DOE/ER/61029--9

DE92 019169

Study of the Atmospheric Chemistry of Radon Progeny in Laboratory and Real Indoor Atmospheres

Progress Report for the Period

July 1, 1991 to June 30, 1992

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July 1992

Prepared for

THE U.S. DEPARTMENT OF ENERGY
AGREEMENT NO. DE-FG02-90ER61029

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Abstract

This report covers the second year of the 28 month grant current grant to Clarkson University to study the chemical and physical behavior of the ^{218}Po atom immediately following its formation by the alpha decay of radon. Because small changes in size for activity in the sub-10 nm size range result in large changes in the delivered dose per unit exposure, this behavior must be understood if the exposure to radon progeny and its dose to the cells in the respiratory tract are to be fully assessed. In order to pursue this general goal, two areas of radon progeny behavior are being pursued; laboratory studies under controlled conditions to better understand the fundamental physical and chemical processes that affect the progeny's atmospheric behavior and studies in actual indoor environments to develop a better assessment of the exposure of the occupants of that space to the size and concentration of the indoor radioactive aerosol. Thus, two sets of specific goals have been established for this project.

The specific tasks of the controlled laboratory studies are 1.) Determine the formation rates of $\cdot\text{OH}$ radicals formed by the radiolysis of air following radon decay, 2.) Examine the formation of particles by the radiolytic oxidation of substances like SO_2 , ethylene, and H_2S to lower vapor pressure compounds and determine the role of gas phase additives such as H_2O and NH_3 in determining the particle size, 3.) Measure the rate of ion-induced nucleation using a thermal diffusion cloud chamber, and 4.) Measure the neutralization rate of $^{218}\text{PoO}^+_x$ in O_2 at low radon concentrations.

The specific tasks of the exposure studies in occupied indoor spaces are 1.) Initiate measurements of the activity size distributions in actual homes with occupants present so that the variability of the indoor activity size distributions can be assessed with respect to indoor aerosol sources and general lifestyle variations of the occupants, 2.) Initiate a prospective study of the utility of measurement of deposited ^{210}Pb embedded in glass surfaces as a measure of the long-term, integrated exposure of the population to radon, and 3.) Develop the methodology to determine the hygroscopicity of the indoor aerosol so that the changes in deposition efficiency of the radioactive indoor aerosol with hygroscopic growth in the respiratory tract can be assessed.

This report describes the progress toward achieving these objectives.

TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	1
LABORATORY STUDIES	1
Radiolytic Nuclei Formation	1
·OH Radical Production Rate	2
Ion-Induced Nucleation	6
Neutralization Rates	7
REAL ATMOSPHERE STUDIES	7
Exposure Assessments in Occupied Houses	8
Prospective Study of ^{210}Po in Glass	8
Hygroscopic Growth of Particles	10
FUTURE RESEARCH PLANS	13
SUMMARY	14
REFERENCES	14
PROJECT PERSONNEL	16
PUBLICATIONS ARISING FROM THIS CONTRACT	17
PRESENTATIONS OF RESULTS OF THIS CONTRACT	19

INTRODUCTION

The health effects of the radon decay products are strongly dependent upon their behavior in the indoor atmosphere. Particularly for ^{218}Po , the partitioning between the "unattached" state and the "attached" forms, combined with the preexisting aerosol particles, has a significant impact on the calculation of the dose to the lung resulting from a given decay product concentration. Thus, the chemical and physical behavior of the ^{218}Po atom immediately following its formation by the alpha decay of radon must be considered if its health implications are to be fully assessed.

At the time when the alpha leaves the nucleus, there are two excess electrons around the residual polonium nucleus. It is probable that the exiting alpha causes autoionization leading to a positive ion. The recoiling polonium nucleus has a recoil energy of 117 kev with a range in air of approximately 100 microns (Lind, 1961). The ion will be gas stripped to a moderate positive charge and then regain electrons as it slows toward thermal velocity such that at the end of the recoil path, 88% of the polonium atoms have a +1 charge and the remaining 12% are neutral (Wellisch, 1913; Porstendörfer and Mercer, 1979). The study of the neutralization behavior of the polonium ion has been reported by Chu and Hopke (1988).

Other studies resulting from our previous DOE-supported programs have found that the $\cdot\text{OH}$ radicals produced by the deposition of energy into the air by the radon decay can oxidize SO_2 to SO_4^- with observable particle formation (Hopke and Ramamurthi, 1988). Thus, the decay of radon modifies the atmospheric environment in which the radioactive is present. It is the further study of these modifications that are the primary interest of this project.

LABORATORY STUDIES

The principal areas of the laboratory studies are 1.) Examine the formation of particles by the radiolytic oxidation of substances like SO_2 , ethylene, and H_2S to lower vapor pressure compounds and determine the role of gas phase additives such as H_2O and NH_3 in determining the particle size, 2.) Determine the formation rates of $\cdot\text{OH}$, $\cdot\text{O}$, or other oxidative radicals formed by the radiolysis of air following radon decay, 3.) Measure the rate of ion-induced nucleation using a thermal diffusion cloud chamber, and 4.) Measure the neutralization rate of $^{218}\text{PoO}^+$ in O_2 at low radon concentrations.

Radiolytic Nuclei Formation

We have measured the activity size distributions for a number of combinations of SO_2 and H_2O concentrations. Some of our previous studies have now been submitted for publication

(Ramamurthi *et al.*, 1992). In addition, other recent studies were reported in the MS thesis of Mrs. K. Karpen-Hayes (1991). In these studies, the activity size distribution are measured using a manual graded screen array system. In addition, open-face filter samples with long collection periods were obtained to measure the rate of sulfate production. The SO_4^{2-} is then leached from the filters and the amount of SO_4^{2-} quantified using ion chromatography. These results have provided additional insights into the process of radiolytic nuclei production. However, because the process of new particle formation really includes the oxidation of the SO_2 to H_2SO_4 as well as its subsequent ion-induced binary nucleation to small droplets, we are continuing to examine each of these steps separately in order to provide better fundamental understanding of each of the constituent steps in the process.

•OH Radical Production Rate

We are measuring the hydroxyl radical concentrations produced from radon decay with two different ways. One approach uses the chemical amplification system developed by Cantrell *et al.* (1984). The other technique is direct measurement using a laser-induced fluorescence (LIF) system.

In the chemical amplifier system, •OH radicals produced by ^{222}Rn decay in air participate in a series chain reactions with high concentrations of CO (10%) and NO (ppm), resulting in high concentrations of NO_2 . The product NO_2 is measured by luminol chemiluminescence. These reactant gases as well as radon-laden N_2 is mixed into a stream of purified air. The air then passes through a flow-through detector system based on the gas-surface reaction of NO_2 with a 0.01M Na_2SO_3 , 0.05% methanol, 0.001M aqueous luminol solution of pH 12 (Maeda *et al.*, 1980). Contaminant NO_2 in the reactant NO gas is removed by passing the gas stream over FeSO_4 .

Some positive results have now been obtained. Figure 1 shows the relationship between the chemiluminescence signal intensity and Rn concentrations in the gas stream. A straight line is fitted through the data points although there is obvious curvature to the data points. A partial mechanism for the reaction of the hydroxyl radicals and the reagent gases is

For our measurement conditions, the major radical sink procedure is HONO formation,

Figure 1. Chemiluminescence intensity as a function of radon concentration in the reagent gas stream.

and the following reaction is negligible.

The chain length (C.L.) for the series of catalytic reactions for NO_2 to $\cdot\text{OH}$ is a function of $[\text{NO}]$, $[\text{CO}]$, reaction time, initial hydroxyl radical concentration, and rate constants.

$$\text{C.L.} = \frac{\Delta [\text{NO}_2]}{[\cdot\text{OH}]_0} = \frac{k_2 [\text{CO}]}{k_3 [\text{NO}]} (1 - \exp(-\beta)) \quad (5)$$

where

$$\beta = \frac{2k_4 [\text{OH}]_0 t}{\alpha} \quad (6)$$

$$\alpha = \frac{2k_2k_4[OH]_0[CO]}{k_1k_3[NO]^2} \quad (7)$$

The rate constants were given by Cantrell *et al.* (1984) as

$$k_1 = 8.3 \times 10^{-12}$$

$$k_2 = 2.7 \times 10^{-13}$$

$$k_3 = 4.8 \times 10^{-12}$$

$$k_4 = 2.8 \times 10^{-12}$$

The total volume of our gaseous reactor and tube is 1662 cm³, and the gas mixture flowrate is 600 cm³ min⁻¹, so the reaction time is 166 seconds. The chain length has been calculated for the different NO concentrations at [CO] = 10% (Figure 2). If [NO] = 1 ppm, C.L. = 5612; if [NO] = 0.5 ppm, C.L. will reach a maximum value of 8816.

The luminol detection system was calibrated with a NIST standard NO₂ permeation tube yielding the calibration curve given in Figure 3. The regression equation is

$$[NO_2 \text{ (ppb)}] = 0.273 + 0.167x \quad (8)$$

Figure 2. Ratio of [NO₂] to [·OH]₀ as a function of chain length in the chemical amplifier system.

Figure 3. Response of the chemiluminescence detector as a function of [NO₂] in the gas stream.

where x is the signal intensity of the chemiluminescence detector. As a result of this calibration, the chemiluminescence intensity can be used to calculate the OH concentration as follows

$$[OH] = \frac{(0.273 + 0.167x) * 2.5 * 10^{10}}{5612} \text{ (molecules cm}^{-3}\text{)}. \quad (9)$$

The $\cdot OH$ radical production rate as a function of radon concentration is given in Figure 4. The slope for this straight line is 6.64×10^{-4} , so the number of OH radicals produced due to per Rn decay is

$$\frac{664 \text{ OH cm}^{-3}}{1 \text{ kBq m}^{-3}} = 6.64 * 10^5 \text{ OH Bq}^{-1}$$

This value is substantially higher than the value that was reported by Payne *et al.* (1991). They found that the number of $\cdot OH$ radicals equaled the number of ions produced in N₂ or Ar by the alpha decay. According to Lind (1961), that would yield $1.76 \times 10^5 \cdot OH$ per decay. Our work is in air. The presence of O₂ raises the possibility of other reactions such as the production of O(¹D) via radiolysis and its subsequent reaction with H₂O to produce $\cdot OH$. Additional measurements of the

Figure 4. Plot of the estimated $\cdot\text{OH}$ concentration as a function of the ^{222}Rn concentration in the reactant gas stream.

production rate at higher relative humidity are currently in progress.

A laser-induced fluorescence system has been established using an excimer laser system. We are currently determining the sensitivity of the system with $\cdot\text{OH}$ produced by photolyzing CH_3NO_2 . Once the response of the system to known $\cdot\text{OH}$ concentrations has been determined, the effectiveness of ^{222}Rn in producing $\cdot\text{OH}$ radicals will be measured.

Ion-Induced Nucleation

Another area of active development is the construction of a thermal diffusion cloud chamber (TDCC) (Katz, 1970) to study the rates of ion-induced nucleation caused by the ions produced by the radon decay. Previously, we calculated that there is the possibility of ion-induced nucleation occurring for indoor volatile organic compounds (VOCs) at levels observed in indoor air (Daisey and Hopke, 1992). Thus, we plan to make detailed studies of ion-induced nucleation. We wish to confirm the VOC calculations through direct measurement using some of the specific compounds identified in our calculations. Since the ionization produced by the α particle emitted by ^{222}Rn is columnar in nature, we will make our initial studies using the more diffuse ionization produced by

the β particle emitted by ^{85}Kr . ^{85}Kr can be introduced into the chamber is very low concentration. Subsequently we can use radon to determine its ability to induce nucleation.

Yefei Wu (1991) has theoretically investigated the behavior of the TDCC as his MS research problem. According to his calculations, we should be able to examine ion-induced of both VOCs and also the oxidation of SO_2 /binary nucleation of $\text{H}_2\text{SO}_4 - \text{H}_2\text{O}$ in the TDCC.

The TDCC has been constructed. The custom-designed and built heating and cooling system that maintains the top and bottom of the chamber at their appropriate temperatures arrived in January 1992. We have been testing the system and adjusting the flows to provide uniform temperatures across each of the plates. The system was assembled and tested using H_2O with helium as the support gas. We can observe visible nucleation. We are currently assembling the laser/optical system that will permit the quantitative measurement of nucleation rates in the chamber. Measurements of homogeneous nucleation of medium chain length alkanes will then be made to test the quantitative functioning of the chamber. Once we are certain that the chamber is able to function properly, the ion-induced nucleation measurements will be initiated.

Neutralization Rates

We have been corresponding with Professor A.J. Howard of the Department of Physics, Trinity College, Hartford, CT. Professor Howard has developed a system for measuring the neutralization rate for $^{218}\text{Po}^+$ ions under well controlled conditions (Howard and Strange, 1991). He has obtained results in good agreement with our earlier measurements (Shi and Hopke, 1991). It appears that it may be easier to make the critical measurements of the O_2 neutralization rates using their system. We have tentatively planned for Dr Howard and his students to visit us in early August in order to begin these experiments. Thus, the laboratory aspects of the project are underway and making good progress.

REAL INDOOR ATMOSPHERE STUDIES

The specific tasks of the exposure studies in occupied indoor spaces are 1.) Initiate measurements of the activity size distributions in actual homes with occupants present so that the variability of the indoor activity size distributions can be assessed with respect to indoor aerosol sources and general lifestyle variations of the occupants, 2.) Initiate a prospective study of the utility of measurement of deposited ^{210}Pb embedded in glass surfaces as a measure of the long-term, integrated exposure of the population to radon, and 3.) Develop the methodology to determine the

hygroscopicity of the indoor aerosol so that the changes in deposition efficiency of the radioactive indoor aerosol with hygroscopic growth in the respiratory tract can be assessed.

Exposure Assessments in Occupied Houses

The measurement of activity-weighted size distributions in normally occupied homes are made using the automated, semi-continuous, graded screen array system described by Ramamurthi and Hopke (1991). During the past year, we completed a series of measurements in a house in Arnprior, Ontario that had begun in May 1991 and ended in early July 1991. Another series of experiments were performed in a home in Parishville, NY from February to March 1992. Two week long measurements were made in 2 homes in Princeton, NJ in March and June. During the second set of exposure measurements, an additional set of intercomparison measurements were made in collaboration with personnel from the Environmental Measurements Laboratory. In our prior studies, there were differences in the estimated activity in the 1.5 to 15 nm size region. Although these differences do not appear significant in terms of typical aerosol measurements, they result in significant differences in estimated dose because of the very high dose per unit exposure in this size range. Additional chamber measurements are planned for August 1992. The results of several of our previous intercomparison efforts have been reported (Hopke *et al.*, 1992).

The results of four previous exposure measurements made on 3 different houses have been analyzed in collaboration with Dr. A.C. James of Pacific Northwest Laboratory (Wasiolek *et al.*, 1992). We have a considerable quantity of PAEC, size distributions, and radon concentration measurements that are currently being analyzed and a series of additional publications is anticipated. This work will continue to be done in collaboration with Dr. A.C. James to provide the most up-to-date dosimetry possible.

We have identified another home in our area in which 740 Bq m^{-3} was measured during the winter of 1990. However, measurements in this house made in early June showed $<20 \text{ Bq m}^{-3}$ because the house is well ventilated during the summer months. Measurements are planned for the fall after the home has been closed up for the colder winter months. Additional sites for study are currently being sought.

Prospective Study of ^{210}Po in Glass

Prior studies by Dr. R. Lively of the Minnesota Geological Survey (Lively and Ney, 1987) and Christer Samuelsson of Lund University, Sweden (Samuelsson, 1988) on the relationship between

measured ^{210}Po embedded in glass and ambient radon concentrations. When ^{214}Pb or ^{214}Bi is deposited on a surface in a room, they will adsorb to the surface. When the resulting ^{214}Po emits its α particle, the ^{210}Pb nucleus recoils with sufficient energy to embed it about 40 nm into the surface. Thus, it is thus below the surface and is not removed by cleaning, but it is close enough to the surface that the 5.3 MeV α from ^{210}Po can be observed.

All of the previous work has been done using existing glass surface in the house and a year of α measurements. Lively and Steck (1991) has demonstrated the kind of wide variability that can occur even in the annual average radon concentration. Also people are not always certain when the particular piece of glass was placed into its current environment. Thus, some uncertainty exists in the value of the calibration constant. Furthermore, there has not been a study of the variations that might exist within a room or between rooms in the same house. These are the questions our study hopes to address.

Our study plan was to place 150 pairs of 8" x 10" pictures into houses. So far we have placed 144 pair of pictures into homes from California to New York. Another 20 pictures in groups of ten have been placed into two schools (one in Maine and the other in South Dakota). In 50 houses, the pictures are placed in the most lived in room of the house (living or family room). In 50 houses, the pair of pictures go in the master bedroom and in the remaining 50 houses, the pair are split between the two rooms. Within each group of 50 placed in the same room, we have split them into two groups. In the one case, both pictures are placed on an inside wall and in the other, the pair are split between inside wall and outside wall. For the 50 split between two rooms, half have both pictures on inside walls and half have both on outside walls. The pictures are being placed for 18 months. With each picture, there is at least one etched track detector. For 50 individual pictures chosen at random, there are 2 detectors as a QA check. We also have storage blanks and spiked samples.

At the end of 18 months of exposure, the pictures will be shipped back to us. We will count the activity in the glass with pulse ionization chambers and send the alpha track detectors to be counted. We think such a study will give us a much tighter handle on the utility of this method for long term exposure assessment and therefore much improved epidemiology.

The ^{210}Po in the glass will be measured using pulse ionization chambers. We have one of Dr. Samuelsson's pulse ionization chambers. During the summer of 1991, it was tested by Jeffrey Ansley, an undergraduate student at the University of Michigan who participated in our NSF-supported Research Experience for Undergraduates (REU) program.

The program is therefore well underway with over three hundred pictures having been deployed. The first of these detectors will be returned in early 1993. The etched track detectors will be immediately returned for processing, but the pictures will need to sit for about 1 year to permit the in-growth of the ^{210}Po . Each picture will be individually wrapped in aluminum foil to prevent any inadvertent exposure in the laboratory. After the growth time has occurred, each piece of glass will be individually counted for a sufficiently length of time to provide better than 10 % counting statistics. Thus, several more years will be needed to complete this phase of our studies.

Hygroscopic Growth of Particles

The final task of this portion of the project was the development of the tandem differential mobility analyzer (TDMA) system to measure the hygroscopic growth of the indoor aerosol when it encounters > 99% relative humidity within the respiratory tract. A system has been developed that does permit the measurements to be made and a report describing this system has been published (Li *et al.*, 1992). The system has been used to perform a series of measurements on several types of indoor particle sources. Growth studies have been conducted on particles present in cigarette smoke (mainstream and sidestream), and produced by natural gas and propane flames. The particles from cigarette smoke are introduced to first DMA right after they are generated from the burning tip. The aerosols from natural gas and propane gas flame are generated in a 55 gallon steel barrel and then introduced into the first DMA. The average residence time of aerosols in the barrel is about 4 minutes. The residence time of all aerosols in the humidifier is about 20 seconds, approximately equal to the residence time of aerosols in human's respiratory tract. The growth ratios as a function of initial dry particle size are shown in Figures 5-8.

The results are as follows: For mainstream cigarette smoke, measurements were made for dry particle sizes ranging from 50 to 200 nm. The smoke was generated using a MasterFlex peristaltic pump to pull air through the burning cigarette and into the system. An average growth ratio of 1.09 ± 0.06 . For sidestream cigarette smoke, the pump was used to burn the cigarette and the generated sidestream smoke particles were directed into the measurement system. The dry particle size range was 50 - 400 nm and an average growth ratio of 1.04 ± 0.03 was observed.

A natural gas flame was produced using a Meeker burner. The burner produced dry particles in the particle size range of 10 - 200 nm. An average growth ratio of 1.08 ± 0.04 was measured. Propane gas flame was produced using a home repair torch. It produced a dry particle size range of 10 - 200 nm. An average growth ratio of 1.07 ± 0.03 .

Figure 5. Growth of mainstream cigarette smoke particles as a function of initial dry particle size.

Figure 6. Growth of sidestream cigarette smoke particles as a function of initial dry particle size.

Figure 7. Growth of natural gas burner particles as a function of initial dry particle size.

Figure 8. Growth of propane torch particles as a function of initial dry particle size.

The growth ratios for each of the four kinds of particles are found to be greater than 1.0. However, the growth is not significant compared to the maximum system measurement error of 8%. (Li *et al.*, 1992). Also the growth data do not show any clear trend indicating that the hygroscopic growth is below or close to the system detection limit.

FUTURE RESEARCH PLANS

The future plans principally focus about completing the tasks described above. With regards to laboratory studies, we expect to complete all of the OH radical production rates measurements within the next 6 months. It is clear from the results to date that radon decay will not produce a sufficient amount of $\cdot\text{OH}$ to significantly affect indoor air chemistry at ^{222}Rn concentrations typically found in indoor air.

The thermal diffusion cloud chamber is now ready to test its performance. Our initial work will be the duplication of prior homogeneous nucleation results on simple hydrocarbons to insure that the system is performing satisfactorily. Once we are certain that the system is performing, then we will examine the ion-induced nucleation of those organic compounds that had the highest free energy of formation in our calculations of ion-induced nucleation based on both the Suck (1982) and Thompson (1906) theories. Since there are discrepancies between the two theories, the quantitative measurements of the nucleation rates that we will be able to make could help to resolve the discrepancies as well as add to the data base for future theoretical studies. Ultimately, we plan to look at the combined oxidation and nucleation mechanism to study the binary nucleation of water and sulfuric acid.

In our field studies, we have tested the first pulse ionization chamber. We have acquired the counting electronics to allow us to build 6 additional units so that we can efficiently count the glass when it returns from exposure in early 1993 and the ^{210}Po has adequately grown into secular equilibrium with the ^{210}Pb . As the counters come on line, we will be looking for opportunities to use the counters to measure artifacts in conjunction with on-going epidemiological studies. We hope to be able to begin taking data that can subsequently be fully evaluated when our prospective glass study has been completed.

We continue to find houses in which we can make exposure measurements. It appears that we will have additional houses to study during the next year in which we will measure activity size distributions in a series of normally occupied homes. We would like a mixture of various housing conditions (oil versus gas heating, hot water versus forced air, gas versus electric stoves, absence or

presence of wood stove or fireplaces, use of space heaters (electric or kerosene), presence or absence of electrostatic air cleaners and/or humidifier on forced air systems, and effects of central air conditioning). In addition to the activity size distributions, we also plan to measure particle number distributions with the DMA and we hope to be able to measure deposition flux so that the room surface-to-volume ratio can be inferred. These additional measurements are by nature grab measurements, but they can provide the data to estimate the room model parameters necessary to better understand the dynamics of the radon progeny in the house.

We are continuing to use the TDMA system to measure the hygroscopic growth of particles from a number of specific sources. The system is not readily portable. The need for several water baths to produce the temperature-controlled water for the wet-film reactor and for the bubblers used to temperature condition and humidify the sheath air for the second DMA means we will have to study aerosols in the laboratory and a very limited range of houses. We want to make additional laboratory studies of sources like a kerosene heater, and electric motors similar to those found in vacuum cleaners. We also plan to examine several commonly used materials that are aerosolized in their normal use including spray deodorant, PAM spray-on oil, household deodorizer (such as Lysol or Glade air freshener), and one or two pesticide sprays (Raid or Black Flag).

We will move the system to Professor Hopke's home for additional studies of a free standing wood stove (Vermont Casting Model "Resolute"), an open fireplace, an electric broiler cooking meat (Jenn-Aire broiler), a deep-fat fryer, and background aerosol at several times during the year. We will also examine the particles in the presence of a number of people in the room to examine people as an aerosol source.

SUMMARY

We have continued to make progress based on our previous efforts. All of the experimental systems are now on-line, and we are well into the collection of substantial new data characterizing the behavior of the radon progeny both in the laboratory and in the real world.

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CURRENT PROJECT PERSONNEL

Principle Investigator

Philip K. Hopke, Robert A. Plane Professor of Chemistry

Other Professionals

Nathalie Montassier, Research Associate, August 1, 1990 - present.

Graduate Research Assistants

Huiling Ding, Research Assistant, July 1, 1990 - present.

Wei Li, Research Assistant, July 1, 1990 - present.

Feng He, Research Assistant, August 15, 1991 - present.

PUBLICATIONS ARISING FROM THIS CONTRACT

Measurement of the Size Distributions of Radon Progeny in Indoor Air, P.K. Hopke, M. Ramamurthi, and C.S. Li, in *Aerosols: Science, Industry, Health, and Environment*, S. Masuda and K. Takahashi, eds, Vol. 2, Pergamon Press, Ltd, Oxford, 842-847 (1990).

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10 / 15 / 92