Perspective

Air Pollution: Current Challenges and Future Opportunities

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Introduction

The invention of fire may be considered as the origin of man-made air pollution, but it is with the industrial revolution that air pollution became a significant societal issue. Major air pollution episodes, such as those in the Belgian Meuse valley in 1930, Donora, PA in 1948, and London in 1952 exemplify the adverse health effects associated with residential coal burning and industrial activities in populated areas, where meteorological conditions can be conducive to the stagnation of pollutants near the ground. In the 1950s, Haagen-Schmidt of the California Institute of Technology (Caltech) showed that nitrogen oxides (NO_x), and volatile organic compounds (VOC) that were emitted from cars and industries reacted in the presence of sunlight to create ozone, and other constituents of the photochemical smog that affected the Los Angeles basin during sunny days.[1, 2] In the 1980s, the death of forests and lake ecosystems in northern Europe and North America was shown to result from the atmospheric deposition of sulfuric acid (H_2SO_4) and nitric acid (HNO_3), i.e., "acid rain" (or, more exactly, acid deposition because pollutants can be deposited via wet processes —rain and snow—and dry processes— diffusion and sedimentation). These acids are formed in the atmosphere from the oxidation of sulfur dioxide (SO_2) and NO_x, respectively, which are emitted from combustion sources and some industrial activities. More recently, tiny particles of a few microns or less have been identified as being the source of adverse health effects associated with pulmonary and cardiovascular functions. These fine particles (regulated as particulate matter with an aerodynamic diameter less than 2.5 microns, $PM_{2.5}$) can be emitted directly from sources, such as the tailpipes of vehicles, smokestacks and the chimneys of residential homes, or can be formed in the atmosphere from gases, such as H_2SO_4, HNO_3, and semivolatile organic compounds. These gases are formed by the atmospheric oxidation of precursor gases (SO_2, NO_x and VOC). Other forms of air pollution include high concentrations of carbon monoxide (CO) and nitrogen dioxide (NO_2) in areas with intense road traffic, high concentrations of some toxic heavy metals (e.g., lead) and organic compounds (e.g., benzene, butadiene, aldehydes, acrolein). Also, persistent organic pollutants (POP) and some heavy metals (e.g., mercury) that deposit from the atmosphere to ecological systems may then accumulate in the food chain (bioaccumulation). We will not discuss here indoor air pollution, radioactivity, or air pollution problems that are of a global nature, such as the depletion of the stratospheric ozone layer or global climate change.

Some natural phenomena can lead to air pollution as well. For example, before man-made pollution became prevalent, the blue haze that was associated with the Great Smoky Mountains resulted from the oxidation of volatile organic compounds (VOC) released from the heavy vegetation in that area and the subsequent nucleation of the semivolatile oxidation products. Large natural forest fires and volcanic eruptions are also sources of air pollutants with possible large-scale adverse impacts. However, those natural air pollution phenomena tend to be localized in space and/or time and are generally uncontrollable. Man-made air pollution, on the other hand, has become widespread, and is, to a large extent, controllable. Consequently, the study of air pollution has become an important scientific discipline that involves sophisticated experimental techniques and advanced modeling tools.

Air pollution: a complex nonlinear system

The evolution of air pollution from its sources to its areas of impact is governed by transport and transformations. Transport processes include advection with the mean wind flow, convection (e.g., updraft in cumulus clouds) and turbulent diffusion. Transformations occur in a multiphase system that includes the gas phase, particulate matter (that can be solid or liquid or a mixture of both), and a water phase (e.g., cloud and fog droplets). Chemical reactions occur in the gas phase and the condensed phase (droplets, aqueous and organic particles), as well as heterogeneously at the surface of particles, droplets or ice crystals. Mass transfer occurs among these various phases and further complicates the system. The concentration of each pollutant is related to the concentrations of the other pollutants through an intricate web of chemical reactions. The chemical reactions that lead to the formation of several major pollutants, such as ozone, sulfate, nitrate, and some organic compounds are nonlinear, and the responses of these air pollutants to changes in the emissions of precursor pollutants can be, in

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some cases, counter-intuitive. Air pollution is, therefore, a complex system[3] and it is essential to understand air pollution as a whole because an emission control strategy that is designed to reduce the levels of one air pollutant could increase the level of on another air pollutant.

Measurements of the chemical composition of the atmosphere provide valuable quantitative information on the levels of air pollution.[4] Many routine monitoring networks now exist to measure the ambient concentrations of a large array of air pollutants, as well as concentrations in atmospheric precipitation (rain and snow). However, measurements alone cannot provide a complete picture of the state of the atmosphere because they are limited in space and time. Moreover, they do not provide information on the sources of those pollutants and do not tell us how their levels will change as man-made emissions of pollution either decrease (as a result of air pollution control) or increase (because of population and economic growth). Data analysis techniques and mathematical modeling are needed to answer such questions.

A recent review by Seinfeld[5] summarizes the fundamentals of air pollution, and the scientific progress made over the past 50 years. In addition, several excellent books are available that provide detailed information on the history, physics and chemistry of air pollution.[6-10] We focus here on some practical aspects of air pollution science. After an overview of the various tools available to study air pollution, we describe the complex nature of air pollution via a few case studies that highlight various aspects of the problem. Finally, we provide some thoughts on some promising areas of research and development.

Analysis of Air Pollution Data

Data that provide information on air pollution include direct evidence (e.g., smoke coming out of a stack or a satellite image of a dust cloud from the Sahara desert), spatial and temporal patterns of air pollutant concentrations, meteorological information (wind rose, air mass trajectories, etc.), and chemical or physical measurements of air pollutants.[11] Some of those data provide only circumstantial evidence, whereas other data can be treated to provide quantitative analyses. We describe here two major categories of such analyses: receptor modeling techniques that relate air pollutant concentrations to emission sources and indicator species methods that provide information on the directional response (increase or decrease) of air pollutants to changes in emission sources.

Receptor modeling

Receptor modeling is based on the analysis of air pollution data (generally chemical concentrations) at an ambient site (receptor). It is assumed that chemical mass is conserved in the atmosphere; thus, a mass balance can be conducted to identify and apportion the sources of air pollution to the ambient air pollution measurements. Sheldon Friedlander (then in chemical engineering at Caltech, later at UCLA) was the first to introduce this concept with a chemical mass balance analysis (CMB) that was initially applied to airborne particles.[12] Those particles contain a myriad of chemicals that can be identified and quantified through laboratory analysis following their sampling from the atmosphere. By assuming that a limited number of sources with different chemical fingerprints contribute to those particulate concentrations, a matrix calculation can be conducted to characterize the relative contributions of each source to the particulate mass concentration at the measurement site. This approach requires that each air pollution source be assigned a chemical profile (which can be obtained via sampling of the source). The methodology has, of course, some limitations. For example, some particulate species, such as sulfate, nitrate and some organics, are formed in the atmosphere rather than directly emitted from a source; it is not possible then to go back to the air pollution source unless some other tracer is available. Also, some sources may have similar chemical profiles that cannot be clearly separated during the analysis. Finally, measurements have uncertainties that propagate through the analysis and limit the accuracy of the results. Nevertheless, this approach has worked well and has become a standard technique.[13]

Many other techniques have subsequently been developed. One category of techniques that has become widely used is based on factor analysis. Those techniques require a large amount of data to be effective, but they offer the advantage that source chemical profiles are not needed, because they are inferred in the analysis. Then, the analyst must identify which air pollution sources correspond to the chemical profiles. Principal component analysis (PCA) is a well-known factor analysis method that was introduced many decades ago,[14] and is now applied in many scientific disciplines. However, it does not provide a quantitative apportionment of the air pollution to its sources. New factor analysis methods have been developed by Philip Hopke in chemical engineering at Clarkson University and Ronald Henry in civil engineering at the University of Southern California that provide quantitative source apportionment (e.g., positive matrix factorization, edge detection) as shown later in a case study. There are many other techniques that rely on air pollution data to characterize and apportion air pollution to sources.[13, 15] Such techniques can also be coupled with meteorological data to obtain information on the direction from which the air pollution came and also its source areas.

Indicator species

Receptor modeling techniques can provide very valuable information on the current state of air pollution and its apportionment to sources. However, they cannot predict how air pollution will evolve if the relationships between the air pollutants, and their sources are nonlinear. Mathematical models of air pollution are typically used to that end (see later). Nevertheless, it is possible to use measurements of the chemical composition of the atmosphere to obtain information on the chemical regime of the atmosphere, i.e., whether air pollutant levels will decrease, remain unchanged or even increase as emissions change. Such observation-based techniques rely on the measurements of several specific species (indicator species) that indicate the chemical regime of the atmosphere. For example, measurements of hydrogen peroxide and inorganic nitrate can indicate whether ozone formation is limited by NO_x or VOC.[16] Although one must be aware of the limitations of such techniques,[17] indicator species approaches can be used to estimate qualitatively the response of ozone, sulfate and particulate nitrate to changes in precursor emissions. Moreover, they are extremely useful to verify whether mathematical models of air pollution describe the chemical regime of the atmosphere correctly.[18]

Mathematical Modeling of Air Pollution

Mathematical modeling of air pollution (generally referred to, in a more positive tone, as air quality modeling) combines information on the emissions of air pollutants, meteorology, geography (terrain elevation, land use, vegetation cover, etc.), and transformations of chemical species in the atmosphere, to calculate the concentrations of air pollutants and their deposition fluxes to the earth. Clearly, meteorology plays an important role in the understanding of air pollution and its representation with mathematical models. Originally, many air pollution problems were associated with chemically inert or slowly reactive pollutants (CO and SO_2 near sources), and air pollution modeling was conducted primarily by researchers with a meteorological background who focused on the transport and dispersion of those pollutants. As it became clear that some pollutants (e.g., ozone) were not emitted from pollution sources, but were formed in the atmosphere from the reactions of precursor pollutants, air pollution started to be approached as a chemical engineering problem with a combination of chemical reactions, thermodynamics, multiphase mass transfer and transport phenomena.

The first three-dimensional (3-D) mathematical model of photochemical air pollution was developed by John Seinfeld's group in chemical engineering at Caltech.[19] This model has continuously evolved and today provides a detailed representation of ozone and other gaseous and particulate constituents of photochemical smog.[20-22] The conceptual framework consists in a 3-D grid over the atmospheric domain of interest. The spatial resolution of the grid is on the order of a few kilometers for an urban area to several tens of kilometers for regional simulations. In the vertical direction, the spatial resolution is finer near the ground, where the micrometeorological processes change rapidly with height. Emissions of air pollutants are released in the grid cells of the surface layer for most sources (e.g., traffic emissions), as well as in grid cells of higher layers for emissions from elevated stacks. Multiphase chemical transformations and mass transfer are simulated within each grid cell of the atmospheric domain. Transport processes are simulated among the grid cells, and for deposition, from the atmosphere to the surface. Thousands of reactions among hundreds of pollutants occur in the atmosphere. For modeling purposes, reduced chemical mechanisms are used that involve typically on the order of one hundred to a few hundreds reactions.[23] The temporal resolution of such models is typically 1 h for the inputs (i.e., emissions and meteorology) and outputs (air pollutant concentrations and deposition fluxes), but is a few minutes for the calculation time steps.

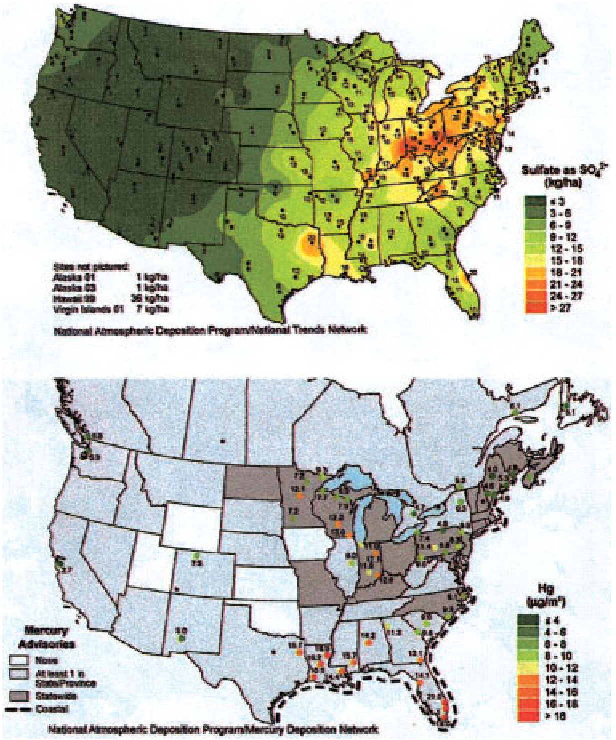
When acid rain became a major air pollution problem, air quality models were extended to larger areas (North America or Western Europe) and augmented with a treatment of cloud processes including aqueous-phase chemistry and precipitation. It became clear that all air pollution problems were interrelated. NO_x and VOC are precursors of ozone and other oxidants (hydroxyl radicals, hydrogen peroxide, etc.); these oxidants, in turn, convert SO_2, NO_x and VOC to H_2SO_4, HNO_3, and oxidized organic compounds that contribute to acid deposition and the formation of particles. Some VOC are toxics and toxic heavy metals, such as mercury react with species, such as SO_2 and O_3. Therefore, emission control strategies for a specific air pollution problem will also affect (positively or negatively) other aspects of air pollution.[24] Consequently, a "one-atmosphere" approach was developed to provide a single computational tool to address a multitude of related air pollution problems. Examples include the air quality models CMAQ (developed by the U.S. Environmental Protection Agency, www.cmascenter.org) and AURAMS (developed by Environment Canada). Such models can simulate photochemical smog, regional haze, acid deposition, nitrogen deposition, and air toxics concentrations and deposition. They typically are applied over domains that range from the continental scale (e.g., North America) to the urban scale using modeling grids that have a coarse resolution of 10 to 100 km for the continental domain and nested grids that have a resolution of a few kilometers for urban areas or sensitive receptor areas (e.g., national parks). Simulations are then conducted for periods that range from a few days for a photochemical smog episode to 1 year for acid deposition, regional haze or air toxics. Although, such simulations used to take a very long time on mainframe computers, they have now become considerably more efficient and a one-year simulation over North America is now conducted over a period of a few days on a cluster of Linux PCs or Macs.

Large experimental field campaigns, as well as satellite data have documented the long-range transport of air pollutants. Intercontinental transport (for example, from Asia to North America and from North America to Europe) is now of increasing concern as the effectiveness of emission control strategies implemented in one country may be limited by the increasing concentrations of air pollutants upwind. To address this issue, air quality models have been extended to the global scale. Simulations of ozone and PM, as well as some air toxics, such as mercury have been conducted to describe the global transport of those air pollutants and provide upwind boundary conditions to continental-scale models. This multiscale approach is now becoming more common as there are no physical boundaries to air pollution.

Case Studies

Acid rain vs. mercury deposition - How chemistry affects the spatial scale of air pollution

As discussed earlier, most of the sulfate in the atmosphere is formed from the atmospheric oxidation of SO_2. Sulfate is removed fairly efficiently by precipitation, leading to acid rain. Mercury, on the other hand, is emitted directly into the atmosphere in gaseous form (elemental, Hg(0), and divalent, Hg(II)) or particulate (divalent) form. Hg(0) is relatively insoluble (typically, only about one-millionth of Hg(0) will be present in cloud or rain drops), whereas most divalent forms (e.g., $HgCl_2$) are very soluble, and consequently, are easily removed by precipitation. Hg(0) is oxidized to Hg(II) by reaction with O_3, OH and some halogen species. Coal-fired power plants are major sources of both SO_2 and Hg (unless those emissions are controlled by scrubbers). It is, however, interesting to note that the spatial patterns of the atmospheric deposition of sulfate and mercury differ significantly (Figure 1). Sulfate deposition appears to be associated with SO_2 source areas, and accordingly, greater sulfate deposition occurs in the northeastern United States, downwind of the industrial Ohio River Valley. On the other hand, there is no spatial gradient for mercury deposition from Minnesota (upwind of the Ohio River Valley) to Pennsylvania (downwind of the Ohio River Valley). Instead, the



National Atmospheric Deposition Program/National Trends Network

National Atmospheric Deposition Program/Mercury Deposition Network

Figure 1. Annual atmospheric wet deposition fluxes of sulfate (top, in kg/ha-y) and of mercury (bottom, in μg/m^2-y) in the United States in 2001 (Source: National Acid Deposition Program and Mercury Deposition Network).

Sulfate deposition is greater in the Northeast whereas mercury deposition is greater in the South.

spatial gradient appears to be extending from the southern latitudes to the northern latitudes. These different behaviors are due mostly to the different atmospheric lifetimes of SO_2 and Hg(0). The oxidation of SO_2 to sulfate occurs slowly in the gas phase (on the order of 1% per h), but rapidly in clouds (on the order of minutes). On the other hand, the oxidation of Hg(0) by OH and O_3 occurs relatively slowly with a half-life on the order of several weeks (oxidation by halogen species can be rapid but it occurs mostly in limited environments, such as the marine boundary layer and the Arctic and Antarctic). As a result, mercury has a long atmospheric lifetime, and its deposition is dominated primarily by oxidant concentrations and global sources.[25] These results exemplify the fact that the relationship between an effect (here, the deposition of air pollutants) and a cause (the emission of the air pollutant or its precursor), depends on meteorology for its transport (by the winds) and removal (e.g., by precipitation), but is also a strong function of its chemistry. Therefore, both transport phenomena and chemistry must be taken into account when analyzing air pollution. It is interesting to note that current mathematical models of atmospheric mercury tend to overestimate mercury deposition in the northeastern United States, i.e., predicting spatial patterns that approach those of sulfate deposition.[25] This suggests that some important chemical mechanism may still be missing in our understanding of mercury chemistry. Therefore, comparing model simulation results to available measurements is a critical step in the development and application of air pollution models.

Particulate matter: A complex mixture of many compounds

Particulate matter (PM) is composed of a large number of chemical compounds. Some PM compounds are directly emitted into the atmosphere (primary PM: dust, soot, fly ash, sea salt, etc.), whereas other PM compounds are released or formed in the atmosphere as gases that subsequently nucleate to form new particles or condense on existing particles (secondary PM: sulfate, nitrate, ammonium, organic compounds, etc.).

Identifying the sources of PM can be a challenging task, particularly when there are large uncertainties associated with the emissions of PM and the formation of secondary PM. The receptor modeling techniques mentioned previously have proven to be very valuable tools. Figure 2 shows the results of an analysis of PM samples collected in Seattle, WA.[26] The analysis used trace metals, as well as different fractions of carbonaceous particulate material (the different fractions were obtained by heating the collected sample at different temperatures under nonoxygenated and oxygenated atmospheres). This approach was successful in apportioning the PM concentrations to gasoline and diesel vehicles, as well as other sources (incinerator, oil combustion, wood smoke, sea salt, soil, paper mill, ferrous metal processing) and PM constituents formed in the atmosphere (sulfate and nitrate).

Reducing the levels of PM is important because PM affects human health, degrades atmospheric visibility and soils materials. To reduce PM, one must target particular components, typically those that constitute the largest fraction of PM. For the components of PM that are directly emitted in the atmosphere as particles, the source apportionment provided in Figure 2 can provide the foundation for an emission control strategy. However, for the components that are formed from gases in the atmosphere, a receptor model cannot predict how the component would respond to a change in the source emissions. It is necessary then to use mathematical models to relate air pollutants to sources, and to develop emission control strategies that are efficient for PM reduction, and also do not adversely affect other pollutants, such as ozone. Those mathematical models must be carefully evaluated against data to ensure that they not only predict the correct PM concentrations and chemical composition, but also will predict the correct response to changes in precursor emissions.

Figure 3 presents the results of model simulations conducted to demonstrate the possible response of PM components to changes in the emissions of precursors.[27-29] Here, we show the changes in particulate sulfate, nitrate and organic compounds resulting from changes in the emissions of their respective precursors, SO_2, NO_x, and VOC. What appears is that the reduction in a precursor leads to a reduction in the corresponding oxidation product (for example, a 50% reduction in SO_2 emissions across the United States leads to reductions in ground-level sulfate concentrations of up to 2 μg/m^3). However, it also appears that other PM components may increase. For example, a decrease in SO_2 emissions leads to an increase in nitrate concentrations in many areas because some of the ammonium that is associated with sulfate becomes available to form ammonium nitrate. A decrease in VOC emissions leads to increases in sulfate and nitrate concentrations because more oxidants become available to oxidize SO_2 to sulfate and NO_x to nitrate, and also, less gaseous organic nitrates are formed making particulate inorganic nitrate formation more likely.

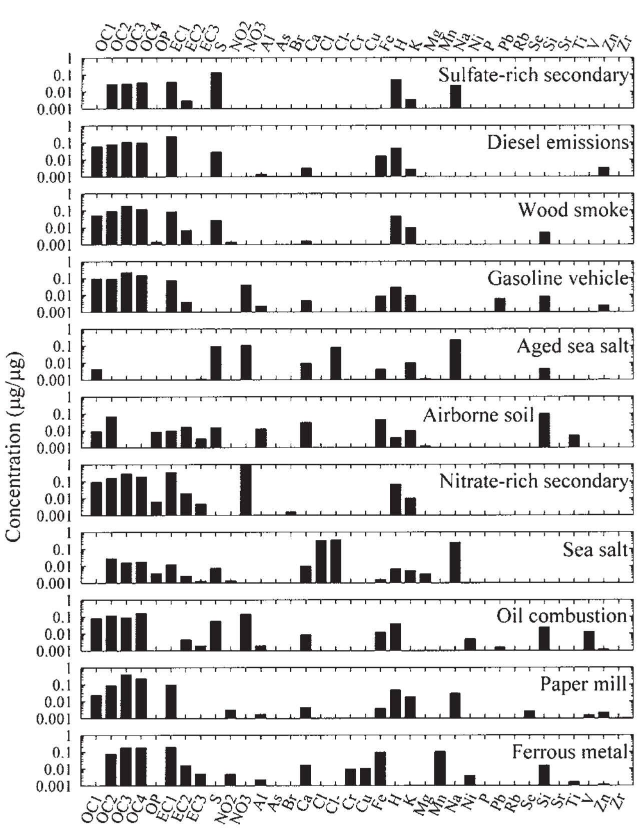


Figure 2. Air pollution source profiles deduced from atmospheric particulate matter samples collected in Seattle, WA.

Thirty-six chemical species measured in 384 samples collected from 1996 to 2000 were analyzed using factor analysis. The identified sources and their average contributions to the particulate concentrations were (from top to bottom) sulfate-rich secondary aerosol (26%), diesel emissions (22%), wood smoke (16%), gasoline vehicles (10%), aged sea salt (4%), airborne soil (7%), nitrate-rich secondary aerosol (5%), sea salt (4%), oil combustion (3%), paper mill (2%) and ferrous metal processing (1%) (Source: Aerosol Science & Technology: Factor analysis of Seattle fine particles. (38):724-738. Copyright 2004. Mount Laurel, NJ. Reprinted with permission.“[26]).

These results show that the control of air pollution is a difficult problem because of the intricate relationships that exist among the air pollutants and the various precursors.[24, 30, 31] It is, therefore, desirable to use several techniques to investigate an air pollution problem (for example, combining mathematical modeling of air pollution with receptor modeling techniques and indicator-species techniques) to ensure that the uncertainty of the overall analysis is minimized.

Intercontinental transport: combining experimental and modeling approaches

Ozone became a major problem in urban areas when gasoline-powered vehicles became widespread. As metropolitan areas have grown, ozone has indeed become a regional problem and even a global problem. Ozone concentrations before the industrial era were on the order of 10 parts per billion (ppb) (i.e., 10 molecules of ozone per billion molecules of air). Today, global background ozone concentrations are about 15 to 35 ppb,[32] and 1-h ozone concentrations can exceed 200 ppb in the most polluted urban areas, such as Los Angeles, Houston (www.epa.gov) or Mexico City.[33] For reference, the EPA national ambient air quality standards for ozone are 120 ppb for 1-h time averaged concentrations and 80 ppb for 8-h time averaged concentrations. Other air pollution problems typical of urban areas include carbon monoxide (CO) and PM.

It has become increasingly apparent that urban air pollution can be exported to long distances at regional and even global scales. Intercontinental transport of “urban” air pollutants, such as ozone, PM and CO has become evident via observational and modeling studies.[34-38] Observational evidence of intercontinental transport is obtained at ground-level sites, aloft with instrumented aircraft and over the vertically integrated atmosphere via satellites. Several global models of air pollution have been developed and are now used to simulate the intercontinental transport of air pollution. Figure 4 presents a comparison of CO concentrations vertically integrated through the atmosphere as obtained from satellite measurements and from a model simulation.[37] Two major conclusions can be drawn from this figure. First, the model simulation reproduces the major spatial pattern of global CO concentrations well, although there remain some areas of uncertainties (e.g., overestimation in India). Second, there is some clear evidence of intercontinental transport across the Pacific Ocean from Asia to North America, and across the Atlantic Ocean from North

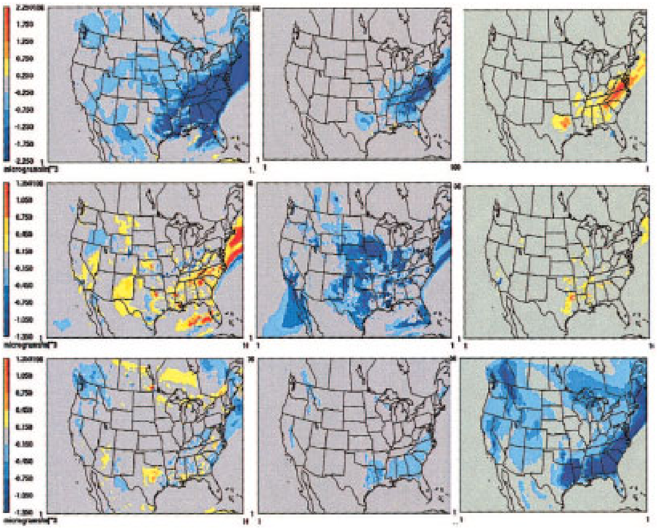


Figure 3. Simulation of the changes (in $\mu g/m^3$) in the atmospheric ground-level concentrations of fine particulate sulfate (top row), nitrate (middle row) and organic compounds (bottom row) due to 50% reductions in emissions of SO_2 (left column), NO_x (center column), and VOC (right column) with the CMAQ-MADRID-1 model[27] for July 1-10, 1999 over the contiguous Untied States (Source: adapted from Pun et al.[28] and EPRI[29]).

Decreases in emissions of a precursor lead to decreases (blue colors) in the associated pollutant (i.e., along the main diagonal from top-left to bottom-right showing the effect of SO_2 on sulfate, NO_x on nitrate, and VOC on organic compounds). However, they may lead to increases (yellow and red colors) in the other pollutants.

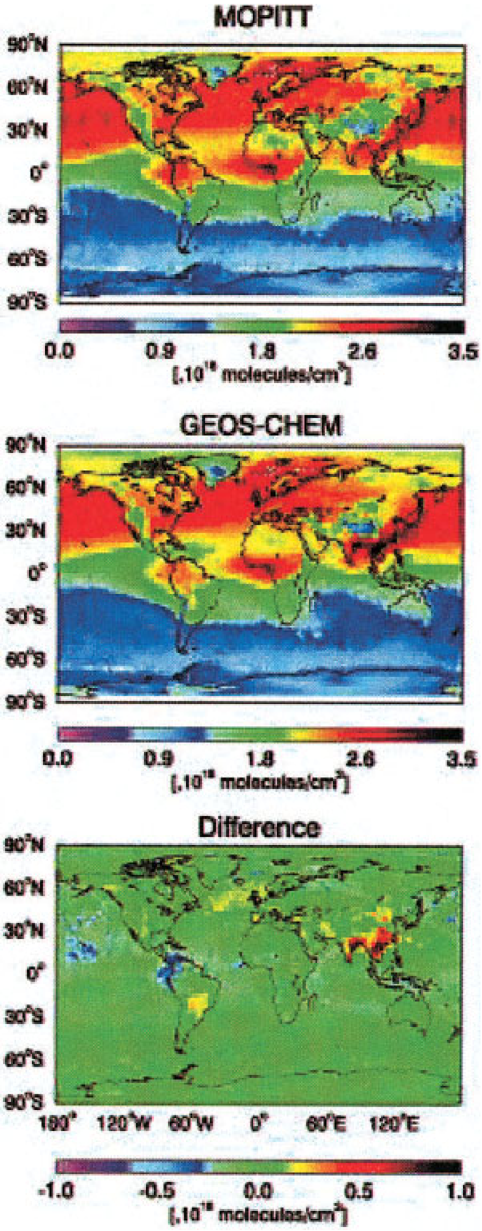


Figure 4. Mean atmospheric concentrations of CO (vertically integrated) measured by the MOPITT satellite (top) and simulated with the GEOS-CHEM model (middle) for the 20 February - 10 April 2001 period.)

Differences between the model simulation and the satellite data are shown at the bottom. Transport of CO over the Pacific and Atlantic oceans appears in both the satellite data and model simulation (Source: Heald, C.L., D.J. Jacob, A.M. Fiore et al., "Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft, and model perspective," *J. Geophys. Res.*, 108, doi: 10.1029/2003JD003507 (2003)[37], Copyright (2003) American Geophysical Union, reproduced by permission of American Geophysical Union).

America to Europe. Similar efforts are being carried out for other pollutants, such as ozone and PM in order to better quantify the possible influence of continents on each other in terms of air pollution export.

Perspectives: future developments

The techniques used to analyze air pollution will continue to evolve and will allow us to better understand, manage, and forecast air pollution at various local, urban, regional, continental and global scales. Some of the most promising areas of research and development are summarized next.

Air Pollution Forecasting. Air pollution forecasting has been conducted by air quality agencies for many decades using statistical models that relate air pollution levels to meteorology and air pollution levels on previous days. Although those empirical relationships have produced reasonable forecasts in many cases, they are limited in their applications to small areas and predictable conditions. The recent ability to conduct meteorological and air quality simulations relatively fast now offers the possibility to use such models as the basis of air quality forecasts. However, several challenges must be addressed. First, the equations that govern the motion of air masses in meteorological models are chaotic,[39, 40] which makes long-range weather forecasting a very difficult problem (a small initial error grows as the simulation proceeds); the forecasting of air pollution will carry the same limitations as weather forecasting. Second, the uncertainties in the emissions of air pollution sources, our limited understanding of certain air pollution phenomena (e.g., physics of the planetary boundary layer, cloud microphysics and cloud/aerosol interactions[41]), and the multiscale nature of air pollution must be addressed in a satisfactory fashion. Considerable progress has been made over the past few years and several organizations including the French Ministry for Ecology and Sustainable Development (www.prevair.org), Environment Canada, the U.S. Environmental Protection Agency (EPA)/National Oceanic and Atmospheric Administration (NOAA),[42, 43] and private firms[44] now use (or plan to use) mathematical models for air quality forecasting. Comparison of model simulation results with actual observations of air pollution suggests that air quality models have now reached a stage of maturity (at least for ozone if not yet for PM) that makes their use for forecasting possible.[44, 45] Figure 5 presents the results of an air quality forecast conducted with the CHIMERE model,[46] and available from the French system Prév'air on the web site of the INERIS (Institut National de l'Environnement et des Risques Industriels; www.prevair.org). Clearly, the accuracy of such air quality forecasts will improve as data on meteorology, air quality and emissions can be incorporated into such simulations in real-time.

Scientific advances in understanding atmospheric aerosols. The simulation of PM probably presents the greatest challenge in air pollution at the moment because of the complex nature of atmospheric particles.[18] Currently, we understand reasonably well the formation of the secondary inorganic component of PM, and the remaining uncertainties are due primarily to uncertainties in the meteorological fields (e.g., clouds and precipitation, vertical mixing) and the emission inventories. The largest uncertainties are associated with the secondary organic component of PM, because it consists of hundreds of chemical compounds that have not been completely identified.[47, 48] Ongoing efforts in model development include the detailed description of particles according to their original chemical composition and their evolution as new material condenses on those existing particles,[49] and the develop-

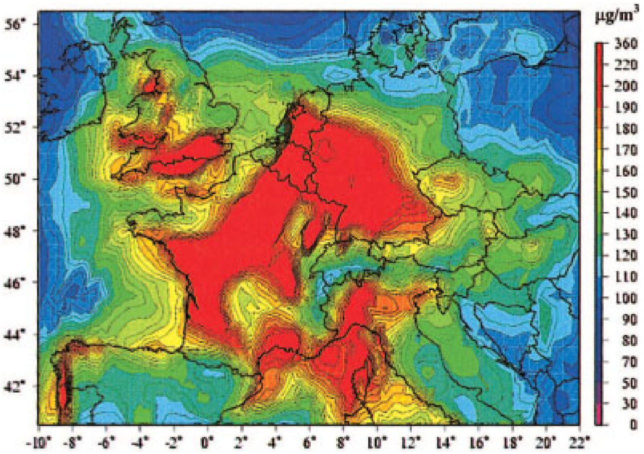


Figure 5. Air pollution forecast of ozone (µg/m³) over western Europe on August 8, 2003 using Prév'air (Source: www.prevair.org).

Unusually high temperatures led to high ozone concentrations during that period.

ment of new thermodynamic models of gas/particle equilibrium that will treat inorganic and organic compounds jointly.[50] Measurements of individual particles have provided valuable insights into the chemical composition of atmospheric PM,[49] and measurements of the evolution of the size-resolved particle chemical composition shed some new light on the processes that govern the particle size distribution.[51, 52] Nevertheless, we are not able today to characterize the full chemical composition of atmospheric particles. There is a dire need to obtain a more complete representation of the organic compounds present in atmospheric PM, because until such information becomes available, it will not be possible to ensure that the models used to simulate PM are indeed simulating the correct processes.

Use of satellite data. Satellite data have started to be used more routinely over the past few years to provide a more complete picture of the concentrations of some air pollutants. These data are being used to better evaluate the performance of air quality simulation models and to study the long-range transport of air pollutants. The forthcoming years should see an increased use of satellite data providing continuous two-dimensional mapping of vertically-integrated concentrations of CO, O_3, NO_2, SO_2, HCHO and PM.

Data assimilation. Inverse modeling has been used to identify the areas of the emission inventory that may be incorrect.[53] The simulation of meteorology has for some time now used the assimilation of meteorological data (e.g., winds and temperature) during the simulation to improve the quality of the model simulation. Similar techniques are starting to be applied in air quality modeling.[54] However, more progress is needed in this area to improve the accuracy of air quality simulations by assimilating actual observations of air pollutant concentrations, particularly aloft.[55] Assimilation of cloud and precipitation data will also greatly improve the meteorological predictions.

Multiscale modeling of air pollution. The importance of intercontinental transport for many pollutants will require the increased use of global models. On the other hand, the adverse effects of air pollution need to be assessed at the local scale (urban area, ecological system). Therefore, the use of modeling systems that include nested grids from the global scale to the regional or urban scale[25, 56] will become more widespread. Moreover, some atmospheric processes are typically not resolved at the spatial scales of the 3-D grid models (e.g., chemical reactions occurring in stack plumes, dispersion of pollutants from a roadway). Subgrid-scale treatments of such processes should be included to provide a more complete and accurate global-to-local representation of air pollution.[57]

Combining meteorology, emission and air quality models. Historically, meteorological, emission and air quality models have been applied in sequence. Over the past decade, meteorological and air quality models have started to be either run in an interactive mode (i.e., air pollution feeding back into the meteorology),[56] or integrated into a single model[55, 58] (thereby, avoiding having different formulation of atmospheric physics in the two models). As the need for rapid and accurate air quality forecasts increases, the simulations of meteorology, air pollutant emissions and air quality are likely to become more integrated within new modeling systems (examples include the Weather Research and Forecast Model with Chemistry, WRF-CHEM, being developed by NOAA and EPA and MOCAGE[55] developed by Météo France).

Combining various modeling techniques to analyze air pollution. Receptor models (i.e., observation-based techniques) and mathematical models of air pollution have historically been developed and applied by different researchers. As it becomes increasingly clear that the uncertainties associated with individual techniques cannot be totally eliminated, it is desirable to use several of those techniques in combination to address complex air pollution problems. A recent success story of such a combined approach to problem solving was the source apportionment conducted for regional haze in Big Bend National Park in Texas.[59] The study focused on sulfate particles because they contributed the most to atmospheric visibility impairment in the park. Two different air quality models were applied along with several receptor modeling techniques to estimate contributions of source areas (selected from the contiguous United States and Mexico) to particulate sulfate concentrations in the park. After several iterations of model evaluations, comparisons and critiques, the results of the different modeling techniques were found to be in general agreement.

Computational resources. As more information keeps being integrated in the models of air pollution, the computational requirements will keep increasing. Currently, the most demanding air pollution simulations, such as the detailed simulations conducted by Jacobson et al.,[60] and Kleeman et al.[49, 61] are performed on clusters of several tens of Linux PCs. As the efficiency of computers continues to increase, the amount of scientific detail that will be integrated into our representations of the polluted atmosphere will also increase. Finding a suitable compromise between accuracy and computational efficiency will continue to be a challenge for the scientific community.

The role of chemical engineers. Air pollution science is a multidisciplinary field that involves an interplay among health scientists, meteorologists, chemists, engineers and other scientists. As discussed earlier, chemical engineers have played a pivotal role in the development of advanced techniques to analyze and simulate air pollution. The chemical engineering discipline is ideally suited to study air pollution because of its unique combination of transport phenomena, thermodynamics, and chemical reactions. It is uniquely qualified to deal with the integration of the atmospheric transport processes governed by meteorology and complex chemical reaction mechanisms leading to air pollutant formation in the atmosphere. However, the current number of chemical engineers actively involved in air pollution science appears to be less than one would expect based on the relevance of their possible contributions. As new measurement techniques and modeling tools continue to be developed, chemical engineers can contribute to both the fundamental scientific progress that will occur over the coming years, and the application of those techniques to resolve practical air pollution problems.

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