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Computed Tomography and Optical Remote Sensing: Development for the Study of Indoor Air Pollutant Transport and Dispersion

A.C. Drescher
(Ph.D. Thesis)

June 1995

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**Computed Tomography and Optical Remote Sensing:
Development for the Study of Indoor Air Pollutant
Transport and Dispersion**

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ABSTRACT

This thesis investigates the mixing and dispersion of indoor air pollutants under a variety of conditions using standard experimental methods. It also extensively tests and improves a novel technique for measuring contaminant concentrations that has the potential for more rapid, non-intrusive measurements with higher spatial resolution than previously possible.

Experiments conducted in a sealed room support the hypothesis that the mixing time of an instantaneously released tracer gas is inversely proportional to the cube root of the mechanical power transferred to the room air. The constant of proportionality is determined and the relationship is used to predict mixing times for two indoor air flow scenarios: forced ventilation and strong natural convection. Predicted mixing times agree well with observation. The empirical relationship helps bridge the gap between the common but mostly untested assumption of instantaneous, perfect mixing and complex computational models for simulating pollutant transport and dispersion.

One table-top and several room-scale experiments are performed to test the concept of employing optical remote sensing (ORS) and computed tomography (CT) to measure steady-state gas concentrations in a horizontal plane. Various remote sensing instruments, scanning geometries and reconstruction algorithms are employed. Reconstructed concentration distributions based on existing iterative CT techniques contain a high degree of unrealistic spatial variability and do not agree well with simultaneously gathered point-sample data. A new reconstruction method, Smooth Basis Function Minimization (SBFM), is developed. SBFM treats concentration distributions as continuous and representable by a superposition of asymmetric smooth functions — bivariate Gaussians are used in this thesis. SBFM reconstructions of synthetic and experimental data show much better agreement with measured concentration profiles than reconstructions based on standard techniques.

The dispersion of tracer gas in a chamber is examined using a scanning open-path FTIR and SBFM computed tomography. Interpolation of successive path-integral concentration measurements for each ray yields input data for CT reconstructions at specific times during the experiment. This allows one to follow the evolution of the tracer gas concentration distribution in time. The results demonstrate significant potential for CT/ORS as a fast, accurate and relatively non-invasive technique to measure time-varying gas concentrations in a variety of applications.

*Dedicated to my mother and father,
Gisela and Heinz Drescher,
whose dedication as parents has opened up
a world of opportunities for me.*

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CHAPTER 1

INTRODUCTION

1.1 Indoor Air Quality and the Study of Pollutant Dispersion

Indoor air quality is of great importance for human well-being. In the industrialized world, people tend to spend most of their time indoors. They are exposed to numerous air pollutants emitted indoors, brought in from the outdoors with ventilation air, or from the soil by natural or forced ventilation (Nero, 1988). A major pollutant of indoor origin is environmental tobacco smoke. Unvented wood-, gas- or coal-fired stoves and heaters produce carbon monoxide, sulfur dioxide, oxides of nitrogen and particles. Volatile organics emitted from building materials, office machines and consumer products are often implicated in sick building syndrome studies (Mendell, 1994). Microbial organisms and products they emit can cause allergic reactions and even illness (Miller, 1992). In occupational settings, workers are exposed to a variety of chemicals used in manufacturing and chemical synthesis. Major pollutants brought into buildings from the outdoors include radon from radium-rich soils, ozone from polluted urban air and a host of particulate and gaseous contaminants, especially in areas with a high concentration of traffic or heavy industry.

In addition to posing health risks to humans, indoor air contaminants constitute a threat to sensitive equipment (Weschler and Shields, 1991) and works of art (Nazaroff, 1989). Due to the health risks and potential material damage associated with exposure to indoor air contaminants, it is important to understand the factors that control indoor air pollutant concentrations. Among these factors are transport and mixing.

The underlying processes leading to transport and mixing are fairly well understood. The fate of contaminants can be described by the fundamental conservation equations of mass, momentum and energy, combined with knowledge about generation,

reaction and scavenging processes in the indoor environment. However, owing to the complexity of most indoor settings and that of the differential equations themselves, accurately formulating and solving the conservation equations is often extremely difficult and time-consuming. In many analyses of indoor pollutant behavior, perfect and instantaneous mixing is therefore assumed. This assumption is generally justified, especially when forced ventilation is supplied according to existing guidelines, and accurate enough for non-critical applications. But there is evidence that the assumption is incorrect in various contexts of importance to human well-being. These situations almost always involve localized indoor sources of air contaminants. Several examples are given below.

Cigarette smoking is one of the most ubiquitous sources of indoor air pollution. Environmental tobacco smoke (ETS), inhaled by non-smokers and smokers alike, is now estimated to cause 3000 excess lung cancer deaths among non-smokers and respiratory tract infections in between 150,000 and 300,000 infants annually in the United States (U.S. Environmental Protection Agency, 1992). Many public places, such as restaurants, bus terminals and airplanes, are divided into smoking and non-smoking sections in an effort to protect non-smokers. There is often no physical barrier separating the two sections. If the entire area were perfectly mixed there would be no difference between smoker's and non-smoker's exposure to ETS, so this is clearly not an appropriate working hypothesis in these cases (Oldaker and Conrad, 1987). Just how different are ETS concentrations in the two sections? What factors influence the difference? How can the difference be maximized? Experimental tools permitting the measurement of spatially resolved concentrations would be helpful to resolve such questions.

Tuberculosis (TB) is an airborne communicable disease of renewed concern in the United States. The past few years have seen an increase in reported cases. Health-care workers are especially at risk from inhalation of *mycobacterium tuberculosis* exhaled by patients. Measures to reduce the potential for exposure and infection of people in health-

care facilities housing TB patients include recommending the use of very high ventilation rates and portable HEPA air filters in the TB patients' rooms (U.S. Department of Health and Human Services, 1994; Lobascio et al., 1994). There is evidence that mixing is by no means complete and instantaneous under these conditions (Miller-Leiden et al., 1995). To design ventilation and filtration systems that minimize disease transmission, studies are required that allow the assumption of complete mixing to be relaxed.

Several studies have been reported in which measurements of personal exposure to indoor contaminants have not been equal to single point measurements of indoor concentrations (Rodes et al., 1990; Budiansky, 1980). Proximity to pollutant sources and effects associated with body temperature and breathing may contribute to this discrepancy. Assuming complete mixing does not account for these effects.

Various modeling approaches have been made to relax the assumption of complete mixing slightly. The simplest is the use of a mixing factor, which represents the effective fraction of ventilation air that is completely mixed in the room (Ishizu, 1980). Another is the use of a multi-compartment model, in which each imaginary subsection of a room is assumed to be well-mixed and fractional air exchange rates exist between compartments (Özkaynak et al., 1982). A slightly more sophisticated model assumes turbulent puff diffusion of a point-source pollutant (Cooper and Horowitz, 1986). To verify such models, one must be able to make measurements of pollutant concentrations at many points in the indoor space. However, since this involves a substantial experimental effort, less than 10 sampling points are usually reported in such studies. The conclusions are therefore often rather weak and limited. A measurement method offering good spatial resolution would be very useful.

A thorough understanding of transport and mixing is important in another context: energy use. Heating, cooling and moving air for the purpose of ventilating buildings constitutes a major end use of primary energy. A central function of building ventilation is preventing excessive accumulation of air contaminants that are emitted indoors. Heating,

ventilation and air conditioning (HVAC) systems must also provide thermal comfort. Traditionally, these systems are designed to achieve close to perfect mixing in the ventilated space. In situations where either the pollutant source(s) or the area to be heated (cooled) is limited in extent, creating a well-mixed environment is an inefficient means of achieving the desired effect.

As concern about energy use has grown over the past few decades, new and different types of HVAC systems have been developed to improve ventilation effectiveness and efficiency. These new systems, such as displacement ventilation, do not produce an even distribution of contaminants and temperature. To study, understand and optimize such systems, one requires tools to test and model them. Presently, both the modeling and the experimental measurement tools are inadequate, as described below.

Substantial progress has been made in the past two decades in the numerical modeling of air movement and pollutant dynamics in buildings (Bauman et al., 1983). Due to the development of computational fluid dynamics (CFD), relatively accurate predictions of air *motion* are now possible. However, versatile CFD codes require workstations to run (even so, a code often must run for several hours to several weeks to model the behavior of a non-trivial system), and they generally can only treat quasi steady-state situations accurately (Post, 1994). Transient, 3-D phenomena, such as turbulence, are still too complex to predict. Yet, these phenomena are instrumental in dispersing pollutants and heat. Simplifying assumptions are often made to enable the calculation of transient or stationary-state phenomena in a reasonable amount of time. It has, however, thus far been difficult to thoroughly test the accuracy of numerical model predictions because of limitations in experimental measurement techniques.

Experimental studies are needed to better characterize forced and natural convection air flow in buildings and to validate numerical models (Fisk et al., 1988; ASTM, 1983). Tracer gas studies are widespread in indoor environmental research to help understand and characterize contaminant and heat transport and dispersion

(Yoshizawa, 1992). This type of research is especially common in the study of ventilation and air-conditioning systems (Mundt, 1993; Heiselberg and Bergsoe, 1992). However, experiments reported thus far are hampered by an inability to track pollutant or tracer gas concentrations quickly and accurately in space and time, as described in Section 1.2.

The research that makes up the bulk of this thesis was born out of the realization that by combining two fairly well established techniques one might be able to develop a new method for indoor pollutant and tracer gas concentration measurements that was faster, less intrusive and had better spatial resolution than existing methods. Such a new method would provide us with a tool to more effectively study the factors that govern pollutant transport and dispersion in the indoor environment, and to improve ventilation system design. One of the techniques making up this new method is optical remote sensing (ORS), which is becoming increasingly important as a tool for monitoring emissions and ambient pollutant concentrations outdoors (Air & Waste Management Association, 1992). The other technique is computed tomography (CT), developed and used extensively in the field of medical imaging to map slices of the brain and body (Kak and Slaney, 1988; Herman, 1979). By combining the rapid, non-invasive measurement abilities of optical remote sensing with the mapping capabilities of computed tomography, it was anticipated that a powerful tool could be developed with which to undertake more sophisticated experiments in indoor air research.

1.2 Gas Concentration Measurement Methods

Three basic methods exist for measuring gas concentrations in ambient air: (1) air is collected in the space of interest and analyzed elsewhere, (2) a sensor is placed at the point at which the concentration is to be determined, and (3) the attenuation of radiation (most commonly, infrared) passed through the air is measured and related to the concentration. All three methods were used at one time or another for the work reported in this thesis.

Method 1 has the advantage that the concentration can usually be determined quite accurately by instruments such as a gas chromatograph/mass spectrometer or a photo-acoustic detector, if care is taken to ensure sample integrity up to the time of measurement. However, this approach suffers three main disadvantages: (1) there is a time delay associated with moving the air to the measuring instrument, (2) the air flow and the concentration profile close to the sampling point may be disturbed by the action of sampling, and (3) the *spatial* sampling density is limited because many sampling points entail a great deal of equipment, such as tubing, sampling pumps and/or sampling bags.

Method 2 is especially convenient for measuring personal exposure. It is also useful for making concentration measurements at fixed locations, but problems similar to those of method 1 are encountered: (1) even 'real time' passive samplers usually have a response time on the order of minutes, since they are limited by the rate of molecular diffusion to the sensing element, (2) the relatively high cost of a real time passive sampler limits the number that can be used for any individual study, thus limiting spatial resolution, and (3) frequent readjustment of the zero and span concentrations is required because these monitors are prone to drift.

Non-intrusive measurements of gas concentration can be made with remote sensing technologies (method 3). Such optically based methods have recently begun to be widely applied for sensing outdoor pollutants. Their advantage over the two methods mentioned above is that measurements can be made very rapidly without interfering significantly with the air being analyzed. One technique, called differential absorption lidar (DIAL), is capable of range-resolved concentration measurements on the scale of hundreds of meters to hundreds of kilometers, with a typical spatial resolution of approximately 30 m (Air & Waste Management Association, 1992). However, for distances less than approximately 300 meters, present remote sensing devices yield only

path-integrated concentrations (total number of molecules in a beam path)¹. This greatly limits their applicability in indoor environments, where, if conditions are assumed to be well-mixed (so that spatially resolved information is not required), a single point sample measurement would not only suffice, but would be far cheaper and easier to employ. However, in a plane, spatial resolution of the information contained in integrated ORS concentration measurements can be achieved by gathering data along intersecting beam path and applying computed tomography.

1.3 Computed Tomography Methods

The word 'tomography' is derived from the Greek, *tomos* meaning 'slice', or 'section', and *graphein* meaning 'to write'. Computed tomography refers to a class of techniques in which the spatial distribution of some parameter defined in a plane is computed from an integrated measurement of that parameter (or of a closely related physical quantity) along various paths in the plane. Parameters of interest in the past have included temperature (Uchiyama et al., 1985), density (Faris and Byer, 1986), velocity (Sato and Shiraki, 1984) and concentration. Computed tomography is applied in fields as diverse as radiology, electron microscopy, geophysics, radio astronomy, holography, fluid dynamics and combustion research. The concept of inverting integrated data to obtain a distribution has even been extended to three dimensions (Colsher, 1977). This thesis is concerned specifically with the measurement of gas concentrations in ambient indoor air in a horizontal plane.

At the outset of the research presented in this thesis, there were two major types of CT reconstruction methods: analytic and iterative (Brooks and Di Chiro, 1975). Analytic methods, such as two-dimensional Fourier reconstruction, are limited to one basic type of beam geometry. Sets of parallel rays (or of rays fanning out from a single point at the

¹DIAL cannot measure concentrations at distances less than approximately 300 meters because the backscattered signal is too strong at these short distances, saturating the detector.

edge of the measurement plane), with each set completely covering the area of reconstruction, are rotated with respect to one another about a point near the center of the reconstruction plane. The line integral of the parameter of interest is measured along each ray in the set. The resulting concentration profile is described by an analytic function based on the sinusoidal functions. Because of the limitations on possible scanning geometries, this method does not appear feasible for indoor air applications on the scale of several meters.

Iterative methods are characterized by a discretized plane of reconstruction and an iterative approach to the final solution from an arbitrary first guess. The three main techniques are the algebraic reconstruction technique (ART), the iterative least squares technique (ILST) and the simultaneous iterative reconstruction technique (SIRT). Iterative methods are completely flexible with respect to scanning geometry and allow constraints and other prior knowledge about the concentration distribution to be incorporated into the reconstruction process. They are therefore ideally suited to applications in many different environments. Their performance is, however, very sensitive to the ray scanning geometry used. Iterative methods are computationally more intensive than analytic methods. They may also be underdetermined, that is, a variety of reconstructions may be possible for a single set of input data. Which reconstruction the iterative method converges on may depend on the first guess or the details of the iteration process.

This dissertation research has led me to develop a technique, named smooth basis function minimization (SBFM) which is, in effect, a combination of the iterative and analytic techniques. The underlying concentration distribution is modeled as an analytical function (based on bivariate Gaussians rather than sinusoids). Yet the characteristics of this function are determined by means of a quasi-iterative procedure aimed at minimizing the discrepancy between measured ray integrals and those calculated for the presumed underlying distribution. In the thesis I have employed ILST and ART in addition to

SBFM. Details of the individual reconstruction methods are found in the chapters in which they are employed.

1.4 Objectives of this Research

The idea of coupling ORS and CT for air pollutant measurements on the scale of meters or tens of meters had been previously investigated *theoretically* for two different applications: (1) remote outdoor air pollution measurement on an urban scale (Wolfe, 1980; Wolfe and Byer, 1982) and (2) industrial hygiene (Todd, 1990,1992). Wolfe tested a limited number of highly artificial concentration profiles using an analytical technique (the convolution backprojection method) and a fan beam geometry. Todd simulated concentration profiles composed of a superposition of bivariate Gaussians with rotational symmetry. She generated path-integral concentration data based on these profiles for a variety of iterative computed tomography algorithms (Todd and Ramachandran, 1994a) and scanning geometries (Todd and Ramachandran, 1994b). All calculations were based on a discretized plane of measurement. The scanning geometries explored in her work required at least two (and as many as forty) sources and detectors.

The theoretical investigations mentioned above were relatively extensive. Yet, no experiments had been undertaken using the coupled methods in their proposed applications. Having never been tested, the conclusions from previous work about the potential of CT/ORS for ambient concentration measurements were weak. One goal for this thesis research was to test the concept of CT/ORS experimentally and thereby to contribute to its maturation. In parallel to the development of CT/ORS, a second goal was to improve the current understanding of indoor pollutant dispersion by performing experiments using conventional point-sampling methods. The specific aim was to determine quantitatively how the rate of pollutant mixing in a room is affected by characteristics of the air flows within it.

1.5 Outline of the Thesis Contents

The thesis begins, in Chapter 2, with an experimental investigation into a process that often affects mixing in indoor environments: forced convection. A set of experiments is described, in which the time for an instantaneously released puff of carbon monoxide to mix evenly throughout a room is correlated with the mechanical power of air motion added to the room by blowers. The results are found to be consistent with a mixing model for stirred reactors based on turbulence theory and proposed by Corrsin (1957). The semi-empirical relationship between mechanical power and mixing time is applied to a typical ventilation scenario and a strong natural convection scenario. In both cases the predicted mixing time agrees well with observation. The experiments described in Chapter 2 were performed with conventional, passive personal CO monitors used as point samplers. Spatial and temporal resolution were adequate to determine mixing time, but were insufficient to yield information about detailed transport and dispersion mechanisms.

Chapter 3 describes our first step in the *experimental* CT/ORS development work. A bench-top, proof-of-principle experiment was devised and conducted to evaluate the performance of CT/ORS under ideal sampling conditions. This experiment involved the detection of a well-controlled and contained plume of sulfur hexafluoride (SF_6) and the measurement of the SF_6 concentration within and around its cross-section using infrared absorption and the ART algorithm. The CT reconstruction was compared with point sample data gathered in the same plane. Tests of the CT program and scanning geometry using a variety of synthetic, noise-free data had predicted excellent results. The reconstruction of the experimental data was much less ideal due to fluctuations in concentration at the plume boundaries coupled with fundamental characteristics of ART. However, the plume was correctly located and the average SF_6 concentration within the plume matched that of the point sampled data to within 30%.

The next stage of experimental CT/ORS testing involved a room-scale chamber experiment under slightly less controlled, but still steady-state conditions. This experiment

is described in Chapter 4. An open path Fourier transform infrared spectrometer (OP-FTIR) was used to gather path-integral concentration data, and the algebraic reconstruction technique ILST was used for CT reconstruction. The experiment entailed moving the optical source and detector for each new ray position and required three hours to generate a single scan of the room cross-section. The long scanning time and the relatively sparse spatial coverage of the infrared rays resulted in a reconstruction that was poorer than would have been possible under more favorable sampling conditions. Experiments reported in Chapters 5 and 6 are based on the lessons learned in the experiment of Chapter 4.

Chapters 5 and 6 report a separate, extensive set of room-scale experiments and the development of a new CT reconstruction technique. In these experiments, which were performed in a larger chamber, the ORS system (OP-FTIR) and scanning geometry were reconfigured to allow much more rapid scanning, on the order of a few minutes per scan, and better coverage of the plane of reconstruction. Eleven steady-state experiments, involving between 9 and 15 scans each, and one non-steady dispersion experiment were completed. The new scanning geometry employed in this set of experiments resulted in ART-based CT reconstructions that were well-matched to the measured ray integral concentration data but not unique. They were also unphysical, being characterized by a high degree of spatial variability, and did not match the simultaneously gathered point concentration data. ART was not appropriate for the type of scanning geometry employed because it assumed no spatial correlation between different areas in the reconstruction plane.

A new CT reconstruction method was developed to incorporate the knowledge that gaseous pollutant concentrations in air are continuous. This method, called smooth basis function minimization (SBFM), was applied to the data sets from the experiments above and found to yield reconstructions that showed better agreement than ART with concentration maps generated from the point-sampled data. SBFM assumes that the

concentration profile underlying the measured path-integral concentrations can be represented by a superposition of asymmetric, bivariate smooth functions, e.g., Gaussians. Chapter 5 describes the SBFM routine developed in this thesis and compares SBFM with ART for two synthetic and two of the eleven experimental data sets. SBFM is also shown to yield a superior reconstruction of the SF₆ plume described in Chapter 3.

In Chapter 6 the nine remaining steady-state experiments and the non-steady dispersion experiment are analyzed using SBFM. For the steady-state cases, CT reconstruction was performed on time-averaged path-integral concentrations. Each steady-state experiment resulted in a different concentration profile. In every case, satisfactory reconstructions were achieved using SBFM with asymmetric Gaussians. For the time-varying experiment, consecutive path-integral concentration measurements for each ray were interpolated to generate a complete set of ray integrals at specific times. Reconstructions performed on these sets show an evolution of the tracer gas distribution that is plausible based on the experimental conditions and whose features agree well with point-sampled concentrations, both qualitatively and quantitatively.

Chapter 7 summarizes the important findings and examines the possibilities for future research and other applications of the CT/ORS method, such as outdoor monitoring, industrial hygiene and leak detection.

1.6 References

- Air & Waste Management Association (1992) Optical Remote Sensing: Applications to Environmental and Industrial Safety Problems, SP-81, Papers from an International Specialty Conference, Pittsburgh, PA.
- ASTM (1983) American Society for Testing and Materials: Standard test method for determining air leakage rate by tracer dilution, ASTM E 741-83, Annual Book of ASTM Standards, Section 4, Volume 04.07.

- Bauman F., A.J. Gadgil, R. Kammerud, E. Altmayer, and M.W. Nansteel (1983)
Convective heat transfer in buildings: Recent research results. *ASHRAE Trans.*,
89(1A), 215-232.
- Brooks R. A. and Di Chiro, G. (1975) Theory of Image Reconstruction in Computed
Tomography, *Radiology* 117, 561-572.
- Budiansky, S. (1980) Indoor Air Pollution, *ES&T*, 14(9), 1023-1027.
- Colsher, J. G. (1977) Iterative Three-Dimensional Image Reconstruction from
Tomographic Projections, *Computer Graphics and Image Processing*, 6, 513-537.
- Cooper, D. W., and Horowitz, M. (1986) Exposures from Indoor Powder Releases:
Models and Experiments, *Am. Ind. Hyg. Assoc. J.*, 47(4), 214-218.
- Corrsin, S. (1957) Simple theory of an idealized turbulent mixer, *A.I.Ch.E. Journal*, 3,
329-330.
- Faris, G.W. and Byer, R.L. (1986) Quantitative optical tomographic imaging of a
supersonic jet, *Optics Letters* 11, 413-415.
- Fisk, W. J., Prill, R. J. and Seppanen, O. (1988) Commercial building ventilation
measurements using tracer gases, LBL Report # LBL-25612, Lawrence Berkeley
Laboratory.
- Herman, G. T. (1979) Image Reconstruction from Projections: Implementation and
Applications, Springer-Verlag, New York.
- Heiselberg, P. and Bergsoe, N. C. (1992) Measurement of Contaminant Dispersion in
Ventilated Rooms by a Passive Tracer Gas Technique, proceedings of *Room Air
Convection and Ventilation Effectiveness*, American Society of Heating, Refrigerating
and Air-Conditioning Engineers, Inc., 427-431.
- Ishizu, Y. (1980) General Equation for the Estimation of Indoor Pollution, *ES&T*,
14(10), 1254-1257.
- Kak, A. C., and Slaney, M. (1988) Principles of Computerized Tomographic Imaging,
IEEE Press, New York.

- Lobascio, C., Nazaroff, W. W., and Derby, M. W. (1994) Effectiveness of Portable HEPA Filter Units in Controlling Airborne Tuberculosis Transmission, Proceedings of the Fourth International Aerosol Conference, August 29 - September 2, American Association for Aerosol Research, Volume 1, 142-142.
- Mendell, M. J. (1994) Non-specific Symptoms in Office Workers: A Review and Summary of the Epidemiologic Literature, *Indoor Air*, 4(1), 63-72.
- Miller, D. J. (1992) Fungi as Contaminants in Indoor Air, *Atmospheric Environment*, 26A(12), 2163-2172.
- Miller-Leiden, S., Lobascio, C. et al. (1995) Update on Efficacy of In-Room Air Cleaners for Controlling Tuberculosis Exposure, presented at the *1994-1995 Industrial Hygiene Forum Series*, Center for Occupational and Environmental Health—Continuing Education Program, University of California, April 4, 1995.
- Mundt, E. (1993) Contaminant Distribution in Displacement Ventilation - Influence of Disturbances, proceedings of the *6th International Conference on Indoor Air Quality and Climate*, Helsinki, Finland, Volume 5, 201-206.
- Nazaroff W. W. (1989) Mathematical Modelling and Control of Pollutant Dynamics in Indoor Air, Ph.D. dissertation, California Institute of Technology, Pasadena, CA.
- Nero, A. V. (1988) Controlling Indoor Air Pollution, *Scientific American*, 258(5), 42-48.
- Oldaker III, G. B., and Conrad, F. C. Jr. (1987) Estimation of Effect of Environmental Tobacco Smoke on Air Quality within Passenger Cabins of Commercial Aircraft, *Environ. Sci. Technol.*, 21, 994-999.
- Özkaynak, H., Ryan, P. B., Allen, G. A. and Turner, W. A. (1982) Indoor Air Quality Modeling: Compartmental Approach with Reactive Chemistry, *Environment International*, 8, 461-471.
- Post, M. P. (1994) Airflow Models Gaining Clout, *ENR*, October 20th, 22-25.

- Rodes, C. E., Kamens, R. M., and Wiener, R. W. (1991) The Significance and Characteristics of the Personal Activity Cloud on Exposure Assessment Measurements for Indoor Air Quality, *Indoor Air*, 2, 123-145.
- Sato, T., and Shiraki, M. (1984) Tomographic Observation of Flow in a Water Tank, *J. Acoust. Soc. Am.*, 76(5), 1427-1432.
- Todd, L. (1990) Optical Remote Sensing and Computed Tomography in Industrial Hygiene, Ph.D. dissertation, University of North Carolina, Chapel Hill, NC.
- Todd, L.A. (1992), Optical Remote Sensing/Computed Tomography Systems for Workplace Exposure Assessments; Optical Remote Sensing/Computed Tomography Beam Geometries for Monitoring Workplace Gases and Vapors, proceedings of *Optical Remote Sensing Applications to Environmental and Industrial Safety Problems*, Air and Waste Manage. Assoc. SP81, 356-360; 390-393.
- Todd, L.A., and Ramachandran, N. (1994a), Evaluation of Algorithms for Tomographic Reconstruction of Chemical Concentrations in Indoor Air, *Am. Ind. Hyg. Assoc. J.*, 55(5), 403-417.
- Todd, L.A., and Ramachandran, N. (1994b), Evaluation of Optical Source-Detector Configurations for Tomographic Reconstruction of Chemical Concentrations in Indoor Air, *Am. Ind. Hyg. Assoc. J.*, 55(12), 1133-1143.
- Uchiyama, H., Nakajima, M., and Yuta, S. (1985) Measurement of Flame Temperature Distribution by IR Emission Computed Tomography, *Applied Optics*, 24(23), 4111-4116.
- U.S. Department of Health and Human Services (1994) Guidelines for Preventing the Transmission of Mycobacterium tuberculosis in Health-Care Facilities, *Morbidity and Mortality Weekly Report*, 43(RR-13), Public Health Service, Centers for Disease Control and Prevention (CDC), Atlanta, Georgia 30333.

U.S. Environmental Protection Agency (1992) Respiratory Health Effects of Passive Smoking: Lung Cancer and other Disorders, Office of Research and Development, U.S. EPA, December, 1992.

Weschler, C. J., and Shields, H. C. (1991) The Impact of Ventilation and Indoor Air Quality on Electronic Equipment, *ASHRAE Transactions*, 97(1), 455-463.

Wolfe, D.C. (1980) On the Application of Optical Computed Tomography to Remote Air Pollution Measurement, Ph.D. dissertation, Stanford University, Palo Alto, CA.

Wolfe, D. C. and Byer, R. L. (1982) Model studies of laser absorption computed tomography. *Applied Optics*, 21, 1165-1178.

Yoshizawa, S. (1992) Spatial Distribution of Indoor Pollutants in Japanese Buildings - Results of Measurements of CO₂, CO, NO_x, and Biological Particles, proceedings of *Room Air Convection and Ventilation Effectiveness*, American Society of Heating, Refrigerating and Air-Conditioning Engineers, Inc., 225-232.

CHAPTER 2

MIXING OF A POINT SOURCE INDOOR POLLUTANT BY FORCED CONVECTION*

2.1 Abstract

The mixing rate of pollutants emitted from indoor sources influences the effectiveness of pollutant removal by building ventilation and the potential variability of exposure for a given release scenario. Quantitative information is scant on the mixing rate and the factors that govern it. In this chapter I present mixing data for an instantaneously released tracer gas, carbon monoxide, in a sealed, unoccupied room under a range of forced air flow conditions, in which the flow is induced by blowers. The resulting mixing times, from 2 to 42 minutes, are related to the mechanical power of the air jets produced by the blowers. Mixing times are found to correlate well with the inverse of the cube root of power, in accordance with theoretical predictions and experimental observations for mixing in chemical reactors. The exposure index, defined as the time-averaged concentration at a point relative to the time-averaged concentration for the room as a whole, is presented for three experimental conditions, yielding quantitative information on the appropriateness of the well-mixed hypothesis under various flow conditions. In general, the exposure period following instantaneous release of a point-source pollutant must be much greater than the mixing time for the assumption of uniform mixing to hold. The correlation between mixing time and power input is used to predict the mixing time from the mixing action of a supply air jet for a typical ventilation scenario. The resulting predicted mixing time, $\tau_{\text{mix}} \sim 7$ min, is substantially lower than the time scale for removal

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by ventilation, $\tau_{\text{vent}} \sim 48$ min. Under these conditions, complete mixing of an instantaneous release, point-source pollutant would be approximately attained within the interior space well before the pollutant would be thoroughly removed by ventilation.

2.2 Introduction

Investigations of indoor air quality are commonly based on the approximation that pollutants are evenly distributed throughout the interior space. This approximation offers great advantages. In monitoring studies, if the air in a room can be assumed to be well mixed, the pollutant concentrations at all points can be approximated by measurements made at any one point. In modeling studies, the assumption of complete mixing leads to governing equations that are systems of either ordinary differential equations or algebraic equations. Although some systems are intractable analytically, numerical solutions to these systems of equations are straightforward, particularly in contrast with the systems of partial differential equations that must be solved if pollutant concentrations vary substantially with position.

Thus, the widespread use of the approximation that indoor air is well mixed is a logical outcome of the benefits that it yields. Furthermore, this approximation has proven sufficiently accurate in many circumstances to have served as a key ingredient for advances in our understanding of indoor pollutant dynamics over the past few decades. Nevertheless, evidence abounds that the assumption of complete mixing should not be applied in all cases. For example, a recent study showed that the levels of respirable suspended particles and nicotine were 40% and 65% lower in no-smoking sections of restaurants than in the smoking sections (Lambert et al., 1993). Pollutant concentrations in a kitchen with the oven turned on have been shown to be highly dependent on the sampling location (Ozkaynak et al., 1982). Personal sampling has revealed consistently higher exposure than predicted from indoor area monitoring assuming complete mixing (Rodes et al., 1991). The concept of improving ventilation effectiveness to reduce the

energy costs of providing building ventilation depends on removing pollutants from indoor air at concentrations that are higher than the spatial average (Skäret and Mathisen, 1982), a condition that defies the well-mixed approximation.

The preceding considerations provoke several questions:

- How much time is required for pollutants released from a localized indoor source to become thoroughly mixed in indoor air?
- How does the time required for mixing vary with indoor air flow conditions?
- Under what circumstances is it appropriate to apply the approximation of complete mixing of indoor air?

The aim of this chapter is to contribute information towards answering these questions. An earlier study by Baughman et al. (1994) explored mixing induced solely by natural convection. In this chapter, I investigate mixing under forced convection conditions. The main themes of the chapter are (a) quantifying the time required for an instantaneous, point source pollutant release to become well mixed under forced convection flow conditions; (b) relating the mixing time to the rate of power supplied to room air flow; (c) exploring the implications of incomplete mixing on indoor pollutant exposure; and (d) comparing predicted mixing time with the ventilation time for a typical residential setting.

2.3 Experiments

The test space, shown schematically in Figure 2.1, is the same one employed in the experiments of Baughman et al. (1994): an unoccupied room within an indoor air quality research house which was constructed to approximate modern building practice with respect to thermal conditions and to have a low infiltration rate. The room volume is 31 m³ (3.53 m × 3.74 m × 2.36 m high). One wall, adjacent to the outdoors, has two windows, each with an area of about 1.25 m². Both windows were covered from the outside with panels of aluminum sheet metal to prevent sunlight from entering the room

and inducing strong natural convection. The two interior doors to the room were closed and sealed during the tests. The registers from the central forced-air heating system to the room were sealed to isolate the convective flow being studied in this experiment from that induced by ventilation. The infiltration rate, previously measured by tracer gas decay, is in the range 0.03 to 0.08 room volumes per hour (Baughman et al., 1994).

The experiments were conducted during the period from February 8-18, 1994, between 13:00 and 18:00 hours; thermal monitoring of the experimental site during the days immediately preceding the experiment confirmed that the uncontrolled rate of change of air temperature, both inside and outside the house, was relatively low during this period. Before each experiment, the room was sealed and the thermal conditions and flow were allowed to stabilize for variable periods, ranging from 10 minutes for the strongest flow to 1 hour for the weakest flow. At the start of each experiment, 1.5 liters of pure carbon monoxide (CO) was pumped from a gas bag (exterior to the test room) through 9.5 mm tubing into the room over a period of 20 seconds. The CO entered the room through a 30 cm³ plastic cylinder perforated with 13 regularly spaced, 1-cm diameter holes. Thus, the gas was exhausted in all directions at approximately 7.4 cm s⁻¹. The discharge cylinder was located 1 m from two of the interior walls and 0.7 m above the floor.

Carbon monoxide concentrations were monitored as a function of time under 14 different conditions (summarized in Table 2.1). In all cases, air movement induced by infiltration was minimized by sealing the doors and windows. In the quiescent case (Case 1), no sources of heating, cooling or forced-convection mixing were present in the room. In all other cases, as many as five essentially identical squirrel-cage blowers were operated to produce air jets that would cause more rapid mixing of the carbon monoxide in the room air. The blowers were started before the room was sealed for each experiment. Blowers were connected to variable AC power units, and all units operating during a given experiment were set to the same voltage so that the flow rate of air through each

blower was approximately the same. By using different voltage settings and by varying the number of blowers, different combinations of total air flow rate and average air jet velocity could be achieved. Overall, both the total air flow rate and the average velocity were varied through an order of magnitude. The range of air circulation through the fans was 0.2 to 2 room volumes per hour. Each experimental condition, except Case 10, was run twice, one week apart, to evaluate reproducibility.

To direct and straighten the air flow, each blower was fitted with a plastic pipe (60 cm long, 5 cm inner diameter) attached to the outlet. The blower outlets were positioned parallel to one another, 3 cm above the ground, as shown in Figure 2.1. The jet of air that was exhausted from each blower was oriented parallel to the ground and diagonally across the room. In one experiment (Case 10) this arrangement was changed to test whether there was any marked effect of placing the blowers in a different location and creating a less ordered jet of air. In this case, the three blowers used were placed parallel to the ground at a height of 3 cm, but their outlets were positioned towards each other with an angle of 120 degrees between adjacent blowers. The air outlets in this case were all 10 cm from the point of "collision" of the air jets, which in turn was directly beneath the CO source, thus presumably creating highly turbulent airflow without a dominant mean velocity direction close to the source.

The airflow through each blower was measured separately as a function of AC voltage setting before the start and again after the completion of all experiments by the following method. Pure CO was fed through 2 mm tubing directly into the blower inlet at a known, well-controlled flow rate, by means of a peristaltic pump. The concentration of CO was measured close to the end of the pipe attached to the blower outlet using one of the CO monitors described below, with a special extension to direct air from within the pipe onto the sensing element. The CO concentration was found to be uniform in the exhaust air. Thus, knowing the inlet flow rate of pure CO and the outlet concentration, the air flow rate could be determined by mass balance.

In the mixing experiments, as shown in Figure 2.1, the carbon monoxide concentration was measured at 9 locations in the room with passive, datalogging CO monitors (Draeger, Model 190). The choice of the number and location of monitors was based on an analysis of the mixing experiment data gathered by Baughman et al. (1994). I found that by using the concentration data from only these 9 of the original 41 sampling sites in those experiments, the same mixing times would be obtained with an error of no greater than the minimum of 1.5 minutes or 6%. Thus, the mixing times for the present experiments should have been well determined by using 9 appropriately placed CO monitors.

The monitors employed in this study measure CO concentrations in the range of 1 to 999 ppm with a precision of the larger of 3% or 2 ppm. The response time is less than 60 seconds to reach 90% of the true concentration. I employed the CO monitors in the "datalogging mode," in which 1-minute average concentrations were acquired and stored in memory. The data were downloaded to a computer for subsequent processing at the end of each afternoon of experiments.

Ten thermistors provided measurements of air and surface temperatures every 10 seconds during the runs. One thermistor was mounted at the center of each wall surface and on the ceiling; the remaining five measured air temperature along a vertical line near the center of the room. The thermistors were calibrated against a mercury thermometer by placing them and the bulb of the thermometer in a Styrofoam box through which room air was blown at an uncontrolled temperature between 284 K and 292 K over a period of 17 hours. All temperatures measured during the experiments were also in this range. The response of the thermistors was found to be well described by a linear function over this range. The precision of the temperature measurements is estimated to be ± 0.1 K.

An air velocity transducer (TSI, Model 8470) was placed in the center of the room to register air speed once every second. This sensor was intended to provide information on the magnitude and frequency of the fluctuations in air speed (characteristics of the

turbulence) during each experiment. However, upward airflow induced by the heated tip of the sensor alone was large enough (4 cm s^{-1}) to mask the true air speeds in many of the experiments with low air flow. Thus, the air speed data could not be used. Nevertheless, in the future it would be worth attempting to link mixing time to turbulence parameters if air speed could be determined accurately at several locations in the room. There is evidence that mixing time could be directly correlated with the root mean square of the fluctuating velocity (Corrsin, 1957). Such a correlation, if general enough, could be a valuable tool for assessing mixing conditions in real settings using only air speed measurements.

2.4 Data Analysis Procedure

2.4.1 Definition and Calculation of Mixing Times

The mixing time, as defined by Baughman et al. (1994), is the period from the instantaneous release of a point source pollutant until the relative standard deviation of concentrations at well distributed points throughout the room diminishes to 10% or less. (Although the concept of mixing time would apply to any space, this particular definition is only useful for rooms in which the mixing time is much shorter than the characteristic removal time of pollutants from the space; otherwise, a uniform concentration (to within 10% or less) will not be attained.) For the present experiments, the mixing time, $\tau_{\text{mix.expt}}$, was evaluated as the period from the midpoint of the interval of CO injection into the room (which had a duration of 20 s) until the time at which the standard deviation of pollutant concentrations at all nine monitoring points dropped permanently to below 10% of the mean concentration.

Because the CO monitors had different zero concentration offsets and, in one case, a different span calibration, all concentration measurements taken by each monitor were first corrected by subtracting that monitor's reading prior to CO injection and then normalized with respect to that monitor's final, stable concentration reading for each

experiment. The corrected, normalized concentrations were then used to plot the relative standard deviation of concentration as a function of time, and the mixing time, $\tau_{\text{mix.expt}}$ was determined from the graph.

The limited response speed (one minute for 90% response to a step change) and the averaging time (one minute) of the CO monitors introduce an uncertainty in the experimental evaluation of the mixing time that I estimate to be 40 s. This corresponds to a relative uncertainty of 14% for the median mixing time of 4.7 minutes. For the shortest mixing time, 2.0 minutes, the uncertainty due to limited time resolution is 33%.

To relate mixing induced by forced convection to the intensity of air movement generated by the blowers, it is important to account for the influence of natural convection on mixing. Toward this end, I conducted two experimental runs under quiescent conditions, i.e., with air movement and mixing induced only by natural convection due to air and surface temperature differences within the room. For the other experiments, it was assumed that the mixing times due to forced convection alone, $\tau_{\text{mix.fc}}$, and due to natural convection alone, $\tau_{\text{mix.nc}}$, combined in the same fashion as the resistance of resistors in parallel. Thus, the following relationship was used to compute the mixing time due to forced convection:

$$\frac{1}{\tau_{\text{mix.fc}}} = \frac{1}{\tau_{\text{mix.expt}}} - \frac{1}{\tau_{\text{mix.nc}}} \quad (2.1)$$

Such a relationship has been employed in the study of mixing in agitated tanks, when the overall "circulation time" is linked to the axial, radial and tangential components of the circulation pattern (Tatterson, 1991, p. 208).

As shown in Table 2, the mixing times for the two experimental runs executed under quiescent conditions were different, with the case in which the average maximum temperature difference (ΔT) was greater having the lower mixing time. This observation is consistent with the notion that greater temperature differences lead to larger buoyancy

forces, resulting in faster air motion and more rapid dispersion of airborne pollutants. As an estimate of the effect of temperature difference on mixing due to natural convection, I assumed a linear relationship between $\tau_{\text{mix.nc}}$ and the average maximum temperature difference during an experimental run. As this is only a first order approximation, it was not expected to be completely accurate. However, the resulting error in $\tau_{\text{mix.fc}}$ should be relatively small. While ΔT varied by a factor of 4 (from 0.25 K to 1.0 K) in the experiments, the predicted variation in $\tau_{\text{mix.nc}}$ was only approximately 30% (from 45 minutes to 32 minutes). Also, the range of ΔT values to which the linear relationship was applied did not extend far beyond the two values of ΔT used to generate the relationship.

2.4.2 Correlating Mixing Time with Power Input

Energy is required to move air and disperse pollutants. It therefore seems appropriate to correlate pollutant mixing time due to forced convection with mechanical power generated within the room. Corrsin (1957) used turbulence theory to hypothesize a relationship between mixing time and mechanical power:

$$\tau_{\text{mix.fc}} \sim \frac{M^{1/3} L^{2/3}}{P^{1/3}} \quad (2.2)$$

where M is the mass of air in the room, L is a characteristic dimension of the flow system, and P is the mechanical power input. This relationship can also be derived using the Buckingham Pi Theorem (Bird et al., 1960), assuming constant viscosity and no major changes in the characteristics of the flow in the range of P of interest.

Of course, the air mass is equal to the air density, ρ , times the room volume, V . For a room shaped approximately as a cube, as was the case in these experiments, the characteristic room dimension may be represented by the cube root of the volume. Thus, one may write

$$\tau_{\text{mix.fc}} \sim \frac{\rho^{1/3} V^{5/9}}{P^{1/3}} \quad (2.3)$$

or

$$\tau_{\text{mix.fc}} = c \times \beta \quad (2.4)$$

where $\beta = \rho^{1/3} V^{5/9} P^{-1/3}$ and c is a proportionality constant that is to be determined empirically. In this chapter, β will be called the power parameter.

Corrsin (1957) arrives at equation (2.2) only after making two important assumptions: locally isotropic turbulent flow and time stationarity. Neither of these conditions was verified for the experiments presented here. Nor are they necessarily strictly satisfied for most indoor environments, though Zhang et al. (1990) provide evidence that air turbulence in the core of a room is reasonably uniform for simple ventilation scenarios. Thus, equation (2.4) should be regarded as a working hypothesis, based on physical reasoning, and subject to verification by experimental testing.

In experiments by Biggs (1963), mixing times for a tracer dispersed by various types of turbines in a tank reactor were correlated with turbine rotation speed, N , and impeller diameter, D , as

$$\tau_{\text{mix}} \sim N^{-0.843(\pm 0.061)} D^{-1.93(\pm 0.194)} \quad (2.5)$$

The power overcome by the impeller, P , is proportional to N^3 and D^5 (Tatterson, 1991, p. 21). Thus,

$$P^{-1/3} \sim N^{-1.0} D^{-1.67} \quad (2.6)$$

Comparing the two equations above one can see that mixing time and the inverse of the cube root of power are nearly proportional for stirred tanks.

In applying equation (2.4) to my experimental data, the rate of power input, P , was evaluated by means of the following equation

$$P = 0.5 \rho \times \sum_{\text{all blowers}} \left(\int_A v^3 dA \right) \quad (2.7)$$

where A is the cross section of the air jet in the pipe attached to a blower outlet and v is the air velocity parallel to the pipe walls. I determined the velocity profile by making pitot tube measurements at 35 points across the pipe approximately 5 cm from the air outlet. From those data, the integral in equation (2.7) was evaluated as a discrete sum. The normalized profile was observed to be almost flat, typical of turbulent pipe flow, and was nearly identical at all settings and for all blowers.

For each experimental run involving blowers, the value of c was computed using equation (2.4), using the value of $\tau_{\text{mix.fc}}$ evaluated from the measured mixing times and equation (2.1), and β as evaluated from its definition and the results of air flow measurements from the blowers (equation (2.7)). The global mean value of c for all experiments, denoted ζ , was computed as the weighted average of values from individual experiments. The weighting factor was taken to be the inverse of the square of the estimated error in the term $\tau_{\text{mix.fc}}/\beta$. The means of determining this error and the error in ζ is described in the appendix to this chapter.

2.4.3 Exposure Index

The exposure index, as defined in Baughman et al. (1994), is a measure of time integrated concentration at a given point relative to the completely mixed condition. Thus, an exposure index of 1.0 at a point means that the same exposure as measured would have resulted under well mixed conditions. An exposure index greater than one implies that exposure would be greater than obtained by assuming well mixed conditions, and vice-versa.

The exposure index was calculated as a function of exposure ending time, t_e , for all concentration sampling points:

$$EI(\bar{x}, t_s, t_e) = \frac{1}{t_e - t_s} \int_{t_s}^{t_e} \frac{C(\bar{x}, t')}{C_{\text{mix}}(t')} dt' \quad (2.8)$$

where $C(\bar{x}, t')$ is the tracer gas concentration at position $\bar{x}$ at time t' , $C_{\text{mix}}(t')$ is the spatial average concentration of the tracer gas within the room and t_s is the starting time for exposure, generally taken to be the time (or very close to the time) of the tracer gas release. For the experiments reported here, $C_{\text{mix}}(t')$ was taken to be constant, since the characteristic time for ventilation with outdoor air was much longer than the duration of the experimental runs.

2.5 Results and Discussion

2.5.1 Mixing Times

Table 2.2 summarizes the measured experimental and derived forced convection mixing times for all 14 experimental conditions and 27 runs. The experimental mixing times ranged from 2 minutes to 42 minutes. As expected, the mixing process was slowest for the quiescent conditions and generally became faster with increasing air flow rate.

Under quiescent conditions, mixing times were approximately half those recorded under nominally similar conditions by Baughman et al. (1994). There are several plausible reasons for this difference:

1. The time average of the maximum spatial temperature difference was only 0.3 K in the experiments of Baughman et al., compared with 0.85 K and 0.5 K in the present work;
2. The temperature distribution in the experiments of Baughman et al. reveals a much more stable temperature profile, stratified with the highest temperatures close to the ceiling and the lowest temperatures near the floor. During my quiescent experiments there was no apparent stable stratification: the outside wall surface temperature was highest, and the ceiling and floor air temperatures were intermediate;
3. The tracer gas source in the experiments of Baughman et al. was buoyant (neutrally buoyant gas injected above a heated filament). Thus, after injection,

the gas would tend to rise to the ceiling and would be inhibited from spreading to the rest of the room by the stable stratification of the air. My tracer gas was neutrally buoyant and was injected in all directions, thus spreading out to the rest of the room from the very beginning.

The alternative placement of blowers tested in experimental run 10 did not result in a significantly different mixing time, compared to the two experimental runs, 9a and 9b, performed under the same flow rate and velocity conditions. However, since only one alternative blower placement was tested, this result does not allow us to conclude that the results are generally valid for all types of point-source locations and arrangements of air flow.

It would be desirable to conduct similar experiments with air flows more directly representative of real ventilation systems. Supply air grids are generally located either in or close to the ceiling, or near the bottom of a wall. The decisions concerning blower position and orientation in these experiments was guided by expediency, augmented by the expectation that these factors would not significantly influence the results. The blowers employed in this study had to be positioned for horizontal air flow because at other orientations they did not work reliably at low supply voltage (used to generate the low air flow rates). Placing the blowers on the floor (rather than near ceiling height) was simplest, as no extra support equipment was required and the blowers' power supplies were easily accessible for adjusting flow rate. The tracer gas was neutrally buoyant, and the temperature data reveal that there was no significant stratification during the experiments. Therefore, it seems unlikely that the results would have been different if the blowers were positioned near the ceiling or vertically along one of the walls.

2.5.2 Power Input Correlation

Table 2.2 shows the value of the power correlation constant, c , for each experiment. The weighted average value of c for all experiments was 17.6 ± 0.6 . The

extent to which the theory fits the experimental data is revealed in Figure 2.2, where $\tau_{\text{mix,fc}}$, the experimental forced convection mixing time, is plotted against the power parameter, β . A line with slope ζ passing through the origin is also shown. Points plotted at the same value of β represent the replicate experiments performed at each condition. The error bars do not symbolize truly statistically derived errors, and so are only a rough guide to the confidence in the individual measurements. Nevertheless, the data support the hypothesis that the forced-convection mixing time is proportional to β , and, therefore, inversely proportional to the cube root of the rate of power input.

2.5.3 Exposure Index

I report values of the exposure index for three different experiments: (1) Test 1 from Baughman et al. (1994), conducted under quiescent, and nearly isothermal conditions; (2) Test 3 from Baughman et al. (1994), where mixing was driven by natural convection induced by a 500 W electric heater; and (3) run 9a from the present study, which had a mixing time of 5.7 minutes (see Table 2.2). For all three cases, box plots of the distribution of exposure indices versus time were generated, showing the 10th, 25th, 50th, 75th and 90th percentiles, as well as data outside the 10th and 90th percentile limits. The value of t_s used in equation (2.8) was 1 minute for the experiments of Baughman et al. and 0 minutes for experimental run 9a. The average concentration, $C_{\text{mix}}(t')$, was calculated at a time at least 10 minutes after the mixing time, when concentration of the tracer gas was essentially uniform throughout the room.

Figures 2.3 (a) - (c) illustrate how the exposure index varies in space and time during the mixing experiments. In each graph, the experimental mixing time is marked by a dashed vertical line. For points close to the source, exposure indices a few minutes after injection of the tracer gas were of the order of 10, being larger for the cases with slower mixing, as expected. With time, exposure indices at all points tend towards 1.0: as expected, the well-mixed assumption becomes a better one for longer exposure times.

However, even at $t_e = t_{\text{mix,expt}}$, exposure calculations based on the well-mixed hypothesis can be in error by a factor of 2 or more: the lowest EI values are approximately 0.5 and the highest are approximately 2.5 at this time.

2.5.4 Potential Implications for Mixing in Residential and Office Environments

The well-mixed hypothesis is commonly and successfully applied in buildings in which a mechanical ventilation system is operating. In fact, in designing a typical ventilation system, one seeks to establish uniform indoor air temperatures through mixing induced by the kinetic energy discharged with the supply air. It is of interest to consider whether uniform indoor air mixing would be expected, according to the correlation between mixing time and power developed in this chapter. To address this issue, the time scale for mixing (τ_{mix}) was compared with the time scale for ventilation (τ_{vent}). If $\tau_{\text{mix}} \ll \tau_{\text{vent}}$, then the assumption of uniform mixing would appear to be well founded.

A typical mechanical ventilation scenario specified by ASHRAE (1993) has a supply-air grill with an area of 0.15 m^2 and a discharge air velocity of 1.7 m/s . The supply air flow rate is $918 \text{ m}^3 \text{ h}^{-1}$. Applying equation (2.7), the mechanical power input is estimated to be 0.44 W . A typical supply rate for a central forced air ventilation system is 5 air changes per hour, with 75% recirculated; thus, the specified flow rate would serve a room volume of 184 m^3 . The corresponding value of the power parameter β (see equation (2.4)) is 25.3 seconds. Multiplying β by 17.6, the value of ζ derived from our experiments, the approximate mixing time would be $\tau_{\text{mix}} \sim 7.4 \text{ minutes}$. With an outdoor air ventilation rate of 1.25 h^{-1} , the characteristic time for ventilation would be $\tau_{\text{vent}} \sim 48 \text{ min}$. Since the mixing time is significantly less than the ventilation time, the assumption of complete mixing appears reasonable for exposure time scales significantly larger than τ_{mix} . Note that these calculations only apply to cases where supply air has the same temperature as the room air. Buoyancy associated with temperature differences between the supply air

and the room air would either impede mixing or promote it, depending on the sign of the temperature difference and on the height of the discharge register.

2.5.5 Correlation of Mixing Time with Power for Strong Natural Convection

Heated bodies may constitute a significant source of mixing energy. A fraction of the thermal energy released by a heated body is converted into organized, bulk air flow (due to buoyancy) that can be considered similar to air motion produced by mechanical means, as far as mixing power is concerned. The rest of the thermal energy is converted into random molecular motion reflected by a rise in temperature of the surrounding air. The fraction of the thermal energy that is converted into organized bulk flow depends on the geometry of the body. This fraction is expected to be small, but it is difficult to predict theoretically for most real heated bodies found in indoor environments, since the geometries tend to be complex.

I was interested in exploring whether the correlation between power and mixing time presented in this chapter would also apply to thermally induced flows. To this end, the experimental configuration used in Tests 3 and 4 of Baughman et al. (1994) was reestablished, with buoyant air flow induced by means of a 500 W electric heater. Temperatures and air speeds were measured near the heater (Drescher and Lobascio, 1993). The heater raised the air temperature from approximately 22 °C at the inlet to approximately 78 °C at the outlet. The air flow rate through the heater was estimated to be approximately 19.8 L s⁻¹ at an average velocity of 50 cm s⁻¹. From these parameters, I estimated that the mechanical power of the heated plume generated by this flow was approximately 2.7 mW, while thermal energy was transferred to the air at approximately 540 W. (There is possibly some error here, since we cannot expect the total power to be more than the 500 W heater specification. This error is most likely a result of overestimating the final temperature of the heated air. This temperature was measured close to the heating elements with a mercury thermometer. Most likely a fraction of the

temperature registered by the thermometer was due to radiant heat from the heating elements. However, it is also possible that the heater specification did not accurately reflect the actual power drawn by the heater). Thus, the fraction of thermal energy that was converted into bulk flow and thereby contributed directly to mixing was extremely small, approximately 5×10^{-6} . Using equation (2.4) with $c = 17.6$, as determined from the forced-convection experiments, the predicted forced convection mixing time is 14.8 minutes, in remarkable agreement with the experimentally observed values for replicate experiments of 13 and 15 minutes. Appendix II (Section 2.10) provides further details on this comparison.

2.6 Conclusion

The experimental results presented in this chapter substantiate the hypothesis that mixing time is correlated with mechanical power input and provide a quantitative, albeit tentative, relationship between these parameters. Additional experiments are needed to explore the general validity of relationships such as equation (2.4). With improvements in gas sensing technologies, such experiments will become increasingly simple to perform.

These results also show that exposure calculations based on the assumption of instantaneous, perfect mixing can be substantially in error if the pollutant source is highly localized and the exposure period of interest is of the same scale, or shorter, than the mixing time. Applying the correlation results to a typical ventilation scenario, we see that the mixing time would normally be significantly smaller than the ventilation time, so that the assumption of complete mixing is reasonable for time scales of the order of the ventilation period or longer. However, not all buildings are ventilated according to typical mechanical ventilation design practice, and so, it is not justified to assume that complete mixing holds in every indoor environment.

Because the exact patterns of air motion and mixing due to natural and forced convection are highly complex, pollutant dispersion is hard to predict, even in cases, such

as the present experiments, in which only a few sources of mixing energy are present. Despite their empirical nature, simple working models such as the one developed here relating power and mixing time, are useful and need to be investigated further. Such models bridge the gap between computational fluid dynamics models, which require enormous computing capacity to handle realistic three-dimensional dispersion simulations, and the assumption—commonly unsubstantiated—of complete and instantaneous mixing. An understanding of mixing processes at an intermediate level of complexity helps build intuition for solving practical pollutant and heat dispersion problems and is needed to further develop effective and energy-efficient indoor air quality control techniques.

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2.8 References

ASHRAE (1993) ASHRAE Fundamentals, Atlanta, Georgia, American Association of Heating, Refrigerating and Air Conditioning Engineers, p. 31.10.

- Baughman, A.V., Gadgil, A.J. and Nazaroff, W.W. (1994) Mixing of a point source pollutant by natural convection flow within a room *Indoor Air*, 4, 114-122.
- Bevington, P.R. (1969) Data Reduction and Error Analysis for the Physical Sciences, New York, McGraw-Hill.
- Biggs, R.D. (1963) Mixing Rates in Stirred Tanks, *A.I.Ch.E. Journal*, 9(5), 636-640.
- Bird, R.B., Stewart, W.E., and Lightfoot, E.N. (1960) Transport Phenomena, New York, Wiley, p. 185.
- Corrsin, S. (1957) Simple theory of an idealized turbulent mixer, *A.I.Ch.E. Journal*, 3, 329-330.
- Drescher, A.C. and Lobascio, C. (1993) Theoretical Analysis and a Small Experimental Study of Turbulent Mixing in a Room, unpublished final project report for ME 263 at the University of California, Berkeley, May 10th, 1993.
- Lambert, W.E., Samet, J.M. and Spengler, J.D. (1993) Environmental tobacco smoke concentrations in no-smoking and smoking sections of restaurants, *American Journal of Public Health*, 83, 1339-1341.
- Ozkaynak, H., Ryan, P.B., Allen, G.A. and Turner, W.A. (1982) Indoor air quality modelling: Compartmental approach with reactive chemistry, *Environment International*, 8, 461-471.
- Rodes, C.E., Kamens, R.M. and Wiener, R.W. (1991) The significance and characteristics of the personal activity cloud on exposure assessment measurements for indoor contaminants, *Indoor Air*, 1, 123-145.
- Skäret, E. and Mathisen, H.M. (1982) Ventilation efficiency, *Environment International*, 8, 473-481.
- Tatterson, G.B. (1991) Fluid Mixing and Gas Dispersion in Agitated Tanks, New York, McGraw-Hill.

Zhang, J.S., Christianson, L.L., Wu, G.J. and Riskowski, G.L. (1990) Room air flow and turbulence characteristics in a 1/4th scale test room, *Proceedings of 5th International Conference on Indoor Air Quality and Climate*, Toronto, Canada, Vol. 4, 249-254.

2.9 Appendix I: Error Analysis

Throughout this appendix, errors in a given quantity are denoted by a "Δ" in front of the quantity. All formulas for derivation and propagation of errors are based on standard error analysis as described in Bevington (1969).

2.9.1 Errors in the Determination of the Forced Convection Mixing Time ($\tau_{\text{mix.fc}}$)

From equation (2.1),

$$\frac{1}{\tau_{\text{mix.fc}}} = \frac{1}{\tau_{\text{mix.expt}}} - \frac{1}{\tau_{\text{mix.nc}}} \quad (2.9)$$

Thus,

$$\Delta \frac{1}{\tau_{\text{mix.fc}}} = \sqrt{\left(\Delta \frac{1}{\tau_{\text{mix.expt}}} \right)^2 + \left(\Delta \frac{1}{\tau_{\text{mix.nc}}} \right)^2} \quad (2.10)$$

Also,

$$\Delta \tau_{\text{mix.fc}} = \tau_{\text{mix.fc}} \times \frac{\left(\Delta \frac{1}{\tau_{\text{mix.fc}}} \right)}{\left(\frac{1}{\tau_{\text{mix.fc}}} \right)} \quad (2.11)$$

From (2.11),

$$\Delta \tau_{\text{mix.fc}} = \tau_{\text{mix.fc}}^2 \times \Delta \frac{1}{\tau_{\text{mix.fc}}} \quad (2.12)$$

Equivalent equations to (2.12) can be written for $\tau_{\text{mix.nc}}$ and $\tau_{\text{mix.expt}}$.

$$\Delta \frac{1}{\tau_{\text{mix.nc}}} = \frac{\Delta \tau_{\text{mix.nc}}}{\tau_{\text{mix.nc}}^2} \quad (2.13)$$

and

$$\Delta \frac{1}{\tau_{\text{mix.expt}}} = \frac{\Delta \tau_{\text{mix.expt}}}{\tau_{\text{mix.expt}}^2} \quad (2.14)$$

$$\Delta \tau_{\text{mix.nc}} = \Delta \tau_{\text{mix.expt}} = 40 \text{ seconds} \quad (2.15)$$

Combining (2.10), (2.12), (2.13), (2.14) and (2.15) yields the result

$$\Delta \tau_{\text{mix.fc}} = \tau_{\text{mix.fc}}^2 \sqrt{\frac{(40\text{s})^2}{\tau_{\text{mix.nc}}^4} + \frac{(40\text{s})^2}{\tau_{\text{mix.expt}}^4}} \quad (2.16)$$

2.9.2 Errors in the Determination of the Power Parameter, β

The only significant error in β is due to inaccuracies in the blower calibrations. The flowrate through each blower was measured twice at each setting used in the experiments. The average value was taken to be the true flowrate. The error was taken to be the difference between the two flowrate measurements divided by $\sqrt{2}$, or 2% of the average value, whichever was greater. The 2% alternative error was used because the accuracy of the CO monitor was no better than 2%. Only 3 out of the 18 pairs of measurements were affected by this 2% rule.

The fractional error in β for each experiment, with a particular setting of the blowers, was derived to be

$$\frac{\Delta \beta}{\beta} = \frac{\sqrt{\sum_{\text{all blowers}} (\Delta Q \times v^2)^2}}{\sum_{\text{all blowers}} Q \times v^2} \quad (2.17)$$

where Q is the flowrate and v is the average velocity of air leaving the blower pipe at that setting.

2.9.3 Errors in c for each Experiment, and Error in ζ

The fractional error in c was simply taken to be the square root of the sum of squares of fractional errors in $\tau_{\text{mix.fc}}$ and β . The error in ζ was calculated using equation (2.18) below.

$$\frac{1}{(\Delta\zeta)^2} = \sum_{\text{all experiments}} \frac{1}{(\Delta c)^2} \quad (2.18)$$

2.10 Appendix II: Further Detail on the Mixing Time Calculation for Strong Natural Convection (Section 2.5.5)

Using the correlation derived from the set of experiments presented in this paper, I calculated a forced convection mixing time, $\tau_{\text{mix.fc}}$, for the heater in the room of 14.8 minutes. The two experiments conducted by Baughman et al. (1994) resulted in experimental mixing times of 13 minutes and 15 minutes. The forced convection mixing time calculated for these cases should be converted to $\tau_{\text{mix.expt}}$ for proper comparison with Baughman's results. However, this is not straightforward, since Baughman's heater experiments resulted in a stable temperature stratification along the vertical centerline of the room, which would tend to suppress rather than enhance mixing. By using a ΔT of 1 K, which was characteristic of the ΔT in the horizontal plane midway between the roof and the ceiling during Baughman's heater experiments, and the correlation between ΔT and $\tau_{\text{mix.nc}}$ used for the experiments presented here, one arrives at a $\tau_{\text{mix.nc}}$ of 29 minutes and a predicted experimental mixing time of 9.8 minutes for the heater experiments. However, the difference between the ceiling and floor temperatures was approximately 4 K. Even at a ΔT of 0.3 K with stable stratification (Tests 1 and 2 of Baughman et al.), we have seen (in Section 2.5.1) that the difference in $\tau_{\text{mix.nc}}$ can be a factor of two when compared with unstratified scenarios (Experiments 1a and 1b of this chapter). Therefore,

the true predicted $\tau_{\text{mix.expt}}$ for the heater experiments is probably much closer to 14.8 minutes than to 9.8 minutes. It thus compares favorably with the experimental mixing times measured by Baughman et al.

Table 2.1 Summary of experimental conditions

Case	Experimental Condition ^a			Total Flow Rate (m ³ h ⁻¹) ^b	Average Velocity (m s ⁻¹) ^b
	B	Q	v		
1	0	0	0	—	—
2	1	1	1	60	9.4
3	2	1	0.5	58	4.0
4	3	1	0.33	59	3.0
5	5	1	0.2	63	1.9
6	1	0.8	0.8	49	7.6
7	2	0.8	0.4	51	4.0
8	4	0.8	0.2	50	1.9
9	3	0.6	0.2	39	2.0
10 ^c	3	0.6	0.2	39	2.0
11	1	0.4	0.4	24	3.8
12	2	0.4	0.2	25	2.0
13	1	0.2	0.2	14	2.2
14	1	0.1	0.1	6	0.9

^a B represents the number of blowers operating during an experiment; Q represents the nominal volumetric flow rate provided by the blowers, normalized by 60 m³ h⁻¹; and v represents the nominal discharge velocity from the blower, normalized by 9 m s⁻¹.

^b Actual flowrate and discharge velocity, as measured.

^c Blower exhausts configured to face each other.

Table 2.2 Summary of experimental results

Case	ΔT^a (K)	Power (Watts)	$\tau_{\text{mix.expt}}$ (minutes)	$\tau_{\text{mix.fc}}$ (minutes)	c (—)	σ_c^b (—)
1a	0.85	—	35.0	—	—	—
1b	0.5	—	42.0	—	—	—
2a	0.5	1.0	2.8	3.0	17.8	6.3
2b	0.55	1.0	2.0	2.9	25.4	6.6
3a	0.95	0.18	2.5	2.7	11.2	3.7
3b	1.0	0.18	2.2	2.4	12.8	3.8
4a	0.6	0.11	4.0	4.4	17.3	3.3
4b	0.8	0.11	3.9	4.4	17.6	3.3
5a	0.6	0.045	3.7	4.1	12.1	2.6
5b	1.0	0.045	3.6	4.1	12.2	2.5
6a	0.4	0.57	3.8	4.2	28.8	5.6
6b	0.5	0.57	4.7	5.3	36.7	5.9
7a	0.6	0.16	4.0	4.4	20.3	3.8
7b	0.6	0.16	4.0	4.4	20.3	3.8
8a	0.65	0.038	3.9	4.3	12.2	2.4
8b	0.75	0.038	5.0	5.8	16.3	2.7
9a	0.4	0.031	5.7	6.6	17.3	2.6
9b	0.3	0.031	6.7	7.8	20.7	2.8
10	0.4	0.031	6.9	8.2	21.6	2.9
11a	0.6	0.072	4.1	4.6	15.9	2.9
11b	0.55	0.072	5.0	5.7	19.8	3.0
12a	0.3	0.021	8.0	9.7	21.5	3.1
12b	0.3	0.021	7.8	9.4	22.2	3.2
13a	0.25	0.013	11.6	15.4	16.9	3.3
13b	0.25	0.013	7.3	8.6	30.2	5.4
14a	0.75	0.0011	12.7	19.3	16.5	2.5
14b	0.5	0.0011	14.8	22.9	19.5	2.9

^a Maximum point-to-point temperature difference observed during experiment, averaged over time period up to complete mixing.

^b Estimated uncertainty in the mixing power constant, c.

AIR SAMPLING POINTS

- 60 cm height
- 120 cm height
- 180 cm height

VELOCITY SAMPLING POINT

- 120 cm height

BLOWER LOCATIONS

- 3 cm height

TEMPERATURE SAMPLING POINTS

- ceiling surface
- air @ 190 cm height
- air @ 145 cm height
- air @ 100 cm height
- air @ 50 cm height
- air @ 1 cm height
- wall surface

Figure 2.1 Schematic diagram of the test room.

Figure 2.2 Forced convection mixing time, $\tau_{mix,fc}$, for each experiment as a function of the power parameter, β . The line has slope 17.6, which is the value of ζ .

Figure 2.3 Summary of exposure indices as a function of time for three experiments. The experimental mixing time, $\tau_{\text{mix,expt}}$, is indicated as a dashed vertical line. (a) Test 1 (Baughman et al., 1994): quiescent conditions, and 41 sampling points; (b) Test 3 (Baughman et al., 1994): 500W heater, and 41 sampling points; (c) Experimental run 9a from the current study, with an experimental mixing time of 5.7 minutes, and 9 sampling points (for clarity, only every third minute is plotted).

CHAPTER 3

BENCH-SCALE EXPERIMENT USING ART AND AN IDEAL SCANNING GEOMETRY

3.1 Abstract

I describe a proof-of-principle experiment on the bench-top scale to test the feasibility of computed tomography and optical remote sensing (CT/ORS) for remote gas concentration measurements in indoor air. A computed tomography program based on the algebraic reconstruction technique (ART) and a parallel-ray geometry was written for the proof-of-principle experiment. When tested on noise-free synthetic data, it performed very well. A steady, vertical plume of sulfur hexafluoride (SF_6) was generated for the experiment. Point-sample concentrations of SF_6 were measured in a horizontal cross-section of the plume. An infrared source-detector combination was designed, assembled, calibrated and used to measure path-integral SF_6 concentrations in the same horizontal cross-section. The path-integral data were inverted using the CT program to produce a map of SF_6 concentration in the measurement plane. This map agreed with the point-sampled data fairly well, but not as well as would have been expected based on the individual projections of the plume. I attribute the disagreement to the interaction between ART and errors in measured path-integral concentrations at the edge of the plume, where temporal and spatial variability in SF_6 concentration were large.

3.2 Introduction

The purpose of the research described in this chapter was to explore, experimentally and on a relatively small scale, whether computed tomography coupled with optical remote sensing (CT/ORS) could be developed into a new tool for studying pollutant transport and dispersion in indoor air. To this end, I designed a bench-top proof-of-principle experiment to compare the steady-state SF_6 concentration in a

horizontal plane measured by two methods: infrared sensing of path-integral concentrations with subsequent CT reconstruction, and extractive point-sampling of air with subsequent analysis for SF₆ content.

I investigated a variety of numerical reconstruction methods to determine which would be most appropriate for the purpose of this project. Several computed tomography algorithms exist, the most common being analytic, back-projection and iterative techniques (Brooks and Di Chiro, 1975). Since the iterative techniques are the most flexible with respect to beam geometry, one such technique, known as the algebraic reconstruction technique, or ART, was selected for use in this study. The method essentially involves dividing the space under consideration into pixels, making an initial guess of the concentration in each pixel and refining this estimate with each iteration by comparing measured path-average concentrations to those calculated from estimated pixel concentrations. Details of ART and my implementation of it are given in the appendix to this chapter (Section 3.8).

3.3 Algorithm Testing with Synthetic Data

To determine whether two-dimensional gas concentration profiles could be adequately reconstructed, a computed tomography program based on ART was written and first tested on synthetic data. A variety of concentration profiles were simulated on grids of different sizes, from 11×11 to 15×15 square pixels. These profiles include an annular plateau (11×11 , Fig. 3.1), a bivariate Gaussian (12×12 , Fig. 3.2), an asymmetric distribution of peaks of various heights (12×12 , Fig. 3.3a) and a circular plateau (15×15 , Fig. 3.4 (a)) offset from the center of the grid. These hypothetical concentration profiles were used to generate synthetic ray-integral concentration data for the CT program.

Figure 3.5 shows a simplified view the scanning geometry superimposed on the discretized plane of reconstruction for the 15×15 grid (the scanning geometries for the

11 × 11 and the 12 × 12 pixel grids are analogous). The scanning geometry consists of 15 sets of parallel rays, called projections. Each projection contains 15 rays and is rotated 12 degrees with respect to its adjacent projections. This type of ray pattern is considered ideal, because it results in even coverage of pixels in the plane of reconstruction (Todd and Ramachandran, 1994): all pixels are sampled approximately equally. The ray width used was half of the pixel width. The initial guess for the concentration profile was a concentration of zero everywhere. Concentrations were also always constrained to be positive.

The following parameter was defined to quantify the extent to which the ray-integral data of a reconstructed profile match the input data:

$$\sigma_{\text{ray}} = \sqrt{\frac{\sum_{r=1}^{\text{nrays}} \left(\text{PIC}_r - \sum_{p=1}^{\text{npix}} a_{p,r} C_{p,\text{CT}} \right)^2}{\sum_{r=1}^{\text{nrays}} \text{PIC}_r^2}} \quad (3.1)$$

where nrays is the number of rays used, npix is the number of pixels in the plane of reconstruction, PIC_r is the known path-integral concentration for ray r , $a_{p,r}$ is the area of pixel p intercepted by ray r , and $C_{p,\text{CT}}$ is the concentration in pixel p calculated by the CT program. Thus, the second summation in the numerator of equation (3.1) is the path-integral concentration for ray r calculated from the reconstructed pixel concentrations. The value of σ_{ray} corresponds approximately to the fractional RMS difference between input (or measured) path-integral concentrations and corresponding reconstructed values. The CT program was terminated when σ_{ray} changed by less than one percent from one iteration to the next.

Another parameter was defined to quantify the agreement between the original test map and the final reconstruction:

$$\sigma_{\text{pix}} = \sqrt{\frac{\sum_{p=1}^{\text{npix}} (C_{p,\text{true}} - C_{p,\text{CT}})^2}{\sum_{p=1}^{\text{npix}} C_{p,\text{true}}^2}} \quad (3.2)$$

where $C_{p,\text{true}}$ is the concentration in pixel p in the original test profile. The parameter σ_{pix} is thus analogous to σ_{ray} but is evaluated on a pixel by pixel basis, rather than on a ray by ray basis. The best possible reconstruction would have a σ_{ray} and a σ_{pix} of zero.

For all synthetic profiles tested, the reconstructions showed excellent agreement with the original concentration distributions. Visually, the reconstructions for the concentration test maps shown in Figures 3.1 and 3.2 were indistinguishable from the original distributions. The reconstruction of the Gaussian profile did, however, have errors as high as 65% at the lowest concentrations. For both Figures 3.1 and 3.2, σ_{ray} was on the order of 10^{-7} while σ_{pix} was around 10^{-4} . The fact that σ_{pix} is so much bigger than σ_{ray} reflects the fact that there are multiple solutions to the ray integral equations (though they are quite close to the 'true' solutions in these cases). The program happened to converge on solutions that were not the 'true' solutions but that are equally valid as far as matching ray integral concentrations is concerned. Only about 20 iterations were needed to reach the final reconstruction in each case. Convergence was rapid because the profiles possessed a high degree of symmetry.

The more random, asymmetric distribution shown in Figure 3.3 (b) converged more slowly (177 iterations) and, when compared with the original test map (Fig. 3.3 (a)), shows slight differences in concentration near the center of the grid, despite the still

extremely low σ_{ray} value of 5.5×10^{-6} . These differences (the fact that the program converged to an alternate solution to the ray integral equations) are reflected in the value of σ_{pix} again being approximately three orders of magnitude higher than σ_{ray} . This problem of non-uniqueness is also quite clear in Figure 3.4. Here, the reconstruction (Fig. 3.4 (b)) shows alternating slightly higher and slightly lower concentrations at the peak, instead of the flat plateau evident in Fig. 3.4 (a). Nevertheless, clearly ART was still capable of close to perfect inversion of noise-free ray integral data from a variety of underlying concentration profiles. I was thus confident that ART was adequate for testing CT/ORS on the bench-top scale.

3.4 Experiments

An apparatus was assembled to generate a concentration distribution as similar to that shown in Figure 3.4 (a) as possible. The apparatus is shown, in simplified form, in Figure 3.6. A gas stream of 8.1 % sulfur hexafluoride (SF_6) in nitrogen was emitted upward through a circular funnel (10 cm diameter) suspended 3 cm below a larger, inverted funnel. The SF_6 gas stream flowed directly from a compressed gas cylinder through a rotameter into the small funnel. After entering the lower part of the small funnel through 8 mm diameter tubing, it passed through a 1 mm mesh grid and a 5 mm thick piece of foam stretching across the diameter of the funnel. These two barriers were put in place to create a pressure gradient and thereby distribute the flow uniformly across the diameter of the funnel. The larger (24 cm diameter), inverted funnel acted as a miniature exhaust hood. It was connected to a squirrel cage blower (located in a laboratory fume hood) by means of a 6 m section of vacuum tubing. The blower sucked air through the large funnel into the laboratory fume hood.

The average velocity of the gas flow out of the lower funnel was approximately 5 cm/s. The Reynolds number, Re , based on the diameter of this funnel was therefore on the order of 300. Air was drawn into the upper funnel at around 20 cm/s ($\text{Re} \sim 2700$).

Because of the higher velocity into the upper funnel, it was expected that the SF₆ would be effectively contained within a 10 cm diameter cylinder even after leaving the lower funnel. The flow of gas in the 3 cm space between the two funnels was expected to be laminar but possibly somewhat unstable due to the higher Re number in the upper funnel.

A horizontal plane, 18 cm by 18 cm, and located half way between the two funnels, was chosen to be the measurement plane in which point sampling and IR sensing were to be done. The plane was discretized into square pixels of width 1.2 cm resulting in a 15 × 15 pixel grid. All surface plots of measured or calculated SF₆ as a function of position in the plane (Figures 3.9 and 3.13) have x and y axes graduated in 'pixel units', units of 1.2 cm.

3.4.1 Point Sampling

The SF₆ concentration profile in the measurement plane was determined by point sampling of gas as follows. A sampling probe was put into place as shown in Figure 3.7. The probe had an inside diameter of 2 mm, and the sampling end was filed down to minimize the area obstructing the fluid flow. A peristaltic pump continuously drew gas into the probe tip at 10 cc/min. Air drawn into the probe at this flow rate had a velocity close to that of the gas flowing from the lower funnel. Gas was sampled from this continuous probe flow into plastic syringes (30 cc or 10 cc, depending on the expected concentration at the probe location) from a sampling point placed in-line between the peristaltic pump and the probe. At each sampling location of the probe, the syringe was first flushed 3 times with approximately 5 cc of sample air. After 30 seconds the entire volume of the syringe was drawn. Drawing the entire volume took approximately 20 seconds for a 30 cc syringe and 10 seconds for a 10 cc syringe. At an average sampling rate of approximately 80 cc/min, the sampling rate was thus several times the isokinetic

sampling rate¹. Finally, the syringe was capped and the sample analyzed between 1 and 5 hours later.

The funnel assembly was immobile. The sampling assembly, however, was connected to a rotation stage which allowed 360° rotation around the funnels. After determining that no SF₆ could be detected outside the 5 cm radius of the space directly above the lower funnel, the SF₆ gas plume was sampled at six different angles as follows (see Fig. 3.5). The first sampling point was located exactly at the edge of the plume, a distance of 5 cm from the center of the plume. For the first three angles (-90°, 30° and 150°) the probe was then moved radially inward 2 mm at a time, and, after a delay of 1 minute, a sample was taken at each of these locations, up to a distance of 3.2 cm from the center. Finally a sample was taken at the center of the plume. On a separate day, the angles -180°, -60° and 60° were sampled in an analogous fashion, except that the increments were 3 mm and the last sampling point before going to the center was at 2.6 cm.

The syringe samples were analyzed for SF₆ concentration using a Miran 1A General Purpose Analyzer. This instrument detects SF₆ based on the absorption of infrared radiation at 10.6 μm. Each syringe sample was injected into the Miran's 5.6 liter, air-filled chamber. The concentration in the syringe, C_s, was determined using the equation (3.3) below:

$$C_s = C_M \times \frac{V_M}{V_s} \quad (3.3)$$

where C_M is the concentration of SF₆ in the chamber of the Miran, V_M is the Miran's chamber volume (5.6 liters) and V_s is the syringe volume. Syringe volumes between 10 cc

¹In isokinetic sampling the speed of air flowing into the probe is equal to the speed of the gas flow from which one is sampling. Sampling at or close to the isokinetic sampling rate minimizes the disturbance of the air stream being sampled. The exact air speed of the gas plume in the sampling plane was unknown, but it was between 5 cm/s and 20 cm/s, the air speeds out of the lower and into the upper funnel, respectively.

and 30 cc were used, depending on the expected SF_6 concentration in the plume, to optimize the response of the Miran. The Miran was calibrated for chamber concentrations between 0 and 60 ppm once before and once after analyzing a set of syringe samples. Drift in the detector's response was found to be negligible (Leiserson, 1995). A cubic spline function was fit to the calibration points. This function was used to determine C_s from the response of the detector when analyzing syringe samples.

The SF_6 concentration as function of distance from the plume center is plotted for all six angles in Figure 3.8. The positioning of the sampling probe was accurate to within approximately 0.5 mm, since it was achieved using a ruler graduated in 1 mm intervals. The error in the concentration values is estimated to be approximately 3%. It is a combination of sampling errors accumulated in transferring the sampled gas from the probe to the Miran via the syringe, and the error in the Miran measurement itself. The six concentration values measured at the plume center have an average of 8.1 ‰ (matching the concentration of SF_6 in the cylinder) and a relative standard deviation of 2.8 %.

The first observation is that the actual plume of SF_6 gas was less than 5 cm in radius. Significant amounts of SF_6 start to appear around 4.2 cm from the plume center. This is consistent with the fact that air was being sucked into the upper funnel faster than it was being emitted from the lower funnel. Air drawn into the upper funnel from around the lower funnel would tend to compress the gas stream from the lower funnel and speed it up. The maximum concentration is reached at approximately 3.2 cm from the plume center. Thus, the transition zone from pure air to pure cylinder gas is approximately 1 cm wide. The effective plume radius (average distance from the plume center at which the SF_6 concentration is 4 ‰) is 3.9 cm. Assuming the gas stream left the lower funnel at a radius of 5 cm and a speed of 5 cm/s, a mass balance would predict the speed of the gas flow in the plane of reconstruction to be approximately 8.2 cm/s. This is reasonable, given that the reconstruction plane is half way between the lower and the upper funnel.

The second observation is that there is a distinct difference between the two sets of angles. The set of (-90° , 30° and 150°) is indicative of a plume that has close to perfect circular symmetry, while the second set seems to suggest a plume shifted in the -60° direction. The transition from zero to the maximum concentration is closer to the center of the plume than expected in the traverse at -180° . Since the two sets of angles were measured on different days, it is possible that some slight change in air flow might have effected this change, even though care was taken to reproduce the conditions of the experiment exactly.

Figure 3.9 (a) shows the point sample measurements as distributed in the measurement plane. The point samples were interpolated and are plotted at measurement plane grid points in Figure 3.9 (b) for qualitatively better visual interpretation. By comparing Figure 3.4 (a) with 3.9 (b), we again see that the experimental SF_6 plume is narrower than but otherwise similar to that assumed in generating the former plot.

3.4.2 Ray Integral Measurements and CT Inversion

The purpose of the experiment described in this section was to gather path-integral SF_6 concentrations in the measurement plane by means of optical remote sensing, and then to tomographically invert these data to predict SF_6 concentrations for all pixels in the plane. The apparatus for remote optical detection of SF_6 comprised a source of infrared radiation collimated into a single beam and located at a fixed distance from an infrared detector sensitive to radiation near $10.6\text{ }\mu\text{m}$. Sulfur hexafluoride has a strong absorption band at $10.6\text{ }\mu\text{m}$. SF_6 gas in the beam path therefore reduces the intensity of the radiation arriving at the detector. The reduction in intensity can be related to the path-integral SF_6 concentration by means of a calibration curve for the detector.

A single-ray experiment was set up to calibrate a liquid-nitrogen-cooled HgCdTe detector (Graseby Infrared, HCT 100B) for the range of path-average SF_6 concentrations of interest. The apparatus is shown schematically in Figure 3.10. A heated NiCr filament

(Buck Scientific, PE-12CIWL) placed in a ceramic tube provided the source of radiation. The ceramic tube was situated in a cardboard box insulated with foam pads. The insulated box was necessary to prevent air currents from cooling the source and causing fluctuations in radiation intensity. The beam was collimated with a lens, passed through a 6 mm diameter iris and chopped at 350 Hz by a rotating chopper wheel (Stanford Research Systems, SR540). The chopper wheel passed through a light gate which was electronically connected to a lock-in amplifier (Stanford Research Systems, SR510). The lock-in amplifier also picked up the signal from the infrared detector, which has a filtered window (narrow bandpass filter centered at $10.6\text{ }\mu\text{m}$) and was located approximately 80 cm from the source. By comparing the signal from the detector with that from the chopper wheel, the lock-in amplifier was able to isolate from the background infrared radiation that part of the infrared signal arriving at the detector that was due to the infrared source. The lock-in amplifier sampled the filtered signal at a frequency of 10 Hz. The detector was calibrated by placing a plastic box fitted with special infrared-transparent windows in the beam path and filling it with various concentrations of SF_6 , from about 0.03% to 2%. The path length of light through the box was 7 cm.

Four separate calibrations of the detector were performed. The results are summarized in Figure 3.11. The results were averaged to generate a single set of data points. A cubic spline fit produced an analytical function used to determine path-integral SF_6 concentrations from the relative detector signal in the CT experiment. The lowest detector signal (corresponding to maximum absorption) expected in scanning the plume was around 65 %o-cm (8.1 %o SF_6 multiplied by a path length of around 8 cm through the center of the gas plume). This path-integral concentration was well within the range covered by the calibration curve.

For the CT proof-of-principle experiment, the optical bench and all of its attachments apart from the calibration gas box were mounted on a translation stage (Arrick Robotics, X-18) fixed onto a rotation table (Arrick Robotics, RT-12). Both the

translation stage and the rotation table were moved by stepper motors (Arrick Robotics, MD-2). The two funnels shown in Figure 3.6 were mounted in a position that would create a gas plume with the center positioned in the measurement plane as shown in Figure 3.5. Using the rotation and translation stages, the optical bench was moved in such a way as to scan the entire plane of reconstruction with the 15×15 parallel ray geometry. Parallel rays were centered 1.2 cm apart. The area of CT reconstruction was thus 18 by 18 cm, with the SF_6 plume slightly offset from the center, towards the $+90^\circ$ direction (see Figure 3.5).

The area was scanned once in the absence of SF_6 to measure the optical interference of the wires suspending the lower funnel from the upper one. The data gathered in the presence of the plume was then corrected for this effect. Within a projection, the optical bench would be moved to a new position every 10 seconds. The movement itself would take approximately 2 seconds. If the change in signal from one position to the next was large, several more seconds were needed for the detector/lock-in amplifier assembly to respond to the change. Therefore, only the last 4 seconds of every 10 second interval were averaged to determine the path integral SF_6 concentration at that position.

A typical scan (one projection) is shown in Figure 3.12. The effect of the lower funnel's suspending wires blocking the beam can be seen at 40-50 seconds, 80-100 seconds and 130-140 seconds. At 30-40 seconds and 140-150 seconds the wires that were, at these positions, very close to the 6 mm core of the infrared beam seem to be leading to a higher signal, probably from reflection, back into the core, of infrared radiation scattered out of the core. The lowest recorded, unblocked relative signal was 0.23, corresponding to a path average SF_6 concentration of 65 $\text{‰}\cdot\text{cm}$. This implies a path of approximately 8 cm through a uniform concentration of 8.1 ‰ SF_6 . Thus, this feature of the path integral data is consistent with the point sampled data. Moreover, the path integral concentration is at a minimum for roughly 65 seconds (from 55 seconds to 120

seconds). This represents a distance of 6.5 pixels or 7.8 cm (1.2 cm per pixel). Beyond this core, the path integral concentration drops off sharply to zero (detector signal of 1.0) within the adjacent 15 seconds or 1.8 cm. This is also consistent with the point-sampled data. Thus, it would appear that the computed tomography inversion should lead to a concentration profile very similar to that shown in Figure 3.9 (b).

The actual concentration profile generated by the CT program is shown in Figure 3.13. Clearly, it is quite different from Figure 3.9 (b). The location and approximate average concentration within the plume match that expected and predicted by the point-sampled data. The cross-section of the plume is also approximately circular, as expected. However, there is a great deal of variability in concentration between adjacent pixels, even within the 6 cm core of the plume, and there are pixels with a non-negligible concentration where we expect no SF_6 at all (well outside the 5 cm radius of the funnel). The CT program converged after only 2 iterations, with $\sigma_{\text{ray}} = 0.21$. Three or more iterations produced no significant reduction in σ_{ray} . This quick convergence and the lack of improvement thereafter are signs of inconsistency in the set of path integral concentrations due to errors in certain ray measurements. Reconstructing a perfectly consistent data set with ART usually leads to a continuous halving of σ_{ray} every X number of iterations, where X depends on the features of the underlying concentration profile. The final value of σ_{ray} is high compared to the values attained with the synthetic data. It indicates that the average difference between measured path integrals and those of the CT reconstruction was roughly 20%.

The scattergram shown in Figure 3.14 summarizes the comparison between point-sampled and CT-reconstructed SF_6 concentrations. The error bars indicate the range of concentrations observed when several sampling points were located in a pixel. In these cases, the y-coordinate of the plotted point is the average of the point-sampled concentrations in that pixel. The point that is farthest from the 45° line (at $x = 6.8, y = 0$) and the point that is third farthest from the 45° line (at $x = 7.0, y = 3.0$) belong to the

traverse at -180° , in which the point-sample concentrations show the plume border to have been exceptionally close to the plume center (see Figure 3.8). Thus, the point-sample measurements here are suspect. The point that is second farthest from the 45° line (at $x = 8.2$, $y = 2.4$) belongs to the traverse at -150° . In this traverse, the first seven sampling points (going inward from the edge of the plume) went diagonally through pixel (5,11). Judging from the variation in concentration through this pixel, it is centered on the transition zone of the plume. This makes it a highly inadequate area for CT reconstruction, which is forced to assume a constant concentration in each pixel.

How can we explain the fact that ART converged to the 'noisy' profile shown in Figure 3.13 in spite of the fact that the minimum concentration and plume width evident in individual scans of a projection were more consistent with the profile shown in Figure 3.9 (b)? The answer most likely lies in the path integral measurements made at the boundaries of the plume:

- (1) Where spatial gradients in concentration are high, the assumption required by ART, that concentrations are constant within a pixel, is violated. This introduces a discrepancy between measured ray integrals, which are continuous integrals in a continuous space, and those ray integrals calculated from the assumed pixel concentrations.
- (2) Where spatial gradients in concentration are high, errors in equipment positioning are more critical. This problem was exacerbated in our experiment because of the need to correct for the optical interference of the suspending wires, which were less than a mm in thickness, but could profoundly impact the signal at the detector (see Fig. 3.12). Slight imperfections in the repeatability of ray positioning between the first scan (without SF_6) and the second scan probably led to inaccuracies in correcting for this effect.

- (3) Time variability was a third significant factor in creating inconsistencies in the path-integral concentration data set. Fluctuations in concentration near the boundary of the plume produced signals with relative standard deviations up to 25%. This type of fluctuation is to be expected for fluid systems that are not laminar. It is evident in Figure 3.12, between 125 and 130 seconds. Time variability was a problem in this experiment because the time scale of fluctuations appears to have been between the averaging time for a single ray position and the time for an entire scan of the measurement plane. Thus, neither was the time spent at a single ray position sufficient to capture a good average of the path integral concentrations near the plume boundaries, nor was the time for the entire scan short enough to avoid seeing time variability in the data at all. This problem could have been overcome by sampling rays passing largely through the plume boundaries for a longer time.

Due to the inherent nature of the ART algorithm (which treats all pixels along a ray independently but equally), the errors (inconsistencies) in the path-integral concentration measurements at the boundary of the plume were spread out across the entire measurement plane. This led to the unanticipated features of the reconstruction: non-zero concentrations outside the limits of the plume (limits that are clearly visible in the individual scans), and, in the core of the plume, lower and more variable concentrations than one would expect from the individual scans.

The following quantity was calculated to compare the measured point-sample concentrations with the concentrations predicted by the CT reconstruction of the path-integral concentrations:

$$\sigma_{\text{pnt}} = \sqrt{\frac{\sum_{p=1}^{\text{npixpnt}} (\overline{C_{p,\text{meas}}} - C_{p,\text{CT}})^2}{\sum_{p=1}^{\text{npixpnt}} (\overline{C_{p,\text{meas}}})^2}} \quad (3.4)$$

where npixpnt is the number of pixels in which individual point sample measurements were taken (npixpnt = 22) and $\overline{C_{p,\text{meas}}}$ is the average of all point sample concentrations measured in pixel p. As one can see from Figures 3.9 (a) and 3.14, often there were several point-sample concentration measurement locations within a single pixel.

The value of σ_{pnt} for the reconstruction was 0.48. This value is higher than one would expect on comparing Figure 3.13 with Figure 3.9 (b) — there does not seem to be a 50% difference between the two profiles. One reason for the high value of σ_{pnt} is that the calculation for this experiment is based largely on a comparison at the plume boundaries, where, as I have already pointed out, errors in both methods of sampling are most likely to occur. On closer examination of the data, however, one also finds that three pixels are responsible for the σ_{pnt} value being excessively high. These are the three pixels mentioned in the paragraph above, summarizing Figure 3.14. If these three pixels are left out of the calculation of σ_{pnt} , the value becomes 0.25. This value is much more consistent with the general level of discrepancy between Figures 3.13 and 3.9 (b).

3.5 Conclusion

This study was undertaken to investigate whether optical remote sensing in conjunction with an existing computed tomography routine, ART, could be a valid method for measuring local tracer gas concentrations in a plane. The computer program written based on ART produced excellent reconstructions given noise-free synthetic data. Results of experimental testing of the method were not as satisfying. Point-sample concentration

measurements taken in and around the cross-section of a relatively well-controlled plume of SF_6 indicated that the plume was approximately circular in cross-section, had a diameter of approximately 8 cm and a core concentration of 8 ‰. These features were also evident from IR projections of the plume. In spite of these features being present in the projections, the computed tomography algorithm produced a reconstruction that had a wider, flatter and more 'bumpy' concentration peak, and non-zero concentrations outside the physical boundaries of the real plume.

Properties of the inversion technique, ART, were responsible for the unexpected, and unphysical, reconstruction produced. The primary weaknesses of ART in this context are: (1) artificial discretization of space, making it difficult to accurately capture steep concentration gradients, and (2) complete independence of pixels, making it possible to distribute errors in certain ray integral measurements across the entire measurement plane in ways that are unphysical. In this experiment, the optical interference of the funnel's suspending wires, and spatial and temporal variability at the plume boundary led to the noise that created the difficulties for ART.

If one is only interested in a rough approximation to the concentration distribution, ART is a useful technique with many attractive features. Any ray geometry can be used, constraints on pixel concentrations can easily be build into the routine, and the routine converges quickly and unambiguously, given a certain set of constraints. It is thus simple and versatile — yet it does not always do justice to the information contained in the individual projections when measurement error is present. The new method of reconstruction I have developed, SBFM, which is based on a continuous space and does not allow the extent of unphysical oscillations produced by ART, is presented in Chapter 5. The results of its application to the experimental data of this chapter can be found in Section 5.7. There I show that SBFM produces a reconstruction with approximately the same σ_{ray} and σ_{pnt} as ART. However, the SBFM reconstruction is far more consistent than the ART reconstruction with the overall underlying concentration profile.

3.6 Acknowledgments

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3.7 References

- Brooks R. A. and Di Chiro, G. (1975) Theory of Image Reconstruction in Computed Tomography, *Radiology*, 117, 561-572.
- Kuhl, D. E., Edwards, R. Q., Ricci, A. R., et al. (1973) Quantitative Section Scanning Using Orthogonal Tangent Correction, *J. Nucl. Med.*, 14, 196-202.
- Leiserson, K. (1995) Point Sampling in an SF₆ Column: Results and Guidelines, unpublished report, prepared under the supervision of Professor W. Nazaroff, Environmental Engineering Program, Civil Engineering Department, University of California at Berkeley.
- Todd, L.A., and Ramachandran, N. (1994), Evaluation of Optical Source-Detector Configurations for Tomographic Reconstruction of Chemical Concentrations in Indoor Air, *Am. Ind. Hyg. Assoc. J.*, 55(12), 1133-1143.

3.8 Appendix: Explanation of the Algebraic Reconstruction Technique (ART)

Computed tomography reconstruction using ART was implemented for the applications in this chapter according to the following scheme: The reconstruction plane is divided into $n \times n$ square pixels, where n is either 11, 12 or 15, depending on the data set being analyzed. A pattern of rays crossing through the plane is superposed on the pixel grid. This pattern consists of n sets of parallel rays (a set of parallel rays is referred to as a projection), with each set rotated by $180^\circ/n$ from the next. The first projection is at 90° from the horizontal (x -axis). The width of each ray is half the linear dimension of a pixel. The length of each ray is four times the linear dimension of the entire plane of reconstruction.

The concentration is assumed to be constant within a pixel and zero outside the pixel grid. It is furthermore constrained to be greater than or equal to zero. The path-integral concentration for each ray is calculated as a strip sum, with contributions from each pixel weighted by the area of that pixel intersected by the ray. The process for arriving at the final concentration value in each pixel is described below.

Initially, all pixels are assigned a concentration of zero. From one iteration to the next, pixel concentrations are changed according to the following scheme, applied to all rays, one by one. For each pixel the ray passes through,

$$C_{p,CT}^{new} = C_{p,CT}^{old} + \lambda \times (PIC_{r,true} - PIC_{r,old}) \times \frac{A_{p,r}}{SSA_r} \quad (3.5)$$

where $C_{p,CT}^{new}$ is the new concentration in pixel p , $C_{p,CT}^{old}$ is the old concentration in pixel p , λ is a constant called the "relaxation parameter", $PIC_{r,true}$ is the known (synthetic or measured) path-integral concentration of ray r , $PIC_{r,old}$ is the path-integral concentration of ray r calculated from the old pixel concentration concentrations, $A_{p,r}$ is the area of

pixel p intersected by ray r , and SSA_r is the sum of squares of $A_{p,r}$ for all pixels that ray r passes through. Equation (3.5) embodies the ART algorithm.

By subtracting $C_{p,CT}^{old}$ from both sides of equation (3.5) and multiplying both sides by the total area of each pixel, A_p , we arrive at equation (3.6) shown below.

$$A_p \times (C_{p,CT}^{new} - C_{p,CT}^{old}) = \lambda \times (PIC_{r,true} - PIC_{r,old}) \times \frac{A_{p,r} \times A_p}{SSA_r} \quad (3.6)$$

The left-hand side of equation (3.6) is the total number of gas molecules (per linear dimension in the vertical direction) added to pixel p . The term $(PIC_{r,true} - PIC_{r,old})$ on the right-hand side represents the excess (deficit) of gas molecules of the ray passing through the grid of **computed** pixel concentrations. Thus, this algorithm effectively takes the excess (deficit) of gas molecules of the ray passing through the grid of **computed** pixel concentrations, and distributes it to the pixels in proportion to the ratio $\lambda \frac{A_{p,r} \times A_p}{SSA_r}$.

Since we always have $A_p \geq A_{p,r}$, applying equation (3.5) to all pixels along a ray tends to result in an over-adjustment of the ray-integral concentration for $\lambda = 1$. After testing ART several times using different values of λ on the order of 1, it was found that the value of λ only influenced the rate of convergence of the algorithm, not the final solution. Henceforth, λ was set equal to 1 in all implementations of ART, including those reported in Chapter 5.

After ART has been applied to a ray, the path-integral concentration is recalculated for all rays (based on the changed pixel concentrations), and equation (3.5) is applied to the next ray. This process is repeated until all rays have been treated. Rays are processed consecutively from one side to the other within a projection. Treating all projections constitutes a single iteration. Projections are treated in the following order: the first projection (at 90°), the projection closest to 180° , the projection at $90^\circ + 180^\circ/n$, the projection at $180^\circ + 180^\circ/n$, the projection at $90^\circ + 180^\circ/n + 180^\circ/n$, etc. This order of

treatment has been reported in the literature (Kuhl et al., 1973) to achieve the fastest possible convergence. This was confirmed in trial tests performed on synthetic data in which I varied the order of projection processing.

Figure 3.1 Annular plateau synthetic concentration profile on 11×11 grid. The ART reconstruction is indistinguishable from the test profile.

Figure 3.2 Bivariate Gaussian synthetic concentration profile on 12×12 grid. The ART reconstruction is indistinguishable from the test profile.

Figure 3.3 (a) Random peaks synthetic concentration profile on 12×12 grid.

Figure 3.3 (b) Reconstruction of random peaks synthetic profile by computed tomography.

Figure 3.4 (a) Cylindrical plume synthetic concentration profile on 15×15 grid.

Figure 3.4 (b) Reconstruction of synthetic cylindrical plume by computed tomography.

Figure 3.5 Schematic of the ray scanning pattern superposed on the plane of reconstruction for the 15×15 pixel grid. Only three of the fifteen ray projection angles are shown. Most rays are omitted for clarity. Also shown is the cross-section of the lower funnel and the radial segments along which point-sample SF_6 concentrations were measured, as described in Section 3.4.

Figure 3.6 Schematic of the experimental apparatus for generation and IR scanning of the SF_6 gas plume. The distance between the IR source and the detector is 80 cm.

Figure 3.7 Schematic of the apparatus for point sampling of SF_6 .

Figure 3.8 Point-sampled SF_6 concentrations in the measurement plane as a function of distance from the plume center (the 'plume center' is directly above the center of the mouth of the lower funnel) for all six angles of traverse.

Figure 3.9 (a) Point-sampled SF_6 concentrations in the measurement plane. At the plume center, the average of all six measurements is plotted.

Figure 3.9 (b) Surface plot of interpolated point-sample SF_6 concentration measurements.

Figure 3.10 Schematic of IR detector calibration apparatus. The distance between the IR source and the detector is 80 cm.

Figure 3.11 Results of the four calibrations of the HgCdTe detector.

Figure 3.12 A typical projection of the SF_6 plume. The IR beam was moved by 1.2 cm, to a parallel position, every 10 seconds.

Figure 3.13 CT reconstruction of the concentration in a cross-section of the SF_6 gas plume based on the measured path-integral SF_6 concentrations.

Figure 3.14 Point-sample SF₆ concentration measurements compared to pixel concentrations in the CT reconstruction. Where several points were sampled within a pixel, the average concentration is plotted. In these cases the lower segment of the error bar indicates the lowest concentration and the higher segment of the error bar indicates the highest concentration sampled within that pixel.

CHAPTER 4

TRACER GAS CONCENTRATION MEASUREMENT IN A CHAMBER USING AN FTIR WITH A PARALLEL RAY GEOMETRY AND ILST COMPUTED TOMOGRAPHY*

4.1 Abstract

The work described in this chapter explores for the first time the feasibility of computed tomography (CT) reconstructions of pollutant concentrations in a real room setting on the scale of several meters. A remote sensing Fourier transform infrared spectrometer was mounted on a movable base in a controlled ventilation chamber. A passive tracer was released from a point source into the room under constant ventilation conditions. A series of experiments was conducted gathering multiple path-averaged concentration measurements in a horizontal plane for CT reconstruction. Simultaneous readings were obtained with a multiple point sampling array for later comparison to the CT reconstructed concentrations. Good qualitative agreement between the reconstruction and point sample data was obtained. Limitations encountered due to the temporal resolution, size, and geometry of the experimental apparatus are clearly surmountable with better instrumentation.

* Most of this chapter was taken from the paper: Yost, M.G., Gadgil, A.J., Drescher, A.C. et al. (1994) Imaging Indoor Tracer-Gas Concentrations with Computed Tomography: Experimental Results with a Remote Sensing FTIR System, *Am. Ind. Hyg. Assoc. J.*, **55**(5), 395-402. Minor changes in wording were made.

4.2 Introduction

Steady advances in infrared detection and quantitative analysis of infrared spectra for airborne pollutants have led to the development of a new class of direct reading instruments, based on the remote sensing Fourier transform infrared (RS-FTIR) spectrometer. These instruments use an infrared beam to probe the air, and are capable of detecting a wide variety of contaminant species in situ at concentrations as low as a few parts per million over path lengths of many meters (Simonds, 1994; Grant and Menzies, 1983). Such instruments recently have been scaled down to "transportable" size and are being used for concentration measurements of airborne gases in workplaces and outdoors (Levine et al., 1989). A fundamental feature of RS-FTIR instruments is that they provide an integrated measurement of the contaminant mass in the beam path. This normally is converted to a path average molar concentration by dividing the measurement (e.g. ppm-meters) by the known path length. A consequence of this spatial averaging is that some quantities of interest such as the peak concentration along the beam path cannot be obtained without additional information on the spatial distribution of contaminants in the beam.

The aim of the research described in this chapter was to demonstrate by experiment the use of RS-FTIR detection of a tracer gas coupled with computed tomography (CT) to measure pollutant gas concentration on the scale of several meters. In this case, the concentration distribution of the gas of interest, acetone, was much less controlled than in the table-top experiment described in Chapter 3. Also, because the scale of sampling was so much larger and the equipment therefore had to be moved greater distances, not nearly as many ray paths could be used if the total scan time was to remain reasonable. Both of these factors made this a more challenging experiment for CT reconstruction.

4.3 Materials and Methods

An RS-FTIR spectrometer system was mounted on a movable base in a controlled ventilation chamber. A tracer was released from a point source into the room under constant ventilation conditions. A series of experiments were conducted gathering multiple path-averaged measurements in a 2-dimensional plane for CT reconstruction. The measurement plane was midway between the floor and ceiling of the room, 1.37 m above and parallel to the floor. Simultaneous readings were gathered with a multiple point sampling array in the same plane for later comparison to the CT reconstructed concentrations.

4.3.1 Test Chamber

The test chamber illustrated in Figure 4.1 (a) was designed to provide ventilation in a horizontal plug-flow arrangement. The chamber is described in detail elsewhere (Yost et al., 1992) so only a brief description follows. The 143 m³ working section had two plenums covering opposing side walls to distribute the supply and exhaust air. Other room walls were sealed to reduce leakage. Two industrial blowers provided push-pull mechanical ventilation and had speed regulation of 1 percent around the set point. The fans were set for a volume flow rate through the chamber of about 142 m³/min, giving a residence time for air in the room of 1 minute (60 air changes per hour) and a mean velocity of approximately 10 cm/s.

The chamber was equipped with a custom-designed computer controlled sampling system that sequentially monitored air concentrations at 14 points. Each sample point was connected by polyethylene tubing to a 3-way solenoid valve and exhaust manifold. Air samples were drawn continuously at a rate of 5 cc/s from each sample line into the exhaust manifold to minimize the delay in response time due to the dead volume in the tubing. The samples passed into a flame ionization detector (FID, Century Instruments, Arkansas City, Kansas) which provided a 1.0 mV/ppm analog output signal that was sampled and

stored in a computer. The FID had a range of 0.2 to 2000 ppm with a response time (0 to 90% of full scale) of about 4 seconds.

Acetone vapor was the tracer gas used in this study. The vapor was generated by passing 1 l/min dry nitrogen through liquid acetone in a 250 ml glass bubbler kept in a water bath set at 35°C. Temperature regulation of the water bath over time was better than 0.1°C. The bubbler output was connected to a 4 l glass mixing chamber and then passed through polyethylene tubing to a 2-cm-diameter porous-ceramic spherical diffuser effectively creating a point source. A needle valve controlled the flow of nitrogen gas; the flow rate was measured by a rotameter calibrated with nitrogen. The source was positioned 60 cm upstream of the geometric center of the room along the inlet to outlet center-line axis, 2.5 cm above the measurement plane (see Figure 4.1 (a) and 4.1 (b)). An FID sample port in the exhaust air duct monitored the generation rate of the source, which was essentially constant during these experiments.

4.3.2 RS-FTIR Description

FTIR data were collected using a Nicolet ST-1 FTIR spectrometer equipped with a liquid-nitrogen-cooled mercury-cadmium-telluride detector (Nicolet Inst. Corp., Madison WI). The optical bench was modified by Nicolet for remote optical sensing as described elsewhere (Xiao et al., 1991). The IR source (1300 K filament) and swinging mirror interferometer (2 cm⁻¹ resolution) were mounted together in a cabinet on a tripod, from which emerged a 5-cm diameter IR beam with a coaxial 1.5 mW visible laser beam to assist with aiming. The separate tripod-mounted detector was fitted to a 20-cm diameter Cassagrain telescope which collected the IR beam. Sample scanning, data collection, and spectral analysis were performed using an IBM-compatible 386 personal computer.

The RS-FTIR was mounted on a specially constructed translating table attached to a floor-mounted rotational platform on a central pivot (Figure 4.1 (a)). The source-interferometer and detector were arranged in a straight line 3-meter beam path on the

translating stage. A remotely controlled traversing system powered by DC motors positioned the translating table (and beam path) at 30 cm fixed intervals in order to gather a set of parallel ray paths forming one projection; the platform was then rotated to a new position and the traverse repeated to gather a projection at another angle.

The multipoint FID sample array was positioned along a midline of the rotating platform. Eleven sample points were arranged at 30 cm intervals along a line perpendicular to the beam path and translating stage. The sample points were scanned continuously by the computer sampling network. Three additional ports monitored the inlet air, exhaust air, and a 125 ppm span gas standard. These additional samples furnished a check on the room background, source emission rate, and the detector calibration. Each port was sampled for 10 seconds, followed by a 10 second delay between samples to preclude any switching artifacts.

Each experimental run consisted of data gathered for 4 angular projections (90, 45, 0, and -45 degrees). Each projection angle resulted in a corresponding fixed position for the FID array aligned perpendicular to the beam path. The entire FID array was scanned every 5 minutes and nine complete array scans were stored in a 45-minute sampling period for a single projection angle. The individual 10 second readings from the FID sample points were averaged together over the 45 minute period for each projection angle, and four of these 45 minute periods constituted an experimental run spanning 180 minutes. Simultaneously with the FID data collection, the RS-FTIR was positioned at one of the 9 ray path locations along the projection and averaged over a 5 minute interval to ensure a representative measurement of the path-integral acetone concentration at that position. At the completion of each projection scan, the platform rotated to a new angle and a new set of ray scans was gathered.

4.3.3 Calibration

The flame ionization detector was calibrated over a range of 1 to 1500 ppm using commercially available gas cylinders (Matheson Gas Products). Two factory analyzed primary gas standards, 100 ppm and 1500 ppm ($\pm 1\%$) acetone in air were diluted immediately prior to calibration in hydrocarbon free (HC-free) air to create working standards. Dilutions were prepared by measuring a volume of HC-free air into 20 l Tedlar bags with a calibrated dry test meter, and then injecting a volume of the primary gas into the bags with a gastight syringe to obtain intermediate concentrations of 5, 10, 50, 500, and 1000 ppm.

Quantitative analysis of IR spectra were performed with a least-squares fit program and a spectral library covering the range of 4000 to 600 cm^{-1} at 2.0 cm^{-1} resolution. Least-squares quantification used a 53 ppm acetone plus water and CO_2 reference spectra, in two analysis windows at 2800-3200 cm^{-1} and 1150-1260 cm^{-1} . The spectral library was obtained from tests with a 10 m gas cell. The choice of this library and the use of the least-squares algorithm for analysis are described elsewhere (Ying and Levine, 1989).

4.3.4 Computational Methods

The FID point data were interpolated using SURFER (Golden Software, Golden, CO) to produce a 2-dimensional uniform grid of concentration values. A kriging algorithm was used to find minimum variance estimates of these grid values. Kriging is a geostatistical technique for estimating a grid of values from a set of arbitrarily positioned point measurements that uses the spatial correlation between points to find minimum variance estimates of intermediate grid points (Jones et al., 1986).

Computed tomography (CT) comprises a variety of different numerical methods for reconstructing images from projections (Kak and Slaney, 1988). The term "image" is used here in the broadest sense, referring to a spatial distribution of a measurable physical

property, such as density or temperature (Hertz, 1985) in a two-dimensional "slice" of matter. The relative performance of different CT methods depends on the type of reconstruction algorithm being used, the characteristics of the distributions being reconstructed, the sampling density and the noisiness of the projection data.

For this study, the reconstruction was achieved as follows: an initial guess of the concentration distribution was improved upon in successive iterations by comparing computed ray averages resulting from the estimated distribution with the measured ray averages. A dimensionless relaxation parameter was used to regulate the speed of convergence and ensure stability against numerical oscillations. The only constraint placed on the reconstruction was that pixel concentrations could not be less than zero. Thus, after every iteration, pixels whose concentrations were negative, were assigned a value of zero.

There are two major parts in an iterative CT algorithm: (1) a *formula* to calculate corrections to the current estimates of pixel concentrations based on the discrepancy between computed and measured ray-average concentrations; (2) an *order* for updating the computed pixel and ray average concentrations. Pixels were updated sequentially, as done in the iterative least squares technique (ILST) (Brooks and Di Chiro, 1975). The formula used to calculate new pixel concentrations was:

$$C_{p,i+1} = C_{p,i} + \frac{\gamma}{N_p} \sum_{r=1}^{r=N_p} (PAC_{r,m} - PAC_{r,i}) \frac{l_{r,p}}{L_r} \quad (4.1)$$

where

$C_{p,i+1}$ = the concentration in pixel p at the i+1th iteration

γ = relaxation parameter

N_p = number of rays passing through pixel p

$PAC_{r,m}$ = measured path-average concentration along ray r

$PAC_{r,i}$ = calculated path-average concentration along ray r at the i th iteration

$l_{r,p}$ = the segment length of ray r passing through the pixel p

L_r = the total length of ray r .

Based on the work of Peterson (1986), where the equivalent formula is referred to as ART1, and tests of this and several similar formulas applied to real and synthetic data (Fraser, 1989), the above formula was expected to produce reasonable reconstructions while being robust to input noise.

Estimates of pixel concentrations were updated according to ILST, that is, pixel by pixel from the first to the last, recalculating ray integral concentrations only after all pixels had been updated. The computed tomography program written specifically for the geometry of this experiment was thus based on both the ART1 formula for calculating the corrections and on the ILST correction scheme for updating the pixel estimates. The computer program was first tested on synthetic Gaussian distributions and on simulated profiles having steep concentration gradients similar to those expected in the real data. Good results were achieved, even when randomly generated noise with a coefficient of variation between 10% and 50% was added to the synthetic ray average concentration data.

The experimental sampling plane covered by rays was divided into square pixels with a width equal to the distance between parallel rays (0.3 m). This resulted in 93 pixels (9 by 9, plus 3 pixels along the middle section of each edge), each having between 2 and 6 of the 36 rays passing through it (Figure 4.1 (b)). A goodness-of-fit parameter, σ_{ray} , was defined to quantify the overall extent of agreement between the average concentration along a specific ray path of the reconstructed image (PAC_r , calculated ray average concentration) and that measured for the physical system ($PAC_{r,m}$, measured ray average concentration):

$$\sigma_{\text{ray}} = \sqrt{\frac{\sum_{\text{all rays}} (\text{PAM}_{r,m} - \text{PAC}_r)^2}{\sum_{\text{all rays}} (\text{PAM}_{r,m})^2}} \quad (4.2)$$

A reconstructed distribution with a lower value of σ_{ray} has a better overall agreement between the average concentrations along the rays and measured data. However, a low σ_{ray} value does not guarantee a close match between pixel by pixel reconstructed concentrations and those in the measured physical system (see Section 4.5).

4.4 Results

The experiments produced two data sets, one consisting of 36 beam-path-averaged values (9 ray paths for each of 4 projection angles) and the second consisting of 44 point values from the FID array (Figure 4.1 (b)). The rays were time averaged over 5 minute intervals while the point values were time averaged over 45 minute intervals. The measured relative standard deviation in average concentration for an individual ray was typically 30%. The variation in concentration at individual points was quite large, the relative standard deviation of FID values being typically around 100%. Figure 4.2 (a) shows a surface plot of the kriged FID grid values representing the plume as captured by the FID array.

Applying the tomographic reconstruction to the FTIR ray average data resulted in the concentration distribution shown in Figure 4.2 (b). The main features of the acetone plume (the source and the direction of advection) are clearly recognizable, including the region of slightly increasing concentration downwind of the source which was apparent in the kriged data. However, the relatively high value of σ_{ray} , 0.4, which was the best achievable value over a wide range of relaxation parameters and number of iterations,

indicates that the reconstructed concentration distribution was based on an internally inconsistent (or noisy) set of ray average values (see Discussion).

To test the ability of the CT program to reconstruct a concentration profile where the concentration values are known, the kriged FID data of Figure 4.2 (a) were used to calculate ray averages and produce a "synthetic" data set of concentrations corresponding to the 93 pixels of the CT grid. When these ray average concentrations were used to reconstruct the spatial distribution of acetone concentrations, the program produced the image illustrated in Figure 4.2 (c). Though clearly much more comparable to the FID map (Fig. 4.2 (a)) than the reconstruction from the measured path averages (Fig. 4.2 (b)), some disagreement at the level of individual pixels remains. However, the low value of σ_{ray} , 0.002, reflects the fact that almost all ray averages of the reconstructed picture match those of the kriged data to better than 0.6 percent.

4.5 Discussion

Figures 4.2 (a) and 4.2 (b) show similarity between the CT reconstruction and the concentration map generated from the FID array measurements. The CT reconstruction correctly located the position of the source and reproduced the steep concentration profile surrounding it. However, notable differences also appear: the CT image shows a trailing plume while the FID image has none, and the peak height at the source is almost double in the CT reconstruction relative to that in the FID image. These differences raise the issues of whether the FID image or the CT image show the "true" picture and what factors may have contributed to the discrepancy.

There are substantial limitations in both the CT projection data and the FID data as obtained in this experiment. For example, to ensure that the concentration pattern remains absolutely stable, the CT data should be collected so that the instrument would not change the airflow. However, in order to produce the ray path geometry shown in Fig. 4.1 (b), the source and detector had to be moved from place to place within the chamber.

Different positioning of the source and detector might well have changed the airflow pattern enough to result in changes in the acetone concentration profile in the measurement plane during the 3 hour scanning period. Such changes would have resulted in inconsistencies between rays in the CT input data.

Modifications to the airflow in the measurement plane owing to the withdrawal of air through the FID sampling array probably had little effect, because a fairly low flow rate was used for sampling through each tube (5 cc/s). Velocity changes at the sample inlet should drop off quickly to below background levels within a few mm of the FID sample tube inlet.

On the other hand, *positioning* the FID sample array represents a limitation. Ideally, the point samples would be in a plane coincident with the CT scan at the same height as the source. Of course, it is impossible to have a sampling point coincident with the source. In our experiments, the IR beams were approximately 5 cm (2 in) in diameter. To avoid blocking the beams with the FID sampling tubes, the tubes were positioned so their tips grazed the bottom edge of the beams. This placed the plane of the FID sampling tubes about 2 cm below the plane of the beam centerlines, and about 5 cm below the center of the source.

To explore the influence of the small vertical separation (2 cm) between the FID sampling plane and the FTIR sampling plane, transport near the point source was modeled based on isotropic eddy diffusion in a uniform flow (Roberts, 1923). Under these conditions the concentration field around the source can be described by the equation (Seinfeld, 1986):

$$C_r = \frac{\dot{m}}{4\pi K r} e^{-\frac{u(x-r)}{2K}} \quad (4.3)$$

where

C_r = concentration at r (mg/m³)

$r = \sqrt{x^2 + y^2 + z^2}$ distance from point source (m)

$\dot{m}$ = mass generation rate (mg/min)

u = air velocity (m/min) (aligned with x axis)

K = eddy diffusivity (m^2/min)

x = distance in x direction (m)

Employing an approach similar to Roach (1981), the effective diffusivity was estimated by calculating K from the above equation using 9 kriged FID concentration values located within a 40 cm radius of the source. All calculations assumed a source emission rate of 1 g/min (based on a saturated source gas) and an air velocity of 6 m/min (based on exhaust air concentration readings). Combining the estimates yielded an average value for the effective diffusivity of $1.28 \text{ m}^2/\text{min}$. Using this average value of K , the calculated concentration for the FID peak 5 cm below the source was 466 ppm, and for the FTIR peak 3 cm below the source was 813 ppm. These calculated values agree quite well with the observed peaks for the FID array (472 ppm) and the CT reconstruction (880 ppm). Thus the placement of the FTIR beam measurements in a plane slightly closer to the source compared to the FID array probably accounts for the difference in the peak concentration.

The results from the synthetic CT reconstruction of the FID data (Fig. 4.2 (c)) appear to closely match the original FID image (Figure 2), both in terms of the peak height and peak shape, with some overall broadening of the peak and lowering of the peak concentration. This strongly suggests that the differences between Figure 4.2 (a) and Figure 4.2 (b) are not due to some inadequacy in the algorithm, but rather to inconsistencies in the input data. This also is indicated by the much higher values of σ_{ray} in the CT image (Fig. 4.2 (b)) compared to the CT image of the FID data (Fig. 4.2 (c)).

Three factors limit the CT reconstruction accuracy: noise, the non-uniqueness of the CT solution, and the spatial sampling frequency. Noise in this sense represents time and space variability in the concentration pattern due to a variety of factors. Changes in

the airflow pattern and vortex shedding from the instrument in the airflow, positioning errors, and measurement errors all contribute to variability in the path-averaged concentrations. Position errors introduced by limitations in the mechanical system were generally small, typically less than 3 mm, and would produce relative errors in position on the order of a fraction of a percent.

It was anticipated that the effects of temporal fluctuations in concentration at fixed positions in space would be reduced by averaging each ray over time. I compared the timescale of the averaging to estimates of the frequency of vortex shedding from objects (cylinders) in the airflow. For an air velocity of 10 cm/s and cylinders of 1 to 50 cm in diameter (the range of obstacle sizes in this experiment), we estimated the frequency of vortex shedding between 0.04 and 1.5 Hz (Schlichting, 1979). This indicates that the longest timescales are on the order of 25 s, far shorter than the 5 minute averaging times for the rays. A more important source of error in the projection data may have been changes in the air flow and plume dispersion pattern as the instrument was rotated to different angles, rather than eddy shedding.

The effect of changing the dispersion pattern during the experiment is to present the reconstruction algorithm with an inherently inconsistent set of data; this is equivalent to trying to reconstruct projections sampled from several different spatial distributions of contaminant. Such data cannot necessarily be assembled to create an image that simultaneously satisfies all ray averages. This lack of consistency manifests itself in the inability to converge further after a certain number of iterations, despite relatively poor agreement between calculated and measured ray average concentrations. The high value of σ_{ray} for Figure 4.2 (b) is an indication that a single spatial distribution of acetone satisfying the measured set of path average concentrations probably did not exist. In contrast, the reconstruction of the synthetic projections derived from the kriged FID data produced a low σ_{ray} value that constantly decreased with an increasing number of

iterations, reflecting the fact that there was at least one distribution that matched the given path-average projection data.

Spatial resolution limits the FID sampling data, perhaps to a more severe degree than the RS-FTIR. If each FID sample point only reflects concentrations within the immediate few cm of surrounding space, relatively small horizontal shifts in the location of the plume could cause dramatic changes in the concentrations measured by the FID. The RS-FTIR however, would still capture the plume because of the much larger sampling path. Thus, spatial-temporal variability in the plume location within the measurement plane is a possible contributor to differences between the measurements.

It is important to point out that if changes in the dispersion pattern did occur as a result of different instrument positions at different times during the experiment, both the kriged map and the CT reconstruction would give an inaccurate picture. Like the CT data, the kriged data were sampled systematically from different instrument locations and at different times, and could reflect different underlying spatial distributions. Kriging these data would tend to add sampled points from the different spatial distributions together to produce a map, while CT reconstruction would tend to average the distributions. The two techniques will produce different concentration maps from the data, but one map is not necessarily more correct than the other.

The second important limit on reconstructing the actual concentration distribution stems from the fact that the system of equations was underdetermined. In theory, complete determination of the image would be possible only if the number of rays matched the number of pixels and the rays were distributed appropriately. Since the reconstruction matrix size is approximately the square of the number of rays per projection (ignoring the edges), this implies that the number of projection angles should at least equal the number of rays per projection. In iterative computed tomography, if the number of projections in the input data is small, the reconstruction tends to predict concentration peaks that are lower than the true maxima (Todd, 1992).

The limited number of projections (four, rather than the necessary minimum of nine) in this experiment leads to the fact that even perfectly consistent data, namely those ray averages calculated directly from the kriged FID data, did not lead to a “perfect” reconstruction. When comparing Figures 4.2 (a) and 4.2 (c), we see that the reconstructed image (Fig. 4.2 (c)) has peaks that are broader and lower than the input data, despite convergence to a low σ_{ray} value. Thus, unless the conditions for uniqueness of the solution to the equations represented by the ray averages are met, a low σ_{ray} value does not guarantee that a true concentration distribution has been determined. Rather, a low σ_{ray} value merely indicates convergence to one of many possible solutions.

Underdetermination does not provide a likely explanation for the discrepancy between Figures 4.2 (a) and 4.2 (b). In contrast to expectation, the peak in the FTIR reconstruction (Fig. 4.2 (b)) is higher and narrower than the FID data would suggest. Also, σ_{ray} is relatively high for Figure 4.2 (b), indicating that no single distribution matched even the limited ray data that was provided. This is the opposite of the problem of underdetermination, where there are too many solutions so the program fails to converge to the true one.

A third limit on CT reconstruction is the spatial sampling frequency, referring to the number of rays per unit of projection length (i.e. the sample frequency f in rays/m). Intuitively, one recognizes the importance of the sampling frequency when trying to capture a steep concentration gradient. For example, to measure a situation where the concentration changes from near background to a peak over a distance of $d = 0.3$ m, one must sample at intervals smaller than d . Sampling over much larger intervals will result in a poor reconstruction of the actual concentration, either by missing the peak or underestimating the gradients. In general, to satisfy the Nyquist criterion we need $f \times d > 2$, where d is the smallest dimension to be resolved in the physical system. The CT results presented here had a sampling frequency of 3 rays/m, which despite the correspondingly limited spatial resolution appeared to reasonably capture a large concentration gradient

and correctly identify the peak location. The important point remains however, that the sampling frequency is not simply an arbitrary quantity dictated by the desired image resolution but one which ultimately depends on the physical distribution under study.

One motivation for choosing a remote optical sensing system is its non-obtrusive nature. The system used here could not fully realize this goal because of limitations imposed by the sizes of the room, the instrument and the test apparatus. A second motivation is the potential for rapid sampling. Although the RS-FTIR gathers data very quickly (up to 2 readings per second), rapid scanning could not be achieved due to limitations in the positioning system. In fact, the ray path sampling procedure in this experiment was quite slow, comparable in speed to the conventional FID sampling technique. Better RS-FTIR sampling technology, such as a scanning system employing moving mirrors, multiple mirrors, or multiple sources and detectors, as suggested by Todd and Leith (1990) could more fully realize the potential for unobtrusive and rapid sampling.

4.6 Conclusion

In this chapter the application of CT to an important class of problems has been demonstrated: in situ measurements of a gaseous species concentration profile in the vicinity of a constantly emitting source. Whether dedicated to outdoor readings, workplace situations, or to indoor air measurements the problems outlined above must be addressed. The issues of indeterminacy and spatial resolution are essentially technical limitations, in that both can be solved by gathering more data. These factors may have contributed to the discrepancies in the reconstruction data. However, the effects of sample point location and temporal variability remain a likely explanation for the disagreement between the FID point data and the CT image, as suggested by the high value of σ_{ray} for that reconstruction. Interestingly, there are two ways to address temporal variability in the projection data; one is to quickly sample all rays simultaneously obtaining a "snapshot" view. The other approach is to measure time-averaged

concentrations over long periods. The latter method was attempted with only limited success, suggesting that more rapid sampling may be necessary.

In summary, this work demonstrates for the first time the feasibility of CT reconstructions of pollutant concentrations in a real room setting. Good qualitative agreement between the reconstruction and point sample data was obtained. The limitations encountered due to the temporal resolution, size and geometry of the experimental apparatus appear surmountable with better instrumentation.

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4.8 References

- Brooks R.A. and Di Chiro, G. (1975) Theory of Image Reconstruction in Computed Tomography. *Radiology*, 117, 561-572.
- Fraser, M. (1989) Unpublished results of summer study at Indoor Environment Program, 1 Cyclotron Rd. Bld 90, Lawrence Berkeley Laboratory, Berkeley, CA.

- Grant, W.B. and Menzies, R. T. (1983) A survey of laser and selected optical systems for remote measurement of pollutant gas concentrations, *J. Air Pollution Cont. Assoc.*, 33, 187-194.
- Hertz, H.M. (1985) Experimental Determination of 2-D Flame Temperature Fields by Interferometric Tomography, *Optics Communications*, 54 (3), 131-136.
- Jones, T.A., Hamilton, D.E. and Johnson, C.R. (1986) Contouring Geologic Surfaces with the Computer, Van Nostrand Reinhold, New York, NY.
- Kak, A.C. and Slaney, M. (1988) Principles of Computerized Tomographic Imaging, New York, NY, IEEE Press.
- Levine S.P., Ying, L.S., Strang, C. and Hong-Kui, X. (1989) Advantages and disadvantages in the use of Fourier transform infrared (FTIR) and Filter IR (FIR) spectrometers for monitoring airborne gases and vapors of industrial hygiene concern, *Appl. Ind. Hyg.*, 4(7), 180-187.
- Peterson, J.E. (1986) The Application of Algebraic Reconstruction Techniques to Geophysical Problems, Ph.D. Dissertation, Dept of Geophysics, University of California, Berkeley, CA.
- Roach, S.A. (1981) On the Role of Turbulent Diffusion in Ventilation, *Ann Occ. Hyg.* 24(1), 105-132.
- Roberts, O.F.B. (1923) The Theoretical Scattering of Smoke in a Turbulent Atmosphere, *Proc. Royal Soc. Series A*, 104, 640-654.
- Schlichting, H. (1979) Boundary Layer Theory, 7th. ed. McGraw-Hill, NY, New York .
- Seinfeld, J. (1986) Atmospheric Chemistry and Physics of Air Pollution, John Wiley, NY, New York.
- Simonds, M., Xiao, H. and Levine S. P. (1994) Optical Remote Sensing for Air Pollutants — Review, *Am. Ind. Hyg. Assoc. J.*, 55, 953-965.
- Todd, L. and Leith, D. (1990) Remote sensing and computed tomography in industrial hygiene, *Am. Ind. Hyg. Assoc. J.*, 51, 224-233.

- Todd, L.A. (1992) Optical Remote Sensing/ Computed Tomography Beam Geometries for Monitoring Workplace Gases and Vapors. Proceedings of *Optical Remote Sensing Applications to Environmental and Industrial Safety Problems*, Air & Waste Management Association , SP81, 390-393.
- Xiao, H.K., Levine, S. P., W.F. Herget, D'Arcy, J.B., Spear, R.C. and Prichett,T. (1991) A transportable remote sensing infrared air monitoring system, *Am. Ind Hyg. Assoc. J.*, 52, 4375-5381.
- Ying, L.S. and Levine, S.P. (1989) Fourier Transform Infrared Least-Squares Methods for the Quantitative Analysis of Multicomponent Mixtures of Airborne Vapors of Industrial Hygiene Concern, *Anal. Chem.*, 61, 677-683.
- Yost, M.G., Xiao, H.K. , Spear, R.C. and Levine, S.P. (1992) Comparative Testing of a FTIR Remote Optical Sensor with Area Samplers in a Controlled Ventilation Chamber. *Am. Ind. Hyg. Assoc. J.*, 53, 611-616.

Figure 4.1 (a) Isometric view of the test facility, showing the RS-FTIR mounted on the translating table attached to the rotating base. The FID array was arranged perpendicular to the beam path and rotated with the base. The source was suspended from the ceiling at a fixed location 60 cm upstream from the geometric center of the room. (b) Plan view of the ray path geometry and CT grid used for the reconstructions. The dark dots mark locations corresponding to the FID array sampling points for each projection angle. The arrows indicate the direction of movement for the translating stage when scanning one of the 4 projection angles (0 , 90 , ± 45 degrees) defined by the position of the rotating base.

Figure 4.2 (a) Acetone concentrations obtained from kriging the FID data.

Figure 4.2 (b) Acetone concentrations reconstructed from ray path averages from RS-FTIR data. ILST algorithm used for CT reconstruction, 1000 iterations, relaxation parameter = 6, σ_{ray} at convergence = 0.4.

Figure 4.2 (c) Acetone concentrations reconstructed from ray path averages calculated from the kriged FID data (Figure 4.2). ILST algorithm used for CT reconstruction, 1000 iterations, relaxation parameter = 10, σ_{ray} at convergence = 0.0024.

CHAPTER 5

NOVEL APPROACH FOR TOMOGRAPHIC RECONSTRUCTION OF GAS CONCENTRATION DISTRIBUTIONS IN AIR: USE OF SMOOTH BASIS FUNCTIONS AND SIMULATED ANNEALING*

5.1 Abstract

Optical remote sensing and iterative computed tomography (CT) can be applied to measure the spatial distribution of gaseous pollutant concentrations. Chamber experiments were conducted to test this combination of techniques using an open path Fourier transform infrared spectrometer (OP-FTIR) and a standard algebraic reconstruction technique (ART). Although ART converged to solutions that showed excellent agreement with the measured ray-integral concentrations, the solutions were inconsistent with simultaneously gathered point-sample concentration measurements. A new CT method was developed that combines (1) the superposition of bivariate Gaussians to represent the concentration distribution and (2) a simulated annealing minimization routine to find the parameters of the Gaussian basis functions that result in the best fit to the ray-integral concentration data. This method, named Smooth Basis Function Minimization (SBFM), generated reconstructions that agreed well, both qualitatively and quantitatively, with the concentration profiles generated from point sampling. I present an analysis of two sets of experimental data that compares the performance of ART and

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SBFM. I conclude that SBFM is a superior CT reconstruction method for practical indoor and outdoor air monitoring applications.

5.2 Introduction

In the previous chapter I described the first test of indoor air concentration measurements in a large horizontal plane using the computed tomography/Fourier transform infrared spectrometer (CT/FTIR) method in a chamber. The test used a scanning geometry comprising four sets of parallel rays, with each set rotated 45° with respect to the previous one. Although ideal for iterative CT reconstruction, this approach suffered from limited temporal resolution. Only one source and detector were available for those experiments. Both had to be moved substantially to produce the desired ray pattern. The resulting scan time was 3 hours. During this time, fluctuations in the concentration pattern, and possibly changes in the airflow and dispersion due to the variable positioning of the equipment, produced inconsistent ray-integral concentration measurements. Spatial resolution was also limited due to the relatively large distance between parallel rays. Thus, the CT reconstruction in this study was not entirely satisfactory. While the peak concentration was correctly located, most of the ray-integral concentrations of the reconstructed profile did not agree well with the experimental ray-integral data.

During October and November 1993, I undertook a series of experiments designed to improve the CT-FTIR method by using a steerable FTIR instrument in conjunction with fixed mirrors and retroreflectors at the perimeter of a test chamber*. With this system, the total time for a single scan was reduced from three hours to just over six minutes.

*The experiments were conducted in collaboration with Doo Yong Park, Michael Yost, Steven Levine, Robert Spear, Jyun De Wu and Zhou Yi.

By necessity, the ray geometry differed substantially from that employed in the previous chapter. This new ray geometry, though permitting us to largely overcome the problem of temporal variability, created false spatial variability in the reconstructed concentration profiles when used in conjunction with the standard algebraic reconstruction technique (ART) suggested by Todd and Leith (1990) and Todd and Ramachandran (1994). Because the rays in the new geometry were less "independent" than would be an equal number of rays in parallel projections, they produced a more underdetermined system of equations when using the number of pixels necessary to accurately capture spatial variability in the concentration profiles.

Using two of the experimental runs as illustrative examples, this chapter describes how CT reconstructions were first attempted with ART, how and why they failed, and then introduces a new reconstruction method that overcomes the difficulties of ART. The new method is based on modeling the concentration distribution with smooth basis functions (using bivariate Gaussians in this case). For the two experimental runs considered, I compare measured point concentration data, concentration profiles generated by ART and concentration profiles generated by the new smooth basis function minimization technique (SBFM). The full set of experiments are described in detail in Chapter 6.

5.3 Method

5.3.1 Experiments

Experiments were conducted in a 6.4 m $\times$ 7.3 m $\times$ 2.4 m high chamber located within a larger building (Fig. 5.1). The chamber had a wooden frame covered by a thick polyethylene liner. One of the short walls had two 46 cm $\times$ 6.4 m openings centered at heights of 67 cm and 1.75 m. Along the opposite side of the chamber, in the ceiling, was a 92 cm wide slot. All openings were covered with coarse-fibred furnace filter material.

The ceiling slot was connected to a fume hood which drew air through the chamber at a flow rate of $\sim 5600 \text{ m}^3 \text{ h}^{-1}$, i.e. approximately 50 room volumes per hour. The center of the fume hood was not exactly in line with the centerline (parallel to the y-axis) of the chamber, but was 50 cm away from the centerline in the positive x direction. The time constant in a tracer gas decay experiment was found to be just under 3 minutes for a point at the center of the chamber, corresponding to an effective air exchange rate of 21 room volumes per hour. The chamber was clearly not well-mixed, and so we could expect concentration gradients when releasing a steady stream of tracer gas.

In both experiments considered in this chapter, a buoyant mixture of 10% sulfur hexafluoride (SF_6) in helium was released at a constant rate from a 2.5-cm diameter, porous ceramic sphere, placed 60 cm above the floor. Once steady-state conditions were established, SF_6 concentrations were measured over a period of one hour in a horizontal plane 1.3 meters above the floor. Two measurement methods were employed and these will be referred to as "point-sample" measurements and "ray-integral" or "path-integral" measurements, respectively. (1) Air samples were collected continuously into gas sampling bags at 26 points and were subsequently analyzed for SF_6 by a photoacoustic detector (Brüel & Kjær (B&K), Model 7620). During each experiment, the photoacoustic detector was used to sample air from five additional points in the plane (and one point in the fume hood, sampling the chamber exhaust air). Air from each of these points was drawn continuously, at a flow rate of 1 l/min, through a 2.3 l glass jar and sampled from the jar by the detector once every six minutes. For the present analysis, SF_6 concentrations at these five points were time-averaged over the duration of the experiment. (2) Path-integral SF_6 concentrations were measured over 56 intersecting ray paths using a centrally placed, steerable OP-FTIR system, 4 large retroreflectors and 52 flat mirrors placed along the perimeter of the chamber. (See Figure 5.1) Nine complete sweeps (of all 56 rays) were gathered over the course of one hour.

5.3.2 Data Processing

For each orientation of the FTIR, the mean, μ_r , and variance, σ_r^2 , of the ray-integral concentrations for the nine measurements were computed. Half of the CT reconstructions were based on the measured time-average ray-integral concentrations, μ_r . The other half were based on synthetic, noise-free ray-integral concentration data.

Figure 5.2 summarizes how the point-sample data and the ray-integral data were processed. Six maps of SF₆ concentration as a function of position in the plane, designated by letters (a)-(f), were generated for each experiment. For the first five maps, the measurement plane was divided into 195 (13 × 15) square pixels (0.22 m² each). The point sample data, (a), were kriged (Jones et al., 1986) using the graphing program SURFER (Golden Software, Golden, CO) and the value at the center of each pixel was plotted in graphs marked (b). The kriged point sample data were also used to generate "synthetic ray integrals" (strip sums, based on the assumption of a uniform concentration in each pixel) for the 56 ray paths used in the experiment. These synthetic ray-integral concentrations were entered into the ART-CT program to test the algorithm under noise-free conditions and compare the resulting concentration maps, marked (c), to those based on the measured ray-integral concentrations, marked (d). SBFM-CT reconstructions were also performed on the synthetic and the measured ray-integral data. Resulting concentration maps are denoted (e) and (f), respectively.

A figure of merit was defined to quantify the overall difference between the various CT reconstructions and the measured point-sample data:

$$\sigma_{\text{pnt}} = \left(\frac{\sum_{\text{pt}=1}^{31} (C_{\text{pt},m} - C_{\text{pt},\text{CT}})^2}{\sum_{\text{pt}=1}^{31} (C_{\text{pt},m})^2} \right)^{1/2} \quad (5.1)$$

where $C_{pt,m}$ is the measured point-sample concentration at a given location and $C_{pt,CT}$ is the reconstructed concentration at that location. A value of zero for the parameter σ_{pnt} would indicate perfect agreement between the measured point-sample data and the concentrations at those same points calculated by the CT programs.

An equivalent parameter was defined for the CT reconstructions based on synthetic ray integrals calculated from the kriged point concentration data:

$$\sigma_{pix} = \left(\frac{\sum_{p=1}^{195} (C_{p,k} - C_{p,CT})^2}{\sum_{p=1}^{195} (C_{p,k})^2} \right)^{1/2} \quad (5.2)$$

where $C_{p,k}$ is the concentration in a pixel obtained from kriging and $C_{p,CT}$ is the pixel concentration calculated by the CT reconstruction program.

5.4 Point Sample Data

Figures 5.3 (a) and 5.4 (a) show the time-averaged SF_6 concentrations at the sampling points in the plane of measurement for Experiments 1 and 2, respectively. The location of the SF_6 source and the general direction of air flow in each case is also shown. It appears that in Experiment 1, one major and one minor concentration peak were present, while for Experiment 2 only one major peak can clearly be distinguished. This interpretation is consistent with the results of flow visualization tests using smoke as a tracer. Smoke released near the SF_6 source point in Experiment 1 was swept immediately up through the measurement plane by the incoming air stream and was dispersed further downstream. This transport pattern would be expected to produce a high concentration of SF_6 just downstream of the source, and a secondary, broader peak in concentration further downstream, as the SF_6 is dispersed back down into the measurement plane. By contrast,

in Experiment 2, smoke released near the SF₆ source was immediately dispersed significantly in all directions while slowly being advected in the general direction of the exhaust hood.

Kriged point-sample data are presented in Figures 5.3 (b) and 5.4 (b). The broadness of the dominant peak in Figure 5.3 (b) and the local minimum visible near position ($x = 5.5$, $y = 13.5$) in Figure 5.4 (b) are artifacts of the kriging interpolation. There is no evidence for these features in the point-sample data. This point should be kept in mind when comparing these figures with the CT reconstructions of the experimental ray-integral concentration data.

5.5 Art Reconstruction

5.5.1 Method

The ART algorithm iteratively adjusts concentrations in pixels along each ray in sequence, so that the difference between the measured ray integral concentration and that calculated using the assumed pixel concentrations is minimized (Brooks and Di Chiro, 1975). ART effectively weights all rays equally in the reconstruction process, as opposed to, for instance, weighting rays by their measurement uncertainty. In the present implementation, the initial guess for the concentration profile was a uniform distribution. A ray-average concentration for each ray was first obtained by dividing the ray-integral concentration by the area of the ray in the measurement plane. The mean of all 56 ray-average concentrations was then taken to be the first guess for the concentration in each pixel.

After each iteration, the value of a parameter called σ_{ray} (defined in analogy with σ_{pnt} and σ_{pix}) was calculated:

$$\sigma_{\text{ray}} = \left(\frac{\sum_{r=1}^{56} (\text{PIC}_r - \text{PIC}_{r,\text{CT}})^2}{\sum_{r=1}^{56} (\text{PIC}_r)^2} \right)^{1/2} \quad (5.3)$$

where PIC_r is the “true” path-integral concentration for ray r ($\text{PIC}_r = \mu_r$ for FTIR data, or PIC_r = calculated value from kriged point concentration data), and $\text{PIC}_{r,\text{CT}}$ is the path-integral concentration calculated from the reconstructed profile. For ART,

$$\text{PIC}_{r,\text{CT}} = \sum_{p=1}^{195} a_{p,r} C_{p,\text{CT}}, \text{ where } a_{p,r} \text{ is the area of pixel } p \text{ intercepted by ray } r.$$

The parameter σ_{ray} quantifies the overall agreement between the “true” path-integral concentrations, PIC_r , and those calculated from the reconstructed concentrations. For the kriged data, the ray integral, PIC_r , was calculated as a strip sum using the experimental ray width as the width of the strip. The iteration process was terminated either when the fractional change in σ_{ray} from one iteration to the next was less than 0.5% or when the value of σ_{ray} fell below 0.001.

The order in which ART was applied to rays to re-evaluate the pixel concentrations along them was found not to influence the final solution significantly. The only constraint applied to the reconstruction process was that pixel concentrations could not be less than zero.

5.5.2 Results

Figures 5.3 (c) and 5.4 (c) show the concentration profiles produced by applying ART to the synthetic ray-integral data generated from the kriged point-sample measurements. The very low values of σ_{ray} , seen in the first column of Table 5.1, show that the reconstructions generated by ART are valid solutions given the synthetic ray-

integral concentrations. In fact, the value of σ_{ray} can be made to be as low as desired (without changing the qualitative features of the concentration map generated) by increasing the number of iterations. However, since the input data for these reconstructions came from a known concentration profile and were noise-free, one would expect a perfect match between frames (b) and (c). In other words, Figures 5.3 (b) and 5.4 (b) each definitely represent a solution to the respective synthetic ray inversion problem solved by ART. It is apparent from Figures 5.3 (c) and 5.4 (c) that ART did not converge to these "true" solutions. The peak of the concentration was correctly located in each case, but the peak magnitude is incorrect. In addition, many secondary artifacts, such as minor peaks and ridges, not present in the original kriged maps, appear in the ART reconstructions. The overall discrepancy is manifested in the relatively high values of σ_{pix} shown in the first column of Table 5.1: 0.62 for Experiment 1 and 0.45 for Experiment 2.

By starting the ART reconstruction with different first guesses or by manipulating the algorithm in other ways the program could be made to converge to other solutions which are equally valid but distinct in detail. Only if the first guess is very close to the true solution does ART converge to the true solution. These unsatisfactory features are a consequence of a fundamental problem: ART embodies a seriously underdetermined system. With 195 unknowns (pixel concentrations) and only 56 equations (ray integrals), problems with indeterminacy were to be expected. Previous simulation work (Todd and Leith, 1990; Todd and Ramachandran, 1994) has shown that, under favorable conditions, good reconstructions can be obtained for an underdetermined system with the ratio of the number of pixels to the number of rays being as large as 6.7. In those reported cases, the reconstructed pixel concentrations matched the true distribution well. However, the concentration profiles used in the work of Todd and coworkers were less challenging (in the sense that they mitigated the problem of indeterminacy) because they were composed of narrow Gaussian peaks centered within the reconstruction plane. Complete, narrow,

and distinct peaks pose a relatively easy challenge for ART, because only a small fraction of the rays pass through the peak, thus limiting the extent to which the detected species can be spatially dispersed in the reconstruction into so-called ghost images. In contrast, the broad peaks evident in the present kriged experimental data are intercepted by almost all rays. This feature magnifies the indeterminacy problem by increasing the number of independent ways in which the species mass can be allocated amongst pixels without affecting the ray-integral concentrations.

The problem of indeterminacy can be mitigated by using more favorable ray geometries, such as parallel projections. However, if the scanning time is to remain short, this approach requires the use of many sources and detectors or ample steerable optics. Presently, the cost of both of these options is prohibitive.

Another approach to lessen the impact of indeterminacy is to make individual pixels larger, thereby reducing the number of pixels and correspondingly decreasing the number of unknowns. However, the number of pixels is also directly related to the spatial resolution of the reconstruction, so that a decrease in the number of pixels may reduce the resolution to the point where the true spatial variation in concentration can no longer be accurately represented. I attempted ART on the measured ray integral concentrations of Experiment 1 with a pixel grid of 7 by 8 (i.e. 56 pixels, equal to the number of rays). ART could not generate a reconstruction with a σ_{ray} below 0.3, indicating that *no* adequate solution to the ray integrals exists at this coarse discretization.

Figures 5.3 (d) and 5.4 (d) show the ART reconstructions of the measured (FTIR) ray-integral concentration data. These concentration maps are even less satisfactory than the ART reconstructions of the synthetic ray-integral data. This is also reflected in the high values of σ_{pnt} : 0.41 for Experiment 1 and 0.93 for Experiment 2 (Table 5.1). Figure 5.4 (d) shows the peak concentration for Experiment 2 to be near the origin. There is no evidence for this in the measured point data, and it is highly unlikely that this was the true

location of the peak concentration given the position of the SF₆ source and direction of air motion (as shown in Figure 5.4 (a)). Nevertheless, the goodness-of-fit parameter σ_{ray} is low (0.026) in this case, tempting one to believe that a correct reconstruction has been achieved. It is clear that applying ART to the reconstruction of gas concentration distributions in air using practical, albeit non-ideal ray geometries can lead to serious misrepresentations.

5.6 SBFM Reconstructions

In view of these problems with ART, I developed an alternative approach that eliminates indeterminacy. This approach recognizes that the solutions can be constrained to a smaller domain of realistic possibilities for a given experimental context by imposing a functional form on the concentration profile. The artificial discretization of space into pixels is not necessary for reconstructions based on measured ray-integral data. The many sharp and mostly unrealistic gradients at pixel boundaries evident in the ART reconstructions are avoided. This approach incorporates the a priori expectation that turbulent air motion will tend to disperse pollutants and thus smooth out concentration gradients. With only a few pollutant sources, no more than three or four local concentration maxima would be anticipated under normal ventilation conditions.

5.6.1 Method

In the newly developed approach, the concentration distribution to be determined (from synthetic or measured ray-integral data) was modeled as a superposition of bivariate Gaussians defined in the x-y plane. The Gaussian was chosen as a basis function because it is easy to calculate and yet versatile enough to satisfactorily represent a broad range of possible concentration profiles. Each Gaussian has six adjustable parameters: x and y coordinates of the peak, standard deviations along its two principal axes, peak height

(maximum concentration) and angle of rotation of the principal axes relative to the x and y axes.

The ideal Gaussian parameters were assumed to be those for which σ_{ray} , as defined in equation (5.3), was minimized. Following standard experimental data fitting theory (Bevington, 1969), an alternative objective of the reconstruction procedure might have been to minimize the *weighted* sum of squares of deviations of the data, PIC_r , from the ray sums calculated using the Gaussian concentration profiles, $\text{PIC}_{r,\text{CT}}$. Thus, for the measured ray-integral data, the reduced chi-squared, defined below, might have been minimized.

$$\chi_v^2 = \frac{1}{(56 - n)} \sum_{r=1}^{56} \frac{1}{\sigma_r^2} (\text{PIC}_r - \text{PIC}_{r,\text{CT}})^2 \quad (5.4)$$

where v is the number of degrees of freedom after adjusting n fitting parameters to match the 56 ray integral concentration values ($v = 56 - n$, where n is six times the number of Gaussians used for the fit). However, after analyzing the two data sets presented here and the steady-state experiments described in Chapter 6, it was found that minimizing the reduced chi-squared often did not lead to the most accurate reconstruction, for the reasons outlined in the two paragraphs below.

For many degrees of freedom, the reduced chi-squared will be close to unity if (1) the correct fitting function is chosen and (2) the measurement errors, σ_r , are Gaussian distributed and accurately estimated. For a set of measurements of an unchanging quantity, the measurement uncertainty would be calculated as the error in the mean (σ_r divided by the square root of the number of observations, N). However, in our case, the measured quantity (the ray-integral concentration for each ray) was itself fluctuating in time. The uncertainty was therefore estimated as σ_r . Consequently, the weighting factor

for the deviation term for each ray-integral concentration is simply the inverse of its variance rather than N/σ_r^2 .

Careful analysis of the data for the 11 steady-state experiments revealed two important factors. (1) For many rays, especially those passing through areas of the plane near the SF_6 source, the deviations in the measured path-integral concentrations from the mean were not Gaussian distributed. Thus, one of the criteria for the proper calculation of χ_v^2 was not satisfied. (2) Moreover, variability in path-integral concentrations of rays having a relatively low μ_r was often disproportionately low. Because the weighting factor for each ray in the calculation of χ_v^2 was the standard deviation *squared*, σ_r^2 , the minimization of χ_v^2 tended to put undue emphasis on the rays with low μ_r . Thus, the peak concentrations in the reconstruction would tend to be much less accurate than when minimizing σ_{ray} . These two factors combined lead to the conclusion that, while calculating χ_v^2 for a reconstructed profile provided useful information about the quality of the reconstruction, it was not the ideal function to be minimized in the SBFM approach.

For SBFM-CT reconstructions using measured ray integrals (profiles marked (f) in our case) the concept of pixels is superfluous and was discarded. Instead, each path-integral concentration, $\text{PIC}_{r,\text{CT}}$, was first evaluated analytically as a line integral along the centerline of the ray. This result was then multiplied by the width of the ray to find the value of the strip integral. Given that the ray width was 43 % of the width of the pixels employed in all other reconstructions, the spatial resolution was approximately doubled by this procedure. For this reason, the graphs shown in Figures 5.3 (f) and 5.4 (f) were plotted at twice the grid density compared to the other surface plots. Moreover, the resulting concentration distribution was no longer a discontinuous function, thus making it a less artificial representation of reality. In this lies the crux of the SBFM method: a model is imposed on the underlying concentration distribution that is closer to reality than that assumed by ART (completely independent concentration values distributed into

pixels). The fact that the domain of permitted concentration profiles in SBFM is much more constrained results in a decrease in the number of degrees of freedom available to the reconstruction and thus a decrease in the number of possible solutions.

The SBFM program was written in Turbo Pascal and based on the Amebsa routine in Numerical Recipes (Press et. al., 1992). It uses a combination of the simplex method and a simulated annealing technique to find the set of Gaussian parameter values that minimizes σ_{ray} . Simulated annealing, ideally, can find the global minimum of a function (rather than a local minimum close to the first guess) by mimicking the gradual cooling of a crystallizing liquid. For such a liquid, the lowest energy state is that of the perfect crystal (global minimum) rather than that of the amorphous solid (local minimum) produced by rapid cooling.

One issue of practical importance is how to select an appropriate number of Gaussians for the reconstruction. In the present study, the SBFM program was first run for each experiment using only one fitting parameter: a constant concentration in the plane of reconstruction. Then the program was run using one Gaussian, two Gaussians and, for Experiment 1, three Gaussians. Each time a Gaussian was added, the parameters of the previously obtained Gaussian peaks were entered into the program as a starting condition to reduce the computation time. The values of σ_{ray} and χ^2_{ν} were noted as a function of the number of fitting parameters. When the relative improvement (decrease) in σ_{ray} or χ^2_{ν} became small compared to the increase in the number of fitting parameters, it was assumed that a reasonable fit had been achieved. The synthetic data were modeled with two Gaussians in the SBFM approach, since this number of Gaussians seemed the minimum number necessary to be able to capture the major features apparent in the kriged maps of concentration.

5.6.2 Results

Figures 5.3 (e) and 5.4 (e) show the results of the reconstruction from synthetic ray-integral data by SBFM using two Gaussians. Clearly, the agreement with the “true” concentration profiles (Figures 5.3 (b) and 5.4 (b), respectively) is superior to the ART reconstructions. This superiority is also evident in that the values of σ_{pix} (Table 5.1, second column) are approximately half of what they are for ART. On the other hand, the σ_{ray} values are more than ten times higher with SBFM than with ART, although still quite good at 0.067 and 0.040, respectively, for the two experiments. The poorer value of σ_{ray} under SBFM reflects the fact that there are fewer degrees of freedom for matching ray integrals relative to ART.

Compared with ART, the maps generated by applying SBFM to the *experimental* ray-integral concentration data (Figures 5.3 (f) and 5.4 (f)) are much more consistent with the measured point-sample data. This is also reflected in the significantly lower σ_{pnt} values of the SBFM reconstructions (see Table 5.1). The near-source differences between point-sample data and SBFM reconstructions may be real, since high spatial and temporal variability close to the SF_6 source, combined with the differences in sampling methods between the bag-sample and ray-integral measurements would lead to somewhat different information contained in each data set. For instance, time variations are completely smoothed in the point-sample data, but they are a source of noise in the ray-integral reconstructions. In addition, for Experiment 1, the peak concentration in the reconstruction occurs at a point between the two point sampling locations closest to the SF_6 source. Perhaps if a higher point sampling density had been used near the source, the measured peak point concentration would have been closer to that predicted by the reconstruction. Given the excellent reconstruction of the peak in the kriged data (comparing Figures 5.3 (e) and 5.3 (b)), I do not believe that any feature of the SBFM significantly distorted the experimental peak.

Figure 5.4 (f) shows a markedly flatter profile with a lower peak concentration than 5.4 (b) and 5.4 (e). Again, I believe this reflects the nature of the measured ray-integral data rather than an artifact of the SBFM routine. When comparing the ray integrals generated from the kriged point sample data to those measured by the FTIR, we find that in the former data the differences among rays are far more pronounced than in the experimental data, i.e. the highest ray-integral concentrations based on the kriged point sample data are up to 30% higher than the corresponding measured ones, and the lowest ray-integral concentrations based on the kriged point sample data are lower than the corresponding measured ones. This observation may be partially due to vertical concentration gradients, which have not been discussed thus far. The cross-section of the FTIR beam had a diameter of approximately 20 cm. Thus, it sampled 20 cm in the vertical as well as the horizontal direction. The sampling for point concentration measurements was done 5 cm above the lower edge of the FTIR beam. Thus, most of the volume scanned by the FTIR was slightly above the plane of the point samplers. Since the SF_6 source was beneath the plane of measurement and the air exhaust was on the ceiling, we expect concentration gradients in the horizontal plane to decrease with height. This would cause the FTIR to sample a more even concentration distribution than the point samplers.

Temporal variability during Experiment 2 probably also contributed to the discrepancy between 5.4 (f) and 5.4 (a)/(b). The four highest concentrations shown in 5.4 (a) were derived from the time-resolved B&K data. The SF_6 concentration at these points exhibited significant variability during the experiments: at two of these points, concentrations steadily decreased from ~ 14 ppm to ~ 0.5 ppm during the experiment, at a third point, the concentration steadily increased from ~ 1 ppm to ~ 12 ppm, and at the fourth point, the concentration fluctuated between 4 ppm and 10 ppm. I believe that these changes occurred owing to varying temperatures inside and outside the building, and the resulting changes in the mixed convection flow in the enclosure. Thus, the highest point

concentrations plotted in Figures 5.4 (a) and 5.4 (b) do not represent truly time-stationary concentrations, and therefore perfect agreement between 5.4 (a)/5.4 (b) and 5.4 (f) should not be expected.

The σ_{ray} values for Figures 5.3 (f) and 5.4 (f) are 1.3 and four times as high, respectively, as the same values for Figures 5.3 (e) and 5.4 (e). There are probably several reasons for this (these reasons also apply to the ART reconstructions). The most obvious is that the former reconstructions are based on experimental data, which have errors associated with them, while the latter are based on internally consistent synthetic data. The error in the experimental data comes partly from the FTIR and partly from the sequential sampling and time averaging process associated with repeated, non-instantaneous scans. One would prefer to reconstruct concentrations based on snapshots of rays throughout the plane (i. e., all ray-integral concentrations would be sampled simultaneously). Such data would not suffer from any temporal fluctuations in the concentration distribution which lead to inconsistencies in the ray-integral data set. Unfortunately, given technological constraints, this objective is prohibitively costly.

Other factors contributing to the inferior σ_{ray} values for the reconstructions of the experimental ray-integral data are the artificial discretization of the reconstruction plane into pixels in ART and the assumption of constant concentration across the ray width in this implementation of SBFM. These factors create a discrepancy between the measured concentration path integral, which is a truly continuous function of position, and the computed integral, which is only piecewise continuous. For regions of low spatial variability in concentration, this discrepancy is likely to be of minor consequence; however, it can have a marked effect in areas of steep concentration gradients. I have observed this effect with SBFM. At the outset of this work, SBFM was applied to the experimental data while still employing a pixel discretization. By switching to line integration, agreement between measured and reconstructed ray integrals was greatly

improved for rays passing through areas of steep concentration gradients (e.g., close to the dominant peak in Figure 5.3 (b)).

5.6.3 Choosing the Proper Number of Gaussians

What is the appropriate number of Gaussian basis functions for modeling the data in a CT reconstruction? Clearly, one Gaussian would be the minimum necessary to represent anything but a perfectly mixed condition. At the other extreme, using as many Gaussians as pixels would constitute a functional equivalent of an ART reconstruction. Between these limits, it seems that the number of Gaussian basis functions should be guided by the principle of Occam's razor: parameters introduced to the model must not be increased beyond necessity. Evaluating the χ_v^2 of a reconstruction helps to determine when this point has been reached, because χ_v^2 is the ratio of the estimated variance of the fit to the variance of the parent distribution, normalized by the number of degrees of freedom n . If the fitting function is a good approximation to the parent distribution and the uncertainties in the measured parameters are correctly quantified, then χ_v^2 should be approximately unity.

Figure 5.5 shows how χ_v^2 and σ_{ray} change with the number of fitting parameters for the two experiments. The first point in each graph represents the case in which the concentration distribution is modeled as a constant. Thereafter, there are six fitting parameters for every Gaussian added to the model. As expected, σ_{ray} decreases each time the number of fitting parameters is increased, but the decrease diminishes for each successive case. χ_v^2 mostly decreases with σ_{ray} , but may increase slightly if σ_{ray} decreases only very slightly (Fig. 5.5 (b)).

For Experiment 1, the value of χ_v^2 decreases to below 2, to 1.2, when using three Gaussians, indicating that 3 Gaussians should be an appropriate number. It is useful also to look at the behavior of σ_{ray} , in which all rays carry equal weight. Between a constant concentration and one Gaussian, σ_{ray} hardly decreases: the single Gaussian produced by

the minimization is broad and flat, thus behaving almost like a constant concentration. Most ray integrals are hardly affected by this relatively minor change in the concentration profile. With two Gaussians, a dominant peak appears. This allows for substantial improvement in the fit for rays passing through the peak, which is reflected in a substantial decrease in σ_{ray} . Adding a third Gaussian allows the secondary concentration peak to be refined, leading to a further significant (20%) decrease in σ_{ray} . An additional Gaussian would not be expected to lead to a further significant decrease, as both σ_{ray} and χ_v^2 are leveling out at this point.

Since the χ_v^2 value for the SBFM reconstruction of Experiment 1 is larger than unity, the overall difference between the ray integrals of the fit and the measured ray integrals is not within the estimated measurement error. Two likely explanations exist: (1) the true concentration profile is not well represented by bivariate Gaussians and a different functional form would be more appropriate; or (2) the true uncertainty in the measured ray integrals is larger than that estimated by using σ_r^2 . The latter factor is likely to be a significant contributor. Drift of the SF_6 plume during the experiment or a change in the airflow pattern within the room due to temperature changes, and thus a drift in the ray integral concentration of some of the rays, would lead to an underestimation of σ_r and thereby a non-ideal χ_v^2 value. In fact, there is evidence of such drift in the time-resolved ray integral data: sets of several adjacent rays manifest similar patterns of change from one sweep of the FTIR to the next. Hence, path-integral concentration values for each ray are not necessarily Gaussian distributed, thereby violating an underlying assumption of chi-squared theory. Nevertheless, the chi-squared approach provides a useful framework for interpreting reconstruction results.

For Experiment 2, both χ_v^2 and σ_{ray} change very slightly in going from one to two Gaussians, so it seems that a single Gaussian is adequate. The values of χ_v^2 are all about one tenth of what one would expect (the best value is around 0.11 instead of unity). Most

of the ray variances were rather large in this experiment. Whereas the mean relative standard deviation of the time-averaged ray-integral concentrations was 15% in Experiment 1, it was 79% in Experiment 2. The quality of the fit, as quantified by σ_{ray} for the final result, is also worse (0.084 for Experiment 1, 0.16 for Experiment 2). The large relative standard deviations in Experiment 2 indicate strong temporal fluctuations in concentration during that experiment, causing the mean path-integral concentration for each ray to be less accurate than in Experiment 1. With the ray *variances* in Experiment 2 being disproportionately larger, however, the χ^2_v value was correspondingly smaller for this experiment. As discussed in the previous paragraph, the χ^2_v value alone is therefore not enough to judge the goodness-of-fit. It must be analyzed in conjunction with the trend in σ_{ray} and detailed characteristics of the input data.

5.7 Reconstruction of Cylindrical Plume Data (Chapter 3) using SBFM

A reconstruction of the experimental data gathered in Chapter 3 was performed using SBFM. In this implementation of SBFM, inverted Gaussians (negative peak height) were allowed. Only by adding negative Gaussians to positive Gaussians could one hope to reproduce the flatness of the plume concentration maximum. Figure 5.6 shows the results using 1, 2 and 3 Gaussians. Adding a third Gaussian hardly reduced the minimum achievable value of σ_{ray} . Two Gaussians were therefore deemed sufficient. The σ_{ray} values in each case are lower than for the ART reconstruction presented in Chapter 3, in spite of the fact that the reconstruction using ART is a much less constrained problem. This reinforces the hypothesis that it is the artificial discretization of space into pixels that lies at the heart of the difficulties encountered with ART.

The value of σ_{pnt} for the 2 Gaussian reconstruction is 0.487, surprisingly close to the ART value of 0.482. The reason for the discrepancy between measured point sample concentrations and those calculated based on the 2 Gaussian representation is different

than it was for ART. With the Gaussian representation, the major errors occur at the base of the plume, for the following reason. In reconstructing a flat-topped, cylindrical plume concentration profile by Gaussians, a compromise must be made between matching the flatness of the top and the steepness of the edge of the plume. Because the largest contribution to the value of σ_{ray} is made by the rays with the highest path-integral concentrations, the minimization of σ_{ray} tends to lead to reconstructions that are more accurate in areas of high concentration. Thus, the reconstructed plume boundary does not drop to zero quite as rapidly as it should. It should also be kept in mind that the calculation of σ_{pnt} in this case is based largely on plume boundary points. It is clear, from comparing Figures 5.6 (b), 3.9 and 3.13, that the SBFM reconstruction is in much better agreement with expected concentrations than the ART reconstruction when including the *entire* reconstruction plane.

5.8 Conclusions

This investigation into measuring airborne tracer gas concentrations using optical remote sensing and computed tomography has led to the following conclusions regarding reconstruction methods:

- (1) Iterative algebraic reconstruction techniques (ART), when used with a ray geometry designed for rapid scanning, result in concentration profiles characterized by unrealistically high spatial variability. Deceptively low values of σ_{ray} make it clear that applying ART to the reconstruction of gas concentrations in air using such ray geometries could lead to unrecognized, significant errors in estimates of the gas concentration profiles.
- (2) The new approach developed here, of modeling the concentration profile with smooth basis functions (SBFM), produces reconstructions that are much more consistent than ART when compared with measured point-sample

concentration data. Bivariate Gaussians constitute a satisfactory basis for representing the concentration profiles generated in the ventilated chamber experiments. The reconstruction of the cylindrical plume was less satisfactory. However, this concentration profile represents a fairly artificial situation, not likely to be encountered in most indoor settings.

- (3) The SBFM approach obviates the need for a tradeoff between spatial resolution and the number of free parameters in the reconstruction, a critical weakness of ART, where the number of pixels determines both the level of spatial resolution and the number of free parameters.
- (4) The minimization of σ_{ray} , when used in conjunction with χ^2_{ν} values, provides a basis for determining the proper number of parameters necessary to represent the data in a manner consistent with the amount of information provided by the rays.

The SBFM routine using analytical integration and executed on a 486 PC (33 SX) requires about two hours for a comprehensive search (annealing) for the global minimum of the goodness-of fit parameter σ_{ray} from a random first guess using two Gaussians. The computing time increases with the number of Gaussians. If one can make a good first guess at the approximate location and height of peaks, the annealing need not be quite as comprehensive and the computing time can be significantly shortened. A first guess might be generated from the trends visible in an ART reconstruction, which can be completed in a matter of minutes. Another option would be to augment the SBFM program with a routine that searches for areas in the reconstruction plane through which many rays with high integral concentration pass, and placing the maxima of the Gaussians there for the first guess. The minimization might also be substantially accelerated by improving the program, including changing to a different programming language, such as C. It is

possible that with a more powerful computer, near real-time monitoring, one goal for CT-FTIR development, could be achieved with SBFM.

Rapid progress has been evident over the past several years both in remote sensing technologies and in understanding the general characteristics of CT reconstruction algorithms for air quality applications. A significant effort remains to demonstrate in a broad range of field tests the circumstances under which coupling CT with remote sensing can yield accurate measurement results. Particular attention is warranted to the problem of reconstructing time-varying concentration profiles, as investigated in Chapter 6. Also, the design of practical monitoring systems for emissions characterization in outdoor air represents a worthwhile, yet unrealized objective. Although still in the early stages of development, the use of computed tomography in conjunction with remote sensing holds substantial ultimate promise for fast and cost-effective gas concentration distribution measurements over a range of spatial scales, with potential application to many indoor and outdoor air quality problems.

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5.10 References

- Bevington P. R. (1969) Data Reduction and Error Analysis for the Physical Sciences, McGraw-Hill, New York, Chapter 10.
- Brooks R. A. and Di Chiro, G. (1975) Theory of image reconstruction in computed tomography, *Radiology*, 117, 561-572.
- Jones T. A., Hamilton D. E. and Johnson C. R. (1986) Contouring Geologic Surfaces with the Computer, Van Nostrand Reinhold, New York.
- Press W. H., Teukolsky S. A., Vetterling W. T., and Flannery B. P. (1992) Numerical Recipes in C, 2nd Ed., Cambridge University Press, Cambridge, MA.
- Todd L. and Leith D. (1990) Remote sensing and computed tomography in industrial hygiene, *Am. Ind. Hyg. Assoc. J.*, 51, 224-233.
- Todd L. and Ramachandran N. (1994) Evaluation of algorithms for tomographic reconstruction of chemical concentrations in indoor air, *Am. Ind. Hyg. Assoc. J.*, 55, 403-417.

Table 5.1 Summary of goodness-of-fit parameters for CT reconstructions.

EXPERIMENT 1				
	Synthetic Ray-Integral Data		Measured Ray-Integral Data	
	ART	SBFM	ART	SBFM
σ_{pnt}	0.53	0.26	0.41	0.11
σ_{pix}	0.62	0.33	-	-
σ_{ray}	0.005	0.067	0.047	0.084
χ^2_{ν}	-	-	-	1.19
Figure	5.3 (c)	5.3 (e)	5.3 (d)	5.3 (f)

EXPERIMENT 2				
	Synthetic Ray-Integral Data		Measured Ray-Integral Data	
	ART	SBFM	ART	SBFM
σ_{pnt}	0.49	0.25	0.93	0.51
σ_{pix}	0.45	0.21	-	-
σ_{ray}	0.002	0.04	0.026	0.16
χ^2_{ν}	-	-	-	0.11
Figure	5.4 (c)	5.4 (e)	5.4 (d)	5.4 (f)

Figure 5.1 (a) Top view schematic of the experimental chamber: M = mirror and R = retroreflector. For clarity, only one mirror and one retroreflector are shown. (b) Top view of the measurement plane, showing pixels (dotted lines), and rays (solid lines) emanating from the centrally located FTIR (with coincident source and detector). Rays in bold are reflected directly back to the center; other rays are first reflected off of a mirror along the chamber perimeter and then off of a retroreflector (R). Pixels are 47 cm on a side. Ray diameter is approximately 20 cm.

Figure 5.2 Summary of data processing approach. Rectangles represent experimental input data; ellipses represent calculations; rectangles with rounded corners represent results. Lower case alphabets in the long horizontal rectangle are keyed to results shown in Figures 5.3 and 5.4.

Figure 5.3 Results for Experiment 1. The x and y axes are in pixel units. (a) Point sample data (source shown as a white dot). The arrow indicates the direction of the ventilation airflow. (b) Kriged point sample data. (c) ART reconstruction using synthetic ray-integral data. (d) ART reconstruction using experimental ray-integral data. (e) SBFM reconstruction using synthetic ray-integral data (2 Gaussians). (f) SBFM reconstruction using experimental ray-integral data (3 Gaussians). Only concentrations up to 18 ppm are shown, so that the low concentrations can be distinguished. The peak concentration of the narrow Gaussian in (f) is 43 ppm.

Figure 5.4 Results for Experiment 2. The x and y axes are in pixel units. (a) Point sample data (source shown as a white dot). The arrow indicates the direction of the ventilation airflow. (b) Kriged point sample data. (c) ART reconstruction using synthetic ray-integral data. (d) ART reconstruction using experimental ray-integral data. (e) SBFM reconstruction using synthetic ray-integral data (2 Gaussians). (f) SBFM reconstruction using experimental ray-integral data (1 Gaussian). Peak concentration of 10.6 ppm in (e) is clipped to show results on the same scale as in other frames. During this experiment, three of the pumps used to draw air into the gas bags failed. Therefore, only 28 time-averaged point concentrations are shown in (a).

Figure 5.5 Effect of number of fitting parameters on quality of fit for SBFM reconstruction based on measured ray-integral data: (a) Experiment 1; (b) Experiment 2. See Equations (5.3) and (5.4) for parameter definition.

Figure 5.6 (a) - (c) SBFM reconstructions using 1, 2 and 3 Gaussians, based on the experimental data from the cylindrical plume experiment described in Chapter 3. Compare to the ART reconstruction (Fig. 3.13) and the point sample data (Fig. 3.9). The peak concentration (not visible due to the scale) in (a) is 10.6 ‰.

CHAPTER 6

MEASUREMENT OF TIME STATIONARY AND TIME DEPENDENT TRACER GAS CONCENTRATION PROFILES IN A CHAMBER USING OPEN PATH FTIR REMOTE SENSING AND SBFM COMPUTED TOMOGRAPHY*

6.1 Abstract

Remote measurement of gas concentrations in indoor air using optical remote sensing (ORS) and computed tomography (CT) has been suggested but not thoroughly investigated experimentally. In this chapter, experiments are presented in which one time-varying and nine different steady-state tracer gas concentration profiles were generated in a horizontal plane in a ventilated chamber and sampled by an open path Fourier transform infrared spectrometer (FTIR) for subsequent CT inversion. CT reconstructions were performed using the smooth basis function minimization (SBFM) technique, described in Chapter 5, and compared with simultaneously gathered point-sample concentration measurements. Agreement between the two sampling methods was qualitatively very good, with concentration profiles showing the same features of peak location and shape. Quantitative agreement was generally good to within 50%. I discuss the sources of discrepancy and suggest directions for future research, especially with regards to monitoring time-dependent processes. With further refinements in reconstruction algorithms and improvements in optical remote sensing hardware, this technique holds promise for rapid and accurate gas concentration measurements.

* This chapter is the basis for the paper: Drescher, A. C., Park, D. Y., Yost, M. G., Gadgil, A. J., Levine, S. P., and Nazaroff, W. W. (1995) Measurement of Time Stationary and Time Dependent Tracer Gas Concentration Profiles in a Chamber Using Open Path FTIR Remote Sensing and SBFM Computed Tomography, to be submitted to *Atmospheric Environment*.

6.2 Introduction

The purpose of this chapter is to describe in detail a set of experiments performed to test the concept of computed tomography used with optical remote sensing (CT/ORS). The aim was to develop an improved understanding of the factors that influence the quality of a CT reconstruction of gas concentrations in air. To achieve this aim, many experiments were conducted, each producing a different tracer gas concentration profile. One of the profiles was purposely allowed to be time-varying. For all experiments, point-sampled concentration measurements were compared with CT reconstructions based on FTIR path-integral data to assess the extent of agreement between the two methods.

The first part of this chapter describes nine of eleven steady-state experiments (the other two are described in Chapter 5). These nine experiments were carried out under constant tracer gas source strength and ventilation conditions. Naturally, however, as is characteristic of any environmental fluid flow system, fluctuations in the concentration profile were inevitable. To minimize the effect of time-variability on the reconstructions, the analysis of these nine experiments was based on 1 to 1.5 hour time averages of the concentration profiles and the path-integral data.

In a different experiment, which is presented in the second part of this chapter (Section 6.4), the chamber was not mechanically ventilated. A burst release of tracer gas was allowed to disperse throughout and out of the chamber over a period of 45 minutes by natural air movement alone. Path-integral concentration data gathered by the FTIR during this period were used to reconstruct the concentration profile at several times during the dispersion process. The results are compared to time-resolved and time-integrated point-sample data gathered in the plane of reconstruction.

Finally, in Section 6.5, the first of the steady-state experiments is reanalyzed in a manner similar to the time-dependent experiment in order to check whether this would affect the results. CT reconstructions are performed for measurements obtained at 6.2 minute intervals during this 90 minute experiment. The range and the average of the

reconstructions are compared to the single CT reconstruction based on the time-averaged ray-integral concentration data.

6.3 Steady-State Experiments

6.3.1 Experimental Method

Eleven experiments were performed in which SF_6 was released at a constant rate from one or two points near the ground in a ventilated room-sized chamber. Figure 6.1 (a) shows the 6.4 m $\times$ 7.3 m $\times$ 2.4 m high chamber. It was located within a larger, mostly empty, uninsulated building without mechanical ventilation, heating or cooling. Details of the chamber are given in Chapter 5.

In each experiment, a buoyant mixture of 10% sulfur hexafluoride (SF_6) in helium was released at a rate of 0.8 l/min from a 2.5 cm diameter, porous ceramic sphere, placed 60 cm above the floor. In three experiments, the same gas mixture was also released in an identical fashion at a height of 90 cm from an additional point in the chamber closer to the exhaust. In these cases the gas was released at 0.4 l/min from each sphere. Once steady-state conditions were established, SF_6 concentrations were measured over a period of between 60 and 90 minutes in a horizontal plane 1.3 meters above the floor. The two measurement methods described in Chapter 5 were employed, yielding "point-sample" measurements and "ray-integral" or "path-integral" measurements. Each experiment was begun either in the late morning or mid-afternoon, when temperature changes were not as rapid as during the rest of the day. The temperature in the chamber was recorded once every minute during an experiment. Air speed was recorded every 10 seconds in the fume hood that provided the exhaust from the chamber, and at one of the air inlets to the chamber.

6.3.2 Data Processing Method

For each orientation of the FTIR, the mean, μ_r , and variance, σ_r^2 , of the ray-integral concentrations measured during an experiment were computed. CT reconstructions were applied to these time-average ray-integral concentrations.

The measurement plane was divided into 195 (13 by 15) square pixels (0.22 m² each). The width of each pixel (47 cm) corresponds to the distance between rays at the edge of the chamber, where they are reflected. Three maps of SF₆ concentration as a function of position in the plane were generated for each experiment. Distances in the plane were measured and plotted in units of pixel length. The first maps, marked (a), show concentrations measured by point sampling in the plane. The point samples were kriged using the graphing program SURFER (Golden Software, Golden, CO) and the value at every half of a pixel width was plotted (profiles marked (b)). Kriging is a geostatistical technique for estimating a grid of values from a set of arbitrarily positioned point measurements. It uses the spatial correlation between points to find minimum variance estimates of intermediate grid points (Jones et al., 1986). CT reconstructions based on the time-averaged path-integral concentrations are shown in the profiles marked (c).

6.3.3 CT Reconstruction Method

The SBFM-CT method is described in Chapter 5. Again, the goodness-of-fit parameter σ_{ray} was used as an optimization parameter:

$$\sigma_{\text{ray}} = \sqrt{\frac{\sum_{r=1}^{56} (\text{PIC}_r - \text{PIC}_{r,\text{CT}})^2}{\sum_{r=1}^{56} \text{PIC}_r^2}} \quad (6.1)$$

where PIC_r is the measured path-integral concentration for ray r (corresponding to μ_r in these experiments) and $\text{PIC}_{r,\text{CT}}$ is the path-integral concentration calculated from the assumed Gaussian concentration distribution. The value of $\text{PIC}_{r,\text{CT}}$ was evaluated analytically as the line integral along the ray centerline multiplied by the width of the ray.

The process of finding the best reconstruction began by minimizing σ_{ray} assuming a constant concentration profile. Then a single Gaussian was used to model the distribution. As a first guess, the Gaussian was assumed to be located in the center of the

reconstruction plane, with standard deviations in x and y equal to half of the width and length of the measurement plane, respectively, and a peak value equal to 1.5 times the best constant concentration found previously. The SBFM program then adjusted the six parameter values — x and y coordinates of the peak, standard deviations along the Gaussian's two principal axes, peak height (maximum concentration) and angle of rotation of the principal axes relative to the x and y axes — so as to minimize σ_{ray} . I then examined the calculated path-integral concentrations for this reconstruction. For each experiment, there were groups of rays whose calculated integral concentrations were significantly lower than the measured ones, indicating areas in the measurement plane where a second peak in concentration was required. The first guess for the position of the second Gaussian was based on these observations. A reconstruction was then performed assuming two Gaussians. The process was repeated for three Gaussians.

For experiments 3,4 and 5 the process of finding the best reconstruction was slightly different from that described above. These were the first experiments to be analyzed. In these cases, the point-sampled concentrations were known before CT analysis was begun. The kriged concentration profiles were used to make a first guess at the Gaussian parameters, starting immediately with two Gaussians. All other experiments were first run "blind" (not knowing what the point-sampled data looked like) with the procedure described in the previous paragraph. After generating what I believed to be the best possible reconstruction, I examined the point-sampled data and tried to improve on the reconstruction using first guesses close to what was observed in the point-sampled data. In most cases, no improvement in the reconstruction could be achieved, but for experiments 6 and 8 a further decrease in σ_{ray} was effected by slight parameter changes of the Gaussian constituting the major peak. This point is discussed further in Section 6.6.

The minimum value of σ_{ray} was noted as a function of the number of adjustable parameters used to model the concentration distribution. The reduced chi-squared was also calculated for each reconstruction:

$$\chi_v^2 = \frac{1}{(56 - n)} \sum_{r=1}^{56} \left\{ \frac{1}{\sigma_r^2} (\text{PIC}_r - \text{PIC}_{r,\text{CT}})^2 \right\} \quad (6.2)$$

where v is the number of degrees of freedom after adjusting n fitting parameters to match the 56 ray integral concentration values ($v = 56 - n$ where n is six times the number of Gaussians used for the fit ($n = 1$ for the constant concentration)). For many degrees of freedom, the reduced chi-squared for a good reconstruction will be close to unity if (1) the correct fitting function is chosen and (2) the measurement errors are Gaussian distributed and accurately estimated. The weighting factor for the deviation term for each ray is the ray variance σ_r^2 . The relative decreases in χ_v^2 and σ_{ray} every time a Gaussian was added were used to judge when the number of free parameters was sufficient to model the underlying concentration distribution. For all but one experiment, the decreases in χ_v^2 and σ_{ray} were significant up to two Gaussians but became negligible when adding a third Gaussian.

As in previous chapters, the overall difference between the final CT reconstructions (figures marked (c)) and the measured point sample data (figures marked (a)) was calculated as follows:

$$\sigma_{\text{pnt}} = \sqrt{\frac{\sum_{\text{pt}=1}^{31} (C_{\text{pt},m} - C_{\text{pt},\text{CT}})^2}{\sum_{\text{pt}=1}^{31} C_{\text{pt},m}^2}} \quad (6.3)$$

where $C_{\text{pt},m}$ is the measured concentration at a point and $C_{\text{pt},\text{CT}}$ is the reconstructed concentration at that point. A value of zero for the parameter σ_{pnt} indicates perfect agreement between the measured point sample data and the concentrations at those same points computed by the CT program.

An analogous quantity, σ_{krig} , was defined to compare the kriged point-sample data (figures marked (b)) with the appropriate CT reconstruction):

$$\sigma_{\text{krig}} = \sqrt{\frac{\sum_{x=0}^{13} \sum_{y=0}^{15} (C_{x,y,k} - C_{x,y,\text{CT}})^2}{\sum_{x=0}^{13} \sum_{y=0}^{15} (C_{x,y,k})^2}} \quad (6.4)$$

where $C_{x,y,k}$ is the concentration of the kriged data at (x,y) and $C_{x,y,\text{CT}}$ is the concentration of the CT reconstruction at (x,y). Both x and y were incremented in steps of 0.5 pixel units.

6.3.4 Results and Discussion

6.3.4.1 General Experimental Conditions

Figure 6.2 is a typical example of some of the quantities measured during an experimental run: air speed, temperature and time-resolved point-sample concentrations in the chamber. Time = 0 indicates the nominal start time of the experiment, the time at which sampling into gas bags was begun. Air flow was clearly turbulent, as evidenced by the fluctuations in air speed at a range of time scales. The average air speed, as driven by the fume hood, was constant. The relative standard deviation in speeds was 3.6 % for this experiment. The maximum deviation from the average was 21%. Temperature rose steadily during the experiment, as it was conducted on a sunny day between 11:00 a.m. and 12:30 p.m.

SF₆ concentrations at the sampling locations shown in Figure 6.2 fluctuated during the experiment, with points downwind and closest to the source fluctuating the most. This was to be expected, since at such locations spatial gradients in concentration are high. Where gradients are high, slight changes in air motion leading to shifting of the gas plume would cause the concentration at fixed points to vary significantly. The two relatively

large drops and subsequent increases in concentration (visible in the three highest concentration profiles) at the end of the experiment occurred simultaneously with a person walking around close to the air inlet to the chamber. These fluctuations in concentration were believed to be caused by this disturbance. Thereafter, we kept the air inlets free of physical disturbance. Nevertheless, relatively large fluctuations in point-sampled concentrations were occasionally observed in some of the other experiments. These were believed to be related to sudden changes in temperature inside the building in which the chamber was located, and wind and pressure fluctuations around the building. This building was open on one side (opening size of approximately 0.3 m by 2 m) to provide the fresh air that was drawn through the chamber. The possibility of fluctuations in source strength having caused the changes in the locally measured concentrations was ruled out: the concentration measured in the fume hood (exhaust air from the chamber) was extremely stable over time.

6.3.4.2 Point Sample Data

Figures 6.3 (a) to 6.11 (a) show the SF_6 concentrations for experiments 1 to 9, respectively, at the sampling points in the plane of measurement. The location of the SF_6 source(s) in each case is also shown. Occasionally a sampling pump or a gas bag would fail. Therefore, some plots contain less than 31 concentration values. The points with percentage values near them are those at which the concentration was sampled continuously into glass jars and measured by the Bruel & Kjaer photoacoustic detector (B&K) every 6 minutes. In these cases the time-average concentration was plotted, and the percentages represent the relative standard deviation over the duration of the experiment. Relative standard deviations tended to range between 10% and 40%, but some were as low as 4% (Fig. 6.5 (a)) and others as high as 91% (Fig. 6.7 (a)).

Figures 6.3 (b) to 6.11 (b) show the kriged point sample data. These surface plots make it easier to visualize the concentration profiles. For all but one of the experiments with a single source of SF_6 , the concentration profile has one sharp major and one broader, minor peak (experiment 3 is the exception). Experiments with two sources

tended to lead to an additional defined peak, although in experiment 6 the extra source did not seem produce an extra peak. All controllable conditions were the same for experiments with equal number and positioning of sources. For such experiments, the concentration profiles produced were similar in general, but distinct in detail. The influence of factors such as outdoor temperature and weather conditions was clearly important. Nevertheless, there is a consistency in the profiles that one would expect, given the fact that the ventilation conditions and the total rate of SF_6 release were the same for all experiments: the mean concentration over the entire measurement plane appears to be fairly constant. As peaks become lower, they tend to become wider. This is especially clear by comparing Figures 6.3 (b), 6.4 (b) and 6.5 (b), and Figures 6.10 (b) and 6.11 (b).

6.3.4.3 CT Reconstructions

Figures 6.3 (c) to 6.11 (c) show what I consider to be the best possible CT reconstructions generated for each experiment. The general features of these concentration profiles agree well with the kriged profiles. The results are described quantitatively by the four parameters plotted in Figure 6.12: σ_{ray} , χ_v^2 , σ_{pnt} and σ_{krig} .

Values of the parameter σ_{ray} ranged from 0.09 to 0.17. These relatively low values indicate good consistency in the set of path-integral concentrations: concentration profiles could be found whose ray integrals matched those measured to within approximately 10 to 20%. This seems to indicate that the use of **time-averaged** ray-integral concentrations generated a valid data set for CT reconstruction. The range in χ_v^2 values is larger, from 0.55 to 2.6. This reflects the difference in time stationarity among experiments. The parameter χ_v^2 is more dependent on fluctuations in the concentration profile during an experiment, because it incorporates the value of σ_r^2 for each ray. However, the fact that χ_v^2 was always on the order of unity is a good sign: the reconstructions are about as good as can be expected, given the uncertainty in the path-integral concentrations themselves.

When compared quantitatively with the point-sampled data, the reconstructions all appear to match the former data to within approximately 50%, with σ_{pnt} values ranging from 0.40 to 0.62. The difference between reconstructions and the kriged data, characterized by σ_{krig} , is closely correlated with σ_{pnt} . This is a reflection of the similarity between interpolation by kriging and the smoothness of a Gaussian basis function. A very close correlation also exists between σ_{pnt} and σ_{ray} , there being an average ratio of 4 between the two. This correlation is a positive sign: it supports the idea that minimizing σ_{ray} is an appropriate way to find the underlying concentration distribution responsible for a certain set of path-integral data.

The greatest contribution to the discrepancy between point-sampled data and reconstructions comes from the areas near the sharp concentration peak(s) in each experiment. This is understandable in light of the following: (1) differences in concentration that are high in absolute value contribute most to the value of σ_{pnt} ; (2) such differences are most likely to occur where concentration gradients are high (near peaks) because in such areas sampling location accuracy is most critical.

There were two aspects of sample-point positioning that are important in explaining the discrepancies near the peaks: (1) positioning in the x-y plane was only good to within approximately 10 cm in each direction because it was done by visually aligning the sampling points with the appropriate mirror along the two closest walls; (2) the height of the sampling points was not at the center of the FTIR beam but 7.5 cm below it, though still within 2.5 cm of the edge of the beam. This was an unfortunate oversight: previous (unrelated) experiments with the FTIR had been done at a slightly lower beam height, and when lifting the beam to coincide with the mirror centers for this experiment we failed to increase the height of the sampling points. Thus, the CT reconstructions were actually based on a measurement plane that had a thickness on the order the beam diameter (20 cm) and was centered slightly above the point-sampling measurement plane. This would explain why the major concentration peaks in the reconstructions tend to be lower and broader than those in the kriged data. Since the SF_6 sources were below the

measurement plane and the general flow of air was upward to the exhaust in the ceiling, concentration gradients would tend to decrease with height as the SF_6 was dispersed into the surrounding air while being transported upwards.

6.4 Time-Dependent Dispersion Experiment

A transient tracer gas dispersion experiment of 50 minutes duration was carried out to test the CT/ORS concept under unsteady conditions. The aim was to generate CT reconstructions that would show the evolution of the tracer gas concentration profile in the measurement plane, from a time shortly after its release to the time at which it was homogeneously dispersed throughout the chamber. It was anticipated that tracer gas concentrations in the measurement plane would change significantly during each complete scan of the FTIR (lasting approximately 6 minutes). Therefore, CT reconstructions could not be based on direct path-integral concentration measurements from individual scans. Instead, successive measurements for each ray were interpolated to generate path-integral concentration values at the specific times during the experiment for which a CT reconstruction was to be performed. Time-dependent and average CT reconstructions were again compared to simultaneously gathered point sample concentration data.

The transient dispersion experiment was carried out in the same chamber and with the same apparatus used for the steady-state experiments. However, for this experiment the chamber was not mechanically ventilated. The nominal start of the experiment ($t = 0$) was marked by the onset of continuous scanning of the measurement plane by the FTIR. Continuous point-sampling by the B&K at five locations in the measurement plane had begun 25 minutes earlier. At $t = 3.5$ minutes, a buoyant mixture of 10% SF_6 gas in He was released from the floor of the chamber at position ($x = 0.5$, $y = 11.5$) at an approximate rate of 2 l/min for 2 minutes. Continuous sampling from 26 points in the measurement plane into 26 gas bags was begun at $t = 7.5$ minutes. Bags were disconnected at $t = 45.5$ minutes. The last FTIR scan was completed at $t = 49$ minutes.

The path-integral concentration data gathered by the FTIR were converted to path-average concentrations by dividing by the area of each ray. When plotting the path-averages as a function of time, as shown in Figure 6.13, two irregularities were discovered: (1) A pair of adjacent rays in the set being reflected from mirrors on the $x = 13$ wall of the chamber (rays 2-8 and 2-9; refer to Fig. 6.1 (b)) was consistently lower in average concentration than the rays on either side of this pair. I cannot explain this discrepancy. Instead of using these clearly erroneous values, values for CT reconstruction were estimated by linear interpolation between the path-integral concentrations of the two rays on either side of the pair. (2) As the well-mixed condition was approached, all rays approached the same path-average concentration except for the very short rays going to the corners of the chamber (1-12, 1-13, 2-14, 2-15, 3-12, 3-13, 4-14, 4-15; see Fig. 6.1 (b)). These rays consistently had an average concentration 20% higher than that of all others. Figure 6.13 shows this effect for rays reflected from the $y = 15$ wall of the chamber (1-12, and 1-13). The short rays were assumed to have been reflected only once, having passed from the FTIR to either a mirror or retro-reflector and back to the FTIR. However, it is possible, due to the divergence of the IR beam, that some of the light returning from the mirror or retro-reflector passed on either side of the FTIR, hit the retro-reflector in the opposite corner of the chamber, returned to the first mirror or retro-reflector and then returned to the FTIR. This would mean that the path length of these beams would have been underestimated, causing their average concentrations to be higher than expected. All path-integral concentrations for these rays were therefore reduced by a factor of ~ 0.8 , such that their path-average concentration under well-mixed conditions was equal to that of their neighbors. Naturally this reduction (a first-order correction) would not be accurate for times close to the beginning of the experiment, since the proper adjustment would depend on the concentration distribution along the respective diagonal of the chamber (second-order correction).

Just under six minutes were required to complete the first scan of the measurement plane by the FTIR. The earliest CT reconstruction was performed for $t = 6$ minutes, since

the first path-integral concentration for each ray had to have been measured before successive measurements could be interpolated. The path-integral concentration data for each ray was linearly interpolated to generate sets of path-integral concentrations for $t = 6, 8, 10, 12, 16, 24, 33$ and 40 minutes. CT reconstructions were performed on these data sets (the 33 minute reconstruction was performed to more accurately estimate the mixing time, as discussed in the final paragraph of this section). The resulting concentration profiles are shown in Figure 6.14 for $8, 10, 12, 16, 24$ and 40 minutes. It was found that two Gaussians were required to adequately represent the concentration distribution for $t < 33$ minutes. At $t = 33$ minutes and $t = 40$ minutes, a single Gaussian was sufficient.

The general evolution of the SF_6 concentration distribution, as shown in Figure 6.14, is what we would expect in an unventilated but leaky chamber: concentrations in different parts of the measurement plane approach each other, and the average concentration decreases with time. The location of the peak concentration is also consistent with our expectation, given the location of the SF_6 release. The reconstructions suggest that SF_6 did not disperse isotropically in the measurement plane. This is plausible in light of the following. Natural airflow in the chamber was most likely not homogeneous, since it was principally governed by two factors whose effect we would expect to vary spatially within the chamber: (1) buoyancy forces induced by natural temperature differences and (2) flow in and out of the chamber openings caused by wind-induced pressure differences inside the exhaust hood, which was vented to the roof of the building. Furthermore, the presence of the FTIR and its support structure, as shown in Figure 6.1 (a), likely impeded the movement of tracer up into the measurement plane at the center of the chamber, initially causing this area to have low SF_6 concentrations. The initial build-up of SF_6 mostly along the area directly beneath the ceiling opening is understandable, given the proximity of the tracer gas release point to the two chamber walls in this vicinity.

Figure 6.15 shows how σ_{ray} (equation (6.1)) for the CT reconstructions decreased with time. Initially, the value of this parameter, which correlates with the expected

accuracy of the reconstruction, is much higher than the value achieved for experiments 1 to 9. This inability to find a good match to the measured path-integral concentration data is probably a combination of (1) inability to accurately interpolate between successive ray measurements in a period of high temporal variability in the underlying concentration profile, and (2) inability of the Gaussian model to adequately describe the concentration profile in the initial stages of dispersion. By $t = 12$ minutes, however, the value of σ_{ray} is already at the lowest level achieved in any steady-state experiment reconstruction. Its improvement beyond this point shows that the interpolation procedure becomes very accurate once the rate of change of concentration has the same sign (negative) everywhere. The asymptotic value of σ_{ray} at $t = 40$ minutes indicates that the precision of path-integral concentration measurements themselves was approximately 3% (once the obvious measurement errors had been corrected, as described in the third paragraph of this section).

Figure 6.16 (a) shows the time-averaged data for this experiment as obtained from the point samples. The integration time for the bag samples was from $t = 7.5$ minutes to $t = 45.5$ minutes. Points at which time-dependent concentrations were measured (numbered 1 to 5) were averaged over this same time period by assuming a linear change in concentration between successive measurements taken at ~ 5 minute intervals at each point (see Figure 6.17 (a)). The kriged data is plotted in Figure 6.16 (b). Here we clearly see the slight inconsistency between the bag samples and the direct B&K point samples, which show up as little sharp peaks and dips in the surface.

Figure 6.16 (c) shows the time-average concentration profile generated by linear interpolation between successive CT reconstructions. It has slightly more structure to it than Figure 6.16 (b), but the overall agreement between the two profiles is very good. It is superior, in fact, to the agreement between CT reconstructions and kriged profiles from any of the steady-state experiments. The σ_{krig} value here is 0.23, as compared to approximately 0.5 for experiments 1 to 9. This suggests that it might have been better to treat the steady-state experiments in a fashion analogous to the dispersion experiment:

interpolate between successive path-integral concentration measurements for each ray, perform reconstructions at specific times during the experiment using the interpolated values, and then average all of the reconstructions generated. Not only might this produce a more accurate estimate of the underlying time-average concentration profile, but it would also generate an estimate of the precision based on the variability among individual profiles produced. The only disadvantage to this procedure is that it takes much longer than calculating a single reconstruction from time-averaged path-integral data. In Section 6.5 I reanalyze steady-state experiment 1 using this procedure.

A point-by-point comparison between the CT reconstructions and the time resolved B&K data (locations marked 1 to 5 in Figure 6.16 (a)) is shown in Figure 6.17. The B&K data (Fig. 6.17 (a)) are plotted as measured, successive points being approximately 1 minute apart. The points in Figure 6.17 (b) are only plotted at times for which a reconstruction was performed. The error bars on Figure 6.17 (b) are based on assigning each concentration value a relative error equal to σ_{ray} (for the reconstruction at that time) multiplied by 4 (the average ratio of σ_{pnt} to σ_{ray} for the steady-state experiments). The decrease in the estimated error with time reflects the increase in agreement between the CT and the B&K data as seen by comparing 6.17 (a) and 6.17 (b). The agreement towards the end of the experiment is excellent.

It is instructive to look closely at the discrepancies between the two graphs at the beginning of the experiment. First, note that sample points 2 and 4, whose concentration tends to be higher in the B&K data than in the CT data, are also distinctly higher in concentration than the bag sample data would indicate, as shown in Figure 6.16 (b). Similarly, points 1 and 3 are clearly lower in concentration when compared to both the CT and the bag sample data. Sample point 5 shows the best agreement among all three measurement methods. It is important at this stage to remember that the B&K point samples have been treated as instantaneous measurements, whereas, due to the sampling from the glass jars, they are actually time-integrated concentrations over approximately the 3 minutes before the B&K measurement was performed. This may explain the delayed

B&K peak in concentration at sample point 2 (in Fig. 6.17 (a)) compared to the CT data at the same location (Fig. 6.17 (b)). Furthermore, the flow rate of chamber air into the glass jars (~ 1 l/min) was approximately 12 times higher than that into the gas bags. Thus, the B&K sampling points effectively sampled the concentration from a larger volume of space. In areas of high average concentration (points 2 and 4, Fig. 6.16 (b)) this seems to have had the effect of increasing the B&K sample concentration over the bag samples, and vice versa, with the smallest discrepancy being in the area of lowest spatial gradient in concentration (point 5). Finally, errors in positioning of the point sampling inlets in the x-y plane, coupled with the ~ 20 cm limit in spatial resolution of the CT reconstructions (concentrations were assumed to be constant across the IR beam width) would be expected to lead to discrepancies between Figures 6.17 (a) and 6.17 (b) in areas of high spatial concentration gradients (such as point 2).

As discussed in Chapter 2, it is frequently of interest, in the study of indoor environments, to characterize the rate of pollutant dispersion by quantifying the mixing time. The mixing time, as defined in Chapter 2, is the time (after release) for the relative standard deviation in concentration of a tracer or pollutant gas as measured at multiple, widely spaced points to diminish to 10%. Based on the measured concentrations at the five B&K sampling points, the measurement plane was well mixed 27.4 minutes after the middle of the SF_6 release period (i.e., at $t = 31.9$ minutes). Based on the 837 (27×31) point concentration values produced by each CT reconstruction, the mixing time was 29.6 minutes ($t = 34.1$ minutes). Thus, the CT and B&K results agree well with respect to mixing time.

6.5 Time-Dependent Analysis of a Steady-State Experiment

Experiment 1 (Fig. 6.3) was selected to explore the effect of using the time-dependent analysis technique on the CT reconstruction of a nominally steady-state experiment. The path-average concentration measurements (path-integral concentration divided by the ray area in the horizontal plane) for each ray during this 90 minute

experiment are shown in Figure 6.18. Clearly, the underlying concentration profile was not time-independent, as already evidenced by the point-sample concentration data shown in Figure 6.2. The rays passing through areas of high SF_6 concentration exhibit substantial fluctuations, the degree of fluctuation increasing with the proximity to the peak concentration shown in the steady-state reconstruction (Fig. 6.3 (c)). The fluctuations were on time scales as short as (and very likely also even shorter than) the time between measurements for each ray. Interpolation between individual measurements for each ray is therefore likely to be inaccurate.

There is also some evidence of the optical effect for the short rays discussed in Section 6.4. These rays again tended to have slightly higher path-average concentrations than expected. Ray 3-4 also tended to be significantly lower in path-average concentration than its nearest neighbor rays, and I suspect that concentration measurements along this ray were in error. However, for the purposes of this analysis, no measured data were altered as they were for the time-dependent experiment.

The time between successive measurements for each ray was 6.2 minutes. Based on these data, the path-integral concentration data for each ray were linearly interpolated, and CT reconstructions based on the interpolated data were performed for $t = 6.0, 12.2, 18.4, 24.6, 30.8, 37.0, 43.2, 49.4, 55.6, 61.8, 68.0, 74.2, 80.4$ and 86.6 minutes. Examples of several of the individual reconstructions are shown in Figure 6.19. The parameter σ_{ray} varied from 0.12 to 0.19 for the 14 reconstructions performed. The value of σ_{ray} for most of these reconstructions was slightly higher than the value attained by the reconstruction (plotted in Figure 6.12) based on the time-averaged path-integral concentration data, $\sigma_{\text{ray}} = 0.13$. This is probably due to the fact that the time-variability in this experiment was large enough so that linear interpolation of path-integral concentration measurements for each ray was less accurate than using a single time-average (as done in the steady-state analysis presented in Section 6.3).

The 14 individual reconstructions were averaged to generate the concentration distribution shown in Figure 6.20 (a). The standard deviation as a function of position is

plotted in Figure 6.20 (b). The average reconstruction is indistinguishable from the single reconstruction based on the steady-state analysis and shown in Figure 6.3 (c). Thus, at least in this particular case, applying the time-dependent analysis does not yield a more accurate final reconstruction. It is, however, useful to have information on the time variability in the underlying profile in the form of the standard deviation distribution. Here we see that the largest error is associated with the peak concentration, the relative error at the peak being approximately 20%. This is consistent with the level of disagreement visible when comparing the average CT reconstruction with the kriged point-sampled concentrations (Figure 6.3 (b)).

6.6 Conclusions

I have shown that using SBFM-CT to reconstruct tracer gas concentration profiles from time-averaged path-integral concentrations gathered by means of an OP-FTIR is a feasible method for determining the steady-state distribution of gas in a plane. For the nine time-stationary experiments conducted, all of which had different concentration distributions, CT reconstructions agreed well with simultaneously gathered point-sample concentrations. The number of Gaussians needed to describe the concentration distribution was generally one more than the number of SF₆ sources in the chamber, but for two of the two-source experiments, only two Gaussians were required.

Measurement of the evolution of the concentration distribution in a time-dependent tracer gas dispersion experiment was also shown to work well using SBFM-CT. Accurate CT reconstructions were made possible by interpolating successive path-integral concentrations for each ray to generate reconstructions at specific times in the continuous scanning process. The agreement between the time-averaged reconstruction and the kriged concentration profile from simultaneously gathered point-sample data was twice as good as for the time-stationary experiments. This suggests that even situations with a constant source strength and ventilation might best be analyzed by performing

several reconstructions at successive times, using interpolated path-integral concentration values for each ray, and then averaging these reconstructions.

Contrary to initial expectations, re-analysis of a steady-state experiment using this technique did not result in better agreement with point-sampled data. At least part of the reason for this is probably that the level of noise in the underlying concentration distributions in the steady-state experiments was much greater than in the dispersion experiment, which was largely characterized by ray-integral concentrations that decreased smoothly and monotonically with time. The forced ventilation in the steady-state experiments resulted in time variability in the form of eddies and turbulent fluctuations. Interpolation is therefore likely to be inaccurate in these cases. As ORS technologies improve, the time for a complete scan may be reduced significantly, thus increasing the accuracy of CT reconstructions based on interpolation.

Bivariate Gaussians proved satisfactory as a model for the concentration distributions. They are attractive, because they can be analytically integrated, which is accurate and saves computing time. However, a more accurate description of concentration profiles might be possible with Gaussian-like functions whose width in $+x$ and $-x$, and $+y$ and $-y$ could be different. The enforced symmetry of bivariate Gaussians limits our ability to describe certain types of concentration distributions well.

Minimizing σ_{ray} appears to be an appropriate method for finding the best possible reconstruction. The difficulty still lies in ensuring that the minimization routine (simulated annealing, in this case) has actually led to the lowest possible value of σ_{ray} . I have found that in certain cases a significantly better value of σ_{ray} could be attained by recommencing the minimization at a slightly different first guess of the major Gaussian peak (for instance, assuming a narrower but higher maximum). The parameter space appears to have many narrow and deep minima close to one another in the general vicinity of the global minimum. This can perhaps be explained by the fact that narrow peaks are intercepted by few rays. Thus, σ_{ray} is not very sensitive to small changes in the parameters of this peak. Simulated annealing is good at converging to the vicinity of the global minimum from

areas of the solution space which have local minima but where the parameters are quite different from those near the global minimum. However, by the time the parameter set reaches the vicinity of the global minimum, the annealing temperature is low, and thus the program quickly converges on the nearest local minimum. Possibly some form of systematic search near the global minimum should be added at the end of the simulated annealing routine to ensure that the true global minimum is found.

Simulated annealing is time-consuming because it is a completely blind search method. The technique I employed manually, of assessing the spatial distribution of discrepancies between calculated and measured ray-integral concentrations, and changing the first guess or estimating the position of additional Gaussians based on that knowledge, should be built into the minimization routine to make it more efficient. To pick the appropriate number of Gaussians and their characteristics, a completely automated SBFM-based CT program might minimize a quantity that is made up of σ_{ray} and a function that quantifies the degree of spatial correlation between the discrepancies mentioned above. The program would then converge on a solution for which: (1) the differences in path-integral concentrations compared to the measured data were as small as possible, given the inaccuracies in the measured data; (2) the sign of these differences was not correlated over large areas of the plane of reconstruction; and (3) the number of Gaussians was not more than what could be justified by the internal consistency of the set of measured path-integral concentrations (too many Gaussians would tend to result in an ART-like reconstruction, containing unrealistic spatial variability in concentration).

This set of experiments highlighted the advantages and disadvantages of CT/ORS compared with point sampling. Both measurement systems take a long time to set up initially. Putting the mirrors, retroreflectors and the heavy FTIR in place was no easy task. However, once they were set up, little work was required, other than the occasional manual adjustment of a mirror or the computer-aided adjustment of the direction of a ray (to compensate for very slight changes in mirror alignment) before an experiment. The initial positioning of the sampling lines, pumps and bags for the point sampling was also

not quick and easy. Moreover, a great deal of additional manual labor was involved for each experiment in reconnecting and disconnecting gas bags, analyzing them, purging them, starting and stopping the sampling pumps and recharging them. Point sampling also precluded any type of activity in the room, since the numerous sampling lines and their supports occupied so much space below the measurement plane.

The point sampling done directly by the B&K was very convenient, as it gave concentration values in almost real time. However, many more instruments would have been required to sample the entire measurement plane in this way. The FTIR produced an approximate¹ path-integral concentration every 6 seconds. This provided a rough idea of where concentrations in the chamber were high and where they were low. Obtaining a good CT reconstruction with the method outlined in this chapter takes several hours. However, by automating the whole process and using a faster computer, one might be able to shorten the delay between path-integral measurements and reconstruction generation to several minutes.

From the dispersion experiment we learn that unexpected optical effects can lead to errors in measured path-integral concentration that might well go unnoticed if the optical configuration of the experimental environment is not carefully characterized. While remote sensing technology is still not fully matured, it is wise to check measured data for irregularities such as the ones encountered in this study. The advantage of SBFM-CT is that all path-integral concentration measurements are aggregated when minimizing σ_{ray} , so that the effect of gross errors in individual rays is damped somewhat. Individual point measurements do not have this advantage.

The routine use of CT/ORS is still far from being a reality. Yet, I believe that the knowledge gained from the research presented here will help develop the technique to maturity and lead to further valuable insights along the way.

¹The path-integral concentration was approximate, because it was calculated based on a single reference spectrum. In the final analysis, each ray had its own reference spectrum, which was measured several times before every experiment.

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6.8 References

- Brooks, R.A. and Di Chiro, G. (1975), Theory of Image Reconstruction in Computed Tomography, *Radiology*, 117, 561-572.
- Jones T. A., Hamilton D. E. and Johnson C. R. (1986) Contouring Geologic Surfaces with the Computer, Van Nostrand Reinhold, New York.
- Press, W. H., Teukolsky, S. A., Vetterling, W. T. and Flannery, B. P. (1992) Numerical Recipes in C, 2nd. Ed., Cambridge University Press, Cambridge, MA.
- Todd, L.A. and Leith, D. (1990) Remote Sensing and Computed Tomography in Industrial Hygiene, *Am. Ind. Hyg. Assoc. J.*, 51, 1165-1178.

Figure 6.1 (a) Schematic of the experimental chamber: M = mirror and R = retroreflector. For clarity, only one mirror and one retroreflector are shown.

Figure 6.1 (b) Top view of the measurement plane, showing pixels (dotted-line boundaries) and rays (solid lines), emanating from the centrally located FTIR. Rays in bold are reflected directly back to the centrally located detector; other rays are first reflected off of a mirror along the chamber perimeter and then off of a retroreflector (R). Rays are numbered 1-1 to 4-15. Pixels are 47 cm on a side. Ray diameter is approximately 20 cm. The circle represents the rotation of the FTIR in the process of scanning the measurement plane, from ray 1-8 clockwise to ray 1-7.

Figure 6.2 Measurements for experiment 1. Air speed was recorded every 10 seconds inside the chamber, 1 cm (along the y-axis) from the center of the lower air inlet. Temperature was recorded at the center of the chamber every minute. SF₆ sampling point locations are in pixel units. The percentage values refer to the relative standard deviation in concentration from time = 0 to time = 90 minutes, as shown in Figure 6.3. Bag samples and path-integral SF₆ concentration measurements (not shown) were taken from time = 0 to time = 90 minutes.

Figure 6.3 Experiment 1 (90 minutes).

Figure 6.4 Experiment 2 (90 minutes)

(a) Point sample data (SF₆ source shown as a white dot). Percentages near points denote relative standard deviations of time resolved point concentration measurements. (b) Kriged point sample data. (c) SBFM reconstruction (2 Gaussians).

Figure 6.5 Experiment 3 (60 minutes).

(a) Point sample data (SF₆ source shown as a white dot). Percentages near points denote relative standard deviations of time resolved point concentration measurements. (b) Kriged point sample data. (c) SBFM reconstruction (2 Gaussians, 3 Gaussians).

Figure 6.6 Experiment 4 (90 minutes)

Figure 6.7 Experiment 5 (75 minutes).

Figure 6.8 Experiment 6 (75 minutes)

(a) Point sample data (SF₆ source shown as a white dot). Percentages near points denote relative standard deviations of time resolved point concentration measurements. (b) Kriged point sample data. (c) SBFM reconstruction (2 Gaussians).

Figure 6.9 Experiment 7 (90 minutes).

Figure 6.10 Experiment 8 (60 minutes)

(a) Point sample data (SF₆ source shown as a white dot). Percentages near points denote relative standard deviations of time resolved point concentration measurements. (b) Kriged point sample data. (c) SBFM reconstruction (2 Gaussians). The tip of the arrow in Figure 6.10 shows the height of the peak (located between plotted points) of that Gaussian.

Figure 6.11 Experiment 9 (60 minutes).

(a) Point sample data (SF₆ source shown as a white dot). Percentages near points denote relative standard deviations of time resolved point concentration measurements. (b) Kriged point sample data. (c) SBFM reconstruction (2 Gaussians).

Figure 6.12 CT reconstruction results for the nine steady-state experiments.

Figure 6.13 Path-average concentrations as a function of time for rays reflected off of the $y = 15$ wall of the chamber during the dispersion experiment. Ray 1-1 is reflected from the mirror at (0.5, 15.0), ray 1-2 is reflected from the mirror at (1.5, 15.0), and so forth, as shown in Fig. 6.1 (b). Each FTIR scan was begun with ray 1-8 and ended with ray 1-7, with the FTIR rotated clockwise.

Figure 6.14 (a) - (f) CT reconstructions of the SF₆ concentration distribution at various times during the transient dispersion experiment.

Figure 6.15 The value of σ_{ray} for the CT reconstructions of the transient dispersion experiment.

Figure 6.16 (a) Point-sample SF_6 concentrations for the dispersion experiment. The numbered points are those that were connected directly to the B&K (rather than to a gas bag) and whose concentration was numerically time averaged. The white point shows the location of the SF_6 source. (b) Kriged point-sample concentration data. (c) Time average of CT reconstructions over the duration of bag sampling.

Figure 6.17 (a) SF_6 concentrations measured at the five sampling points that were directly connected to the B&K during the dispersion experiment. (b) SF_6 concentrations at these five points as reconstructed from the FTIR-measured path-integral concentration data.

Figure 6.18 Ray integral concentration data for experiment 1. See Fig. 6.1 (b) for the location of individual rays in the reconstruction plane.

Figure 6.19 (a) - (c) CT reconstructions for experiment 1 close to the beginning, middle and end of the experiment.

Figure 6.20 The average and standard deviation of the 14 CT reconstructions based on the time series data of experiment 1. Compare to Figure 6.3.

CHAPTER 7

CONCLUSIONS

7.1 Summary of the Major Findings

Experiments employing blowers were performed in a sealed room to determine how the time for an instantaneously released contaminant to become well mixed in the room air depended on forced convection conditions. The results provided evidence supporting an existing hypothesis, based on turbulence modeling, that the mixing time is inversely proportional to the cube root of the mechanical power transferred to the room air. The constant of proportionality was determined from the experimental results, and the relationship was applied to predict mixing times for two scenarios of interest: forced ventilation and strong natural convection. The predictions agree well with expected or measured mixing times. The semi-empirical relationship found is a useful tool to quickly approximate, given basic airflow parameters, how appropriate the assumption of complete mixing would be for a scenario of interest. This provides knowledge on mixing time one step beyond the assumption of complete and instantaneous mixing commonly used to estimate exposure or pollutant removal time. It was also found that estimates of integrated exposure from the time of contaminant release up to the mixing time for a variety of conditions can be in error by as much as a factor of two if based on the assumption of instantaneous, complete mixing.

Several steady-state experiments were performed to test the concept of combining previously existing iterative computed tomography algorithms (CT) with optical remote sensing (ORS) to measure tracer gas concentrations in a plane. The first experiment was designed and executed on the table-top scale and employed an ideal scanning geometry with a high spatial sampling density. The second experiment was on the scale of several meters and also employed a parallel ray geometry, but with sparse spatial sampling and a

very slow scanning rate. For the third set of experiments, again conducted in a chamber on the scale of several meters, a less ideal scanning geometry was used, but scanning was rapid and reconstructions were based on time-averages of measured path-integral concentrations.

In these three cases, existing CT methods proved either marginally satisfactory or altogether unsatisfactory, the root of the problem in each case lying in the artificial discretization of space. Dividing the reconstruction plane into pixels all of equal size, and enforcing constant concentration within a pixel, effectively lays down a template that favors overall evenness in the concentration profile while allowing high pixel-to-pixel variability. This problem is usually not significant when the input data are noise-free and the scanning geometry guarantees maximum independence of rays. However, it becomes severe when using experimental path-integral measurements and non-ideal (but practical) scanning geometries.

Pixels must be small enough for the approximation of constant concentration in a pixel to be valid everywhere. If they are too large, no acceptable solution to the ray integral equations can be found. The area of highest spatial concentration gradient thus dictates the smallness (and thereby the total number) of pixels necessary to accurately represent the underlying profile. However, in most cases of interest, concentrations in the measurement plane will be fairly even over most of the plane but contain one or more distinct concentration peaks. In the large areas of approximately constant concentration, the pixel density is far too high. Effectively, a problem of indeterminacy has been created: multiple solutions to the ray-integral equations are possible by creating small-scale variability in the areas of low overall concentration. This problem can be diminished by employing more rays or by changing the scanning geometry to make rays more independent (Park et al., 1994). However, one quickly reaches the limit of technical and economic feasibility. A reconstruction method was needed that would decouple the ability

to represent true concentration maxima from the problem of having too many adjustable (free) variables.

A new CT method was developed, which, when applied to the same new experimental data, was shown to overcome the difficulties associated with previous reconstruction algorithms. The new method was called smooth basis function minimization (SBFM). The underlying concentration distribution is modeled as a superposition of a limited number of smooth basis functions. The parameters characterizing these functions are adjusted to minimize the difference between measured path-integral concentrations and those calculated based on the smooth function representation. I employed asymmetric bivariate Gaussians in my implementation of SBFM. Using Gaussians, a variety of real tracer gas concentration profiles created in a ventilated chamber under steady-state conditions could be well represented. Because the inversion of ray-integral data with SBFM is so much more constrained than with the traditional pixel-based approaches, far fewer rays are required to determine a unique solution. This has great practical advantage in terms of hardware requirements and total scanning time. A weakness of SBFM is that it not yet completely automated; further software development is needed to reduce the time required to arrive at the best possible reconstruction.

For the first time, CT/ORS was applied to a time-varying concentration profile. Reconstructions were performed at specific times during the dispersion of an instantaneously released tracer gas by interpolating successive path-integral concentration measurements for each ray. During the first third of the approach towards the well-mixed condition, when temporal variability was greatest, the inability to accurately interpolate path-integral concentrations was evident from the relatively high σ_{ray} of the reconstructions. However, overall, this method of CT/ORS proved to be very effective, since the slow, steady and monotonic decrease in concentration during most of the experiment allowed for accurate interpolation. This was not the case for a steady-state

experiment re-analyzed with the same method. Forced ventilation in this case caused fluctuations in the concentration distribution that precluded accurate interpolation.

In general, the usefulness of computed tomography in conjunction with optical remote sensing for studies of indoor air motion and pollutant dispersion will be influenced by several factors. The applicability of the method will depend, for instance, on whether sufficient path average concentration data can be gathered in a short enough time to accurately follow the changes in the system under investigation. Even stationary-state situations, such as the steady release of a pollutant from a fixed position within a room having a constant ventilation rate, will exhibit fluctuations in concentration about some mean value at every point in space. The time scale of these fluctuations is critical in determining the rate at which ray path-average concentration data must be gathered for computed tomography. The spatial concentration gradients will also affect the feasibility of using this method, because they will dictate the necessary spatial sampling frequency.

7.2 Directions for Future Research

7.2.1 Indoor Air Transport and Dispersion Research

The relationship between mixing time and mechanical power input presented in this thesis is tentative. Further experiments and applications of the relationship to well-characterized scenarios in the literature are needed to determine to what extent the relationship is valid and generalizable. In particular, factors such as (1) room geometry and obstacles in the room, (2) location of the contaminant source, and (3) orientation and placement of the mixing air flows should be investigated. Ideally, the relationship would be refined to include the effects of vertical temperature gradients on mixing time. To quantify the contribution of heated bodies commonly found in indoor environments to mixing, an understanding of the conversion from thermal energy to organized bulk flow must be developed. A theoretical and experimental exploration of the mixing time for a non-instantaneous but finite-time continuous release of inert contaminant would also be of

interest. It might be possible to model such a release as a superposition of burst releases in quick succession with the same total contaminant emission. Such a model could be further extended to situations involving removal of the pollutant by ventilation in cases in which mixing is imperfect.

7.2.2 Refinement of SBFM

SBFM holds promise as the most appropriate reconstruction technique for CT/ORS measurements of ambient gas concentration profiles. However, its present implementation is far from perfected. The technique must be refined and automated so that it can become a practical and reliable tool for near real-time concentration measurements. Simulated annealing should be coupled with (1) an intelligent routine that estimates the location of Gaussian peaks by looking at spatial correlations in discrepancies between measured and calculated path-integral concentrations, and (2) a systematic search for the best parameters in the vicinity of the global minimum of σ_{ray} .

7.2.3 Optical Remote Sensing Hardware

The main barriers to successful, large-scale application of CT and remote sensing in indoor air lie in the hardware for detection and scanning. Ideally, all path average concentrations would be measured simultaneously, essentially producing 'snapshot' profiles of gas distribution. However, this would require a large number of sources and detectors, both of which are expensive at present. Either cheap detection methods must be found, or clever scanning mechanisms must be developed that do not require many sources and detectors and offer rapid scanning with minimum disruption of the air motion being studied. Scanning involves the use of reflecting optics, which are vulnerable to misalignment, especially in field applications. More experimental work is needed to test the robustness of ORS scanning for the purposes of computed tomography reconstruction.

Theoretically, CT/ORS need not be limited to a horizontal plane, and not even to a plane. Clearly, however, the hardware challenge becomes much more formidable when working in a plane other than horizontal or in three dimensions.

7.2.4 Other Applications of CT/ORS

This thesis has focused on CT/ORS for pollutant and tracer gas dispersion studies. Several other useful potential applications exist. Leak detection and location in large indoor chemical storage facilities would be an ideal application. Once installed, a CT-FTIR system could run continuously at very little cost. Pollutant monitoring for exposure assessment or safety alerts is important in industrial hygiene (Timberlake et al., 1994; Ghittori et al., 1993). Todd (Todd, 1990; Todd and Leith, 1990; Todd, 1992; Todd and Ramachandran, 1994 (a), 1994 (b)) has been working on CT/ORS specifically with this application in mind.

There is also an interest in the use of CT/ORS to measure pollutant concentrations in outdoor air, for instance, above hazardous waste sites, to characterize the concentrations of air toxics near an industrial facility, or in urban areas (Fischer et al., 1994; Wolfe, 1980). No experimental work on these applications has been published, but the hardware obstacles will no doubt be much greater than for indoor environments. The effects of (1) an increased scale of measurement (from tens to thousands of meters) and (2) environmental conditions being much less stable outdoors would make it very challenging to gather sufficient data with adequate accuracy for CT reconstruction. However, as ORS hardware improves, such applications may one day become feasible.

7.3 References

Fischer, M. L., Drescher, A. C., Gadgil, A. J., and Yost, M. G. (1994) A System for Rapid Detection and Mapping of Gas Plumes on 100 m Scales; Examination of some Technical and Economic Issues, proceedings of the *International Symposium on*

- Optical Remote Sensing for Environmental and Process Monitoring*, November 7-10, 1994, in McLean, VA, Air and Waste Manage. Assoc.
- Ghittori, S., Marraccini, P., Franco, G. and Imbriani M. (1993) Methylene Chloride Exposure in Industrial Workers, *Am. Ind. Hyg. Assoc. J.*, 54, 27-31.
- Park, D. Y., Yost, M. G., Drescher, A. C., Gadgil, A. J., Zhou, Y., and Levine, S. P. (1994) Evaluation of Beam Path Configurations for Use with a Monostatic OP-FTIR Capable of Rapid Beam Movement for Tomographic Reconstruction of Gas and Vapor Concentrations in Workplaces, proceedings of the *International Symposium on Optical Remote Sensing for Environmental and Process Monitoring*, November 7-10, 1994, in McLean, VA, Air and Waste Manage. Assoc.
- Timberlake, D. L., Marshall, M. and Scott, J. (1994) Evaluation of Chemical Releases and Worker Exposures from Filter Press Operations, *Am. Ind. Hyg. Assoc. J.*, 55, 438-442.
- Todd, L. (1990) Optical Remote Sensing and Computed Tomography in Industrial Hygiene, Ph.D. dissertation, University of North Carolina, Chapel Hill, NC.
- Todd, L. and Leith, D. (1990) Remote sensing and computed tomography in industrial hygiene, *Am. Ind. Hyg. Assoc. J.*, 51, 224-233.
- Todd, L.A. (1992), Optical Remote Sensing/Computed Tomography Systems for Workplace Exposure Assessments; Optical Remote Sensing/Computed Tomography Beam Geometries for Monitoring Workplace Gases and Vapors, proceedings of *Optical Remote Sensing Applications to Environmental and Industrial Safety Problems*, Air and Waste Management Association, SP81, 356-360; 390-393.
- Todd, L.A., and Ramachandran, N. (1994a), Evaluation of Algorithms for Tomographic Reconstruction of Chemical Concentrations in Indoor Air, *Am. Ind. Hyg. Assoc. J.*, 55(5), 403-417.

- Todd, L.A., and Ramachandran, N. (1994b), Evaluation of Optical Source-Detector Configurations for Tomographic Reconstruction of Chemical Concentrations in Indoor Air, *Am. Ind. Hyg. Assoc. J.*, 55(12), 1133-1143.
- Wolfe, D.C. (1980) On the Application of Optical Computed Tomography to Remote Air Pollution Measurements, Ph.D. Dissertation, Stanford University, Palo Alto, CA.