This article was downloaded by: [National Taiwan University]

On: 30 March 2009

Access details: Access Details: [subscription number 908165593]

Publisher Taylor & Francis

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK

Journal of Environmental Science and Health, Part A

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713597268>

Applying open-path Fourier transform infrared spectroscopy for measuring aerosols

Chang-Fu Wu ^{ab}; Yen-Ling Chen ^b; Chih-Chieh Chen ^b; Tzu-Ting Yang ^c; Pao-Erh Chang ^d

^a Department of Public Health, National Taiwan University, Taipei, Taiwan ^b Institute of Occupational Medicine and Industrial Hygiene, National Taiwan University, Taipei, Taiwan ^c Department of Environmental Engineering and Health, Yuanpei University, Hsinchu, Taiwan ^d Industrial Technology Research Institute, Hsinchu, Taiwan

Online Publication Date: 01 January 2007

To cite this Article Wu, Chang-Fu, Chen, Yen-Ling, Chen, Chih-Chieh, Yang, Tzu-Ting and Chang, Pao-Erh(2007)'Applying open-path Fourier transform infrared spectroscopy for measuring aerosols', Journal of Environmental Science and Health, Part A, 42:8, 1131 — 1140

To link to this Article: DOI: 10.1080/10934520701418631

URL: <http://dx.doi.org/10.1080/10934520701418631>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Applying open-path Fourier transform infrared spectroscopy for measuring aerosols

CHANG-FU WU^{1,2}, YEN-LING CHEN², CHIH-CHIEH CHEN², TZU-TING YANG³
and PAO-ERH CHANG⁴

¹Department of Public Health, National Taiwan University, Taipei, Taiwan

²Institute of Occupational Medicine and Industrial Hygiene, National Taiwan University, Taipei, Taiwan

³Department of Environmental Engineering and Health, Yuanpei University, Hsinchu, Taiwan

⁴Industrial Technology Research Institute, Hsinchu, Taiwan

This paper examines the feasibility of using Open-Path Fourier Transform Infrared Spectroscopy (OP-FTIR) to measure aerosols. The extinction spectra of water, ammonium nitrate, and ammonium sulfate aerosols were first simulated with various particle size distributions (geometric mean ranged from 2 to 10 μm ; geometric standard deviation ranged from 1.1 to 2.5) based on the Mie theory. An optimization procedure was developed to retrieve the geometric mean and standard deviation of the aerosols size distributions from the spectra, assuming that the complex refractive index is known. To test sensitivity, we also added 4%, 7%, and 10% noise levels to the spectra and compared the reconstruction results. In the experimental study, water aerosols were generated by a two-fluid nozzle inside a cylindrical chamber (3325 cm^3). The extinction spectrum was collected with a modified FTIR and the size distribution information was retrieved following the same optimization procedure as the one used in the simulation study. The optimization procedure developed in this study reconstructed the size distribution reasonably well for particles with known refractive index (i.e. homogeneous or internally mixed aerosols). The results were robust with the added noise levels up to 10%, after removing inaccurate estimates with the use of the censoring criteria for reconstructed $\text{GSD} < 1.3$, reconstructed $\text{GM} < 2.5 \mu\text{m}$ and $\text{GSD} < 1.5$, and reconstructed $\text{GM} > 10 \mu\text{m}$. With regard to externally mixed aerosols, the reconstructed results were sensitive to the noise within the measuring systems, although most ambient aerosols were internally mixed. The reconstructed size distribution in the chamber experiment had a GM of 3.85 μm and GSD of 1.70. The simulation results were applied to support this reconstruction result. We conclude that OP-FTIR can be used to measure aerosols and screen for the right region for a more detailed aerosol measurement campaign.

Keywords: Particulate matter, OP-FTIR, extinction spectrum, Mie theory, Smooth Basis Function Minimization.

Introduction

Particles and gases need to be measured simultaneously to assess air quality accurately. Most air monitoring instruments measure either gaseous (e.g., using organic vapor monitor) or particulate (e.g., using dust monitor) air pollutants, not both. One technique potentially capable of monitoring both at the same time is Open-Path Fourier Transform Infrared (OP-FTIR) spectroscopy, an optical remote sensing (ORS) technique that is conventionally used for monitoring gaseous air pollutants.^[1–7] When there are aerosols in measurement beam path, baseline features of the spectra change.^[8] This change is usually considered undesirable, but "shearing" of the baseline of the spectra ac-

tually contains important information about the physical and chemical characteristics of aerosols.

With regard to retrieving aerosol size distribution information from OP-FTIR measurements, Arnott et al.^[9] generated water clouds in a 1 m^3 chamber and used FTIR to collect the extinction spectra of the water aerosols. In that same study, droplet size information was retrieved using an iterative algorithm developed based on the Mie theory.^[10] Hashmonay and Yost,^[8] conducting a similar experiment, generated water droplets in a standard shower chamber (0.85 m width, 1.18 m length, 1.53 m height). Assuming a Gaussian size distribution for the aerosols, they calculated the extinction spectrum of water aerosols based on the Mie theory. They then manually matched the calculated extinction spectrum with the measured spectrum by changing the parameters of the size distribution.

The aim of this study was to develop an optimization procedure for reconstructing aerosol size distributions from the OP-FTIR measurements, extending the methodology

Address correspondence to Chang-Fu Wu National Taiwan University, Room 717, No.17, Xu-Zhou Rd., Taipei 100, Taiwan; E-mail: changfu@ntu.edu.tw
Received January 11, 2007.

developed by Hashmonay and Yost.^[8] In this study, we developed an optimization procedure to find a best-fit solution, and then conducted a series of simulations to evaluate how this procedure performed for different aerosols with various size distributions. We also performed an initial chamber experiment to demonstrate the feasibility of this approach.

Theory development

The theory of retrieving aerosol size distribution from the OP-FTIR spectra was discussed in Hashmonay and Yost^[8] and summarized briefly as follows. The light extinction phenomenon due to aerosols is described by the Mie theory.^[10] The associated extinction spectrum can be computed if the extinction efficiency (Q_e) is known. The Q_e is a function of the particle size, light wavelength, and the complex refractive index m at the corresponding wavelengths, and can be calculated using the recurrence procedure described in Wickramasinghe.^[11] The refractive index is usually expressed as a complex number:^[12]

$$m = n - ik \quad (1)$$

where n is the real refractive index (real part), i is $\sqrt{-1}$, and k is related to the absorption coefficient (imaginary part) of the aerosols.

The extinction coefficient (σ_e) is calculated from the Q_e and the size distribution of the aerosols according to the relationship of:^[12]

$$\sigma_{e_i} = \frac{\pi}{4} \sum_j Q_{e_{ij}} N_j d_j^2 \quad (2)$$

where N_j is the number concentration at the j^{th} particle size class; d_j is the mean diameter for the j^{th} particle size class; i is the index number for the wavelength; and j is the index number for the particle size class.^[8] The extinction spectrum, i.e., the OP-FTIR spectrum, is then calculated from multiplying extinction coefficient by OP-FTIR beam path length.

During the retrieval process, the spectra for various size distributions of certain compounds are calculated, assuming the complex refractive index is known. When a specific size distribution results in a match between the calculated spectrum and target spectrum (e.g., spectrum collected experimentally), that size distribution is regarded as the solution.

Method

Simulation investigation

In the first part of this study, we obtained the complex refractive index information from the published literature and simulated infrared spectra on computer, assuming different

aerosol size distributions. We then applied the Mie theory to develop an optimization procedure to reconstruct the infrared spectra and obtain the underlying size distribution information. The total particle number concentration and distribution parameters predicted from the reconstruction procedure were compared to the input parameters that were used to generate those simulated spectra.

Single compound scenario

The complex refractive indices of three compounds were obtained from published literature: water,^[13] ammonium sulfate,^[14] and ammonium nitrate.^[15] The FTIR spectrum for each compound was simulated independently according to Equation 2. The latter two compounds were chosen originally because they are two important anthropogenically produced components of ambient particulate matters. Assuming a log-normal distribution for the aerosols, various distribution parameters (i.e., geometric mean, or GM, ranged from 2 to 10 μm ; geometric standard deviation, or GSD, ranged from 1.1 to 2.5) were applied to obtain the corresponding extinction spectra. All the distributions were assumed to have a total particle number concentration of 10,000 $\#/\text{cm}^3$. Although it is rare to have only aerosols with one pure chemical species in the ambient environment, testing on the single compound scenario helped identify the limitation of our optimization procedure since fewer parameters were involved in the inversion process, compared to testing on mixtures of compounds.

Multiple compounds scenario

Spectra for mixtures of the three compounds were also simulated and tested. The mixed compounds were classified into two categories: external and internal mixtures.^[16] An external mixture is defined as the condition in which each aerosol is chemically pure and aerosols are mixed in space without coagulation. The extinction coefficient of external mixture (σ_{em}) is the sum of the contributions from each pure species (σ_{em_k}):

$$\sigma_{em} = \sum_{k=1}^N \sigma_{em_k} \quad (3)$$

where N ($=3$ in this study) is the total number of species. On the contrary, an internal mixture is defined as the condition in which different chemical species are mixed in each aerosol in a fixed proportion. To calculate its extinction coefficient, the effective refractive index of the mixture is computed using the refractive indices of the individual species based on various mixing rules (e.g., the volume weighted method^[16]).

Optimization procedure

Theoretically, we should be able to reconstruct the N and d information from the measured OP-FTIR spectrum, assuming the complex refractive index is known

(Equation 2). The process of searching for a solution described in the Theory Development section can be done manually or solved by the nonlinear least-squares optimization methods. In this study, we developed an optimization procedure similar to the Smooth Basis Function Minimization (SBFM) method used in radial plume mapping analyses with the OP-FTIR measurements.^[17–19] With this approach, a known smooth basis function (e.g., log-normal distribution) with unknown parameters is assumed to describe the size distribution of the aerosols. The SBFM approach requires a ‘predicted’ extinction spectrum and an error function for minimization. The predicted extinction spectrum can be calculated from Equation 2 with the assumed distribution.

The error function is defined as the sum of square differences between the test and the predicted spectra. It is minimized by optimization functions provided in the Matlab computer software (MathWorks, Inc., Natick, MA) to yield a set of parameters that produce the best-fit between the test and predicted spectra. For single compound and internal mixture scenarios, only three parameters need to be retrieved (i.e., GM, GSD, and particle number concentration). However, for external mixture scenario, nine parameters need to be retrieved (i.e., three parameters for each of the three compounds). Mathematically, the reconstruction performance in the internal mixture scenarios should be similar to the one in the single compound scenarios, since the internally mixed aerosols were also assumed to have one set of known complex refractive index (compared to the three sets of complex refractive indices for externally mixed aerosols). This was also confirmed by our preliminary study results. Thus, for the multiple compound scenarios, only the results from the externally mixed aerosols are presented.

Data analysis

The Pearson’s correlation coefficient (R) values between the test and the reconstructed spectra (R_{spec}) were used to represent the fitness of the reconstructed spectrum. The R values

between the test and reconstructed size distributions (R_{size}) were used to represent the fitness of the reconstructed size distribution. For each compound and size distribution, we also calculated the percent difference between the test and reconstructed total particle number concentrations (N_{p}). In addition, we conducted a series of sensitivity analysis by adding 4%, 7%, and 10% noise levels to the simulated spectra and observed how they affected the reconstruction results.

Experimental investigation

An initial chamber experiment was conducted using a FTIR system (Illuminator, MIDAC, California, USA) with a modified White’s cell. The outer glass shell of the White’s cell was removed so that aerosols generated outside the chamber could enter the chamber. The cylindrical chamber was 3325 cm³ in size (radius = 5.5 cm; length = 35 cm) and was constructed with 0.5 cm thick plexiglass (Figure 1). One 165 cm² opening was located in the center of the chamber floor for aerosols access. A two-fluid nozzle (SU2, Spraying Systems Co., Wheaton, USA) with a sprayed angle of 15° is set up to generate aerosols with the air pressure at 25 lb/in². The spectrum was collected with the sampling regime using 32 co-averaged scans at 2 cm^{−1} resolution. The total beam path length was 440 cm.

The crude spectra contained very fine absorbance features that could not be accounted for using the available complex refractive index information, so we smoothed the measured extinction spectra before subjecting to analysis. The smoothing process, which was performed by calculating the 50-point moving average of the spectra, also reduced the noise levels for better reconstruction results.

Results and discussion

Simulation—single compound

The complex refractive indices and the spectra simulated by assuming that the GM was 5 or 10 μm and GSD was

Fig. 1. The experimental setup of using a two-fluid nozzle to generate water aerosols.

Fig. 2. The complex refractive index of water, ammonium nitrate, and ammonium sulfate and the corresponding extinction spectra with size distributions of $(GM = 5 \mu\text{m}, GSD = 2)$ and $(GM = 10 \mu\text{m}, GSD = 2)$.

2 are shown in Figure 2. The locations of the minima of the spectra (e.g., 950, 1650, and 3500 cm^{-1} in Figs. 2d and 2g) were approximately the same as the locations of the minima of the real part of the complex refractive index (e.g., 950, 1650, and 3500 cm^{-1} in Fig. 2a). These locations were independent of the assumed size distributions, suggesting that these minima features are the signature characteristics of these aerosols. The effects of size can be identified from the overall shape of the baseline features. For all the three compounds, the baseline of the spectra of larger particles (Fig. 2, the third row) had sharper slopes in the 2000 to 3500 cm^{-1} spectral range than the baseline of the spectra of smaller particles (Fig. 2, the second row).

Figures 3a and 3c are examples of the test and reconstructed size distributions for water aerosols, assuming that the test distribution had a GM of 2 and 5 μm , respectively, and both had GSD of 1.5. Their corresponding OP-FTIR spectra are presented in the right column in Figure 3. The R_{spec} and R_{size} levels in both examples were larger than 0.99. The more systematic testing results are shown in Figure 4, in which the x- and y-axes represent the test GM and

GSD of the water aerosols, respectively. The grayscale bar in Figure 4a represents the R_{spec} while the grayscale bar in Figure 4b represents the R_{size} . In ideal conditions (i.e., noise level = 0%), the optimization procedure was able to reconstruct the spectra with good R_{spec} for almost all the test distributions. The R_{size} was generally good for most test distributions except for the distributions with $GSD = 1.1$, in part because the R_{size} parameter for a narrow distribution is relatively sensitive to the shift of the particle size distribution. For example, in Figure 3e ($GM = 9 \mu\text{m}, GSD = 1.1$), the R_{size} is only 0.36 but the reconstructed distribution was quite similar to the test distribution when inspected visually.

Another region worthy of notice is the upper-right corner of Figure 4b. There the R_{size} values were also low. This is because that the Mie theory is generally applied to conditions in which the particle size and the wavelength of the light source are of the same order of magnitude.^[10,12] In our case, the assumed wavelength of the FTIR spectrometer ranged from 2.5 to 25 μm . Therefore, when there was a greater portion of larger particles, the reconstructed size distribution was less than ideal, as can be seen in Figure 3g

Fig. 3. Examples of the test and reconstructed size distributions (left column) and their corresponding spectra (right column) for water aerosols. (a) & (b) test GM = 2 μm , test GSD = 1.5; (c) & (d): test GM = 5 μm , test GSD = 1.5; (e) & (f): test GM = 9 μm , test GSD = 1.1; (g) & (h): test GM = 8.5 μm , test GSD = 2.3.

(test GM = 8.5 μm , GSD = 2.3, $R_{\text{size}} = 0.86$). With regard to total particle number concentrations (Fig. 4c), the $N_{\%}$ was less than 10% in all regions, except in cases in which distributions had GSD = 1.1 or in which distributions had larger particles (i.e. lower-left and upper-right regions).

Sensitivity analysis was performed by adding different levels of noise to the test spectra of water aerosols (Figs. 4d–4l). As expected, the more noise, the greater the decrease in

R_{spec} (Fig. 4, first column). The 5th~95th percentiles of R_{spec} were 0.96~1, 0.90~1, and 0.81~1, respectively, when 4%, 7%, and 10% noises were added to the test spectra. This phenomenon was more apparent for distributions with larger particles (upper-right region), because larger particles have stronger extinctions and the noise was added in proportion to the strength of extinction signals. Regardless of noise levels, the R_{size} and the $N_{\%}$ (Fig. 4, second and third

Fig. 4. The sensitivity analysis results by adding different levels of noises to the test spectra for water aerosols. The grayscale bars in the 1st to the 3rd columns represent the R_{spec} , R_{size} and $N_{\%}$, respectively. The 1st to the 4th rows represent the results from adding 0%, 4%, 7%, 10% noises, respectively.

columns, respectively) did not vary significantly, indicating that the reconstruction algorithm is robust enough even for spectra with a noise level of 10%.

Similar results were observed for ammonium nitrate and ammonium sulfate aerosols. The optimization procedure reconstructed the spectra well for most distributions for noise level of 0% (5th~95th percentile of $R_{\text{spec}} = 0.98\sim 1$). In addition, the R_{size} was larger than 0.98 and $N_{\%}$ was smaller than 10% for most distributions, except in cases in which the distribution was narrow or the distribution had larger particles. The sensitivity analysis confirmed the robustness of the optimization algorithm. That is, the R_{size} and $N_{\%}$ did not vary as fast as the R_{spec} when noise was added to the spectra.

The above analyses also suggest that the R_{spec} alone is not a sufficient indicator for evaluating the quality of the reconstructed size distribution. Combining the results from the three types of aerosols with all noise levels, we found that potentially inaccurate results can be identified using the three censoring criteria: (i) reconstructed $\text{GSD} < 1.3$ for narrow distribution; (ii) reconstructed $\text{GM} < 2.5 \mu\text{m}$ and $\text{GSD} < 1.5$ for distribution with a significant portion of small particles; and (iii) reconstructed $\text{GM} > 10 \mu\text{m}$ for distribution with a

significant portion of large particles. After applying these censoring criteria to the reconstructed distributions, the median values (with 5th~95th percentiles) of the R_{size} and $N_{\%}$ were 0.99 (0.95~1) and 3.3% (0.5%~17.4%), respectively. More stringent criteria could be applied to improve the quality of fit, though this would also limit the applicable aerosol size range.

Simulation—multiple compounds

In Figure 5a can be seen one example of the test and reconstructed size distributions for external mixture. The size distributions of each compound were the same ($\text{GM} = 5 \mu\text{m}$ and $\text{GSD} = 1.5$). The corresponding OP-FTIR spectrum is presented in Figure 5b. As in the single compound scenario, the optimization procedure retrieved the spectrum accurately and produced a good estimate of the aerosol size distribution for all the three compounds ($R_{\text{size}} > 0.99$ and $N_{\%} < 2.1\%$). We also tested the situation in which each compound had different distributions. As can be seen in Figures 5c and 5d, the searching algorithm produced good results ($R_{\text{size}} > 0.99$ and $N_{\%} < 2\%$).

Fig. 5. Examples of the test and reconstructed size distributions (left column) and their corresponding spectra (right column) for externally mixed aerosols. (a) & (b): test GM = 5 μm , test GSD = 1.5 for all compounds; (c) & (d): test GM = 5, 6, 7 μm for water, ammonium nitrate, ammonium sulfate aerosols, respectively. The test GSD is 1.5 for all compounds.

The results from a more systematic testing approach are shown in Figures 6a–6g. The size distributions of each compound were assumed to be the same but the optimization procedure was designed to retrieve three sets of size parameters (one set for each type of aerosols) in each of the simulated spectrum of the externally mixed aerosols. In an ideal situation with a noise level of 0%, the algorithm reconstructed well for all the spectra. The R_{size} (5th~95th percentiles: 0.98~1) was higher than 0.98 and the $N_{\%}$ (5th~95th percentiles: 2%~12%) was smaller than 12% for most size distributions, except for distributions with small GSDs or small particles. Unlike in the single compound scenarios, our sensitivity analysis showed that adding only 4% noises affected the reconstruction quality (Figs. 6h–6n). The 5th~95th percentiles of R_{size} were 0.87~1, 0.73~1, and 0.57~1, respectively, when 4%, 7%, and 10% noises were added to the spectra. This is because the noise was added in the composite spectrum (Equation 3) and the algorithms were not be able to smooth out the noise in the presumed individual compound's spectrum.

In this study, in addition to water aerosols, we also chose to test ammonium sulfate and ammonium nitrate. Although these two aerosols usually exist in the ambient envi-

ronment as small particles and thus may not be suitable for FTIR measurement, they represent some of the few compounds having complete complex refractive indices. Most other compounds (e.g., mineral aerosol particles)^[20] have the complex refractive index only at a few wavelengths. The similarity in results obtained from all the three test compounds provide adequate evidence that the FTIR can be used for aerosol monitoring when the complex refractive index of the aerosols in interest are available at multiple wavelengths. One possible extension of this technique may be its potential use for remote sensing instruments with light sources of shorter wavelength (such as the Ultraviolet Differential Optical Absorption Spectroscopy, or UV-DOAS) to detect submicrometer particles.

Chamber experiment

The spectrum collected using the two-fluid nozzle to generate water aerosols and the reconstructed spectrum is shown in Figure 7b. There were three minima at about 950, 1650, and 3500 cm^{-1} corresponding to the minimum location in the real part of the complex refractive index of water (Fig. 2a). The R_{spec} value was 0.96 and the reconstructed

Fig. 6. The sensitivity analysis results by adding different levels of noises (a–g: 0 %; h–n: 4 %) to the test spectra for externally mixed aerosols. The grayscale bars in the 1st to the 3rd columns represent the R_{spec} , R_{size} and $N_{\%}$, respectively. The 1st & 4th, 2nd & 5th, and 3rd and 6th rows represent the results for the components of water, ammonium nitrate, and ammonium sulfate aerosols, respectively.

parameters were $GM = 3.85 \mu\text{m}$, $GSD = 1.70$, and $N = 2833 \text{ \#}/\text{cm}^3$. As stated earlier, this initial experiment was conducted to demonstrate the feasibility of the optimization procedure. Thus, no non-FTIR measurements were collected to verify the reconstruction results experimentally. The fact that the reconstructed size distribution did not fit the censoring criteria, as proposed in the simulation study, gives evidence of its validity.

We originally attempted to use the Aerodynamic Particle Sizer (APS, TSI, Inc., Shoreview, MN, USA) to measure aerosol size distribution simultaneously. However, APS and most conventional aerosol measuring instruments (such as the impactors or time-of-flight instruments) were found to be unsuitable for measuring volatile aerosols (e.g., the water aerosols), since these instruments usually require the aerosols be sampled into the sensing chamber and the

Fig. 7. (a) The reconstructed aerosol size distribution ($GM = 3.85 \mu\text{m}$, $GSD = 1.7$) in the chamber experiment. (b) The spectrum collected with a FTIR spectrometer and the reconstructed spectrum.

aerosol property might change during the sampling process. This contrasts the in-situ types of measurements from the OP-FTIR for which no sampling of air is required.

To apply this technique in the field, two issues should be addressed. First, availability of known refractive index at multiple wavelengths is crucial for this inversion process. Where such information is unavailable, one could conduct certain calibration experiments (e.g., calibrate the extinction signals against filter samples) to obtain a suitable correction factor. This is done routinely for many conventional aerosol monitoring devices such as the nephelometer or other light scattering instruments. Second, judging from the inherent uncertainty and the $N\%$ error, this system might be better used as a screening tool. For example, an OP-FTIR or UV-DOAS, with multiple scanning beam paths, could scan a wide area first to identify the regions with high aerosol concentrations. Then, a more established aerosol measuring system (e.g. impactors or tapered element oscillating microbalance) can be used at the identified region to obtain more accurate results.

Conclusions

The simulation results showed that the optimization procedure developed in this study reconstructed reasonably well for particles with known refractive index (i.e., homogeneous or internally mixed aerosols). The results were robust with added noise levels up to 10%. We also found that fitness between the test and reconstructed spectra (i.e. the R_{spec}) alone is not a sufficient indicator of the quality of the reconstructed size distribution. The result can be improved, however, by implementing appropriate censoring criteria based on the reconstructed GM and GSD . With regard to the externally mixed aerosols, the results of our reconstructions were sensitive to noise within the measuring systems. Note that in the ambient environments, most aerosols were internally mixed. In the initial chamber experiment, we demonstrated that the optimization procedure could reconstruct the size distribution of water aerosols generated in a controlled environment. Future studies should consider

generating other types of aerosols to test the performance of this system.

Acknowledgments

The authors thank the National Science Council of Taiwan for financially supporting this research under contract NSC94-2320-B002-040.

References

- [1] Grant, W.; Kagann, R.; Mcclenny, W. Optical remote measurement of toxic gases. *J. Air Waste Manag. Assoc.* **1992**, *42*(1), 18–30.
- [2] Childers, J.W.; Thompson, E.L.; Harris, D.B.; Kirchgessner, D.A.; Clayton, M.; Natschke, D.F.; Phillips, W.J. Multi-pollutant concentration measurements around a concentrated swine production facility using open-path FTIR spectrometry. *Atmos. Environ.* **2001**, *35*(11), 1923–1936.
- [3] Ross, K.R.; Todd, L.A. Field evaluation of a transportable open-path FTIR spectrometer for real-time air monitoring. *Appl. Occup. Environ. Hyg.* **2002**, *17*(2), 131–143.
- [4] Yan, L.; Wang, J.D.; Huang, Z.H.; Xu, H.Q.; Zhou, X.T. Monitoring leaking gases by OP-FTIR remote sensing. *J. Environ. Sci. Health A* **2002**, *37*(8), 1453–1462.
- [5] Bacsik, Z.; Mink, J.; Keresztury, G. FTIR spectroscopy of the atmosphere. I. Principles and methods. *Appl. Spectrosc. Rev.* **2004**, *39*(3), 295–363.
- [6] Shores, R.C.; Harris, D.B.; Thompson, E.L.; Vogel, C.A.; Natschke, D.; Hashmonay, R.A.; Wagoner, K.R.; Modrak, M. Plane-integrated open-path Fourier transform infrared spectrometry methodology for anaerobic swine lagoon emission measurements. *Appl. Eng. Agric.* **2005**, *21*(3), 487–492.
- [7] Bacsik, Z.; Komlosi, V.; Ollar, T.; Mink, J. Comparison of open path and extractive long-path FTIR techniques in detection of air pollutants. *Appl. Spectrosc. Rev.* **2006**, *41*(1), 77–97.
- [8] Hashmonay, R.; Yost, M. On the application of open-path Fourier transform infra-red spectroscopy to measure aerosols: observations of water droplets. *Environ. Sci. Technol.* **1999**, *33*(7), 1141–1144.
- [9] Arnott, W.P.; Schmitt, C.; Liu, Y.G.; Hallett, J. Droplet size spectra and water-vapor concentration of laboratory water clouds: Inversion of Fourier transform infrared (500–5000 cm^{-1}) optical-depth measurement. *Appl. Opt.* **1997**, *36*(21), 5205–5216.
- [10] Bohren, C.F.; Huffman, D.R. *Absorption and Scattering of Light by Small Particles*. Wiley, New York, 1983.

- [11] Wickramasinghe, N.C. *Light Scattering Functions for Small Particles, with Applications in Astronomy*. Wiley, New York, 1973.
- [12] Hinds, W.C. *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*. 2nd ed. Wiley, New York, 1999.
- [13] Downing, H.D.; Williams, D. Optical-constants of water in infrared. *J. Geophys. Res.* **1975**, *80*(12), 1656–1661.
- [14] Toon, O.B.; Pollack, J.B.; Khare, B.N. Optical-constants of several atmospheric aerosol species - ammonium-sulfate, aluminum-oxide, and sodium-chloride. *J. Geophys. Res. Oceans Atmos.* **1976**, *81*(33), 5733–5748.
- [15] Jarzembski, M.A.; Norman, M.L.; Fuller, K.A.; Srivastava, V.; Cutten, D.R. Complex refractive index of ammonium nitrate in the 2–20- μ m spectral range. *Appl. Opt.* **2003**, *42*(6), 922–930.
- [16] Ouimette, J.R.; Flagan, R.C. The extinction coefficient of multicomponent aerosols. *Atmos. Environ.* **1982**, *16* (10), 2405–2419.
- [17] Drescher, A.C.; Gadgil, A.J.; Price, P.N.; Nazaroff, W.W. Novel approach for tomographic reconstruction of gas concentration distributions in air: use of smooth basis functions and simulated annealing. *Atmos. Environ.* **1996**, *30*(6), 929–940.
- [18] Hashmonay, R.A.; Yost, M.G.; Wu, C.F. Computed tomography of air pollutants using radial scanning path-integrated optical remote sensing. *Atmos. Environ.* **1999**, *33*(2), 267–274.
- [19] Wu, C.F.; Yost, M.G.; Hashmonay, R.A.; Park, D.Y. Experimental evaluation of a radial beam geometry for mapping air pollutants using optical remote sensing and computed tomography. *Atmos. Environ.* **1999**, *33*(28), 4709–4716.
- [20] Volten, H.; Munoz, O.; Rol, E.; De Haan, J.F.; Vassen, W.; Hovenier, J.W.; Muinonen, K.; Nousiainen, T. Scattering matrices of mineral aerosol particles at 441.6 nm and 632.8 nm. *J. Geophys. Res. Atmos.* **2001**, *106*(D15), 17375–17401.