

13 MODELING OF ADVERSE AIR QUALITY EFFECTS

Often the real goal of an air pollution study is not only to evaluate the concentration field of atmospheric pollutants, but also to quantify their adverse effects. In other words, often one needs to quantify a relation

$$E = f(\mathbf{c}) \quad (13-1)$$

where E represents a quantitative description of an adverse effect (e.g., the reduction in atmospheric visual range) and the vector \mathbf{c} represents the ambient pollutant concentrations. The relation f can vary from a simple semiempirical formulation, based on empirical data, to a complex simulation mechanism of the adverse effects. As an example of a simple relation, long-term ambient concentrations of carcinogenic pollutants measured in a certain region R can be empirically related to the number of extra cancers that they are expected to generate in the next (say) 30 years, in the population P living in the region R . As an example of a complex relation, mathematical models of the human body's respiratory system can be used to assess the short-term and long-term effects of a predefined pattern of exposure to toxic pollutants.

Adverse effects of atmospheric pollutants have been studied by several authors. A full volume (Volume II, Stern, 1977) of the Stern air pollution series is dedicated to this topic. More recently, Volume VI (Stern, 1986), which is a supplement of Stern (1977), integrates Volume II with additional discussion of 1) physical and economic systems, 2) vegetation, 3) acidic deposition on aquatic ecosystems, and 4) human health.

Adverse effects can be divided into

1. short-term and long-term ecological damage to
 - human health (e.g., see Lipfert (1985) for a discussion of a possible connection between mortality and air pollution)
 - animals
 - plants (e.g., see the critical review by McLaughlin (1985) on the effects on air pollution of forests)

2. damage to human "welfare," such as
 - atmospheric visibility impairment
 - odors (e.g., see Poostchi et al. (1986) for a comparison of models used for the determination of odor thresholds)
 - undesired changes in local weather
3. economical damages to
 - materials
 - structures
 - real estate values
 - artistic heritage (e.g., see Tombach (1982) for a discussion of climatological and air pollution factors affecting stone decay)
4. global effects due to
 - CO_2 accumulation
 - stratospheric ozone depletion
 - nuclear winter scenarios

In this chapter, we will discuss the use of mathematical models in some of the above fields, particularly

1. atmospheric visibility
2. CO_2 accumulation and the "greenhouse" effect
3. depletion of stratospheric ozone
4. nuclear winter scenarios

13.1 VISIBILITY IMPAIRMENT

Visibility impairment and its modeling is a significant issue in the U.S., where visibility has been recognized as an important aesthetic value to be preserved, especially in high scenery regions such as the national parks.

Two major topics have been addressed by visibility modeling techniques:

1. plume visibility, models of which simulate the visual effects of a single plume

2. regional haze, models of which address the visibility impairment (mainly a reduction of visual range) caused by large air masses containing high concentrations of fine particles.

13.1.1 Plume Visibility

The objective of a plume visibility model is the evaluation of a plume's impact on human vision in a certain region. This is achieved by a series of simulation modules, as illustrated in Figure 13-1, in which, starting from the plume's emission data, results regarding the plume's appearance are obtained.

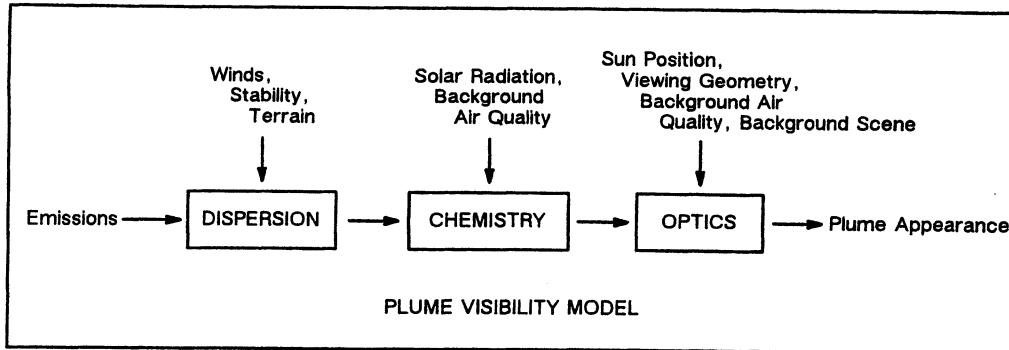


Figure 13-1. The organization of modules in a plume visibility model (from White, 1984). [Reprinted with permission from the American Petroleum Institute.]

Figure 13-2 is a more detailed description of the computational modules of a plume visibility model. The main simulation processes are

1. plume dispersion by atmospheric turbulence
2. plume chemistry, in which NO is converted to NO_2 and SO_2 , NO_2 and organic gases are oxidized into SO_4^{2-} , NO_3^- and organic particles, respectively
3. particle growth
4. atmospheric optics
5. human perceptual effects of visibility impairment

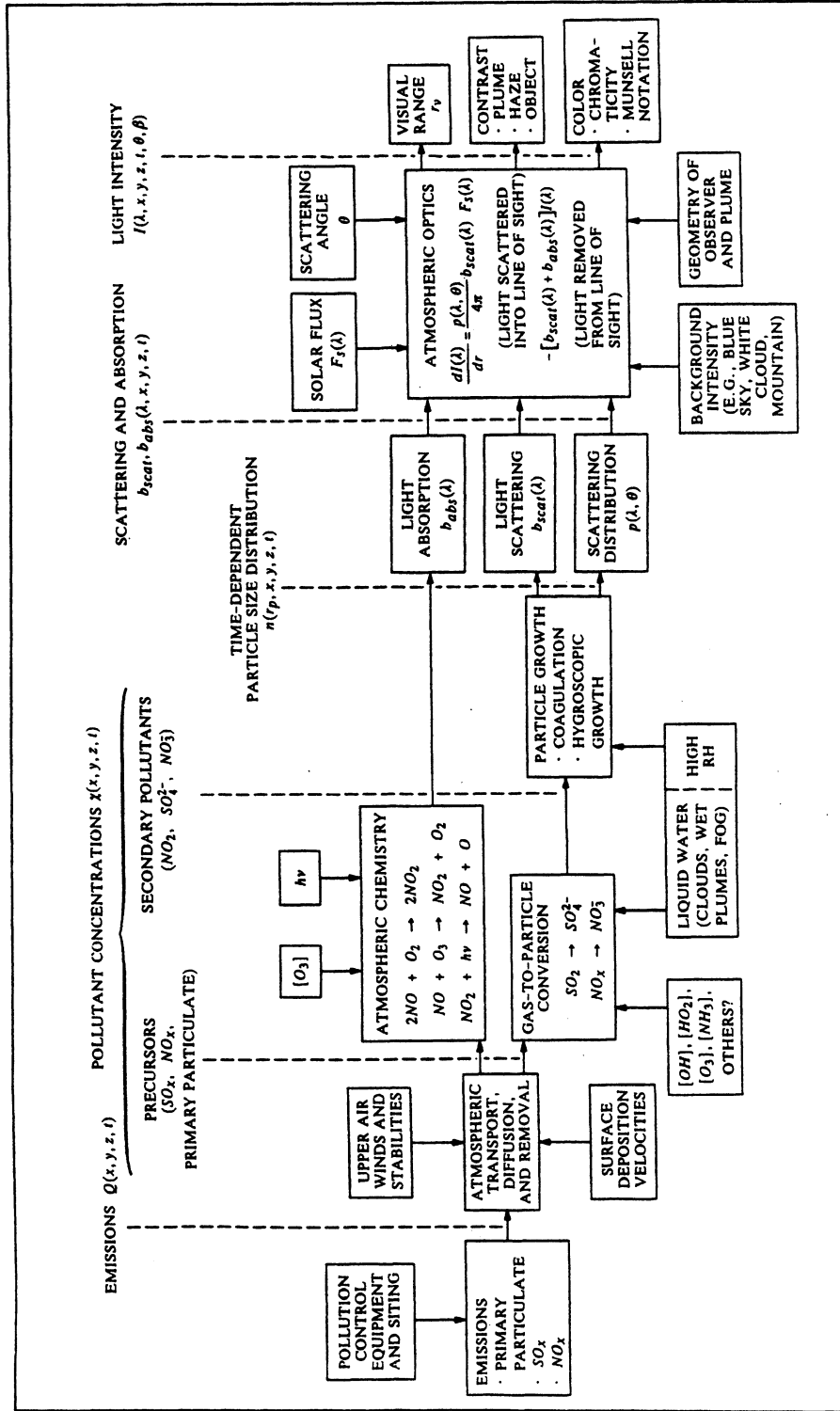


Figure 13-2. Schematic logic flow diagram of the visibility models (from Latimer et al., 1980). [Reprinted with permission from the American Meteorological Society.]

Four visibility packages, among others, are available for plume visibility simulations (White, 1984):

1. the ERT Visibility Model, Version 3 (Drivas et al., 1981)
2. the Los Alamos Visibility Model (Williams et al., 1980 and 1981)
3. the PHOENIX model (Eltgroth and Hobbs, 1979)
4. the PLUVUE Model (Latimer and Samuelsen, 1978; Latimer et al., 1980^(*))

An intercomparison study (White, 1984) tested the performance of these four models against field data collected by the VISTTA measurement program. The U.S. EPA and the Salt River Project (SRP) cosponsored the VISTTA project to collect suitable data near the SRP's Navajo Generation Station in northern Arizona in order to evaluate plume visibility models. According to White (1984), "none of the models is successful in predicting observed variation in plume dispersion or concentration parameters," and "much of the observed variability appears due to short-term fluctuations in source emissions and atmospheric transport." The interesting conclusion of this preliminary intercomparison study points out that future model development should focus on improving the parameterization of plume dispersion which, in the available models, appears to be the major component presently limiting model performance.

13.1.2 Regional Haze

Plume visibility impairment is not the only aspect of the air quality-visibility issue. Especially in the eastern U.S., visibility is impaired by regional haze much more than by single plumes. Regional haze is due to both natural causes (e.g., high relative humidity) and the presence of fine particulate matter (particles $\leq 2.5 \mu\text{m}$ in aerodynamic diameter).

One of the basic assumptions of current (e.g., Latimer and Hogo, 1987) regional haze studies is that the visual range r_v can be computed using the Koschmieder relationship

$$r_v = K/b_t \quad (13-2)$$

where K is the Koschmieder constant, initially estimated as 3.9, but re-evaluated as 3.0 for air quality applications (Tombach and Allard, 1983). The parameter b_t

^(*) A revision of the PLUVUE model, called PLUVUE II, is currently available (Latimer and Ireson, 1988).

is the total light extinction coefficient, which is assumed to be an additive function of each atmospheric contributor to light scattering and absorption. In many cases, it is assumed that

$$b_t = b_s + b_{ns} \quad (13-3)$$

where b_s is the fraction of b_t due to the sulfates and b_{ns} combines the contribution of all other components (nitrates, organics, NO_2 , etc.). The fraction b_s can be related to the average SO_4^{2-} concentration by

$$b_s = \text{const } c_{SO_4} f(RH) \quad (13-4)$$

where c_{SO_4} is the sulfate concentration and $f(RH)$ is a function of the relative humidity RH (e.g., as in SAI, 1984).

One of the major issues in regional haze modeling is the evaluation of the visual range improvements associated with SO_2 emission reductions (e.g., a 50 percent SO_2 emission reduction has often been proposed in the eastern United States). Several authors have addressed this problem in both episodic summertime studies (Ferman et al., 1981; Stevens et al., 1984; Weiss et al., 1982) and annual average calculations (Latimer and Hogo, 1987; Zannetti et al., 1988). Such quantifications, however, are difficult to perform for several reasons: 1) the large uncertainties that even advanced models possess in simulating long-range transport, diffusion, chemistry and deposition of atmospheric sulfur; 2) the difficulty in quantifying the roles that other components, such as non-sulfate-containing fine particles, coarse particles and gases, play in visibility impairment; and 3) the scarcity of suitable field data, since most measurement studies are conducted during episodic conditions, while annual average assessments require input data that represent other conditions as well as episodic.

The effect of SO_2 emission reduction on visual range is complicated by four major factors that cause nonlinear behavior; they are discussed below:

1. Atmospheric light extinction is caused by the concentration of fine particles and other airborne components. SO_2 emission controls will largely affect only the fraction of light extinction that is due to fine particles. (Actually, SO_2 controls will also affect sulfate-containing coarse particles, but, in general, with negligible associated visibility improvements.)
2. The fine particle aerosol is composed of sulfate-containing particles and of particles containing other species (but no sulfates).

SO_2 emission controls will affect only the fraction of the fine particles that contains sulfates. (Actually, SO_2 controls may increase the concentration of nonsulfate particles such as nitrate and chloride; see Pilinis (1989).)

3. Fine sulfur-containing particles are a fraction of the total concentration of sulfur in the atmosphere in both gaseous and particulate form. They are produced mostly by SO_2 -to- SO_4^{2-} chemical transformations that appear to be nonlinear. Therefore, although SO_2 emission controls will decrease proportionately the total ambient sulfur along the trajectories of plumes from controlled regions, the fine sulfates may decrease to a lesser extent because of the nonlinear chemistry.
4. Total ambient sulfur in one geographical area is due to both local emissions and sulfur transported from other regions. SO_2 emission controls will affect only the fraction of sulfur that is transported from the regions affected by the control scenario.

Because of these four factors, the percent of visibility improvement (e.g., the percentage improvement of visual range) is expected to be much less than the percent reduction of SO_2 emissions.

Another complication is caused by the role of water adsorbed by both sulfate and nonsulfate particles. This adsorbed water affects visibility by increasing the mass of fine particles and, consequently, the atmospheric light extinction. However, the water mass is generally unknown, since fine particles are measured at a low relative humidity RH_o (e.g., $RH_o \approx 0.4$), where most of the water is removed from the particles collected on the ambient sampling filters.

Since fine particle concentrations (F) are the sum of sulfate (SO_4)(*) and nonsulfate (NS) species, a simple mechanism for evaluating the role of adsorbed water (Cass, 1979; Tang et al., 1981; Appel et al., 1985) is given by

$$F^{(w)} = K_s SO_4 + K_{ns} NS \quad (13-5)$$

(*) Here we use SO_4 instead of SO_4^{2-} to indicate that the entire sulfate particle (i.e., NH_4HSO_4 or $(NH_4)_2SO_4$, not just the anion SO_4^{2-} , must be accounted for in computing the fine particle concentration. This distinction is important since most aerosol measurements provide the sulfate SO_4^{2-} concentration only.

where $F^{(w)}$ is the concentration of fine particles including adsorbed water. K_s (a term greater than one) represents a suitable “amplification” of the SO_4 concentration to allow for the adsorbed water and, in a similar way, K_{ns} represents the increase for the concentration NS of nonsulfate particles, where

$$K_s = \left(\frac{1 - RH_o}{1 - RH} \right)^{\beta_s} \quad (13-6)$$

and

$$K_{ns} = \left[h_{ns} \left(\frac{1 - RH_o}{1 - RH} \right)^{\beta_{ns}} + (1 - h_{ns}) \right] \quad (13-7)$$

In the formulas above, RH is the ambient relative humidity, h_{ns} is the fraction of NS that is hygroscopic, and β_s, β_{ns} are exponents that need to be evaluated ($\beta = 1$ or 2 have often been chosen). More complex modeling techniques can be used to calculate more precise values of K_s and K_{ns} . For example, Pilinis and Seinfeld (1987) developed and tested a computer code that performs a chemical equilibrium calculation in the sulfate, nitrate, chloride, sodium, ammonium and water system. This code successfully predicted the concentrations of various aerosol species at Long Beach, California. Its application, however, requires detailed air quality and meteorological information that is often unavailable on a regional and annual average basis. Hence, semiempirical relations such as Equations 13-6 and 13-7 must, at least at the present time, be used.

13.2 CO₂ ACCUMULATION AND THE “GREENHOUSE” EFFECT

More carbon dioxide (CO_2) is emitted by anthropogenic processes than any other substance. CO_2 does not show adverse effects to the earth ecology. Several studies, however, point out that CO_2 strongly adsorbs electromagnetic radiation at a wavelength of about $15 \mu\text{m}$, which corresponds to the maximum intensity of the earth radiation (the atmosphere and the CO_2 are transparent to the sun’s radiation, which heats the earth’s surface, but about 30 percent of the sun’s radiation is reflected into space). Therefore, an increase of CO_2 in the global atmosphere can trigger a general increase of the earth’s temperature, an effect that has been called the “greenhouse” effect.

CO₂ measurements indeed show a constant accumulation pattern. Current CO₂ concentrations (above 340 ppm) tend to increase by 1 ppm per year (see Figure 13-3). With the continuous industrial and urban development expected in the next decades, CO₂ concentrations could easily reach, in the next century, values two to three times the current ones (i.e., between 600 and 1,000 ppm).

Several models forecast the meteorological consequences of these possible CO₂ increases. Manabe and Wetherald (1975) predict that a CO₂ concentration of 600 ppm should increase the average temperature of the earth of about 2-3°C with maxima of about 7-10°C at high latitudes. The consequent (Mercer, 1978) melting of the polar glaciers would cause a 5 m increase of the mean sea level. Coastal cities and large sections of Holland and Florida would be covered by

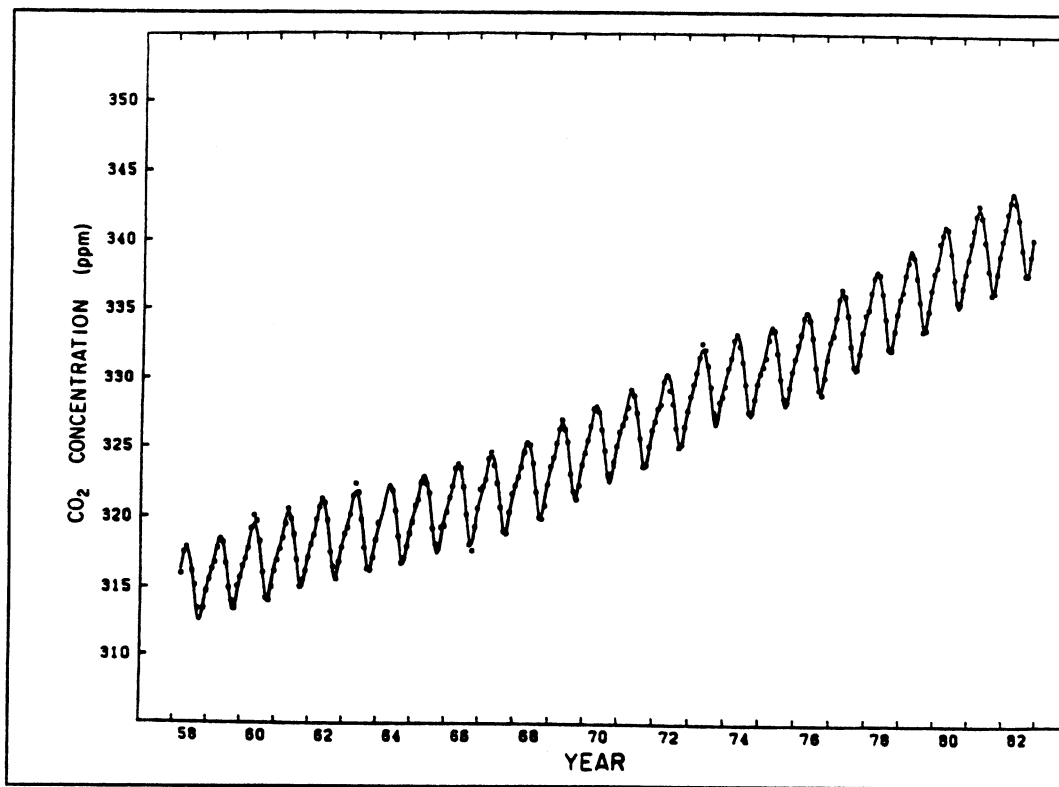


Figure 13-3. Concentrations of atmospheric CO₂ at Mauna Loa Observatory, Hawaii pressed as a mole fraction in parts per million of dry air. The dots are monthly averages. Selected data have been adjusted to the center of each month. The curve represents the fit simultaneously to an exponential function, a linear function and a linearly increasing seasonal cycle (from Bacastow et al., 1975). [Reprinted with permission from the American Geophysical Union.]

water. The above simulations have been confirmed by other studies (Schneider, 1975; Manabe and Wetherald, 1980).

The temperature measurements on the earth, however, have not shown the same systematic increase as the CO_2 concentration, even though worldwide high temperatures in 1988 could be a first measurable sign of the greenhouse effect. On the contrary, the earth's average temperature, after a constant slow increase between 1880 and 1940, decreased in the following 40 years by more than half a degree. These latter data can be interpreted in several ways:

1. CO_2 increase is not, due to "feedback" effects (Newell and Doplick, 1979), really affecting the earth's temperature.
2. The earth's temperature is mainly controlled by variations in the solar radiation (e.g., the Gleissberg cycle, of about 90 years, in the solar activity; Agee, 1980).
3. Increasing concentrations of anthropogenic particles in the atmosphere are increasing the albedo (i.e., the fraction of solar radiation directly reflected by the atmosphere into the space), thus reducing the fraction of the incoming solar radiation that heats the earth's surface.
4. The effects of the CO_2 increase are not yet noticeable.

CO_2 is not the only trace gas that can cause the greenhouse effect. Other trace gases contribute, as shown in Table 13-1, which illustrates trace gas concentrations and trends (observed trends and projected mid-21st century values). Hansen et al. (1988) used a three-dimensional climate model to simulate the combined global climate effects of time-dependent variations of trace atmospheric gases (CO_2 , CH_4 , N_2O , $CFCs$) and aerosols. Among their several conclusions is that the greenhouse effect should be clearly identifiable (i.e., measurable) in the 1990s and that temperature changes will be sufficiently large to have major impacts, especially in the frequency of occurrence of extreme events.

Additional discussion of the CO_2 problem can be found in articles by Idso (1984) and Hileman (1984). Overviews of research activities in this field are discussed by Riches et al. (1985) and Lal and Jain (1989). It must be pointed out that trace gas accumulation is just one of many factors affecting global scale climate. Figure 13-4 illustrates a reconstruction of the mean global temperature in the last 150,000 years. We are currently in the middle of an interglacial period, which should be followed by a natural cooling, unless CO_2 accumulation induces a "superinter-glacial" period.

Table 13-1. Trace gas concentrations and trends: observed and projected. The concentrations are from Dickinson and Cicerone (1986); the decadal trends for 1975 to 1985, showing the percentage of increase in concentrations, are from Rasmussen and Khalil (1986). (From Ramanathan, 1988). [Reprinted with permission from the American Association for the Advancement of Science.]

Gas	Concentrations		Observed trends for 1975-1985 (%)	Mid-21st century
	Pre-1850	1985		
CO ₂	275 ppmv	345 ppmv	4.6	400-600 ppmv
CH ₄	0.7 ppmv	1.7 ppmv	11.0	2.1-4 ppmv
N ₂ O	0.285 ppmv	0.304 ppmv	3.5	0.35-0.45 ppmv
CFC-11	0	0.22 ppbv	103.0	0.7-3.0 ppbv
CFC-12	0	0.38 ppbv	101.0	2.0-4.8 ppbv
Tropospheric O ₃ * (below 12 km)		10-100 ppbv		
CH ₃ CCl ₃	0	0.13 ppbv	155.0	
CCl ₄	0	0.12 ppbv	24.0	

*Values (below 9 km) for before 1850 are 0 to 25% less than present-day; values (below 12 km) predicted for mid-21st century are 15 to 50% higher.

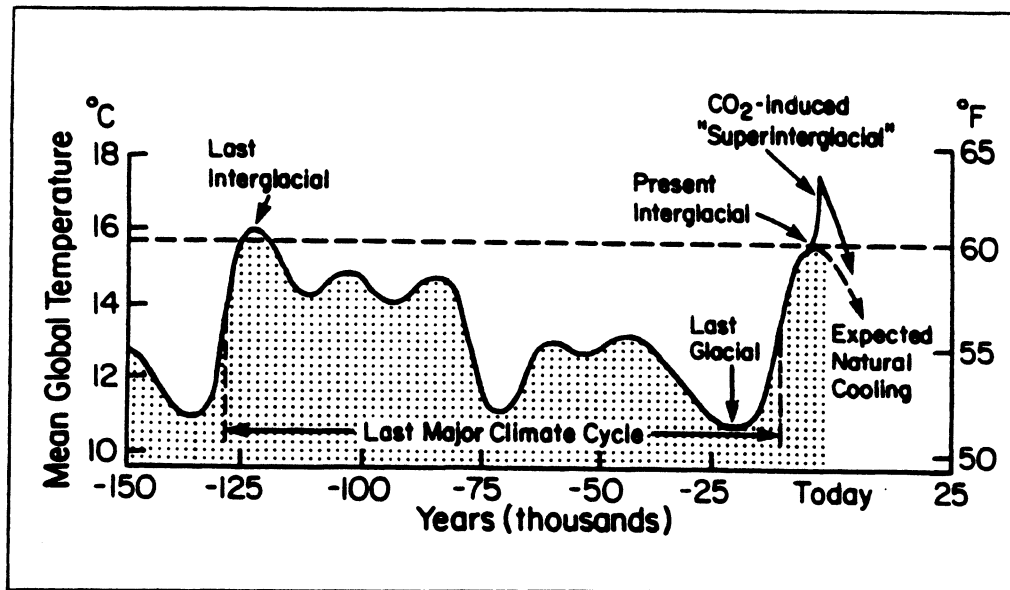
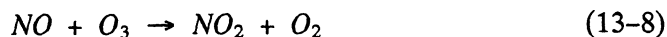


Figure 13-4. Potential effect of carbon dioxide on global climate (from Urone, in Stern, 1986). [Reprinted with permission from Academic Press.]

13.3 STRATOSPHERIC OZONE

The lower stratosphere (i.e., between an altitude of 10 and 20 km) contains an ozone layer that absorbs an appreciable part of the ultraviolet (UV) energy of the solar radiation. Without this ozone layer, the sea-level component of light that directly causes sunburn (erythema), a narrow spectral range centered at $0.297 \mu\text{m}$, would increase its intensity, causing, among other things, an increase in the incidence of skin cancer.

Combustion products from high altitude aircraft and missiles constitute a threat to the ozone shield. In particular, *NO* emissions (Dobbins, 1979) enter the cycle



and consume ozone and the oxygen free radicals in a "catalytic" sequence (i.e., without *NO* destruction). Another catalytic sequence is generated by the hydroxyl *OH*, which is also a product of combustion.

Another chemical threat to stratospheric ozone is posed by compounds that are emitted from human activities at the earth's surface, but that may diffuse to the stratosphere due to their extended lifetime. For example, halocarbons, such as CFC-11 ($CFCl_3$) and CFC-12 (CF_2Cl_2), undergo photolytic decomposition in the stratosphere because of the ultraviolet intensity. This generates atomic chlorine *Cl*, which enters the cycle (Dobbins, 1979)



and consumes both O_3 and *O* in a catalytic sequence (*Cl* is not consumed).

Recent observations of global ozone concentrations have revealed the following aspects of the phenomenon (Tung and Yang, 1988):

1. The year-to-year decline of the October ozone minimum over Antarctica is associated with a decline of the midlatitude ozone maximum.
2. These declines appear to be associated with a cooling of the lower stratosphere in middle and high latitudes.

3. The interannual October mean column ozone changes over Antarctica appear to follow a quasibiennial oscillation. This variation is in phase with the tropical oscillation of the east-west wind.
4. The seasonal spring deepening of the minimum column ozone appears to be associated with an intensification of the surrounding maximum in midlatitudes.
5. There is spatial correlation between total ozone patterns and temperature distributions in the lower stratosphere.
6. In November, the Antarctic ozone hole is filled in.

Initial modeling simulation (NAS Report, 1979) indicated an expected decrease of stratospheric ozone by -1.5 percent (± 1.1 percent) during the period 1970-1978. However, this decrease was not confirmed by ozone measurements during the same period (Reinsel et al., 1981).

More recent data have shown alarming results. For example (Bornstein, 1986a), the British Antarctic Survey measured 0.32 cm of ozone above their station in Antarctica in 1957, 0.32 cm in 1970, 0.30 cm in 1980, 0.24 cm in 1980 and only 0.20 cm in 1984. These data, among others, seem to indicate a deepening of the Antarctic ozone hole, a phenomenon that many theories suggest can be related to the release of the chlorine in fluorocarbons.

Recent results from the Ozone Trend Panel(*) (Kerr, 1988) indicate that, after a large reanalysis of the ozone measurements collected in the last 17 years, the stratospheric ozone is indeed decreasing, sometimes even faster than previously predicted. This decrease, which is shown in Figure 13-5, indicates a year-round effect. These new data and data elaboration strongly indicate anthropogenic chlorine as primarily responsible and raise questions about the sufficiency of the proposed production controls (50 percent) on chlorofluorocarbons (CFCs).

Tung and Yang (1988) have made comprehensive modeling simulations of changes in Antarctic and global ozone. Their simulations have been successful in reproducing the quasibiennial signal in the year-to-year variations. They conclude that atmospheric dynamics is an important component of the phenomenon, even though not the sole cause of the Antarctic ozone hole, since ozone depletion seems more severe than can be accounted for by pure advection phenomena.

(*) A creation of NASA in collaboration with the National Oceanic and Atmospheric Administration, the Federal Aviation Administration, the World Meteorological Organization, and the United Nations Environment Program.

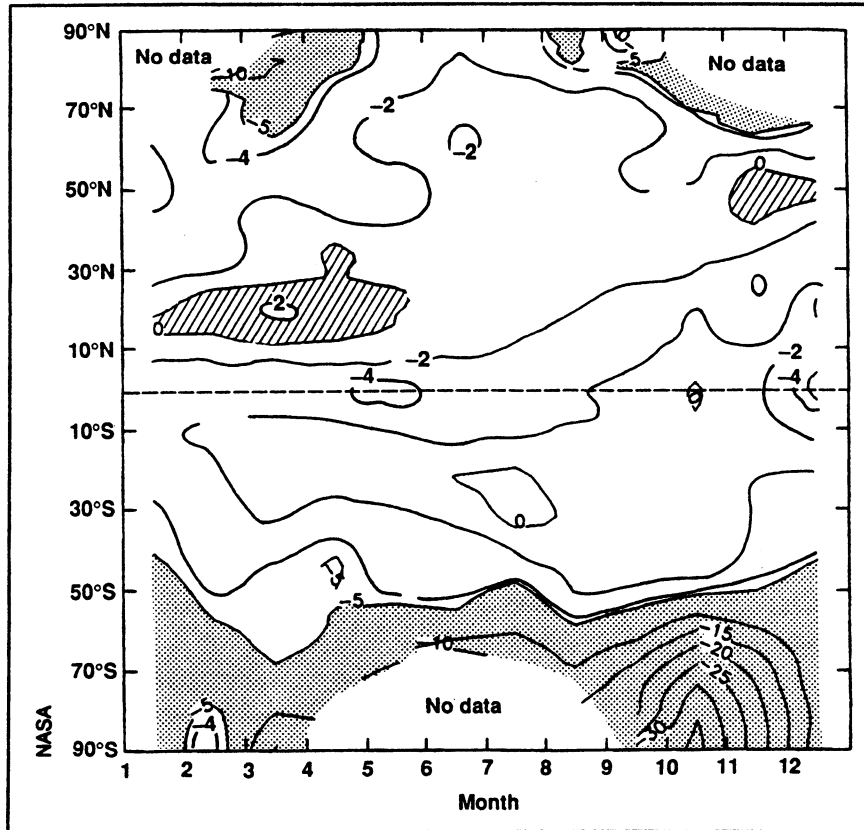


Figure 13-5. The decrease of stratospheric ozone. This is a plot of the percent change in total ozone from 1979-80 to 1986-87 as determined by the satellite-borne Total Ozone Mapper Spectrometer. The tendency toward larger losses at higher, colder latitudes and colder seasons is apparent. The Antarctic ozone hole is in the lower right (from Kerr, 1988). [Reprinted with permission from the American Association for the Advancement of Science.]

13.4 NUCLEAR WINTER

The meteorological effects of a major nuclear exchange have been a subject of debate in the last few years. Crutzen and Birk (1982) first recognized that large fires following a nuclear war may inject large amounts of particles into the atmosphere, whose absorption of the sunlight could generate a substantial and prolonged decrease of earth's temperatures, i.e., a nuclear winter. This suggestion was elaborated by Turco et al. (1983) in what is known as the TTAPS study

(from the initials of the authors). Their conclusion, using a one-dimensional model, was dramatic: a decrease of 30–40°C in land surface temperatures.

More recent simulations using two- and three-dimensional models have provided contradictory results. For example, Malone et al. (1986), using a three-dimensional global model, simulated 1) localized injection of smoke, 2) wind transport, 3) sunlight absorption, and 4) precipitation removal. They found an even longer-lasting “nuclear winter,” due to higher residence time than that normally assumed for the troposphere. However, in contrast with the TTAPS study, Penner et al. (1986), using a two- and three-dimensional model, concluded that little smoke is expected to be injected into the stratosphere, even for very intense fires. They also pointed out that, for intense fires, significant amounts of water vapor are condensed, raising the possibility of early scavenging of smoke particles by precipitation.

Mitchell and Slingo (1988) improved the calculations of the climatic effects of nuclear war by including two important phenomena that were not treated by previous studies: 1) the diurnal cycle of insolation and 2) surface and boundary layer parameterizations, including a four-layer soil model. The results did not change much, since the errors caused by neglecting the two phenomena above were approximately equal and opposite.

It can be concluded (Mitchell and Slingo, 1988; Bornstein, 1986b) that the greatest uncertainty in the “nuclear winter” theory is in the amount of smoke and dust that would be injected into the atmosphere and the parameterization of the deposition phenomena. A typical emission scenario is 180 Tg (million metric tons) of soot. With this emission, virtually all models agree that the sunlight will be blocked. The disagreement is about how cold it would get and for how long. Reviews of the atmospheric effects of a nuclear war are presented by Colbeck and Harrison (1986) and Colbeck (1989).

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