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UNDERSTANDING AIR POLLUTION-THE PAST AND THE FUTURE

Main menu

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Abstract: This paper illustrates the steps involved in understanding air pollution problems through two examples: acid deposition and urban air pollution. We show how modeling acts as an intermediary between fundamental understanding obtained through idealized experiments and observations made in the real world. In understanding acid deposition, comprehensive modeling demonstrated the relevance of laboratory results on the aqueous phase oxidation of SO_2 by H_2O_2 to explaining observations of ambient sulfate as well as sulfur in rain. In the second example, we show that concentrations of air pollutants in complex urban areas can be estimated using relatively simple models based on local values of turbulence and mean flow. We also show that inputs to such models can be estimated using measurements made on towers located in urban areas. We conclude by examining the increasing reliance on comprehensive numerical models, which are likely to become much more important in the future.

Keywords*: Air pollution, dispersion, acid deposition, urban pollution, micrometeorology, numerical models, laboratory experiments.*

INTRODUCTION

Understanding an air pollution problem involves the following interacting steps: 1) Study the fundamental processes that govern the problem, 2) Incorporate this fundamental understanding into a comprehensive or semi-empirical air pollution model, 3) Conduct field or laboratory studies to collect data to evaluate the model, 4) Evaluate the model with data and improve the model, and 5) Use model to conduct sensitivity studies. The last step is critical because the model becomes a surrogate for reality and allows us to conduct numerical experiments that would be impossible in the real world. In this paper, we illustrate the steps involved in understanding an air pollution problem by considering two problems: acid deposition and urban air pollution. In doing so, we examine how modeling has evolved over the past twenty years, and what we can expect to see in the future.

Acid Deposition

Acid deposition refers to the wet and dry deposition of acidifying pollutants, the precursors of which are emissions of SO_2 and NO_2. These pollutants are converted into sulfuric and nitric acids as they are transported over long distances. In the 1970s and 1980s, there was concern in Europe and in the Northeastern United States and Canada was acid deposition was causing damage to lakes and forests. This led to the development of several relatively simple Lagrangian models (Eliassen, A. and J. Saltbones, 1975; Fisher, B.E.A. 1978) that were used to estimate the contribution of sources to acid deposition at receptors. These early models converted the precursor gases to the secondary acidifying products using linear rates that were essentially parameters obtained by fitting model estimates of sulfur and nitrogen deposition to corresponding observations. The understanding of chemistry then indicated that the primary mechanism for conversion of SO_2 and NO_2 to the corresponding acids was the gas phase oxidation by the OH radical (Stockwell, W.R. and Calvert, J. G. 1983). So the wet deposition pathway of sulfur was thought to consist of gas conversion of SO_2 to sulfate followed by efficient removal of sulfate by rain; the dissolution of SO_2 in rain is not an efficient removal mechanism. However, the wet removal rate through the gas phase oxidation of SO_2 was not consistent with the empirically determined rates. Furthermore, the gas phase concentrations of sulfate were underpredicted if the relatively slow oxidation by the OH radical was used in the acid deposition models.

Subsequent understanding of atmospheric chemistry indicated the major role of aqueous phase oxidation of SO_2 by H_2O_2 (Kunen, S.M. *et al.*, 1983; Lind, J.A. *et al*., 1987). The dissolved SO_2 could be rapidly converted to sulfuric acid in cloud droplets, which could explain the rapid removal by rain. It also provided a likely explanation for the gas phase concentrations of sulfate: the sulfate formed in clouds entered the gas phase when non-precipitating clouds evaporated. This fundamental understanding of aqueous phase chemistry from the laboratory had to be incorporated into a atmospheric model before it could be confirmed as the actual explanation of observations. This happened in the 1980s when comprehensive models, such as ADOM (Venkatram, A. and Karamachandani, P. 1988) and RADM (Chang, J.S. *et al*., 1987) were developed. These numerical models incorporated the governing processes, transport, deposition, and chemistry in as much detail as possible in an Eulerian framework. Most importantly, they included cloud models with aqueous phase chemistry. The importance of aqueous phase oxidation of SO_2 was confirmed when predictions of wet deposition and sulfate in air from such models compared well with actual observations (Karamchandani, P. and Venkatram, A. 1992).

Sensitivity studies with the numerical models showed that the central role of aqueous phase oxidation is associated with the concept of oxidant limitation. The wet deposition of SO_2 is controlled by the availability of the primary oxidant H_2O_2. Thus the concentration of SO_2 that is incorporated into clouds and converted into sulfate is limited by the concentration of H_2O_2: 1 ppb of H_2O_2 will remove only 1 ppb of SO_2, so that any excess of SO_2 above 1 ppb is not removed. This has important implications for control of SO_2 emissions to reduce acidic deposition. If the emissions result in concentrations of SO_2 above the atmospheric concentrations of H_2O_2, emission control will have little effect on wet deposition until the SO_2 levels drop below the H_2O_2 concentrations. This concept is illustrated in Figure 2.

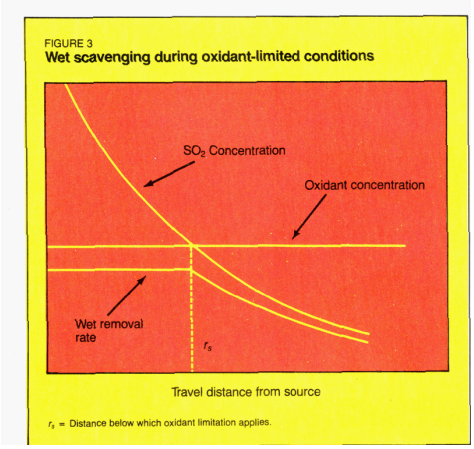

Figure 2: Concept of oxidant limitation

This chronology of model development illustrates how understanding of acid deposition was obtained through an interaction between laboratory results, modeling, and field observations.

Urban Air Pollution Problems

We confine our discussion here to urban pollution problems related to exposure to pollutants at source-receptor distance of tens of meters to kilometers. Although chemistry might be important at these scales, we will assume that transport and dispersion govern pollutant concentrations. Some of the sources of urban air pollution that are important at these scales are small power plants and automobiles. The early urban pollution problems, such as the one that caused the famous London smog in 1952, were related to burning of high sulfur coal in homes. While such combustion related air pollution is still a problem in developing countries, the primary air pollution problem in developed countries is perceived to be the formation of secondary photochemical pollutants over scales of tens of kilometers. Lately, more attention is being paid to exposure to traffic related primary emissions and deliberate releases of toxics in urban areas.

One of the earliest studies of dispersion in urban areas was conducted in St. Louis Missouri in the period 1963-65 (McElroy, J.L. and F. Pooler, 1968), in which a tracer was released at several locations in an urban area and then sampled at distances ranging from 800 m to 16 km. Routine meteorological data were collected to characterize the dispersion conditions during the experiment. The data collected during this experiment formed the basis of the urban dispersion curves (Briggs, G.A. 1973; Venkatram, A. 2005) used until recently in most dispersion models. The major conclusion from this early study was relatively simple Gaussian dispersion models can provide estimates of ground-level concentrations if the appropriate meteorological parameters are used.

More recent urban experiments in Switzerland (BUBBLE experiment, Rotach, M.W. *et al*. 2005) and the US (Salt Lake City (Allwine, K.J. *et al.* 2002; Hanna, S.R. *et al.* 2003), and Oklahoma City (Allwine, K.J. *et al.* 2004), Barrio Logan (Venkatram, A. *et al.* 2004)) have taken advantage of progress in making measurements of flow and turbulence using sonic anemometers. These studies indicate that we can make reasonable estimates of near concentrations beyond 100 m from the source of plume spreads are estimated using local measurements of flow and turbulence. Figure 3 shows a comparison of model estimates with measurements in Barrio Logan, California (Venkatram, A. *et al.* 2005).

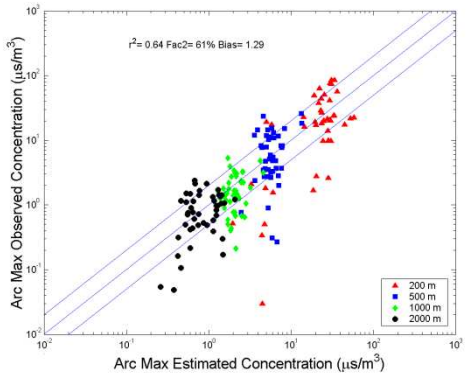

Figure 3: Comparison of tracer concentration measurements made in Barrio Logan with model estimates

This has led to research into estimating urban micrometeorology from measurements made in rural areas or on towers in urban areas. Several studies indicate that measurements of mean wind speed and temperature fluctuations on an urban tower can provide useful estimates of micrometeorology relevant for dispersion (Princevac, M. and Venkatram, A. 2007;

Venkatram, A. and Princevac, M. 2008; Qian, W. *et al.* 2010). Figure 4 shows an example where the measurements were made at three urban towers in Riverside, CA.

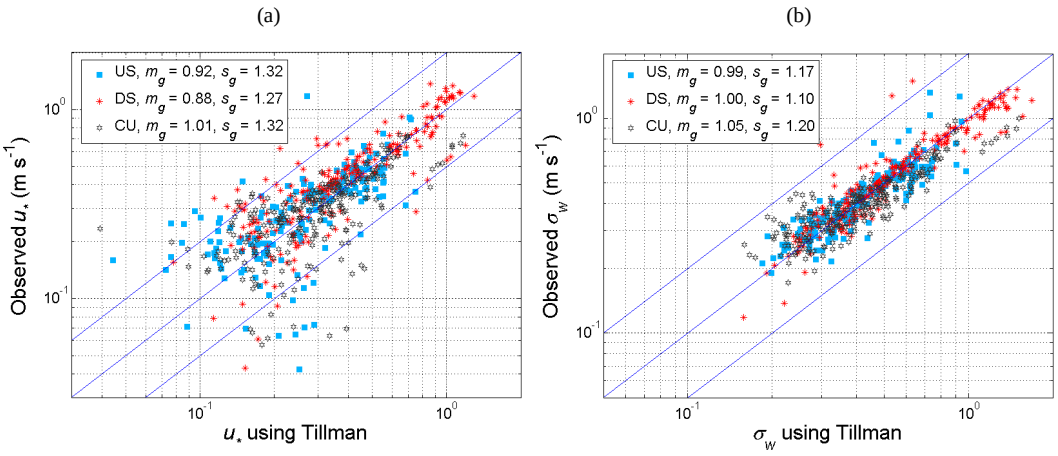

Figure 4: Comparison of estimates of micrometeorological parameters with measurements made at three urban sites in Riverside.

We have made progress in estimating the contribution of traffic related emissions to concentrations of pollutants within tens of meters from the road (Venkatram, A. *et al.* 2008). Figure 5 shows an example of this application (Venkatram, A. *et al.* 2009).

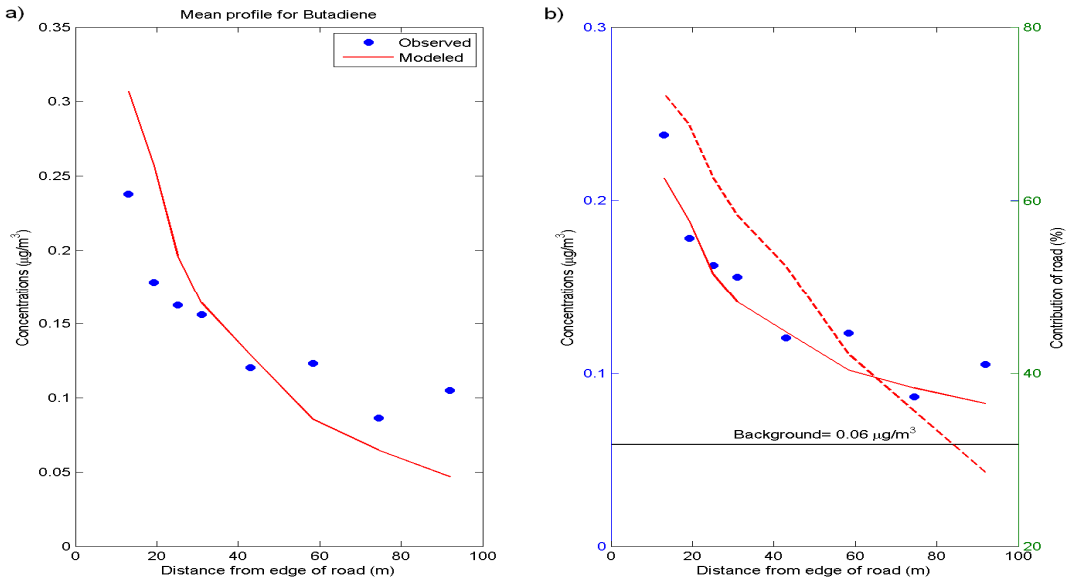

Figure 5: A dispersion model is used to estimate the contribution of traffic to butadiene concentrations measured near a road in Raleigh, NC.

The relatively simple dispersion models described earlier cannot be used to estimate concentrations at small distances from the source where the details of the flow field and turbulence are affected by local building geometry (Rotach, M.W. 1999; Rotach, M.W. *et al.* 2004). Under these circumstances, it might be necessary to use numerical models that solve the mass and momentum conservation equations. Although there have been successes in such modeling efforts, it is not clear that they do much better than the simpler Gaussian dispersion models. This means that there is a great deal of uncertainty in estimating concentrations near buildings. This is especially true for sources such as a small distributed generator (Allison, J.E. and Lents, J. 2002; Carreras-Sospedra, M. *et al.* 2008) located in an urban area. Here the buoyant plume from the source interacts with the complicated flow in the vicinity of the power plant; the flow is governed not only by the immediate building but all the buildings of the source. Under these circumstances, water channels and wind tunnels can provide importance guidance. Figure 6 provides an example of the type of results that can be obtained from such studies (Pournazeri, S. *et al.* 2010). Ultimately such results have to be converted into a model that can be used to estimate concentrations.

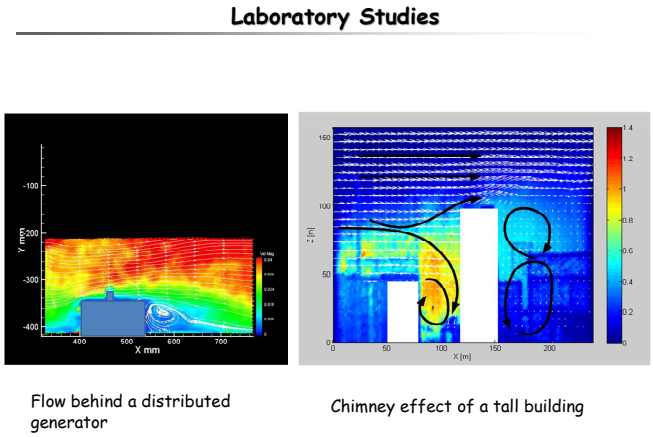

Figure 6: Results from water channel studies on building effects on flows.

The Future

It is clear that current trends in modeling indicate an increasing reliance on numerical models (e.g. Baik, J.-J. *et al.* 2003; Hanna, S.R. *et al.* 2007; Kim, J.-J. and Baik, J.-J. 2004; Smith, W.S. *et al*. 2001), which in principle can handle complex physical processes without the approximations used in simpler models (Cimorelli, A.J. *et al.* 2005; EPA, 1995; Venkatram, A. 1986). However, such models can become so complex that interpreting results can become a mere description of the results rather than a way of obtaining insight into the dominant processes; it might be necessary to use a simpler model to gain insight into results from the complex model. Numerical models are prone to numerical error that can mislead or swamp real effects. Another problem with comprehensive numerical models is that computational resource constraints forces one to use grid sizes that might lead to incorrect physical effects. For example, pollutant emissions that are widely separated in a grid are mixed instantaneously through the grid and thus undergo chemical reactions that do not occur in reality. We encounter similar problems in modeling dispersion of plumes where the grid size rather than turbulence governs mixing. There are plume in grid models (e.g. Karamchandani, P. *et al.* 2006) that purport to solve this problem, but the results are not yet widely accepted. These problems are likely to be solved in the future when computational resources as well numerical methods improve. Until then it is prudent to rely both on simple semi-empirical models and comprehensive models to understand air pollution problems.

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