CHAPTER 7

Air Pollution

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PROBLEMS

REFERENCES

"From a particulate exposure standpoint, a 2-percent decrease in environmental tobacco smoke (passive smoking) would be equivalent to eliminating all the coal-fired power plants in the country."

-Kirk R. Smith, East-West Center, Program on Environment, Honolulu, 1993

7.1 INTRODUCTION

Air pollution is certainly not a new phenomenon. Indeed, early references to it date to the Middle Ages, when smoke from burning coal was already considered such a serious problem that in 1307, King Edward I banned its use in lime kilns in London. In more recent times, though still decades ago, several serious episodes focused attention on the need to control the quality of the air we breathe. The worst of these occurred in London, in 1952. A week of intense fog and smoke resulted in over 4000 excess deaths that were directly attributed to the pollution. In the United States the most alarming episode occurred during a four-day period in 1948 in Donora. Pennsylvania, when 20 deaths and almost 6000 illnesses were linked to air pollution. At the time, Donora had a population of only 14,000, making this the highest per capita death rate ever recorded for an air pollution episode.

Those air pollution episodes were the results of exceptionally high concentrations of sulfur oxides and particulate matter, the primary constituents of *industrial smog* or *sulfurous smog*. Sulfurous smog is caused almost entirely by combustion of

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fossil fuels, especially coal, in stationary sources such power plants and smelters. In contrast, the air pollution problem in many cities is caused by emissions of carbon monoxide, oxides of nitrogen, and various volatile organic compounds, which swirl around in the atmosphere reacting with each other and with sunlight to form *photochemical smog*. Although stationary sources also contribute to photochemical smog, the problem is most closely associated with motor vehicles. A major effect of efforts in the United States to control both sulfurous smog and photochemical smog has been the elimination of those dramatic, peak concentrations of pollution that were responsible for the air pollution episodes just mentioned. In their place, however, is the more insidious problem of morbidity and mortality increases associated with long-term exposure to lower concentrations of pollution. The human toll is much more difficult to document, but estimates place the current excess deaths caused by air pollution (mostly small particles) at several tens of thousands per year in the United States alone.

Much of the work on air pollution in the last few decades has centered on a small set of six substances, called *criteria pollutants*, that have been identified as contributors to both sulfurous and photochemical smog problems. The sources, transport, effects, and methods of controlling these criteria pollutants will be a principal focus of this chapter.

More recently, attention has been shifting toward the characterization and control of a growing list of especially hazardous air pollutants, many of which we are exposed to in our homes and workplaces, where we spend roughly 90 percent of our time. As the quote at the begining of this chapter suggests, modest improvements in indoor air quality can improve public health as much as major reductions in the traditional outdoor sources, which have been the focus of most of the scientific and political efforts of the past 50 years.

In the next chapter we will discuss the emissions and impacts of carbon dioxide, chlorofluorocarbons, and other trace gases that are affecting global climate and causing stratospheric ozone depletion. In a number of ways these gases are so different from the usual air pollutants that they deserve special treatment.

7.2 OVERVIEW OF EMISSIONS

There are many sources of the gases and particulate matter that pollute our atmosphere. Substances that are emitted directly into the atmosphere are called *primary* pollutants, while others that are created by various physical processes and chemical reactions that take place in the atmosphere are called *secondary* pollutants. For example, nitrogen oxides and hydrocarbons emitted when fuels are burned are primary pollutants, but the ozone that is created when those chemicals react with each other in the atmosphere is a secondary pollutant.

The sources of primary pollutant emissions can be conveniently categorized by the processes that create them. Most primary pollutants enter the atmosphere as a result of either combustion, evaporation, or grinding and abrasion. Volatile substances such as gasoline, paints, and cleaning fluids enter the atmosphere by evaporation; dust kicked up when land is plowed and asbestos fibers that flake off of pipe insulation are examples of grinding and abrasion; while automobile exhaust emissions and power plant stack gases are created during combustion. Of these it is combustion that accounts for the great majority of emissions, and it is the gases and particulate matter released when fuels are burned that have been the focus of most of the technical and legislative pollution control efforts.

In its simplest form, we can imagine the complete combustion of a pure hydrocarbon fuel such as methane (CH_4) :

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \tag{7.1}$$

The products of combustion are simple carbon dioxide (CO_2) and water (H_2O) , neither of which had been considered an air pollutant until we realized that the accumulation of CO_2 in the atmosphere was enhancing the earth's natural greenhouse effect (as will be described in the next chapter).

If the temperature of combustion is not high enough, or there is not enough oxygen available, or if the fuel is not given enough time to burn completely, then the fuel will not be completely oxidized and some of the carbon will be released as carbon monoxide (CO) instead of CO_2 . Also, some of the fuel will not be completely burned, so there will be emissions of various partially combusted hydrocarbons that we will represent by (HC). So we can write the following descriptive reaction to represent incomplete combustion of our pure hydrocarbon fuel, methane:

$$CH_4 + O_2 \rightarrow mostly (CO_2 + 2 H_2O) + traces of [CO + (HC)] (7.2)$$

Of course, most combustion takes place in air, not in a pure oxygen environment, and air is roughly 78 percent nitrogen (N_2) and 21 percent oxygen (O_2) . When the temperature of combustion is high enough, some of that nitrogen reacts with the oxygen in air to form various nitrogen oxides (NO_x) . Since this NO_x is formed when combustion temperatures are high, it is referred to as *thermal NO*_x.

air
$$(N_2 + O_2)$$
 + Heat \rightarrow Thermal NO_x (7.3)

So far we have assumed that the fuel being burned was a pure hydrocarbon such as methane. In reality, of course, most fuels have a number of other elements in them, such as nitrogen, sulfur, lead (in gasoline), and other unburnable materials called ash. Burning fuel with these "impurities" in them releases additional NO_{χ} (called *fuel NO₄*), oxides of sulfur (SO₄), lead (Pb), more particulate matter, and ash.

Combining the effects of incomplete combustion, combustion in air, and combustion of fuels that are not pure hydrocarbons yields the following qualitative description of combustion:

Fuel (H, C, S, N, Pb, ash) + air (N₂ + O₂)
$$\rightarrow$$

Emissions (CO₂, H₂O, CO, NO₃, SO₃, Pb, particulates) + Ash (7.4)

Now let's add a simple representation of the photochemical reactions that produce ozone (O_3) and other constituents of photochemical smog. Hydrocarbons (HC) and other organic compounds that readily vaporize are called *volatile organic compounds* (VOCs). VOCs react with NO_x in the presence of sunlight to produce photochemical smog:

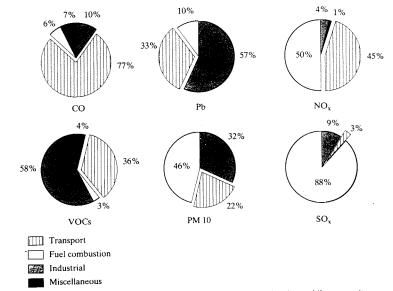
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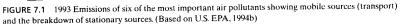
VOCs + NO_x + Sunlight \rightarrow Photochemical smog (O₃ + etc.) (7.5)

To distinguish between the ozone that is formed near the ground by (7.5) from the ozone that exists in the stratosphere (next chapter), the designations *ground level ozone* and *stratospheric ozone* are sometimes used. As we shall see, ground level ozone is harmful to our health, while stratospheric ozone protects our health by shielding us from ultraviolet radiation from the sun.

Reactions (7.1) to (7.5) are greatly simplified, of course, but they do introduce the six principal players in urban air pollution: CO, NO_x , SO_x , Pb, O_3 , and particulate matter (ash and unburned hydrocarbons).

Another way to approach emissions and controls of air pollutants is to categorize the sources as being *mobile* sources or *stationary* sources. Mobile sources include highway vehicles (automobiles and trucks) and other modes of transportation, including railroads, aircraft, farm vehicles, and boats and ships. Stationary sources are often categorized as stationary *fuel combustion*, which includes electric power plants and industrial energy systems; *industrial processes*, such as metals processing, petroleum refineries, and other chemical and allied product manufacturing; and *miscellaneous* sources. Emissions of the principal air pollutants following this categorization are illustrated in Figure 7.1.





7.3 THE CLEAN AIR ACT

Initial efforts on the part of the U.S. Congress to address the nation's air pollution problem began with the passage of the Air Pollution Control Act of 1955. Although it provided funding only for research, and not control, it was an important milestone because it opened the door to federal participation in efforts to deal with air pollution. Up until that time, it had been thought to be a state and local problem. This was followed by a series of legislative actions by Congress that included the Clean Air Act Amendments of 1963, 1966, 1970, 1977, and 1990, all of which are sometimes lumped together and referred to as simply the *Clean Air Act* (CAA). In 1998, the Act will be due for reauthorization.

Much of the real structure to the Clean Air Act was established in the 1970 Amendments. In those amendments, the EPA was required to establish *National Ambient Air Quality Standards* (NAAQSs), and states were required to submit *State Implementation Plans* (SIPs) that would show how they would meet those standards. In addition, the Act required *New Source Performance Standards* (NSPSs) to be established that would limit emissions from certain specific types of industrial plants and from motor vehicles.

Air Quality and Emission Standards

The Clean Air Act requires the EPA to establish both air quality standards (NAAQS) and emission standards (NSPS), and it is important to keep in mind the fundamental difference between the two. Ambient air quality standards are acceptable *concentrations* of pollution in the atmosphere, while emission standards are allowable *rates* at which pollutants can be released from a source.

National Ambient Air Quality Standards have been established by EPA at two levels: *primary* and *secondary*. Primary standards are required to be set at levels that will protect public health and include an "adequate margin of safety," regardless of whether the standards are economically or technologically achievable. Primary standards must protect even the most sensitive individuals, including the elderly and those already suffering from respiratory and cardiopulmonary disorders. NAAQSs are, therefore, conceptually different from maximum contaminant levels (MCLs) that have been set for drinking water. Recall that the Safe Drinking Water Act requires the EPA to balance public health benefits with technological and economic feasibility in establishing drinking-water MCLs.

Secondary air quality standards are meant to be even more stringent than primary standards. Secondary standards are established to protect public welfare (e.g., structures, crops, animals, fabrics, etc.). Given the difficulty in achieving primary standards, secondary standards have played almost no role in air pollution control policy, and in fact they have usually been set at the same levels as primary standards.

National Ambient Air Quality Standards now exist for six *criteria* pollutants: carbon monoxide (CO), lead (Pb), nitrogen dioxide (NO₂), ground-level ozone (O₃), sulfur dioxide (SO₂), and particulate matter. The Clean Air Act requires that the list of criteria pollutants be reviewed periodically and that standards be adjusted according to the latest scientific information. Past reviews have modified both the list of pollutants and their acceptable concentrations. For example, the original particulate standard did not refer to size of particulates, but in 1987 the standard was modified to include only particulates with aerodynamic diameter less than a nominal 10 μ m (PM 10), and in 1997 an additional category of fine particles with diameters less than or equal to 2.5 μ m (PM 2.5) was added. Also in 1997 the ozone standard was tightened from 0.12 ppm to 0.08 ppm.

For a given region of the country to be in compliance with NAAQS, the concentrations cannot be exceeded more than once per calendar year. The law allows states to establish standards that are more stringent than the NAAQS, which California has done. Federal air quality standards along with California's standards are shown in Table 7.1.

For the gases in Table 7.1, the concentrations are expressed two ways—in parts per million by volume (ppm) as well as in mass per unit volume ($\mu g/m^3$ or mg/m³). The volumetric units (ppm) are preferred since those are independent of pressure and temperature. The mass-per-volume concentrations assume a temperature of 25 °C and 1 atm of pressure. The conversion between units was discussed in Section 1.2, and the following example illustrates the procedure.

Besides establishing National Ambient Air Quality Standards, the Clean Air Act also requires the EPA to establish emission standards for mobile sources such as cars

Pollutant	Averaging time	Federal primary	Federal secondary	California	Most relevant effects
Carbon monoxide (CO)	8 hr	9 ppm (10 mg/m ³)	None	9 ppm	Aggravation of angina pectoris; decreased
monoxide (CO)	l hr	35 ppm (40 mg/m ³)	None	20 ppm	exercise tolerance; possible risk to fetuses
Nitrogen dioxide (NO ₂)	Annual mean	0.053 ppm (100 μg/m ³)	Same	None	Aggravation of respiratory disease; atmospheric
	1 hr	None	None	0.25 ppm	discoloration
Ground level ozone (O ₃)	8 hr	0.08 ppm (155 μg/m ³)	Same	0.09 ppm	Decreased pulmonary function; surrogate for eye irritation; materials and vegetation damage
Sulfur dioxide (SO ₂)	Annual mean	0.03 ppm (80 μg/m ³)	None	None	Wheezing, shortness of breath, chest tightness;
	24 hr	0.14 ppm (365 μg/m ³)	None	0.05 ppm	plant damage and odor
	3hr 1hr	None	0.50 ppm 0.25 ppm	None	
PM 10	Annual 24 hr	50 μg/m ³ 150 μg/m ³	Same Same	30 μg/m ³ 50 μg/m ³	Exacerbation of respiratory mean disease symptoms; exces deaths; visibility
PM 2.5*	Annual 24 hr	15 μg/m ³ 65 μg/m ³	Same Same	None None	•
Lead (Pb)	1 month 3 months	None 1.5 μ g/m ³	None Same	1.5 μg/m ³ None	Impaired blood formation: infant development.

4 added in 1997

and trucks. The 1970 Amendments to the Clean Air Act gave the auto industry a fiveyear deadline to achieve a 90 percent reduction in emissions from new cars. At the time it was not even known whether such reductions were technologically possible, let alone how they could be implemented in such a short period of time. This "technology-forcing" legislation predictably led to numerous clashes between Congress and the automobile industry, and the standards were modified and delayed for many years. Later in this chapter the emission controls that eventually were implemented will be described in some detail.

The EPA is also required to establish emission standards for certain large stationary sources such as fossil-fuel-fired power plants, incinerators, Portland cement plants, nitric acid plants, petroleum refineries, sewage treatment plants, and smelters of various sorts. The methods of achieving the emission standards for stationary sources will be explored later in this chapter.

EXAMPLE 7.1. Air Quality Standards Expressed in Volumetric Units

California's air quality standard for nitrogen dioxide (NO₂) is 470 μ g/m³ (at a temperature of 25 °C and 1 atmosphere of pressure). Express the concentration in ppm.

Solution In Section 1.2 the ideal gas law was used to show that 1 mol of an ideal gas at 1 atm and 25 °C occupies a volume of 24.45 L (24.45×10^{-3} m³). The molecular weight of NO₂ is mol wt = $14 + 2 \times 16 = 46$ g/mol

so that

$$NO_{2}) = \frac{24.45 \times 10^{-3} \text{ m}^{3}/\text{mol} \times 470 \times 10^{-6} \text{ g/m}^{3}}{46 \text{ g/mol}}$$
$$= 0.25 \times 10^{-6} = 0.25 \text{ ppm}$$

which agrees with Table 7.1.

Notice that parts per million by volume (ppm) is really a dimensionless volume fraction, independent of temperature and pressure.

The Clean Air Act Amendments of 1977

The goal of the 1970 Amendments was to attain clean air by 1975, as defined by the NAAQS, with allowable extensions in certain circumstances until 1977. For a number of reasons, only about one-third of the air quality control regions in the nation were meeting the standards by 1977. This forced Congress to readdress the problem through the Clean Air Act Amendments of 1977. Besides extending the deadlines, the 1977 Amendments had to deal with two important questions. First, what measures should be taken in *nonattainment areas* that were not meeting the standards? Second, should air quality in regions where the air is cleaner than the standards be allowed to degrade toward the standards, and if so, by how much?

For nonattainment areas, the 1970 Act appeared to prohibit any increase in emissions whatsoever, which would have eliminated industrial expansion and severely curtailed local economic growth. To counter this, the EPA adopted a policy of *emission* offsets. To receive a construction permit, a major new source of pollution in a nonattainment area must first find ways to reduce emissions from existing sources. The reductions, or offsets, must exceed the anticipated emissions from the new source. The net effect of this offset policy is that progress is made toward meeting air quality standards in spite of new emission sources being added to the airshed.

Offsets can be obtained in a number of ways. For example, emissions from existing sources in the area might be reduced by installing better emission controls on equipment that may or may not be owned by the permit seeker. In some cases, a permit seeker may simply buy out existing emission sources and shut them down. Emission offsets can be "banked" for future use, or they can be sold or traded to other companies for whatever the market will bear. In addition to offsets, new sources in nonattainment areas must use emission controls that yield the *lowest achievable emission rate* (LAER) for the particular process. LAER technology is based on the most stringent emission rate achieved in practice by similar sources, regardless of the economic cost or energy impacts.

The 1970 Amendments were not specific about regions that were cleaner than ambient standards required, and in fact appeared to allow air quality to deteriorate to those standards. The 1977 Amendments settled the issue of whether or not this would be allowed by establishing the concept of *prevention of significant deterioration* (PSD) in attainment areas. Attainment areas are put into one of three classes, and the amount of deterioration allowed is determined by the class. Class I areas include National Parks and Wilderness Areas, and almost no increase in pollution is allowed. At the other extreme, Class III areas are designated for development and allowable increments of new pollution are large. Everything else falls into Class II areas, where moderate deterioration in air quality is allowed. In PSD areas, *best available control technology* (BACT) is required on major new sources. BACT is less stringent than LAER, as it does allow consideration of economic, energy, and environmental impacts of the technology, but it can be more strict than allowed by NSPS.

In all PSD areas, the allowable increments of air quality degradation are constrained by the NAAQS. That is, in no circumstance would air quality be allowed to deteriorate to the point where the area is no longer in compliance with ambient air quality standards. To demonstrate compliance with these PSD increments and with air quality standards in general, mathematical models predicting ambient pollutant concentrations must be used for any proposed new source. Such models, which use meteorological and stack emission data to predict air quality impacts, will be described in Section 7.5.

The Clean Air Act Amendments of 1990

The Clean Air Act Amendments of 1990 significantly strengthened the government's efforts to assure healthful air in the United States and it broadened its scope to include control of pollutants that affect a global problem—stratospheric ozone depletion. Principal changes in the Act include the following:

- A new acid deposition control program (Title IV)
- New requirements for nonattainment areas (Title I)

- Tightened automobile emission standards and new fuel requirements (Title II)
- New toxic air pollution controls (Title I)
- · Phase-out schedule for ozone-depleting substances (Title VI).

One of the most important shortcomings of the Clean Air Act before the 1990 Amendments was its inability to deal effectively with acid rain (or, more correctly, acid deposition). As will be described later, acid deposition results from emissions of sulfur dioxide (SO₂) that convert to sulfuric acid, and nitrogen oxides (NO_x) that become nitric acid. The goal of the Amendments is to cut annual SO₂ emissions to half of 1980 levels, establishing a cap of 8.9 million tons by 2000, and to cut NO_x emissions by 2 million tons. The NO_x reductions are to be achieved in the traditional way—that is, by tightening the emission standards for major stationary sources (sometimes referred to as the "command and control" approach), but the SO₂ reductions will result in large part from a new market-based approach.

In addition to specifying certain emission limits for SO_2 , the EPA is administering a more flexible *allowance system*, in which one allowance authorizes the owner to emit one ton of SO_2 . Large coal-fired power plants are not allowed to emit any more tons of SO_2 than the number of allowances they own. If insufficient allowances are owned to cover emissions, the owners are subject to an excess emissions penalty of \$2000 per ton of SO_2 . By controlling the number of allowances that the EPA issues each year, a cap is placed on emissions from these large sources. The intent is for these allowances to be bought and sold or banked in the same way that other commodities are traded. New sources that have no allowances would have to purchase allowances from existing sources or from annual EPA auctions. The idea, of course, is that major sources will find the least expensive ways to cut their emissions and then sell some of their allowances to others who cannot reduce their emissions as cheaply. The goal is a least-cost emission limitation that allows sources the flexibility they need to make the most cost-effective choices.

An even more innovative section of the acid deposition section of the 1990 Amendments includes the creation of a conservation and renewable energy reserve (*The Reserve*). The Reserve is a pool of 300,000 SO₂ allowances that are being given to eligible electric utilities as a reward for customer energy conservation programs and to utilities or independent power producers who build new renewable energy systems (such as wind or solar power). These bonus allowances are equivalent to emissions associated with 150 billion kWh of electricity, which will cut emissions of SO₂ by over 400,000 tons (U.S. EPA, 1994a). In addition, over 400,000 tons of NO_x and over 100 million tons of CO₂ reductions will result from this set aside (see Example 1.11 in Chapter 1).

The 1990 Amendments also address the slow progress being made in nonattainment areas. The Amendments establish a rating system for nonattainment areas based on the extent to which the NAAQSs are exceeded. A given area is designated as marginal, moderate, serious, severe, or extreme, and each category has its own deadlines and control requirements. One element of the new system is the amount of offsetting that is required for new sources. In *marginal* areas, industries must remove 10 percent more emissions from existing sources than the new emissions they intend to release. In *extreme* areas, that offset ratio is 1.5 to 1; that is, a new source must offset 1.5 times as



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Index	Designation	1 hr O ₃ (ppm)	8 hr CO (ppm)	24 hr PM 10 (μg/m ³)	24 hr SO ₂ (ppm)	1 hr NO ₂ (ppm)
0		0	0	0	0	*
50		0.06	4.5	50	0.03	a
100	NAAQS	0.12 ^b	9	150	0.14	a
200	Alert	0.20	15	350	0.30	0.6
300	Warning	0.40	30	420	0.60	1.2
400	Emergency	0.50	40	500	0.80	1.6
500	Significant harm	0.60	50	600	1.00	2.0

^aNo index values reported at concentrations below the Alert level. ^bdoes not yet reflect 1997 change in standard. *Source* EPA, 1994b.

interpolation between the indicated breakpoints. The highest PSI subindex determines the overall PSI. A PSI calculation is provided in Example 7.2.

EXAMPLE 7.2. Determining the PSI

Suppose on a given day the following maximum concentrations are measured:

 1 hr O₃
 0.18 ppm

 8 hr CO
 9 ppm

 24 hr PM 10
 130 μg/m³

 24 hr SO₂
 0.12 ppm

 1 hr NO₂
 0.3

Find the PSI and indicate the descriptor that would be used to characterize the day's air quality.

Solution Using Table 7.3, it can be seen that the ozone level (O_3) yields a subindex over 100; CO yields an subindex of 100; PM 10 and SO₂ are less than 100. There is no subindex for NO₂ since it is below the Alert level (0.6 ppm). The highest subindex therefore corresponds to O₃. To calculate the PSI, we must interpolate. An ozone concentration of 0.12 ppm yields a subindex of 100, while a value of 0.20 ppm corresponds to 200. By interpolation, the measured ozone concentration of 0.18 ppm yields a subindex of

Subindex
$$O_3 = 100 + \frac{(0.18 - 0.12)}{(0.20 - 0.12)} \times (200 - 100) = 175 \text{ ppm}$$

The highest subindex is 175, so the PSI would be 175 and the air quality would be described as unhealthful.

The PSI is human health based and does not specifically take into account the damage air pollution can cause to animals, vegetation, and materials. It also does not take into account the possibility of synergistic effects associated with combinations of pollutants. For example, the combination of sulfur oxides and particulates is thought to be much more damaging to health than the sum of the individual effects, but the current version of the PSI does not account for that magnification.

The number of days that the PSI is above 100 is often used to describe progress that is being made in air quality. Figure 7.3 shows 10-year trend data from 1984 to 1993

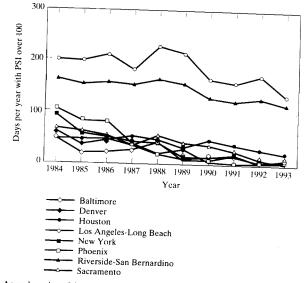


FIGURE 7.3 Annual number of days when PSI was greater than 100 for the most polluted American cities. Data cover only those sites in each metropolitan area that have complete data sets. (*Source*: U.S. EPA, 1994b)

for the most polluted American cities. In every case the number of days with PSI over 100 has decreased over time, with some cities, such as Sacramento, Denver, Baltimore, New York, and Phoenix. having cut their number of exceedances by over 70 percent. The worst cities in 1984. by a wide margin, were in southern California (Los Angeles-Long Beach and Riverside-San Bernardino). In 1993 those were still the most polluted cities (by this measure), with roughly one-third of their days having at least one pollutant exceeding the national ambient air quality standard. Almost always, that pollutant was ozone (O_3).

7.5 CRITERIA POLLUTANTS

Given the ongoing focus of the Clean Air Act, most of the monitoring of emissions, concentrations, and effects of air pollution has been directed toward the six criteria pollutants: ground level ozone (O_3) , carbon monoxide (CO), sulfur dioxide (SO_2) , small particulates (PM 10), nitrogen dioxide (NO_2) , and lead (Pb). The original ambient air quality standards for these pollutants were based on extensive documentation assembled and published by the EPA in a multivolume set of *Air Quality Criteria* documents, from which the name *criteria pollutants* originated.

Figure 7.4 shows the progress that has been made in total emissions associated with these six criteria pollutants over the 25-year period from 1970 (the year the Clean