelters, refineries, large factories, etc.

The spatial distribution of these points is specifically given by the coordinates of the stacks themselves (grid locations are determined within CAMx). Plume rise is determined within CAMx as a function of stack parameters (height, diameter, exit velocity and temperature) and ambient meteorological conditions, so the point source file only provides speciated time-resolved emission rates and stack parameters for each individual source. A single point source file provides the definition of all stacks and their emissions over the entire modeling domain. Plume rise is calculated using the multi-layer stability-dependent algorithm. This approach calculates the momentum and buoyant plume rise energy from the stack, takes the larger of these two values, and determines the dissipation of that energy via mixing with ambient air according to the meteorological conditions through the host model layer. If sufficient energy remains to reach into the next model layer, the calculation for buoyant rise repeats for the meteorological conditions of that layer, and so on, until a layer is found where the plume cannot rise any farther. All emissions from this source are then injected into the grid cell directly above the stack at this layer height. This algorithm was adopted for CAMx because it provides a more realistic handling of stable layers aloft that can trap plume rise, whereas this effect would not be realized based on meteorological conditions at stack top alone.

5.3.5 Transport Fundamentals

CAMx transport algorithm is both mass conservative and mass consistent. Mass conservation refers to the ability to accurately account for all sources and sinks of mass in the model, with no spurious loss or gain of mass during model integration. To be mass conservative, CAMx internally carries concentrations of each species as a density ($\mu mol/m_3$ for gases, $\mu g/m_3$ for aerosols), and solves the advection equations in flux form. This also serves to simplify mass budget accounting, which is used by the various source apportionment and process analysis options.

Gas concentrations are internally converted to volumetric mixing ratio (parts per million, or ppm) for the chemistry and diffusion steps, and when they are written to the average output files. Mass consistency refers to the models ability to transport pollutant mass exactly equivalent to the input atmospheric momentum field. For example, a model that is perfectly mass consistent will preserve a unity pollutant mixing ratio field given constant unity boundary and initial conditions and zero sources and sinks.

Sources of poor mass consistency in air quality models are typically related to the following (in order of importance):

1. supplying meteorology that is inherently inconsistent

2. spatially interpolating or averaging meteorological model fields, especially three-dimensional wind vector fields, to an air quality model grid of different resolution and/or different mapping projection;

3. relying on a nested grid model to internally interpolate coarse grid meteorological parameters to finer grids, especially in cases of varying vertical resolution among the grids;

4. exploiting numerical techniques within the air quality model that are mass inconsistent;

5. exploiting different numerical and/or physical methods in the air quality and meteorological models.

CAMx operates on the map projections and grid systems exploited in several widely used meteorological models (e.g., MM5, RAMS, and WRF) so that translation of meteorological data to CAMx requires as little manipulation as possible. To account for all sources of error while maintaining flexibility on input requirements and grid configuration, CAMx internally minimizes the sources of mass inconsistency as the model integrates forward in time. As discussed below, the model does this in several ways.

1. the transport equations are written and solved in flux form. Gridded meteorological inputs are carried in an Arakawa C arrangement (5.1), which optimizes the calculation of mass flux divergences while ensuring mass conservation;

2. CAMx can accept input meteorological fields (horizontal wind components, pressure, temperature, moisture, vertical diffusion coefficients, cloud parameters) for each individual nested grid, if available from a meteorological model. This reduces errors stemming from internal interpolations from parent to nested grids. Input gridded fields of layer heights, temperature, pressure, horizontal winds, water vapor, and vertical diffusivity are then interpolated in time to the unique time steps for each modeling grid.

3. the grid- and timestep-specific horizontal momentum fields are used to determine a vertical velocity field that balances the local atmospheric continuity equation for the specific grid configuration exploited. Since the vertical grid structure is defined via external inputs, layer interface

heights may be specified as any arbitrary function of space and/or time. This allows the CAMx vertical grid system to exactly match all or a subset of any meteorological model layer structures, whether they are defined in terms of physical height above ground, normalized height coordinates (i.e., *sigma-z*), or normalized pressure coordinates (i.e., *sigma-p*). Therefore, total vertical transport is the combination of resolved vertical advection and mass exchange across undulating layer interfaces.

Transport algorithms Horizontal advection is performed using the area preserving flux-form advection solver or the *Piecewise Parabolic Method* (PPM). These two finite difference schemes were incorporated into CAMx because they provide higher order accuracy with minimal numerical diffusion, yet are equivalent in execution speed compared to other simpler advection algorithms when operating on equivalent time steps. In CAMx, the Bott scheme is allowed to take larger time steps than PPM because Bott remains stable for *Courant-Friedrichs-Levy* (CFL) numbers up to 1 (i.e., the ratio of wind speed to grid spacing per timestep). Time steps are determined for Bott using a CFL number of 0.9, while time steps for PPM are restrained by a CFL number of 0.5. Therefore, the Bott option results in a faster simulation than the PPM option, perhaps at the price of some accuracy.

The Bott and PPM algorithms have been shown to produce significant differences in concentration predictions compared to previously used flux-corrective schemes algorithm. This is a result of the considerably lower amount of numerical diffusion associated with Bott and PPM. The latest advection solvers reduce numerical diffusion to the point where modelers need to be concerned about including appropriate levels of explicit horizontal diffusion. Currently, there is very little information on the appropriate level of horizontal diffusion for Eulerian grid models. We see this issue becoming increasingly important as more accurate advection solvers are utilized in grid models.

Explicit horizontal diffusion coefficients are determined within CAMx using a deformation approach:

$$K_{X/Y} = K_0 + \frac{\Delta x \Delta y}{4\sqrt{2}} \left[\left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right)^2 + \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right)^2 \right]^{\frac{1}{2}}$$

Separate diffusivity components are generated for fluxes in the x- and y-directions since K_X and K_Y are calculated for separate cell faces in the Arakawa C grid arrangement. The value of K_0 is specified according to the approach in MM5:

$$K_0 = 3 \times 10_{-3} \frac{\Delta x \Delta y}{\Delta t}$$

A maximum value of K is set to maintain numerical stability for the given grid-specific timestep. Horizontal diffusion is applied using an explicit simultaneous two-dimensional flux-divergence calculation. Vertical transport and diffusion are both solved using implicit integration approaches. Since implicit schemes are absolutely stable, only one solution step is necessary per driving time step. Explicit approaches require potentially many sub-steps (on the order of 10-100) to maintain a stable solution, which introduces the potential for excessive numerical diffusion. Whereas the vertical advection step is implicitly solved in a single step, the vertical diffusion is implicitly solved over potentially several sub-steps, depending upon the magnitude of the vertical diffusivity relative to the vertical layer depths.

Gridded vertical diffusion coefficients must be supplied to the model for the master grid via input file; the user may optionally supply vertical diffusion coefficients for any or all nested grids.

Pollutant Removal Trace gases and particles are removed from the atmosphere via deposition to the ground. Dry deposition refers to the direct sedimentation and/or diffusion of material to various terrestrial surfaces and uptake into biota. Wet deposition refers to the uptake of material into cloud water and precipitation, and its subsequent transfer to the surface. The efficiency with which wet and dry deposition processes can remove pollutants from the air depends upon the physical and chemical properties of the pollutants, local meteorological conditions, the type of surface on which they are being deposited, and on the frequency, duration, and intensity of precipitation events.

Wet Deposition

Wet deposition is the predominant removal process for particles. Particles act as cloud condensation nuclei; the cloud droplets grow and collect into sufficiently large sizes to fall as precipitation. Particles that are entrained into the cloud, and that exist below precipitating clouds, can also be directly scavenged by precipitation via accretion and impaction processes. The rates of nucleation and impaction depend upon cloud type (e.g., prolonged stratiform vs. vigorous convective development), precipitation rate, and particle size distribution. Wet deposition can also be an important removal process for relatively soluble gaseous pollutants and this occurs through the following series of steps:

- mixing of trace gas and condensed water in common air space;
- absorption of gas molecules by water droplets;
- possible aqueous-phase reactions of the pollutant within water droplets;
- precipitation of droplets to the earths surface;
- diffusion of ambient gases into falling precipitation.

It is important to note that each of the above steps may be reversible, so that the overall wet deposition rate for gases depends on the net results of the forward and backward processes at each step. The rate at which the first and fourth steps proceed depends on the frequency and nature of cloud formation and precipitation events. The rates at which the second, third, and fifth steps proceed depend on the extent to which the pollutant in question dissolves in water and its overall reaction rate once in solution. Cloud water droplets can absorb gases from the air up the limit of their solubility in water. For many pollutants this solubility far exceeds the amount of pollutant present in the air so that the distribution of the pollutant between the air and water droplets is determined by the *Henry's Law constant*, which is defined as the ratio of pollutant concentrations in the liquid-phase to the gas-phase at equilibrium.

- high values for the Henry's law constant (greater than about 10,000 M/atm) indicate a strong tendency to dissolve in water droplets;
- low values (less than about 100 M/atm) indicate a tendency to remain in the air ([45]).

Equilibrium between concentrations of pollutants in the air and in water droplets is usually established on time scales of minutes, so equilibrium conditions can generally be assumed to exist in the atmosphere. If a pollutant partitions significantly into water droplets it can be removed by precipitation. Averaged globally, highly soluble gases that are removed efficiently by wet deposition are estimated to have lifetimes due to rainout of about 5 days (Warneck, 1989). Clearly, on regional or local scales, lifetimes against wet deposition will depend upon the frequency and intensity of rainout events. Gases that are removed less efficiently by wet deposition will have longer lifetimes.

The CAMx wet deposition algorithm was improved with the introduction of the full-science PM chemistry package in version 4.00. The basic model implemented in CAMx is a scavenging approach in which the local rate of concentration change $\partial c/\partial t$ within or below a precipitating cloud depends on a scavenging coefficient Λ:

$$\frac{\partial c}{\partial t} = \Lambda c$$

The scavenging coefficient is determined differently for gases and particles, based upon relationships described by Seinfeld and Pandis (1998). Two components are calculated for gases: (1) direct diffusive uptake of ambient gases into falling precipitation; and (2) accretion of cloud droplets that contain dissolved gases.

Two components are also determined for particles:

1. impaction of ambient particles into falling precipitation with an efficiency that is dependent upon particle size;

2. accretion of cloud droplets that contain particle mass. Each of these processes are described below.

The external environmental inputs to the CAMx wet deposition algorithm include the three-dimensional gridded distribution of cloud and precipitation water contents, with the precipitation contents broken down into liquid, snow, and ice (graupel).

Scavenging rate equations were derived in terms of equivalent liquid precipitation rates, so the input precipitation water contents are internally translated into this metric. The following general assumptions are made in the CAMx scavenging model:

1. rain drops, snow flakes, and graupel particles are each separately represented by a single mean size, mass, and fall speed, which are determined from equivalent liquid precipitation rate;

2. there is no mixed-phased precipitation within a given grid cell the dividing line between liquid rainfall and the two frozen forms is 273 K;

3. snow is only associated with stratiform precipitation, and graupel only with convective precipitation;

4. liquid cloud water is allowed to exist below 273 K a linear ramp function is applied to apportion total cloud water into liquid form between 243-273 K (all cloud water is assumed to be in ice crystal form below 243 K);

5. all gases can directly diffuse into or from liquid rainfall (only strong acids can diffuse into frozen precipitation) at a rate according to the precipitations state of saturation, pollutant diffusivity, and aerodynamic considerations;

6. all gases can dissolve into liquid cloud water, which can be scavenged by all precipitation forms dissolved gasses are in equilibrium with ambient concentrations according to *Henry's Law*;

7. PM is irreversibly scavenged directly by all precipitation forms via impaction, and by uptake into cloud water (liquid and ice) as condensation nuclei that is itself scavenged by all precipitation forms;

8. all in-cloud PM mass exists in cloud water (i.e., no dry aerosols exist in the interstitial air between cloud droplets) all PM species and sizes are hygroscopic and internally mixed.

Dry Deposition

For many compounds, dry deposition can be as important as wet deposition as a removal process. Due to the difficulty of making direct measurements of dry deposition and the need for a suitable model parameterization, dry deposition is often treated as a first-order removal mechanism. The flux of a pollutant to the surface is the product of a characteristic deposition velocity and its concentration in the *surface layer* (i.e., the lowest model layer). Deposition velocities are derived from models that account for the reactivity, solubility, and diffusivity of gases, the sizes of particles, local meteorological conditions, and season-dependent surface characteristics.

For a given species, particle size, and grid cell, CAMx determines a deposition velocity for each landuse type in that cell and then linearly combines them according to the fractional distribution of landuse. The deposition flux is used as the lower boundary condition in the vertical diffusion algorithm. Aerosol size spectra and species-dependent properties needed for the deposition velocity calculations are externally supplied to CAMx for all pollutant species via the chemistry parameters file; gridded landuse is supplied to the master grid and optionally any nested fine grids; the season is determined by the simulation date and location on the globe. Movement of material along a path from the atmosphere, through any plant canopy, and onto the various plant and ground surfaces within and below the canopy is typically modeled by analogy to an electrical circuit. Resistances in serial and parallel arrangements are used to represent the relative ease with which material moves through different portions of the deposition pathway. Each branch of the circuit represents a different path by which material may be deposited. For example, gaseous pollutants may transfer through the lowest layers of the atmosphere partially into a plant canopy, through the stomatal openings on plant leaves and into the plant mesophyll tissue. Alternatively, the material may travel all the way through the plant canopy and deposit on the ground surface.

5.4 CALMET

CALMET [44] is a meteorological model which includes a diagnostic wind field generator containing objective analysis and parameterized treatments of slope flows, kinematic terrain effects, terrain blocking effects, and a divergence minimization procedure, and a micro-meteorological model for overland and overwater boundary layers.

CALMET reads hourly surface observations of wind speed, wind direction, temperature, cloud cover, ceiling height, surface pressure, relative humidity, and precipitation type codes (optional, used only if wet removal is to be modeled). Input files needed are the following:

- **Surface Meteorological Data** that are hourly observations of:
  - wind speed,
  - wind direction,
  - temperature,

  - cloud cover,
  - ceiling height,
  - surface pressure,
  - relative humidity.

- **Upper Air Data**:
  - wind speed,
  - wind direction,
  - temperature,
  - pressure,
  - elevation.

- **Overwater Observations** (optional):
  - air-sea temperature difference,
  - air temperature,
  - relative humidity,
  - overwater mixing height,
  - wind speed,
  - wind direction,
  - overwater temperature gradients above and below mixing height.

- **Geophysical Data**,gridded fields of:
  - terrain elevations
  - land use categories
  - surface roughness length (optional)
  - albedo (optional)
  - Bowen ratio (optional)
  - soil heat flux constant (optional)
  - anthropogenic heat flux (optional)
  - vegetative leaf area index (optional)

Missing values of temperature, cloud cover, ceiling height, surface pressure, and relative humidity at surface stations are allowed by the program. The missing values are internally replaced by values at the closest station with non-missing data. However, one valid value of each parameter must be available from at least one station for each hour of the run.

5.4.1 Grid System

CALMET model uses a grid system consisting of NZ layers of NX by NY square horizontal grid cells. *Grid point* refers to the center of the grid cell in both the horizontal and vertical dimensions. The *cell face* refers to either the horizontal or vertical boundary between two adjacent cells. Horizontal wind components (u and v) are defined at each grid point. The vertical wind component (w) is defined at the vertical cell faces. It is assumed that the orientation of the X and Y axes of the CALMET grid are west-east and south-north, respectively. The grid system is compatible with the usual definition of the u and v horizontal wind components as the easterly and northerly components of the wind, respectively. One commonly used grid system compatible with CALMET is the Universal Transverse Mercator (UTM) Grid.

CALMET model operates in a terrain-following vertical coordinate system.

$$Z = z - h_t \tag{5.1}$$

where:

- Z is the terrain-following vertical coordinate (m),
- z is the Cartesian vertical coordinate (m), and
- h_t is the terrain height (m).

The vertical velocity, W, in the terrain-following coordinate system is defined as:

$$W = w - u\frac{\partial h_t}{\partial x} - v\frac{\partial h_t}{\partial y} \tag{5.2}$$

where:

- w is the physical vertical wind component (m/s) in Cartesian coordinates, and
- u,v are the horizontal wind components (m/s).

Glossary

Aerosol: is a suspension of fine solid particles or liquid droplets in a gas.

CORINAIR: CORe INventory of AIR emissions. CORINAIR was a project performed since 1995 by the then European Topic Centre on Air Emissions under contract to the European Environment Agency. The aim was to collect, maintain, manage and publish information on emissions into the air, by means of a European air emission inventory and database system. This concerns air emissions from all sources relevant to the environmental problems of climate change, acidification, eutrophication, tropospheric ozone, air quality and dispersion of hazardous substances. Before 1995 the CORINAIR project was developed under the CORINE programme of the EU (CO-oRdination dINformation Environnementale, a programme established by Council Decision 85/338/EEC).

EMEP: the Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe, linked to the Convention on Long-range Transboundary Air Pollution (see LRTAP). The main objective of the EMEP programme is to regularly provide Governments and subsidiary bodies under the LRTAP Convention with qualified scientific information to support the development and further evaluation of the international protocols on emission reductions negotiated within the LRTAP Convention.

Emission rate: of a pollutant is the product of the measured pollutant concentration and the measured effluent flow rate.

EPA: U.S. Environmental Protection Agency, (www.epa.gov).

FIP: Federal Implementation Plan.

Plume Rise: the height to which a plume emitted from an elevated source will rise.

PM: Particulate matter, alternatively referred to as particulates or fine particles, are tiny particles of solid or liquid suspended in a gas. The notation itemPM_{10} is used to describe particles of 10 micrometers or less and

$PM_{2.5}$ represents particles less than 2.5 micrometers in aerodynamic diameter.

Puff: dispersion of non-continuous air pollution plumes.

SIP: State Implementation Plan.

SNAP: Selected Nomenclature for sources of Air Pollution - developed as part of the CORINAIR project for distinguishing emission source sectors, sub-sectors and activities.

VOC: Volatile Organic Compounds, are organic chemical compounds that have high enough vapor pressures under normal conditions to significantly vaporize and enter the atmosphere.

Glossary of Pollutants for which reporting is requested or encouraged under the 2002 UNECE/EMEP Reporting Guidelines.

Main pollutants: NO_X: Oxides of Nitrogen nitric oxide and nitrogen dioxide, expressed as nitrogen dioxide (NO_2); *CO*: Carbon Monoxide; *NMVOCs*: Non-Methane Volatile Organic Compounds; SO_X: Sulphur oxides means all sulphur compounds, expressed as sulphur dioxide (SO_2); NH_3: Ammonia

Particulate Matter: $PM_{2.5}$: Ultra-fine particles, having diameter of 2.5 μm or less; PM_{10}: Fine particles, having diameter of 10 μm or less; *TSP*: Total suspended particulate matter.

Priority Heavy Metals: *Cd*: Cadmium; *Hg*: Mercury; *Pbv*: Lead.

Persistent Organic Pollutants (POPs)- Annex I: *Aldrin; Chlordane; Chlordecone; Dieldrin; Endrin; Heptachlor; Hexabromo-biphenyl; Mirex; Toxaphene.*

Persistent Organic Pollutants (POPs)- Annex II: *HCH*: Hexachlorohexane; *DDT*: Dichloro-Diphenyl-Trichloroethane; *PCBs*: Polychlorinated biphenyls.

Persistent Organic Pollutants (POPs)- Annex III: *DIOX*: Dioxins and furans - polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans; *PAHs*: Polycyclic Aromatic Hydrocarbons. Following the POPs Protocol, the following indicator compounds should be used for PAHs: benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-cd] pyrene.]; *HCB*: Hexachlorobenzene.

Additional Reporting. Other Heavy Metals: *As*: Arsenic; *Cr*: Chromium; *Cu*: Copper; *Ni*: Nickel; *Se*: Selenium; *Zn*: Zinc.

Additional Reporting. Other POPs: *PCP*: Polychlorinated phenols; *SCCP*: Short-chained chlorinated paraffins.

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