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Global Atmospheric Changes

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Increasing concentrations of CO_2 and other greenhouse gases in the atmosphere can be directly related to global warming. In terms of human health, because a major cause of increasing atmospheric concentrations of CO_2 is the increased combustion of fossil fuels, global warming also may result in increases in air pollutants, acid deposition, and exposure to ultraviolet (UV) radiation. To understand better the impacts of global warming phenomena on human health, this review emphasizes the processes that are responsible for the greenhouse effect, air pollution, acid deposition, and increased exposure to UV radiation.

Introduction: Climatic Changes and Global Warming

Atmospheric CO_2 levels have increased by 25% since 1850. This is due largely to combustion of fossil fuels and deforestation that releases CO_2 as the litter is burned or decays (*1*). Present average global concentrations of CO_2 are about 350 ppm. In 1850, concentrations were approximately 265 ppm. Over the last 130 years, average global surface temperatures have increased by 0.5°C (*2–5*). Since 1958, when atmospheric levels of CO_2 were first measured at the Mauna Loa Observatory in Hawaii, average global atmospheric concentrations have increased from about 316 to about 347 ppm in 1986 (*6*). A plot of the change in annual average atmospheric CO_2 concentration is given in Figure 1. Energy consumption patterns for the last 30 years project that consumption of fossil fuels will increase worldwide by 0.5 to 2% per year for the next 50 years. With this rate of increase, CO_2 concentrations in the lower atmosphere will double (to about 600 ppm) within the next 50 to 60 years (*7*).

If atmospheric concentrations of CO_2 and other greenhouse gases such as chlorofluorocarbons (CFCs), CH_4, oxides of nitrogen, and CO continue to increase in the atmosphere, global climatic models (GCMs) have calculated that global average surface temperatures will increase 1.5° to 4.5°C, and sea levels will rise 0.1 to 0.3 m by the year 2040 (*8–10*). However, there are limitations to these model projections that are due to a lack of understanding of important environmental phenomena. They include the crude treatments of biological and hydrologic cycles; the neglect of the high heat capacity of the oceans; and the exclusion of regional effects of clouds. Thicker clouds caused by SO_2 and acid aerosols reflect incident solar radiation before it strikes the earth's surface. Finally, current models lack the data and precision to compute temperature and rainfall changes for small regions such as individual states and groups of states.

Even with the limitaitons of GCMs, these projections of temperature and sea level increases are truly extraordinary, essentially because they will occur over such a very short geological time period. From ice core data taken from Antarctica, during the ice age extreme of 18,000 years ago, the average annual surface temperature has been estimated to be only about 5°C colder than it is today, and atmopheric concentrations of CO_2 were about 194 ppm (*11*). These ice core data provide a continuous record of atmospheric CO_2 concentrations over the last 160,000 years. Examining these data, there is an apparent cyclical character to CO_2 concentrations in the atmosphere. The length of this cycle is approximately 100,000 years, and it has been attributed to eccentricities and wobbling in the earth's orbit around the sun (*12*). These results, along with present atmospheric levels of CO_2, are plotted in Figure 2. If the projected changes in CO_2 are true, atmospheric levels of CO_2 in 50 years will be almost twice as high as they have ever been in the last 160,000 years. Surface temperature changes of this magnitude will cause major changes in water resources, sea level, agriculture, forests, biological diversity, air quality, human health, urban infrastructure, and electricity demand (*13–16*).

The Greenhouse Effect

The greenhouse effect is the set of energy transport phenomena that occurs due to the interactions of the sun with the earth's surface and its atmosphere. It should be recognized that because of these interactions, the average temperature of the earth is maintained within a fairly narrow seasonal and diurnal range (about 20–40°C at all latitudes and longitudes). These changes in average surface temperature can be compared with our moon, which has very little atmosphere and has temperature changes from day to night that can range from 150 to 200°C.

Energy that is transmitted from the sun to the earth's surfaces has wavelengths primarily in the ultraviolet (UV) (wavelengths between 200 and 400 nm), visible (400–800 nm), and infrared (IR) (1200–20,000 nm) portions of the electromagnetic energy spectrum (EMS). Ozone that exists in the upper atmosphere (stratosphere) absorbs most of the UV energy, keeping it from striking the earth's surfaces below. This is the protective shield or protective ozone layer in the stratosphere. Once energy from the sun is absorbed by the earth's surfaces and its atmosphere, to complete the energy balance, it is re-radiated back into space. The amount of heat gain must equal the amount of heat loss at

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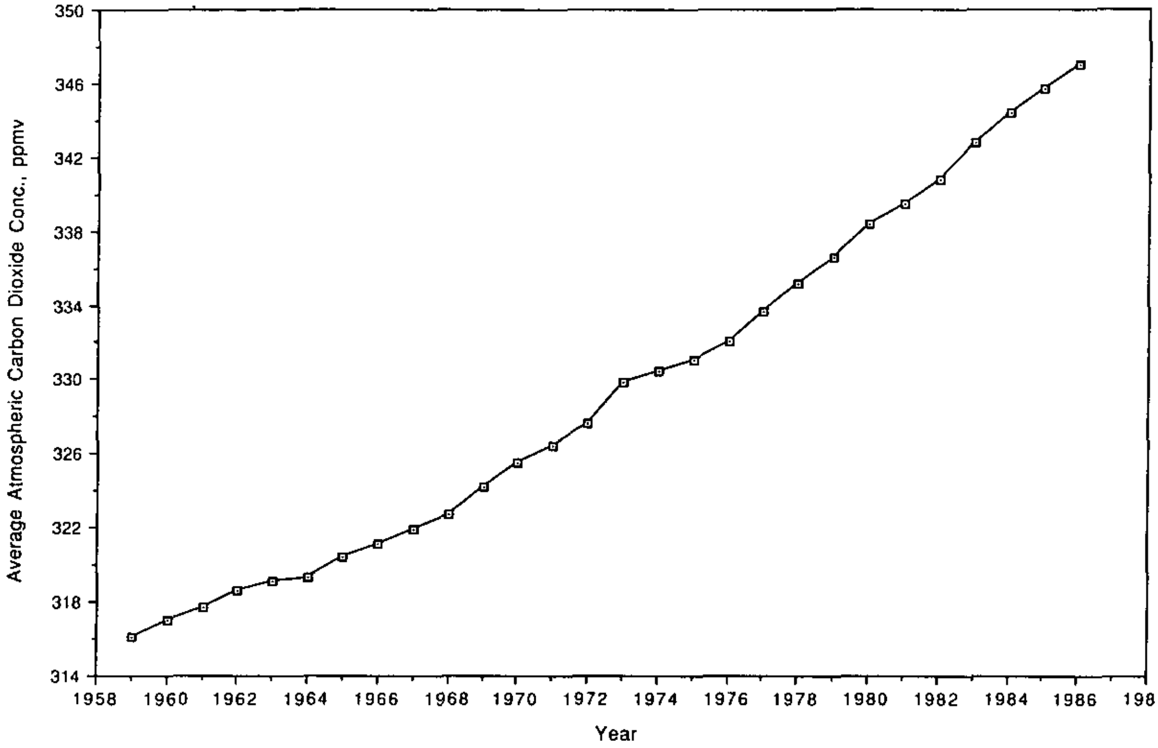

FIGURE 1. Average annual atmospheric CO concentrations at the Mauna Loa Observatory (*6*).

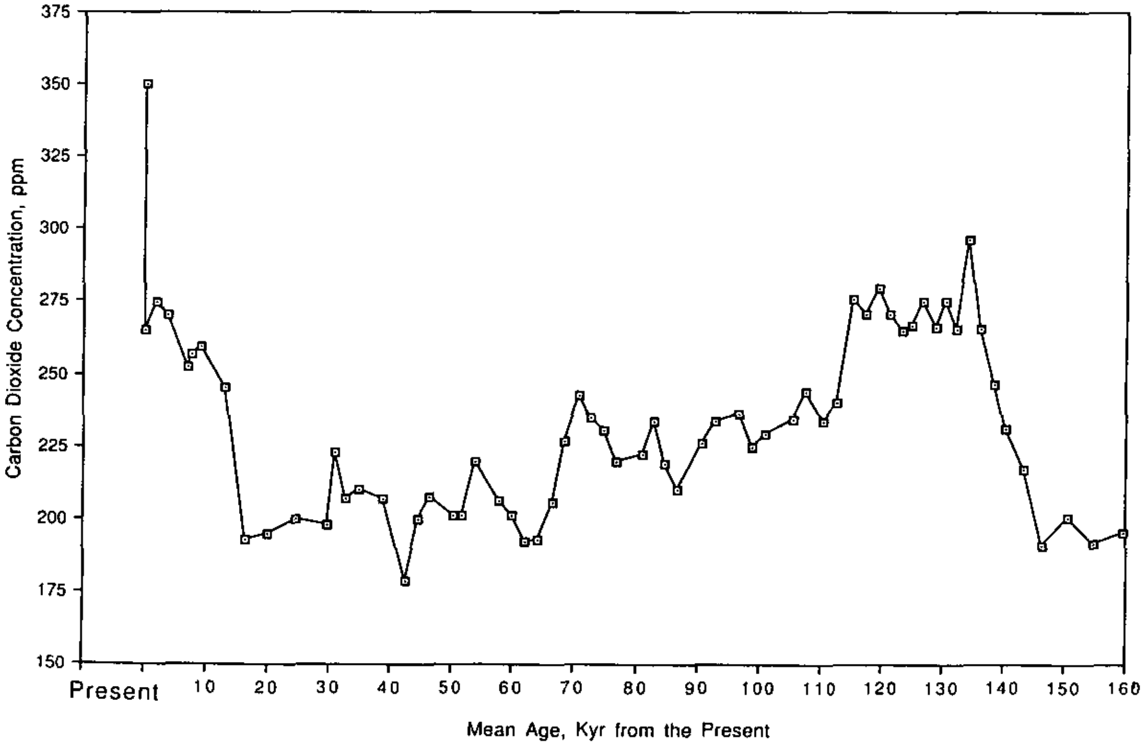

FIGURE 2. Ice core data on CO_2 atmospheric concentrations (*11*)

equilibrium. This heat balance equation is essentially a statement of the first law of thermodynamics, which describes a state of equilibrium but tells us nothing about the rate at which heat is gained or lost from the earth and its atmosphere.

The earth's surfaces emit or re-radiate absorbed radiant energy from the sun at temperatures (285°–300°K) in the infrared (IR) "window" of the EMS (wavelengths between 7000 and 13,000 nm). Gases in the atmosphere that strongly absorb energy in this IR window are CO_2, methane, N_2O, and O_3 present in the troposphere from photochemical smog, CO, and the CFCs. If the earth's atmosphere can be considered as a blanket of insulation, increasing the concentrations of greenhouse gases in the atmosphere has the same effect as raising its R-value (resistance to heat gain or heat loss). The net effect is to slow down the rate of heat loss from the earth's surface back into space and increase the surface temperature.

Human activities that contribute greenhouse gases to the atmosphere are combustion of fossil fuels that produce CO and CO_2, overuse of nitrate fertilizers (aerobic degradation of nitrate produces N_2O), liberation of methane as a result of decay of organic matter, and accumulation of CFCs (from uses as degreasing solvents, in making plastics or from leaky air conditioning equipment). Figure 3 is a pie chart showing the relative contributions of these greenhouse gases to global warming in 1985. The data are from Lashof and Ahuja (*17*) and clearly show that the combustion of fossil fuels, which produces both CO and CO_2, is the main source of global warming potential from greenhouse gases.

Replacement Chemicals for CFCs

Besides existing as greenhouse gases in the troposphere, the CFCs also contribute to the depletion of the ozone layer in the stratosphere. CFC chemicals degrade very slowly in the troposphere (half-lives of 10–50 years) because of the absence of shortwave UV energy. In the stratosphere, however, the CFCs photodegrade at wavelengths in the UV, liberating chlorine atoms that react with ozone (*18*). This reaction with O_3 depletes the O_3 concentration in the stratosphere, allowing more UV energy of shorter wavelengths (UV-B, 280–320 nm; and UV-C, <280 nm) to get through this protective shield and strike the earth's surfaces, plants, animals, and humans. In addition, the absorption of UV radiation by ozone causes the stratosphere to be much warmer than the upper regions of the troposphere, an upper atmospheric temperature inversion. Depletion of ozone may result in a net cooling of the stratosphere that would profoundly affect the heat balance in the troposphere. There changes in the heat balance in the upper and lower atmosphere would ultimately affect surface temperatures.

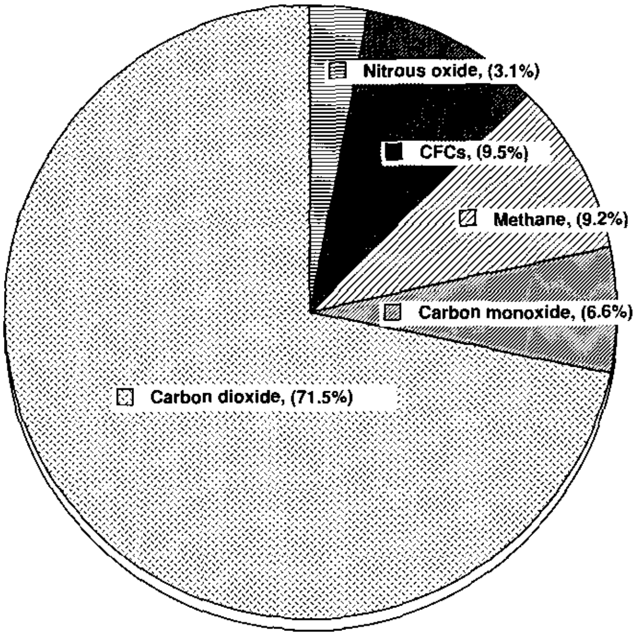

FIGURE 3. Relative contribution of greenhouse gases to global warming in 1985 (*18*).

Fisher et al. (*19*) have evaluated proposed important replacement chemicals for the CFCs, the hydrochlorofluorocarbons (HCFCs). This evaluation process included the determination of the IR absorbance properties of the HCFCs in the IR window region and their estimated global warming potentials (GWPs). In Table 1, chemical formulas and chemical names for CFCs, HCFCs, and several other candidate replacement chemicals are listed. Figure 4 is a graph showing a comparison of atmospheric lifetimes for these chemicals and their GWPs. In the evaluation, the GWPs for replacement HCFCs were compared to the warming potential of CFC-11 (trichlorofluoromethane), which was designated as the reference chemical and given a GWP of 1.0. GWPs were calculated from atmospheric transport models developed by Owens et al. (*20*) and Wang and Molnar (*21*).

From these calculations and the data shown in Figure 4 (*19*), it would appear that the GWPs for the replacement HCFCs are substantially less than the warming potentials of CFC-11 (GWP=1.0) and CFC-12 (GWP=3.4). The presence of hydrogen atoms in their structures makes the HCFCs more reactive in the troposphere than the CFCs. Because of this, the HCFCs remain in the atmosphere for a much shorter time. The major route of degradation is believed to be by reaction with hydroxyl radicals (OH) to produce a water molecule and a HCFC radical (*22*). The HCFC radical then reacts with O_2 to form acid chlorides:

$$OH + RH \rightarrow H_2O + R \qquad (1)$$

$$R + O_2 \rightarrow R'COCl \qquad (2)$$

where R and R′ are chloroflurocarbon radicals. Acid chlorides are very reactive chemically. Biologically, acid chlorides are extremely irritating to skin, eyes, and mucous membranes.

Other possible replacement chemicals for the CFCs are fluorocarbons (FCs). These chemicals have substituted fluorine

Table 1. Chlorinated fluorocarbons (CFCs) and hydrochlorofluorocarbon (HCFC) replacements.

Halocarbon	Formula	Chemical name
CFC-11	CCl_3F	Trichlorofluoromethane
CFC-12	CCl_2F_2	Dichlorodifluoromethane
CFC-113	$CClF_2CClF_2$	1,1,2-Trichloro-1,2,2-trifluoroethane
CFC-114	$CClF_2CF_3$	1,2-Dichloro-1,1,2,2-tetrafluoroethane
CFC-115	$CClF_2CF_3$	Chloropentafluoroethane
HCFC-22	$CHClF_2$	Chlorodifluoromethane
HCFC-123	CF_3CHCl_2	2,2-Dichloro-1,1,1-trifluoroethane
HCFC-124	CF_3CHClF	2-Chloro-1,1,1,2-tetrafluoroethane
HFC-125	CF_3CHF_2	Pentafluoroethane
HFC-134a	CF_3CH_2F	1,1,1,2-Tetrafluoroethane
HCFC-141b	CCl_2FCH_3	1,1-Dichloro-1-fluoroethane
HCFC-142b	$CClF_2CH_3$	1-Chloro-1,1-difluoroethane
HFC-143a	CF_3CH_3	1,1,1-Trifluoroethane
HFC-152a	CHF_2CH_3	1,1-Difluoroethane
CCl_3CH_3	CCl_3CH_3	1,1,1-Trichloroethane

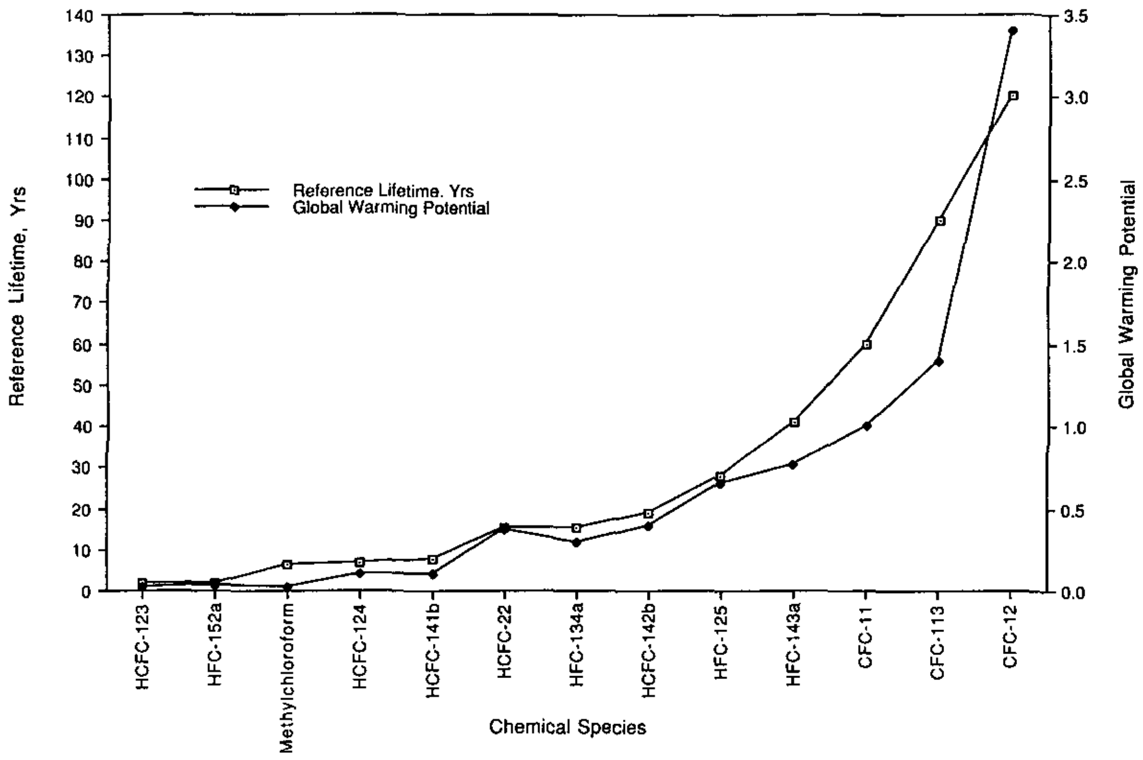

FIGURE 4. Comparison of atmospheric lifetimes of chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) with global warming potentials (*19*).

for chlorine in their molecular structures (*23*). Because of this absence of chlorine, it is claimed that the GWPs of the CFs are zero. In the stratosphere, the photochemical degradation of the CFCs produces chlorine atoms that then react with ozone (*18*). For the FCs, no information was provided on the absorption properties of these chemicals in the IR window, making it impossible to evaluate their potential as greenhouse gases. In addition, there are no data on degradation products in either the troposphere or stratosphere.

Even with the detailed models used to calculate GWPs for the CFCs and HCFCs, Solomon and Tuck (*24*) contend that the estimates of ozone-depleting potentials (ODPs) estimated by Fisher et al. (*19*) probably represent the lower limits of actual ozone depletion by the CFCs or their replacements. The gas-phase photochemical reaction model used in these calculations predicts only a maximum ozone depletion of 5% in the next century based on present CFC use rates. This disagrees strongly with observed ozone losses of 50% during the Antarctic spring (*25,26*) and 3 to 10% in the midlatitudes of the Southern Hemisphere (*27*).

At present, the evaluation of the environmental and health impacts of the replacement chemicals for the CFCs is not complete. Missing data are transport and fate information on the HCFCs and FCs in both the troposphere and the stratosphere and toxicity data on these replacement chemicals and their biologically mediated breakdown products. Because of potential increased exposure to UV radiation due to the depletion of the ozone layer, studies are needed to determine the ODPs of the HCFC replacements. Human health concerns from increased UV exposure are increased incidences of melanomas and nonmelanoma skin cancers (*28*), cataracts and other ocular disorders (*29*), and possible effects on the immune system (*30*).

Global Warming and Urban Air Pollution

The consumption of fossil fuels is projected to increase at an annual rate of 0.5 to 2% for the next 50 years. The combustion of high sulfur coal for electric power generation and combustion of gasoline in motor vehicles will produce not only increasing quantities of CO_2, but also increasing quantities of NO_x, SO_x, particulates, CO, ozone, and other photochemical oxidants. Major efforts and expenditures to control emissions of hydrocarbons and nitrogen oxides and reduce ozone concentrations in urban atmospheres have been modest. Actual reductions amount to approximately 10% between 1979 and 1985 (*31*).

Air pollutants accumulate in urban atmospheres because the capability of the atmosphere to remove these pollutants via dispersive transport becomes overloaded. In this condition, the atmosphere is not able to dilute air emissions from automobiles, power plants, and other mobile and stationary sources at a rapid enough rate to prevent their concentrations from increasing with time at ground level. One of the principal methods of measuring the dispersive capability of the troposphere is to measure the rate of change of atmospheric temperature with altitude above the earth's surface. If the rate of change is negative, e.g., temperature decreases with altitude, then the dispersive capability of the atmosphere is very high, and air pollutants released into the airspace above cities will not accumulate to a high degree. However, if there is a temperature inversion, e.g., temperature increases with increasing altitude, then the capability of the lower atmosphere to remove these pollutants is very poor. In this condition, concentrations of air pollutants can increase to levels that can cause adverse health effects in many vulnerable and healthy members of an urban population.

The atmospheric chemistry of several of these primary pollutants is very important in producing the type of urban air pollution called photochemical smog. Ozone is formed in the atmosphere by the reaction between atomic and molecular oxygen:

$$O + O_2 + M \rightarrow O_3 + M \tag{3}$$

Oxygen atom production is from the photodissociation of nitrogen dioxide:

$$NO_2 + h\nu \rightarrow NO + O \tag{4}$$

where the photon $h\nu$ has a wavelength between 280 and 430 nm, in the visible portion of the EMS. Nitric oxide produced in this reaction reacts rapidly with ozone to regenerate NO_2:

$$NO + O_3 \rightarrow NO_2 + O_2 \tag{5}$$

Ozone concentrations in unpolluted air vary from 20 to 50 ppb. In polluted air, ozone concentrations have been observed to be as high as 400 ppb. When hydrocarbons are present in the atmosphere, particularly in the form of peroxy radicals (RO_2 where the R is an alkyl group), a pathway is available to convert NO to NO_2 without consuming a molecule of ozone. The net result is that ozone is not being used as rapidly, and its concentration increases. The net reaction sequence that allows ozone concentrations to increase is

$$RO_2 + NO \rightarrow NO_2 + RO \tag{6}$$

$$NO_2 + h\nu \rightarrow NO + O \tag{7}$$

$$\underline{O + O_2 \rightarrow O_3} \tag{8}$$

$$\text{Net: } RO_2 + O_2 + h\nu \rightarrow RO + O_3 \tag{9}$$

The combustion of fossil fuels in electrical power plants and automobiles also generates aerosol particles. Prevalent compounds found in urban aerosols are sulfates, nitrates, ammonium ions, trace elements, carbonaceous material, and water. Particles of sizes below 2 mm diameter are largely responsible for severe visibility deterioration and are easily respirable (*32*).

In this sequence of atmospheric photochemical reactions, once this process is initiated, free radicals from hydrocarbon emissions beget more free radicals for as long as there is sunlight to photodissociate NO_2. The net result is that ozone is not being consumed and is allowed to accumulate. A temperature inversion occurring at the same time will greatly reduce the dispersive capability of the atmosphere and concentrations of ozone and other photochemical oxidants will increase at even greater rates at ground level.

Human health effects due to exposure to urban air pollutants have been investigated extensively (*33,34*). Ozone and nitrogen dioxide both reduce pulmonary function and increase morbidity and mortality, especially in people with chronic respiratory diseases. Combustion of fossil fuels also produces emissions of sulfur oxides, suspended particles, and carbon monoxide. Exposure to the combination of SO_x and suspended particles also increases morbidity, especially in people with chronic respiratory diseases (*35*). Carbon monoxide reacts with hemoglobin to form carboxyhemoglobin. Carboxyhemoglobin reduces the oxygen-carrying capacity of the blood and causes increased morbidity and mortality, especially in people with chronic cardiac diseases (*36*).

The use of methanol and methanol–gasoline blends has been proposed as a means of reducing ozone levels in urban areas. Studies with exhaust emissions from alternative fuels (*37*), however, have determined that ozone reductions depend on the presence of other chemical substances in the atmosphere. A more realistic evaluation of whether or not a shift to alternative fuels will reduce urban ozone concentrations includes determining the ratio of the concentration of nonmethane organic chemicals (NMOC) to the concentration of NO_x. If this ratio is high, methanol-fueled motor vehicles may not reduce ozone concentrations.

Along with the concerns for increased exposure to ozone from alternative fuels, more work will be required by automobile and gasoline manufacturers to reduce formaldehyde emissions from methane-fueled vehicles (*37*). In addition, the identification of other exhaust emissions from methanol–gasoline blends in well-tuned and untuned motor vehicles needs to be made. Other exhaust emissions include unburned methanol, unburned gasoline, a wide variety of polynuclear aromatic hydrocarbons, other aldehydes, and possible adducts of aldehydes with amines, polymers of aldehydes, and a wide variety of aliphatic and aromatic hydrocarbons.

Urban air contains very complex mixtures of air pollutants. Complexity is guaranteed not only because of the wide variety of sources and atmospheric formation of these substances, but also because it changes with time and space as well. It is not at all clear what long- and short-term exposure to these mixtures will mean for human health.

Global Warming and Indoor Air Quality

About 80 to 90% of our time is spent indoors in air-conditioned buildings and homes. Because of the longer periods of time spent indoors, indoor air quality and chronic exposure to indoor air pollutants are major human health concerns that are related to ambient air quality and air pollution (*38,39*). Indoor air pollutants consist of tobacco smoke, respirable particles, CO, NO_x, radon, asbestos, biological contaminants, and a large number of volatile organic chemicals such as formaldehyde, benzene, and other volatile organics from insulation, cleaning solutions, and other building materials. Cumulative exposure to these contaminants depends on the rates of accumulation of these substances in different rooms of a home or building and the amount of time a person spends in a particular room. Rates of accumulation depend on release rates of contaminants from materials of construction, infiltration of outside air, and the ventilation of air within buildings. An example of a mathematical model that incorporates these features to determine exposure rates for radon has been presented by Mowris and Fisk (*40*). Personal monitors to determine cumulative exposures to CO (*38*) have shown that exposures to CO can be substantially greater in cars and parking garages. In homes with very little infiltration of outside air, it is not uncommon for concentrations of indoor air pollutants to be two to five times greater than corresponding concentrations found outdoors (*39*).

Global Warming, Combustion of Fossil Fuels, and Acid Deposition

Whereas photochemical smog is usually confined to an urban setting, acid deposition and human exposure to acid aerosols usually occurs at great distances from the sources of the two major precursors of acid rain, sulfur and nitrogen dioxide (SO_2 and NO_2). One of the main sources of these two chemicals is the combustion of high-sulfur-content coal. Mathematical models describing transport, transformation, and deposition of these chemicals have been developed and rates of acid deposition determined (*41*). The models incorporate source strength, atmospheric chemical transformations, and wind and weather patterns over continental distances. The regional impact of acid deposition in North America is due to the broad distribution of emission sources of acid rain precursors in the United States and southeastern Canada. The intensity and greatest impact of acid deposition, however, remains highest near regions of greatest emission density. From the perspective of human health, the reactions that make up the atmospheric chemistry of acid aerosol formation produce very complex mixtures.

Along with the public health impacts of acid aerosols on urban populations (*35,42,43*), acid deposition may be an important mechanism for mobilizing metals by changing the pH in aquatic environments. Recent studies have observed enhanced bioaccumulation of mercury, cadmium, and lead by fish living in low pH surface waters (*44–46*). Studies of mercury, in particular, have found elevated levels of methylmercury in fish from low pH lakes. Decreased pH may stimulate methylmercury production at the sediment–water interface and possibly in the aerobic water column. Decreased pH may also decrease the loss of volatile mercury from lake water and increase mercury binding to particulates in water. These two factors may increase methylation at low pH by enhancing the bioavailability of mercury for methylation. Also at low pH, the rates of mercury methylation reactions are increased. Acidification of lakes also strongly affects the speciation of other metals such as cadmium and arsenic in lake water, changing their solubility and bioavailabilty. In addition, in forested watersheds of North America and northern Europe there appears to be a strong relationship between acid deposition as a source of mobile anions and the enhanced mobilization of aluminum (*47*).

Energy Consumption and Photovoltaic Devices

Over the last several years, advances in photovoltaic (PV) materials have produced high-efficiency crystalline silicon and gallium arsenide solar cells with conversion efficiencies between 22 and 31% (*48*). Manufacturing processes for PV devices are becoming more efficient, with the net effect that the cost of PV-generated electricity has dropped from $15 to approximately $0.30 per kilowatt-hour. With these higher efficiency PV energy conversion systems, the cost of PV-generated electricity is becoming price competitive with electricity generated by conventional fossil-fuel-fired facilities.

PV-generated electricity eliminates emissions of CO_2, NO_2, SO_2, and other air pollutants caused by the combustion of fossil fuels. However, as with the HCFC and FC replacement chemicals for the CFCs, it is essential to evaluate the health and environmental impacts of this emerging technology before it is put into full production. For photovoltaic devices, the basic power element is the solar cell. Each cell has two or more semiconductor layers whose atoms absorb sunlight, converting solar energy directly into electrical energy. The highest solar conversion efficiencies have been obtained with combinations of Group III and V elements in the Periodic Table of Elements. Group III elements that have been effective in this application include gallium (Ga), indium (In), and thallium (Tl). Group V elements include arsenic (As) and antimony (Sb). The highest efficiencies have been achieved with gallium arsenide (GaAs) and its alloys such as aluminum gallium arsenide (AlGaAs), indium gallium arsenide (InGaAs), and copper indium gallium selenide ($CuInGaSe_2$). At the present time, only one study could be found on occupational exposures to the components of PV devices (*48*). An *in vitro* and *in vivo* study in rats indicated that GaAs dissociates into its constituent elements (*49*); however, no additional toxicity information could be found on the other combinations of elements listed above.

Summary

From the Antarctica ice core data, it appears that the average annual global atmospheric CO_2 concentration rises and falls on a periodic basis. The periodicity of this cycle extends over geological time frames of about 100,000 years. As CO_2 concentrations rise and fall, average global surface temperatures rise and fall in a similar fashion and with the same frequency and cycle of change, suggesting a relationship between the processes that cause these changes to occur. The geophysical phenomena that control these changes are not well understood. It is also apparent that a relationship exists between the geophysical processes that affect climate and the atmospheric processes that play key roles in increasing human exposure to harmful chemical pollutants. Again, the exact relationships between these phenomena are not well understood, except in a qualitative manner. However, by increasing the concentrations of greenhouse gases in the atmosphere and by depleting the ozone layer in the stratosphere, these naturally occurring processes and defense mechanisms against increased exposure to UV radiation have been greatly perturbed. Indeed, these processes are being perturbed at a faster rate than can be understood or predicted. In terms of human health, perturbations that are being ascribed to global warming also result in increases in the atmospheric concentrations of the precursors of acid rain and urban air pollutants.

Over 30 years ago, Revelle and Suess (*50*) stated that we were carrying out a great geophysical experiment. Because of our incomplete understandings of these geophysical processes that are the subjects of this experiment, it is not at all certain how the experiment will end and what will be the outcomes for environmental and human health. However, in view of how increased concentrations of CO_2 in the atmosphere implies that concentrations of important air pollutants will increase also because their primary source is the combustion of fossil fuels, it would appear prudent to implement measures to reduce the rate of increase of these substances. In so doing and before alternatives and replacements are put into full service, the opportunity is available to evaluate their human health and environmental impacts.

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