TEMPERATURE EFFECT OF PHASE TRANSITION RADIATION OF WATER

BY

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THESIS

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ABSTRACT

Phase transition radiation of water revealed in the past provides a possible idea to modify weather predictions nowadays into more accurate ones. With existence, characteristic wavelength, and mechanisms of this phase transition radiation presenting by scientists and engineers, temperature effect of such radiation is studied in this research to obtain more knowledge on this phenomenon.

Experiments and computer simulations of transmissivity tests as well as results discussion are performed in this study. A mixture of liquid and vapor water is generated while letting IR radiation at 3.1-4.95 \( \mu m \) passing through. A detector is used to measure photon counts serving as radiation intensity of two conditions, with mixture, and without mixture. Transmissivity at different volume fraction of liquid water is calculated by manipulating measurements of photon counts. The results of Monte Carlo transmissivity simulation at considered conditions match experimental data, indicating that phase transition must be included whenever phase transitions occur. Temperature effect is discussed by comparing experimental and simulation results at different temperatures. Transmissivity at another temperature is expected to predict well using the conditions shown in this research.
To Mom and Dad
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Chapter 1 Introduction and Literature Review

Water is broadly used in science and engineering research including as a cooling medium in industries and a solvent in chemical and biological sciences, due to its high heat capacity and unusual density change. With rising concerns on global climate issue, water plays a key role since it largely exists on Earth surface in three phases. Moreover, weather phenomena such as rain, snow, and thunderstorms are created by its phase transitions. The most common weather phenomenon is water clouds in the sky and properties of water clouds have been explored by researchers. In an initial understanding, water clouds can be assumed to be a plain layer in the air. Supposing the optical constants and the density profile of clouds are known, transmissivity of clouds in the sky can be calculated. Another important factor that is required in calculation is the size distribution of cloud droplets. Mono-disperse droplets represent equal-size cloud droplets, while poly-disperse droplets stand for droplets at different size. Experimentally it is very difficult to determine actual size distribution of cloud droplets; therefore, gamma distribution is often assumed to describe the size distribution of cloud droplets. If gamma distribution is given, some quantities for water clouds such as mean droplet radius, cloud density, and other characters can be deduced. Detailed calculation about quantities of water clouds can be found in [1]. Yet, phase transitions of water cloud, also known as water cycles that can be detected every day are not included in these calculations. It is widely known that phase transition of water is a latent heat exchange mechanism. Due to the large scale of water on Earth and in the sky, these
phase transitions have effects on the atmosphere and the weather. It seems that water phase transitions have been well understood for more than one hundred years. However, about forty years ago, an abnormal infrared phase transition radiation disclosed by scientists has drawn our attention to water phase transition in a quantum view manner. The temperature effect of this radiation is still unclear while such radiation and its temperature dependence may directly contribute to precise weather predictions.

1.1 Uncommon Phase Transition Radiation

In the 1960s, an unusual infrared thermal radiation was reported during water liquid-vapor transition in the sky cloud observation; however, interpretation of this fact was unclear at that time. It is known that at a given temperature in thermodynamic equilibrium, the ideal radiation is blackbody radiation, also called Planck’s radiation, since this kind of radiation obeys Planck’s function. In order to examine phase transition radiation, experiments are usually set up in thermodynamic non-equilibrium. Hence, this phase transition radiation could be called non-Planck’s radiation. It is also expected that this phase transition radiation should occur in an equilibrium system whenever phase transitions take place. The traditional theories and equations for phase transition using the Einstein coefficients, $A_{21}$ for spontaneous emission, $C_{12}$ for absorption, and $C_{21}$ for stimulated emission, are insufficient to describe this certain non-Planck’s radiation. In addition, efforts were made to observe the reality and
characteristic wavelength of water phase transition radiation. Nevertheless, the qualitative expression and mechanism of this radiation were undefined.

This kind of phase transition radiation is of great importance, and needs to be further investigated because it is related to global warming and has an impact on weather prediction. Although the possibility of energy exchange from latent heat to photons is low in a large water mass, extensive water stays in oceans and in the atmosphere, which makes this radiation phenomenon discernible. In addition, its characteristic wavelength is in infrared spectra. Radiation emitted from the Earth to space is also dominant in infrared spectra under normal temperature. Accordingly, it could be expected that phase transition radiation for absorbing or emitting would influence climate prediction and have a contribution to global climate change. The so-called global warming is the result that radiation which comes from the Sun’s reflection and the Earth’s emission are blocked by the atmosphere, so the heat cannot go to space and retaining heat raises the Earth’s temperature. Notwithstanding the fact that water has not attracted much concern in the current global warming issue, it can be conjectured that global warming predictions will be more accurate than today, once the knowledge for water phase transition radiation is solid and included in climate models. Besides, radiation is a very important mechanism of heat transfer. As long as two objects have different temperatures even in a vacuum condition, radiative heat transfer would occur. Usually in most science and engineering, when other heat transfer mechanisms, conduction, convection, or mass transfer, are involved, radiation is neglected by virtue of its comparing small magnitude. Present atmospheric models are
such cases which disregard radiation. Yet a large amount of water on Earth and in the sky increases the possibility of the occurrence of phase transition radiation. Therefore, adjustments to current weather prediction models can be anticipated.

In order to show evidence for the existence of water phase transition radiation and its characteristic wavelength, experiments and observations were made for three different transitions, vapor-liquid, liquid-solid, and vapor-solid [2-6]. In 1968, Nichols and Lamar [2] used an IR line scan instrument to convert IR image to visible image in three spectral regions, 0.5-1.0, 3.0-5.5, and 8.0-14.0 microns. When photograph and infrared view between forming and dissipating cumulus clouds were compared as shown in Fig. 1.1 and Fig. 1.2, the top sides of both clouds are good reflectors for sunlight in visible and near infrared regions. At the same time, surprisingly a phenomenon happened: an unknown strong radiation appeared at 8-14 \( \mu m \) at the bottom of both forming and dissipating clouds. The authors claimed that this strong radiation might come from the cloud itself or radiation from the Earth by cloud scattering. Also in 1968, Potter and Hoffman [3] discovered an abnormal increased infrared radiation from two to one hundred times larger than blackbody radiation from boiling water at 1.537 and 2.10 \( \mu m \), as shown in Fig. 1.3. While the strong emission bands at 1.537 and 2.10 \( \mu m \) were not found in water vapor or \( CO_2 \) bands, Potter and Hoffman used water cluster and water liquid absorption band at 1.44 and 1.91 \( \mu m \) to explain this radiation. The authors called this radiation phase transition luminescence, and asserted that water clusters could radiate latent heat. Then water clusters were free
to combine other molecules to form clusters again. Later, Mestvirishvili et al. [4] in 1976 designed two experiments measuring characteristic wavelength for water phase transition radiation of two situations, liquid-solid and vapor-liquid transitions. Obtaining experimental data on characteristic transition bands, liquid-solid transition at 28-40 microns and vapor-liquid transition at 4-8 microns, the authors declared that releasing latent heat from phase transition could be converted into both radiation and photons. After reaching thermodynamic equilibrium, these photons would generate Planck’s radiation. Peral’mann and Tatartchenko [5] in 2008 obtained their experimental data for characteristic wavelength in three transitions, sublimation at 2.57 microns, condensation at 2.96 microns, and crystallization at 20 microns. When comparing their experimental characteristic wavelength to data collected by Mestvirishvili et al. [4], the authors argued that radiation emission from Mestvirishvili et al. [4] were multi-photon transitions. Recently, Tatartchenko [6] in 2011 analyzed and commented on various infrared images from satellite and radiometer. A fact was demonstrated that at specific wavelengths, infrared characteristic radiation did exist. In order to explain this circumstance, Tatartchenko referred to Bader [7] to construct the scheme of interpretation. It was mentioned that radiation detected by satellite would mostly come from the upper troposphere if humid upper troposphere was present. Yet, several misunderstandings still arise without clear qualitative interpretation of this radiation.
Fig. 1.1 (a) Photograph of forming cumulus clouds. (b) Infrared view of the same clouds. [2]

Fig. 1.2 (a) Photograph of dissipating cumulus cloud. (b) Infrared view of the same cloud at temperature about -5 °C. [2]
Fig. 1.3 Relative intensity from boiling water intensity $I_w$ at 100°C to blackbody intensity $I_{bb}$. [3]

Fig. 1.4 Laser-induced condensation results in atmosphere. Mie backscattering coefficient $\beta_{Mie}$ was measured as a function of height. [13]
In recent years, with the demonstration of existence of water phase transition radiation, research was shifted to seek application of this transition radiation [8-14]. Optical control of crystal growth rate was suggested to be performed by phase transition radiation. By analyzing the relaxation time and the radiation power, Sall’ and Smirnov [8] proposed that this phase transition could happen in a bulk containing $10^5$ particles leading to controlling desired properties and grain size distribution of particles. Ambrok et al. [9] revealed the possibility of modifying the growth kinetics of particle ensemble by providing external selective heat source. Controllable growth rate and particle ensemble evolution could be achieved using different external heating. Moreover, several applications of phase transition radiation were recommended by Tatartchenko [10-12]. First, fog formation and crystallization could be triggered by such transition radiation. Secondly, a good prediction of hailstorm would be obtained due to intensive infrared radiation of hailstorm. Thirdly, infrared laser could be generated from this transition radiation and the power of laser could be amplified with proper mirror geometry settings. Fourthly, detecting radiation offered a method to discover the subsistence of water on other planets. Finally, Earth temperature could be regulated by emitting or absorbing radiation to/from clouds to adjust the greenhouse effect. In fact, Rohwetter et al. [13] seemed to realize the idea of rain formation. Laser pulses at 0.8 $\mu m$ were used to motivate water condensation in laboratory cloud chamber during atmospheric ($T= -24^\circ C$, RH=230%) and sub-saturated atmospheric ($T= 60^\circ C$, RH=75-85%) conditions. Laser-induced condensation was also
accomplished in real atmosphere and a relative increase of *Mie backscattering* with laser ejection at different heights (mostly at 45-75 meters) indicating the occurrence of condensation, compared with reference signal without laser ejection, as shown in Fig. 1.4. With further investigation, Petrarca et al. [14] utilized 100TWatts laser pulses with a photon bath (the beam portion surrounding filaments) to produce water condensation in the air, which was much faster than utilizing typical incident intensity. The power of laser could be above 550 $GW/cm^2$ and the active volume for using laser with a photon bath was three to four order magnitudes larger than using filaments only. Macroscopic effect on laser-induced condensation in the air was expected with such high power laser.

To give qualitative expression of this uncommon phase transition radiation, Wang and Brewster [15] started from the two-level radiative transfer equation in condensation process,

$$\left( \frac{dI_v}{ds} \right) = -n_l L_i \frac{I_v}{c} C_{12} + n_g H A_{21} \frac{I_v}{4\pi} + n_g H \frac{I_v}{c} C_{21}$$

(Eq. 1.1)

where $H$ and $L$ are population distributions of excited state and lower state, $n_g$ and $n_l$ are number densities of excited state and lower state, $I_v$ is specific intensity in frequency basis, $c$ is the light speed, $ds$ is a differentiate path length, and $dw$ is a differentiate solid angle. Population density function $H$ is the convolution of translational energy $F$ and rotational energy $G$,

$$F\left( \frac{1}{\lambda}, T \right) = \frac{2}{\sqrt{\pi}} \frac{hc}{kT} \frac{1}{\lambda} \exp\left( -\frac{hc}{kT} \frac{1}{\lambda} \right)$$

(Eq. 1.2a)
Fig. 1.5 Radiative relaxation during vapor condensation. [15]

Fig. 1.6 Population distribution for different temperature (a) 2-a transition (b) 2-b transition. [15]
\[ G\left(\frac{1}{\lambda_r}, T\right) = \frac{g_{j,r} \exp\left(\frac{-e_{j,r}}{kT}\right)}{Z_r} \]  
(Eq. 1.2b)

\[ H\left(\frac{1}{\lambda_{r,s}}, T\right) = \sum_{\frac{1}{\lambda_r}}^{\frac{1}{\lambda_s}} F\left[\left(\frac{1}{\lambda_{r,s}} - \frac{1}{\lambda_r}\right), T\right] \ast G\left(\frac{1}{\lambda_r}, T\right) \]  
(Eq. 1.2c)

where \( \frac{1}{\lambda_r} \), \( \frac{1}{\lambda_s} \), and \( \frac{1}{\lambda_{r,s}} \) are translational, rotational, and trans-rotational wavenumbers, \( k \) is Boltzmann’s constant, \( g_{j,r} \) is rotational energy degeneracy, \( T \) is temperature, \( e_{j,r} \) is rotational energy, and \( h \) is Planck’s constant. Using the assumption of thermodynamic equilibrium, a new form of radiative equation along a line of sight was derived. While comparing the new derivation with the general radiative equation along a line of sight, the authors defined evaporative absorption coefficient \( K_v \) to account for phase transition in radiative transfer calculation,

\[ K_v = \frac{A_v}{4\pi} \frac{H}{B_v} n_g \]  
(Eq. 1.3)

where \( B_v \) is blackbody intensity in frequency basis. This evaporative absorption coefficient is emerged into typical radiation equation when phase transition is considered,

\[ \frac{dI}{ds} = -\left(\frac{K_v}{K_v} + K_{v}\right)I_v + \left(\frac{K_v}{K_v} + K_{v}\right)B_v \]  
(Eq. 1.4)

where \( K_v \) is non-phase-transition absorption coefficient. As for mechanism of phase transition condensation, they recommended that the radiation relaxation would come
from the transition between excited state (state 2 in Fig. 1.5) and first intermediate state (state a in Fig. 1.5) while different intermediate states represent different numbers of hydrogen bond formation. By defining the energy difference between state 2 and state a, the characteristic wavelength of condensation radiation could be assigned,

\[
\frac{1}{\lambda} = \frac{e_t}{hc} + \frac{e_{J.r}}{hc} + \frac{1}{hc}(-H_{HB} - 3kT)
\]

(Eq. 1.5)

where \( H_{HB} \) is hydrogen bond energy, and \( e_t \) is translational energy. The population distributions at various temperatures for two kinds of transition were shown in Fig. 1.6. The characteristic wavelength matched the result from Mestvirishvili et al. [4] for vapor-liquid transition at 4-8 microns.

1.2 Scattering Theories for Radiation in Attenuating Medium

The most general scattering theory is *Mie scattering theory* that describes the solution of electromagnetic radiation scattered by a sphere. When particle size parameter \( x = \pi d / \lambda \) and relative particle optical constants \( \tilde{n} \) are given, extinction coefficient, albedo, and phase function can be expressed analytically [16]. Although particle size parameter \( x \) is defined by particle diameter, Mie scattering theory is still applicable to non-spherical particles when characteristic length of particle is delineated. Relative particle optical constants are computed by taking the ratio from particle optical constants to surroundings optical constants. When absorption of surroundings is strong, scattering by particles is negligible. Nevertheless, even with analytical equations, Mie
scattering solutions are still too complicated to apply. Therefore, some approximations to Mie theory made Mie theory practical to use.

One approximation for \(|\bar{n} - 1| \ll 1, 2x|\bar{n} - 1| \ll 1\) and \(Q_e \ll 1\) is called Rayleigh-Gans scattering by van de Hulst [17]. It contains characteristics of Mie scattering and yet gives much simpler forms than Mie theory. The other advantage for this approximation is that if refractive index of a particle is close to one and absorptive index is very small, particle size parameter can be arbitrarily chosen. Another approximation is called anomalous diffraction. Several works have been done to reveal its analytical expressions. Evans and Fournier [18] presented a semi-empirical approximation for extinction coefficient in anomalous diffraction region. By introducing a factor for extinction coefficient, the modified anomalous diffraction approximation was valid for all particle size parameters, including Rayleigh and large particles, when optical constants were in the domain, \(1.01 \leq n \leq 2.00\) and \(0 \leq k \leq 10\),

\[
Q_e = Q_v f \quad \text{(Eq. 1.6a)}
\]

\[
Q_v = \text{Re} \left[ 2 + \frac{4e^{-w}}{w} + \frac{4(e^{-w} - 1)}{w^2} \right] \quad \text{(Eq. 1.6b)}
\]

\[
f = 2 - \exp \left( -x^\frac{2}{3} \right) \quad \text{(Eq. 1.6c)}
\]

where \(w\) was given by

\[
w = 2kx + i\rho, \quad x = \frac{\pi d}{\lambda}, \quad \rho = 2x(n-1).
\]
Brewster [16] suggested the extinction and absorption coefficients of anomalous diffraction in different forms, which do not contain complex number calculation, but the extinction coefficient $Q_e$ in Eq. 1.7a yielded same results as $Q_v$ in Eq. 1.6b presented by Evans and Fournier [18],

$$Q_e = 2 + 4u^2 \cos(2\beta) - 4e^{-\rho \tan(\beta)} \left[ u \sin(\rho - \beta) + u^2 \cos(\rho - 2\beta) \right]$$

(Eq. 1.7a)

$$Q_a = 1 + \frac{2}{v} e^{-\rho} - \frac{2}{v^2} (1 - e^{-\rho})$$

(Eq. 1.7b)

where

$$\rho = 2x(n-1), \quad \tan(\beta) = \frac{k}{n-1}, \quad u = \frac{\cos(\beta)}{\rho}, \quad v = 4xk = 2\rho \tan(\beta).$$

Specific to water clouds, Chylek et al. [19] came up with 10th order polynomials for $Q_a$, $Q_v$, $Q_s$, and asymmetry parameter $g$ in long and short wavelength regions as functions of particle radius $r$,

$$Q_a = \sum_{n=0}^{N} a_n r^n$$

(Eq. 1.8a)

$$Q_v = \sum_{n=0}^{N} b_n r^n$$

(Eq. 1.8b)

$$Q_s = \sum_{n=0}^{N} c_n r^n$$

(Eq. 1.8c)

$$g(r)Q_s = \sum_{n=0}^{N} d_n r^n$$

(Eq. 1.8d)

where $a_n$, $b_n$, $c_n$, and $d_n$ are obtained by least-square fitting. In long wavelength
region, 3-25 \( \mu m \), these fittings provided accuracy within 3% and \( 10^4 \)-\( 10^5 \) times faster of computational time. If an error was tolerated, the quadratic fittings were sufficient for calculating these radiative properties. In short wavelength region, 0.5-2.0 \( \mu m \), with a proper droplet size distribution, the fittings were close to Mie theories and more accurate than considered approximations of anomalous diffraction and geometric optics.

1.3 Research Objectives

Increasing knowledge on this issue, theoretical statements, mechanisms, and characteristic properties, some parts of this radiation puzzle have been revealed through previous research. However, scientists and engineers have not reported temperature effect of water phase transition radiation. In order to have a better understanding of water phase transition radiation, experiments and simulations are necessary. Wang [20] developed an experimental setup and a Monte Carlo simulation program to learn more about this radiation phenomenon. This work on temperature effect of water phase transition radiation is inspired by Wang and the purpose of this work is to disclose temperature feature of the puzzle. When the operating temperature is changed, thermodynamic saturated conditions change, which influences water liquid and vapor masses in the experiment. In addition, temperature has an effect on water liquid optical constants, which alters extinction and absorption coefficient of water liquid. Wang’s experimental and Monte Carlo simulated conditions are adjusted in this research. A
new form of extinction coefficient is shown to have a more complete expression. Notwithstanding that the operating temperature of the experiment is set at a high temperature, the modified extinction coefficient is expected to be applied in a broad temperature range.

The temperature of clouds has a big impact in predicting Earth surface temperature. Temperature of clouds is determined by altitude, and temperature can be a driving force to change the altitude of clouds. As pointed out by Thomas and Stamnes [21], by using a simple radiative equilibrium model, different altitudes of clouds gave different Earth surface temperatures for fixed cloud coverage and opacity. Moreover, from thermodynamic point of view, condensation and evaporation take place at all temperatures except at zero degree Kelvin. Once condensation and evaporation happen, there are potentialities for the occurrence of absorbing and emitting phase transition radiation. Considering numerous water phase transitions everyday on the Earth’s surface and atmosphere, accommodating atmospheric models with temperature effect of water phase transition is necessary, which may lead to a more refined weather prediction.
Chapter 2 Experimental Methods and Results

An experiment [20] is set to understand the temperature effect of phase transition radiation of water. Water clouds are generated in a cloud chamber system to provide a condition of vapor-liquid mixture. While infrared radiation passing through chamber with cloud inside, attenuation of infrared radiation is observed by measuring photon counts on an infrared camera. Temperature is made to be uniform on the chamber wall. Transmissivity of homogeneous cloud with occurrence of condensation and evaporation inside the cloud chamber is concerned. The experimental result for transmissivity of 75 °C clouds is presented.

2.1 Experiment Apparatus

The schematic for transmissivity experiment of water clouds is shown in Fig. 2.1. This system can be divided into three parts: IR camera with a focusing lens, cloud chamber, and IR source with an optical shutter.

Fig. 2.1 Schematic for of measurement of water clouds transmissivity. [20]
IR Camera and Focusing Lens

The IR camera and the focusing lens were assigned on the one side of cloud chamber to supply optical data measurement of the experiment. The IR camera was a liquid-nitrogen cooled device, which covered spectral range from 1 $\mu m$ to 5.2 $\mu m$ (Santa Barbara Focalplane, CA). The filter used in this experiment was at 3.1-4.95 $\mu m$ prescribing by rotating filter wheel on the camera. The computer software, WinIR3, associated with camera was installed and focal plane array 240×304 pixels were set for this experiment. The IR camera could show different colors on the image according to the temperatures it detected. The frame rate at 63.13 Hz and integration time at 0.2004 msec were used during experiment. The operating temperature of the camera is less than 90 K and calibration for camera must be done every time before employment. An external focusing lens was put between the IR camera and the cloud chamber to collect radiation from cloud chamber and infrared source. The position of camera and external focusing lens were determined by the alignment test. In the alignment tests, a heating object was placed to substitute IR source and adjusted the position of camera and external lens until a glow bar was shown in the camera image.

Cloud Chamber

Cloud chamber was regarded as the place to have phase transition radiation of water. The chamber was made of aluminum in a cylindrical T-shape (Al tee tubing: O.D. 4’’ (10.16 cm), 16 gauge, 16’’ (40.64 cm) long). High thermal conductivity of
aluminum could maintain temperature uniformity over the chamber. Two IR windows were settled on the two horizontal openings of cloud chamber and were silicon wafers with only one side polished. The unpolished side of windows working as a diffusive surface was positioned facing the interior cloud chamber. An aluminum piston was located on the vertical opening of cloud chamber. Teflon tape was twined on the piston to prevent leakage between cloud chamber and atmosphere outside the chamber. Communicating tube was built on the cloud chamber to detect pressure change of cloud chamber, served as a manometer. Nichrome ribbon was coiled on the exterior chamber surface and connected to a DC power supply. Temperature of chamber could be regulated by changing heating rate from power supply to sustain an approximately uniform temperature on chamber surface. Fiberglass insulation was covered on the exterior chamber surface after coiling nichrome ribbon.

Fig. 2.2 Thermocouples locations on the inner surface of cloud chamber. [20]
Fig. 2.3 Temperature of inner wall as labeled in Fig. 2.2 in test 1210-1.

Fig. 2.4 Averaged wall temperature and medium temperature in test 1210-1.
Since there were four openings on the chamber, two horizontal openings for IR windows, one vertical opening for Al piston, and one small opening for manometer, it was necessary to examine sealing situation for the chamber. Small amount of liquid water was supplied into cloud chamber, and the four openings were sealed. While the heating process of the cloud chamber, liquid water evaporated into vapor water with a slightly change of pressure. The Al piston was moved manually to balance the pressure change. Cooling process of chamber was proceeded to let water vapor condense. Condensation of water showed a more noticeable pressure change than evaporation. Cloud chamber was untouched for more than 12 hours to test whether the chamber could maintain the pressure change or not. Proper sealing situation was decided until almost no pressure balance to surroundings happening in the chamber. Therefore, one parameter of transmissivity experiment, liquid water mass, could be controlled.

As for temperature measurement, there were six thermocouples (Watlow, K-type) for the cloud chamber. These thermocouples were connected to computer by a recorder box (BNC- 2090, National Instrument) and a data transmitter card (Daqcard-Ai-16e-4, National Instrument). Five of them were allocated along the horizontal inner surface of chamber as shown in Fig. 2.2. The other thermocouple was suspended into cloud chamber at position 5 in Fig. 2.2 to show radial temperature variation. This suspended thermocouple was placed not to touch chamber surface and also not to interfere light path in the experiment. Since data was measured by IR camera using photon counts, objects inside the chamber, which might block light would lead to a different outcome on data measurement. Efforts had been made to provide an uniform temperature
distribution along chamber surface. The difference between maximum and minimum temperature readings from chamber wall was less than 1.5 °C in most experiments. In some tests, this difference was bigger than 1.5 °C but still less than 1.8 °C. Average of readings from five thermocouples represented chamber wall temperature, while the suspended thermocouple gave temperature reading of the cloud medium. During the heating and cooling process, chamber medium temperature was always less than averaged wall temperature. Throughout heating stage, heat was supplied to cloud chamber by nichrome ribbon on the outer surface of chamber; accordingly, the hanging thermocouple that was away from heating supply had lower temperature reading. In the course of cooling, the dangling thermocouple was close to IR window that did not have fiberglass insulation protection. Therefore, a lower temperature from suspended thermocouple was expected. The averaged wall temperature could be regarded as maximum temperature and the medium temperature was behaved as minimum temperature during experiment and data calculation. For all tests, the difference between maximum and minimum temperature was less than 3 °C. A temperature distribution of chamber wall temperature is shown in Fig. 2.3 and a comparison of averaged wall temperature and medium temperature is shown in Fig. 2.4. The other temperature variation that might appear was in azimuthally direction. It was pointed out by Wang [20] that azimuthally temperature distribution processed better temperature uniformity than horizontal direction. Consequently, thermocouples along azimuthally direction were taken out in the experiment. The current and voltage of DC power
supply were modified to support heat to cloud chamber. When heat was supplied to the chamber, several mechanisms contributed to heat loss such as conduction and convection from insulation to ambient, as well as radiation through IR window. For a typical experiment in winter, voltage at 24V and current at 1.38A were set to give a steady operating chamber temperature at 75 °C.

Fig. 2.5 Comparison between IR source and blackbody intensity. Both intensities are normalized respectively by their intensity difference between 323-368 K. [20]

**IR Source and Optical Shutter**

The third part of experiment device is the IR source with an optical shutter located on the other side of cloud chamber horizontally. From radiative heat transfer viewpoint, a practical way to make an object emit blackbody radiation is to have isothermal
enclosure with a small hole letting radiation leave. Radiation that leaves from the small cavity would approach blackbody radiation even if emissivity of the enclosure is not equal to one. Additionally, the inner surface of enclosure should be a rough surface instead of an optically smooth surface. Smooth surface could not maintain enough reflection inside the enclosure, since multi-reflection is another key factor to generate blackbody radiation [16, pp. 12-15]. Therefore, a Teflon-coated aluminum cavity was built to offer radiation close to blackbody radiation. Aluminum was also used for the source because of its high thermal conductivity which could produce isothermal condition as described in cloud chamber. In order to probe this radiation from the IR source, Wang [20] used a pair of calcium fluoride plano-convex lenses (Thorlabs, 25.4 mm diameter, 100 mm effective focal length) and the IR camera to measure this radiation. Intensity measured from camera was compared to Planck’s radiation at a temperature range from 323 K to 368 K and the normalized result is shown in Fig. 2.5. The result conveyed a message that this aluminum enclosure could bring about blackbody-like radiation. The operating temperature of IR source could be chosen arbitrarily if no change of chemical composition and phase states in the chamber happened. A typical operating temperature of IR source is around 130 °C, and the maximum radiation range of the source at this temperature is in infrared region. An optical shutter made from aluminum with anodized in black was placed between the cloud chamber and the IR source to work as a switch of IR source. With or without IR source during data measurement was an important factor; details of data accessing will
be presented in later sector. The positions of IR source and shutter as well as the IR camera and focusing lens were also determined in the alignment test.

2.2 Experiment Procedure

The idea of this experiment was to measure transmissivity when water clouds appeared inside the chamber as compared to the transmissivity without water clouds existing in the chamber. Transmissivity data at different volume fractions of water liquid was introduced. When water vapor-liquid mixture arose inside the chamber, it was anticipated that the measured transmissivity data must contain information of phase transition radiation. With such experiment setup, temperature effects of phase transition radiation could be examined. Transmissivity calculation was presented by Wang [20]. Volume fraction of water liquid was related to water liquid mass, water vapor mass, density and chamber volume. Details for each experiment step and calculation will be explained in the following sections.

Initial Mass of Water Vapor in the Air

Before any operations for experiment, it was necessary to know how much water vapor already existed in the air. One experimental purpose was to create water clouds inside the chamber; hence, initial mass of water vapor must be taken into account. Such information could be obtained from relative humidity of air, so a hygrometer was used for this measurement. Transmissivity experiment was hold indoor in a college town, so relative humidity would not be zero as if in a desert. Moreover, on a rainy day, with
indoor air-conditioner or heater, relative humidity would not be one. Typical relative humidity of the lab environment is 20%-50%. Assuming water-vapor is an ideal gas, specific volume of initial water vapor inside the chamber at surroundings temperature \( T_s \) could be calculated from relative humidity \( RH \) and saturated specific volume of water vapor at surroundings temperature \( v_{g,\text{sat}}(T_s) \),

\[
v_g(T_s) = \frac{v_{g,\text{sat}}(T_s)}{RH}
\]  
(Eq. 2.1)

where subscript \( g \) represents water in vapor phase. During the heating and cooling processes of the cloud chamber (evaporation and condensation of water), pressure inside the cloud chamber maintained at ambient pressure, together with proper sealing (no water mass exchange). Therefore, water vapor partial pressure remained unchanged from surroundings temperature \( T_s \) to operating temperature \( T \) (75 °C in this experiment).

\[
P_g(T_s) = P_g(T)
\]  
(Eq. 2.2)

Then, specific volume of water vapor in cloud chamber at operating temperature \( T \) could be expressed as,

\[
v_g(T) = v_g(T_s) \frac{T}{T_s}
\]  
(Eq. 2.3)

The initial mass of water vapor could be shown by taking ratio between chamber volume at operating temperature \( T \) and \( v_g(T) \),
\[ m_i = \frac{V(T)}{v_s(T)} \]  

(Eq. 2.4)

**Heating and First Data Record**

After initial mass of water vapor was considered, cloud chamber was heated up to operating temperature 75 °C to a steady state, which means temperature of the chamber was maintained at 75 °C within tiny fluctuation and first data of transmissivity experiment should be recorded. When optical shutter was closed, the data of photon counts from IR camera \( R_{w_0,c} \) (\( w_0 \) represents without water clouds, and \( c \) stands for closed shutter) was noted as background signal which contained radiation from optical shutter, cloud chamber, and other radiation noises from surroundings. When optical shutter was opened, reading from IR camera \( R_{w_0,o} \) (\( o \) for opened shutter) included radiation from IR source, optical shutter, cloud chamber, and other radiation noise from surroundings. Difference between \( R_{w_0,o} \) and \( R_{w_0,c} \) gave information of IR source intensity for chamber without cloud at operating temperature. This difference value was regarded as the reference intensity in transmissivity calculation.

As for data acquisition region, a cold spot suggested by Wang [20] from camera image was selected. This cold spot was a reflection of liquid nitrogen dewar on the IR window near IR camera, where thermal noise was minimized. The cold spot reflection is shown in Fig. 2.6 and can be recognized easily on camera image. Readings from image were recorded by choosing an appropriate area inside the cold spot. Whereas
uniform photon counts were distributed in the cold spot, an area of camera image could be chosen arbitrarily to some extent. The computer software, WinIR3, would provide averaged photon counts once an area was picked.

![Reflection of cold spot used as data record region.](image)

Cloud Formation and Second Data Record

In order to generate clouds inside the chamber, some operations should be performed, adding water liquid as well as letting water evaporate and condense. Adding more water was essential because initial water vapor from air could not reach a saturation condition for cloud formation. After recording first data of relative intensity without water clouds, distilled water liquid was poured into cloud chamber. Typical added water liquid mass $m_a$ would be from 0 to 1.3 gram, because water mass from this region would show an apparent change and a trend for transmissivity data. Next step would be heating the chamber with dispensed water liquid and initial water vapor to a temperature higher than evaporation temperature. Within a closed chamber system,
when evaporation of water took place inside cloud chamber, a pressure rise could be identified with care. This temperature should be held for a while to let all water inside evaporate. Then, heating rate would be decreased to make chamber cool down. Absolutely turning off heating was not allowed during cooling process, because rapid cooling of the chamber possessed a high possibility of enabling water condense on IR windows, on which had no fiberglass insulation as mentioned before. Another opening on the chamber which was connected to manometer would be a place to have water condense. Thus, additional heat must be supplied to manometer and IR windows openings during cooling cloud chamber not to let condensation happen. The other opening, Al piston, was not a major concern as appropriate insulation was installed and also the piston was treated as the boundary of cloud formation.

There were some phenomena need to be clarified during cooling process. At the time of chamber cooling, once temperature was lower than dew point temperature, condensation would occur inside the chamber. When water condensation happened in the chamber, a pressure drop could be observed easily compared to evaporation. Dew point temperature was calculated from water steam table to make sure that the operating temperature of chamber 75 \( ^\circ C \) was below dew point temperature to form clouds. Another aspect should be deliberated was supersaturation of water clouds. This transmissivity measurement was conducted in a college town without specific aerosol treatment inside the laboratory, so homogeneous condensation nuclei were unlikely achieved and cloud condensation nuclei were enough to prevent clouds supersaturation. In addition, low cooling rate also reduced the possibility of supersaturation. Moreover,
Kelvin formula describes the relation between critical supersaturation and droplet radius at a given temperature and a given surface tension from droplet curvature effect [22]. Smaller droplet radius would have higher critical supersaturation. For droplet radius at 0.01 \( \mu m \), equilibrium supersaturation is about 12.5%; for droplet radius at 1 \( \mu m \), equilibrium supersaturation is about 0.12%. Typical radius of cloud droplets at 5-15 \( \mu m \), supersaturation from Kelvin curvature effect is around 0.01%. Furthermore, if a specified amount of solute (NaCl) is contained in the medium to induce condensation, Köhler curves were used to describe supersaturation as a function of droplet radius. For a large droplet as cloud droplet, Köhler effect is even smaller than Kelvin curvature effect. As a result, supersaturation of cloud droplet was neglected in this study.

When homogeneous water clouds existed inside the chamber and the temperature of chamber was maintained at 75 °C steadily, second data should be marked. It is the same idea as first data recording to measure relative intensity when optical shutter was closed and opened. Camera readings with cloud inside the chamber with opened shutter was noted as \( R_{w,c} \), while data with closed shutter was recorded as \( R_{w,o} \). (w represents with water; \( c \) and \( o \) stand for closed and opened shutter respectively) Difference between \( R_{w,o} \) and \( R_{w,c} \) indicated IR source intensity for chamber with clouds at desired temperature. This relative intensity value suggested a concept that how much IR source radiation had disappeared when water clouds subsisted. Besides, chamber volume \( V(T) \) should also be registered by measuring the position of the Al piston.
Transmissivity and Water Liquid Volume Fraction Calculation

In order to turn the measured raw information into useful data, transmissivity and volume fraction of water liquid, some manipulations have to been carried out. Transmissivity of water clouds at operating temperature 75 °C in spectral region 3.1 μm to 4.95 μm was defined as,

\[
\tau = \frac{R_{w,o} - R_{w,e}}{R_{wo,o} - R_{wo,e}} \quad \text{(Eq. 2.5)}
\]

To calculate volume fraction of water liquid, how much water vapor mass was still inside cloud chamber without condensation should be computed first,

\[
m_v = \frac{V(T)}{v_{g,\text{sat}}(T)} \quad \text{(Eq. 2.6)}
\]

With these water mass values, initial water mass \(m_i\), added water liquid mass \(m_a\), and existing water vapor mass \(m_v\), volume fraction of water liquid inside the chamber could be obtained,

\[
f_v = \frac{m_i + m_a - m_v}{\rho_l(T)} \frac{1}{V(T)} \quad \text{(Eq. 2.7)}
\]

where \(\rho_l(T)\) is water liquid density at target temperature. Error range of transmissivity is determined by measuring photo counts from IR camera in first and second data record. As for volume fraction, maximum and minimum temperature of cloud chamber defined earlier, averaged wall temperature and medium temperature, were used in Eq. 2.6 to get two \(v_{g,\text{sat}}(T)\) values which would provide a span for volume fraction.
Fig. 2.7 Water condensation on the IR window close to the IR camera.

### 2.3 Experiment Observation

**Water Condensation**

As stated beforehand, water condensation must be avoided except inside the chamber, and there are some matters that should be marked. When part of water mass did not condense inside cloud chamber to form cloud, calculation of liquid volume fraction would be wrong. Therefore, checking water condensation on the four openings on the chamber and chamber wall was required for every experiment. For the manometer and the piston, these two could be removed from chamber to observe condensation by eyes. For the two IR windows, IR camera image could be used to examine whether condensation occurs or not. While choosing an area on image to record photon counts, standard deviation was also calculated and shown in the image.
When water condensation happened, it was easy to see the IR window image shown in Fig. 2.7, which was close to IR camera by changing the position of focal lens in front of camera. In this experiment, since the medium temperature, 75 °C, is higher than surroundings temperature, condensation could be found easily on the IR window close to IR camera when water $f_w$ is larger than $0.8 \times 10^{-4}$. This condensation situation would lead to wrong data measurement because homogenous cloud inside the chamber is broken and energy extinction primary comes from water condensation on the IR window. Therefore, efforts had been made to avoid condensation on the IR window close to the camera by carefully installing insulation protection and conductive heating around the window. With these improvements, the difference between medium temperature and wall-averaged temperature in Fig. 2.4 dropped from less than 3 °C to less than 2 °C. Another IR window close to IR source would not be a place to have water condensation because IR source was nearby to make the window to be relative warmer region. Condensation on chamber wall was successfully overcome because of experiment design: nichrome ribbon was installed on outer surface of chamber to heat cloud chamber. As noticed formerly, chamber wall temperature was always higher than medium temperature when heating and cooling. Accordingly, condensation might come into existence at a relative colder region (medium) instead of a relative hotter region (wall).
Camera Photon Counts

Although due to bad pixels in the camera image, it was not easy to notice the existence of water clouds inside the chamber on IR camera image, photon counts of image from chamber still offered useful information to learn more about water phase transition radiation and its temperature effect. Photo counts from camera image could be adjusted by changing settings in WinIR3 software. The settings in “video oscilloscope” could be modified to give different photon counts. A typical reading for first data record (without water clouds) was around 2500 with a standard deviation around 13 and difference for first data record was 70-80. In some experiments, the difference values were dropped to 60, so IR source temperature was altered to retrieve this difference value back to 70-90. Wang [20] tested the effect of different IR source temperatures at 104 °C and 125 °C. For the same liquid volume fraction, the transmissivity results from two IR source temperatures were nearly unchanged. Consequently, IR source temperature could be decided arbitrary as long as water clouds inside the chamber was not affected. A common temperature of IR source in this work was set 120-140 °C depending on surroundings temperature.

Fig. 2.8 Calibration result using two-temperature method.
IR Camera Calibration and Alignment Test

Every time before conducting experiment, camera calibration had to be done. Calibration could be performed by following the instruction in the WinIR3 software manual. The idea of camera calibration was that by providing an object with known temperature to camera, camera could measure photon counts of this object to set as a calibration reference. Regular calibration for IR camera in transmissivity test was to prepare objects at two different temperatures for the camera. One temperature calibration could also be done, but yielded not so good calibration result. Typical calibration result is shown in Fig. 2.8. The most suitable material for these objects is a metal with a high thermal conductivity to keep it isothermal and with black surfaces to make them close to blackbody.

If the positions of IR camera and focusing lens were changed, the alignment test must be executed. IR source and optical shutter were moved out and replaced by a hot
object, while making no changes on cloud chamber. Projection of glow bar on cold spot as shown in Fig. 2.9 could be obtained by modifying positions of IR camera, focusing lens, and heated object. After alignment test with clear appearance of glow bar, the position of the heated object determined the positions of IR source and optical shutter.

**IR Radiation Contribution of IR Camera**

There were several sources of infrared radiation contributing to photon counts data of the IR camera. When there was no water clouds, radiation came from IR source, two IR windows and cloud chamber at respectively temperatures. In addition, reflection of surroundings radiation from the polished surface of IR window facing IR camera would offer another radiation contribution. Moreover, background noises and electronics in the laboratory may affect radiation detected by IR camera. When IR camera was slightly moved so the cold spot would be not in the image, photon counts dropped much and an uniform distribution of photon counts were found. These uniform photo counts included radiation from the chamber, background noise, and reflection of IR window. Moreover, a comparison was made when cloud chamber was at surroundings temperature and a temperature few degrees higher than surroundings temperature. These examinations ensured that IR reading of camera mainly came from radiation from IR window.
Fig. 2.1 Mass absorption coefficient of water liquid and vapor at room temperature. Circular points for liquid and rhombus points for vapor. [20]

**Water Vapor Absorption**

Absorption of IR radiation inside the chamber came from water liquid rather than water vapor; therefore, suspended water clouds inside the chamber was regarded to be responsible for radiation extinction. In some experiments, water liquid mass was controlled to be low which could not reach saturation condition such that no water clouds were generated in cloud chamber. When no cloud occurred, transmissivity went to unity. Then temperature was decreased a little bit to make condensation happen, and transmissivity fell to ~0.65. This showed that in the spectral region in this work, water vapor and air had nearly no contribution to absorption, while water liquid was a major concern of radiation absorption. Rusk, Williams, and Querry [23] suggested mass
absorption coefficient $\kappa [cm^2/g]$ of water liquid and While et al. [24] presented mass absorption coefficient of water vapor at room temperature. The comparison of two mass absorption coefficients were shown in Fig. 2.10. It was clear that in the working spectral region, mass absorption coefficient of water liquid is almost 3 orders magnitude larger than mass absorption coefficient of water vapor. Therefore, water vapor absorption was neglected in this work, and the emphasis would be put on water liquid and phase transition.

2.4 Experiment Results

Transmissivity data of water clouds for cloud chamber at $74.8 \pm 1.5 \, ^\circ C$ (simply 75 $^\circ C$) in spectral region 3.1-4.95 $\mu m$ is plotted in Fig. 2.11 as a function of volume fraction of water liquid $f_v$. The range of volume fraction is chosen between 0 and $2 \times 10^{-4}$ to show distinct variation of transmissivity. When water liquid volume fraction is small, transmissivity is very sensitive. In such a small interval of volume fraction at 0-0.2 $\times 10^{-4}$, transmissivity $\tau$ varies from 1 to 0.65 to around 0.4. As described in last section, when small amount of water was added to cloud chamber causing small volume fraction, transmissivity jump from 0.65 to 1 could also be found by increasing cloud chamber temperature a few degree Celsius, maintaining that extinction of IR radiation mostly came from water liquid absorption. In some measurements, condensations on IR window close to camera and manometer were observed, provoking wrong determination of water liquid mass in cloud chamber. Those data that acted like
a shift in transmissivity plot were omitted in the transmissivity plot. Except for the alteration of transmissivity at tiny volume fraction, another transition of $\tau$ is found at volume fraction $0.1\times10^{-4} - 0.5\times10^{-4}$. In this small volume fraction region, transmissivity is also sensitive to $f_v$; therefore, more experimental data were acquired. When volume fraction reaches $0.6\times10^{-4}$, transmissivity decreased gradually as $f_v$ increases.

![Transmissivity for cloud chamber at 75 °C in 3.1-4.95 μm](image)

Fig. 2.11 Transmissivity for cloud chamber at 75 °C in 3.1-4.95 μm.
Chapter 3 Simulation and Result Discussion

Monte Carlo simulation for transmissivity experiments is presented in this chapter. Radiative heat transfer theories used in Monte Carlo simulation as well as the conditions and ideas for transmissivity simulation are also articulated. Comparisons between simulated results and experimental results imply the existence of phase transition radiation. A complete form of extinction coefficient replaces the extinction coefficient defined in [20] and is used in simulations. While proper probability constant of Einstein’s spontaneous emission coefficient $a_{21}$ is given, the method and idea of this simulation are expected to clarify temperature dependence of water phase transition radiation.

3.1 Radiative Heat Transfer Theories

Before going to computer simulation, thermal radiative heat transfer theories and equations for water clouds need to be constructed to form a base of simulation. As explained before, strong water liquid absorption coefficient and data from transmissivity test make it obvious that water clouds behave as a participating medium in cloud chamber, and the rest of gas such water vapor, nitrogen, and oxygen would have nearly no contribution to radiation extinction. Extinction of energy from two contributions, volumetric and surface, is shown in this section and the concepts and equations will be used in Monte Carlo simulation. Other simulation conditions such as
scattering phase function, optical constants, and a polynomial fitting of $H/B$ are also presented in this section.

**Extinction from Water Droplet Volumetric Contribution**

Volumetric extinction for water droplet is one energy depletion mechanism out of two, and extinction and absorption coefficient are described by Mie scattering theory. This work adopts the approximations for Mie theory to reduce simulated computational time. The modified anomalous diffraction approximation (MADA) by Evans and Fournier [18] is selected to express volumetric extinction efficiency. The applicable conditions for MADA is when optical constants at $1.01 \leq n \leq 2.00$ and $0 \leq k \leq 10$, as well as no restriction on particle size parameter $x$. In the transmissivity experiments, since optical constants of water clouds sit in this range, it is appropriate to apply MADA and equations for extinction efficiency are shown in Eq. 1.6, where additional subscript $v$ in Eq. 3.1 indicates that these properties are volumetric contribution,

$$Q_v = Q_v f$$  \hspace{1cm} (Eq. 3.1a)

$$Q_v = \text{Re} \left[ 2 + \frac{4e^{-w}}{w} + \frac{4(e^{-w} - 1)}{w^2} \right]$$ \hspace{1cm} (Eq. 3.1b)

$$f = 2 - \exp \left( -\frac{2}{3}x^3 \right)$$ \hspace{1cm} (Eq. 3.1c)

where $w$ was given by

$$w = 2kx + i\rho = \frac{\pi d}{\lambda}.$$
As for volumetric absorption efficiency, the equation is presented by Brewster [16] shown in Eq. 1.7b,

\[ Q_{av} = 1 + \frac{2}{v} e^{-v} - \frac{2}{v^2} (1 - e^{-v}) \]  
(Eq. 3.2)

where

\[ v = 4\alpha k = 2\rho \tan(\beta) \].

Volumetric scattering efficiency \( Q_{sv} \) is the difference between extinction efficiency \( Q_{ev} \) in Eq. 1.6 and absorption efficiency \( Q_{av} \) in Eq. 1.7b,

\[ Q_{sv} = Q_{ev} - Q_{av} \].  
(Eq. 3.3)

Droplets of water clouds were assumed mono-dispersed and in spherical shape; hence, volumetric extinction, absorption, and scattering coefficient \( K_{ev}, K_{av}, K_{sv} \), are able to obtained respectively and a summation equality is attained due to mono-dispersed assumption,

\[ K_{ev} = \frac{1.5Q_{ev}f_v}{r} \]  
(Eq. 3.4a)

\[ K_{av} = \frac{1.5Q_{av}f_v}{r} \]  
(Eq. 3.4b)

\[ K_{sv} = \frac{1.5Q_{sv}f_v}{r} \]  
(Eq. 3.4c)

\[ K_{ev} = K_{av} + K_{sv} \]  
(Eq. 3.4d)

where \( r \) is water droplet radius, and \( f_v \) is volume fraction of water liquid. The relationship between water droplet radius and liquid volume fraction is,
\[ f_v = \frac{4}{3} \pi r^3 N_0 \]  

(Eq. 3.5)

where \( N_0 \) is number density of water droplets also called cloud condensation nuclei (CCN) in the literature. At given water liquid volume fraction, once CCN is known water droplet radius could be derived. Without special aerosol treatment in the laboratory where transmissivity tests were executed, heterogeneous condensation nuclei could be achieved. As regular heterogeneous condensation nuclei occupied in laboratory air, CCN can be assumed nearly constant.

**Extinction from Water Droplet Surface Contribution**

Another contribution of attenuating energy during transmissivity analysis comes from the surface of water droplets, and part of this surface contribution comes from single surface effect of cloud droplet. Phase transition of water is treated to proffer part of energy diminishment that appeared on the interface between water liquid and air. Considering the condition for experiment was settled at thermodynamic equilibrium, it is valid to apply evaporative absorption coefficient in Eq. 1.3, and this coefficient only counts for single surface absorption. Surface phase transition has no contribution on scattering, so extinction from surface contribution all originates from absorption. The term and symbol for Eq. 1.3 are redefined here. Single surface absorption coefficient \( K_{ss} \) is,

\[ K_{ss} = \frac{A_{31} H}{4\pi B_v n_g} \]  

(Eq. 3.6)
where $A_{21}$ is Einstein’s coefficient for spontaneous emission, $H$ is population distribution of gas state, $B_v$ is blackbody intensity in frequency basis, $n_g$ is number density of gas state, and subscript $ss$ represents single surface. In [20], Wang relates Einstein’ coefficient to binary collision rate due to interface phase transition, and define $A_{21}$ as,

$$A_{21} = a_{21}N_0r_d^2 \frac{8\pi kT}{m_g},$$  \hspace{1cm} (Eq. 3.7)

where $a_{21}$ is probability constant of $A_{21}$, $N_0$ is CCN, $r_d$ is radius of water droplet, $m_g$ is molecule mass of water. Thence, single surface absorption becomes,

$$K_{ss} = \frac{a_{21}H}{4\pi B_v}n_gN_0r_d^2 \frac{8\pi kT}{m_g}.$$  \hspace{1cm} (Eq. 3.8)

Accompanying single surface absorption cross-section per particle $C_{ss}$ and single surface absorption efficiency $Q_{ss}$ are,

$$C_{ss} = \frac{K_{ss}}{N_0} = \frac{a_{21}H}{4\pi B_v}n_gr_d^2 \frac{8\pi kT}{m_g};$$  \hspace{1cm} (Eq. 3.9)

$$Q_{ss} = \frac{C_{ss}}{\pi r_d^2} = \frac{a_{21}H}{4\pi^2 B_v}n_g \frac{8\pi kT}{m_g}.$$  \hspace{1cm} (Eq. 3.10)

The derivation for single surface absorption coefficient can be found in [20, Appx. D].
Surface phase transition does not happen only on a single surface, so to make surface contribution more complete, multi-surface absorption must be investigated. Considering Fig. 3.1, when a unit of incident energy hits a water droplet at an arbitrary surface, three phenomena happen on the surface. Part of this unit energy would be reflected by the surface, part of energy would be absorbed due to phase transition absorption, and the rest of energy would transmit through the first surface. An energy balance on a surface is anticipated,

\[ 1 = R + \tau + f_s \]  

(Eq. 3.11)

where \( R \) is surface reflection, \( \tau \) is surface transmission, \( f_s \) is surface absorption. Water liquid droplet is able to absorb energy since its optical constant \( k \) is not zero; therefore, internal transmittance \( T \) is included when keeping track of the unit energy.
Whenever energy runs into droplet surface, reflection, absorption, and transmission come into being. Surface absorption coefficient is represented by counting total surface absorption in Fig. 3.1 described as,

\[ Q_{as} = f_s + \tau T f_s + \tau^2 T^2 R f_s + \cdots \]  

(Eq. 3.12)

where subscript \( s \) in \( Q_{as} \) stands for surface contribution. Exploiting the sum of linear progression, Eq. 3.12 can be rewritten as,

\[ Q_{as} = f_s + f_s \frac{\tau T}{1 - TR} . \]  

(Eq. 3.13)

It is clear that the first term on right hand side in Eq. 3.13 is the first surface absorption or single surface absorption defined in last section, and second term accounts for all surface absorption other than the first surface. The parameters in Eq. 3.13 are,

\[ f_s = Q_{as} \]  

(Eq. 3.14)

\[ T = 1 - Q_{as} \]  

(Eq. 3.15)

where \( Q_{as} \) is in Eq. 3.7, and \( Q_{as} \) is in Eq. 3.2. In order to calculate surface reflection term \( R \), several steps need to be gone through. Taking advantage of Fresnel relations, directional spectral reflectivity can be achieved. The equations of Fresnel relations in [16, pp. 130] which does not contain complex number calculation are,

\[ R_\perp = \frac{(n_1 \cos \theta_1 - u)^2 + v^2}{(n_1 \cos \theta_1 + u)^2 + v^2} \]  

(Eq. 3.16)

\[ R_\parallel = \frac{\left[ (n_2^2 - k_2^2) \cos \theta_1 - n_1 u \right]^2 + \left[ 2n_2 k_2 \cos \theta_1 - n_1 v \right]^2}{\left[ (n_2^2 - k_2^2) \cos \theta_1 + n_1 u \right]^2 + \left[ 2n_2 k_2 \cos \theta_1 + n_1 v \right]^2} \]  

(Eq. 3.17)
where $R_\perp$ and $R_\parallel$ are perpendicular and parallel interface reflectivities, $n_i$ is refractive index of air assuming constant at 1.003 in this case, $n_2$ is refractive index of water liquid, $k_2$ is absorption index of water liquid, and $\theta_i$ is incident angle from air to water liquid. Intermediate parameters $u$ and $v$ are given by,

$$2u^2 = \left(n_2^2 - k_2^2 - n_1^2 \sin^2 \theta_i\right) + \sqrt{\left(n_2^2 - k_2^2 - n_1^2 \sin^2 \theta_i\right)^2 + 4n_1^2k_2^2};$$  
(Eq. 3.18)

$$2v^2 = -\left(n_2^2 - k_2^2 - n_1^2 \sin^2 \theta_i\right) + \sqrt{\left(n_2^2 - k_2^2 - n_1^2 \sin^2 \theta_i\right)^2 + 4n_1^2k_2^2}.$$  
(Eq. 3.19)

Directional spectral reflectivity is to take average value of perpendicular and parallel reflectivities,

$$\rho_\lambda = \frac{1}{2}(R_\perp + R_\parallel)$$  
(Eq. 3.20)

The reflectivity for a spherical particle subject to collimated irradiation is equivalent to the hemispherical reflectivity of a flat surface to diffuse irradiation. [16, pp. 308]

Accordingly, the surface reflectivity $R$ can be shown,

$$R = \frac{1}{\pi} \int_{2\pi} \rho_\lambda \cos \theta_i d\Omega = 2\int_0^{\pi/2} \rho_\lambda \cos \theta_i \sin \theta_i d\theta_i$$  
(Eq. 3.21)

where $d\Omega$ is differential solid angle for a sphere and can be expressed as,

$$d\Omega = \sin \theta d\theta d\phi$$  
(Eq. 3.22)

where $\theta$ is polar angle, and $\phi$ is azimuthal angle in polar coordinate system while azimuthally symmetry is assumed in Eq. 3.21. Combining Eq. 3.13 through Eq. 3.22, surface absorption coefficient is,
$$Q_{as} = Q_{asv} + Q_{ass} = Q_{asv} + Q_{asv} \left(1 - R - Q_{asv}\right) (1 - Q_{av}) \over 1 - (1 - Q_{av}) R$$

(Eq. 3.23)

where $Q_{ass}$ represents absorption due to multiple surface other than the first surface.

The results for $Q_{asv}$ and $Q_{ass}$ are shown in Fig. 3.2 and Fig. 3.3 at two limiting volume fractions. The cloud condensation nuclei CCN value is chosen at $6.22 \times 10^4$ cm$^{-3}$, probability constant $a_{21}$ is $3 \times 10^{-8}$, and wavelength is at 3.1-4.95 $\mu$m in working range of this research. It is clear that $Q_{ass}$ has the same order magnitude comparing to $Q_{asv}$, and even at some wavelength, adding $Q_{ass}$ almost doubles total surface absorption efficiency $Q_{as}$. Associating two contributions, volumetric and surface, extinction efficiency, extinction coefficient, scattering coefficient, and albedo can be acquired,

$$Q_v = Q_{ev} + Q_{as} = Q_{ev} + Q_{sv} + Q_{as}$$

(Eq. 3.24)

$$K_e = \frac{1.5Q_{as} f_v}{d} + \pi r^2 N_0 Q_{as}$$

(Eq. 3.25)

$$K_s = \frac{1.5Q_{as} f_s}{d}$$

(Eq. 3.26)

$$w_0 = \frac{K_s}{K_e}$$

(Eq. 3.27)

These equations are key factors for transmissivity calculation and will be used in Monte Carlo simulation.
Fig. 3.2 Comparison of $Q_{a_{ss}}$ and $Q_{m_{sa}}$ at $f_v = 0.1 \times 10^{-4}$. (a) Single surface absorption efficiency $Q_{a_{ss}}$. (b) Multiple surface except the first surface absorption efficiency $Q_{m_{sa}}$.

Fig. 3.3 Comparison of $Q_{a_{ss}}$ and $Q_{m_{sa}}$ at $f_v = 2 \times 10^{-4}$. (a) Single surface absorption efficiency $Q_{a_{ss}}$. (b) Multiple surface except the first surface absorption efficiency $Q_{m_{sa}}$. 
Scattering Phase Function

Scattering phase function describes directional distribution of scattered energy. When energy undergoes a participating medium with scattering particles, this energy may change its own direction after hitting scattering particles. Phase function is defined as a probability distribution to indicate which direction energy would go after particle scattering. One limiting case is called isotropic scattering characterizing that energy after scattering is equal to all directions, and in this case, phase function $p$ is equal to one. Plots for phase function at different particle size parameters $x$ can be found in [1, pp. 365-372]. This work adopts phase function of Rayleigh-Gans scattering theory named by van de Hulst [17]. Optical constants does not appear in the phase function which only depends on angular angles and $x$. Assuming azimuthal symmetry, phase function of Rayleigh-Gans scattering can be formulated as,

$$P(\theta) = C \left[ \frac{3}{u^3} (\sin u - u \cos u) \right]^2 \left(1 + \cos^2 \theta\right)$$

(Eq. 3.28)

where

$$u = 2x \sin(\theta/2)$$

and $C$ is a normalized constant determined by conservation of scattering energy,

$$\frac{1}{2} \int_0^\pi P(\theta) \sin \theta d\theta = 1$$

(Eq. 3.29)
Table 3.1 Optical constants $n$ and $k$ of liquid water. [25]

<table>
<thead>
<tr>
<th>$\tilde{v}$ [cm$^{-1}$]</th>
<th>$\lambda$ [\mu m]</th>
<th>$n$ (39 °C)</th>
<th>$k$ (39 °C)</th>
<th>$n$ (50 °C)</th>
<th>$k$ (50 °C)</th>
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<tr>
<td>3300</td>
<td>3.030</td>
<td>1.42</td>
<td>0.23</td>
<td>1.41</td>
<td>0.21</td>
</tr>
<tr>
<td>3200</td>
<td>3.125</td>
<td>1.48</td>
<td>0.14</td>
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</tr>
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</tr>
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</tr>
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<td>0.0085</td>
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<td>0.0085</td>
</tr>
<tr>
<td>2000</td>
<td>5.000</td>
<td>1.319</td>
<td>0.007</td>
<td>1.314</td>
<td>0.007</td>
</tr>
</tbody>
</table>

Other Simulation Conditions

Optical constants of water liquid were obtained by Pinkley et al. [25] at different temperatures as shown in Table 3.1. As discussed in Chapter 2, water vapor has almost no contribution to absorb energy; therefore, water vapor optical constants are not concerned in the simulation. To get optical constants of water liquid at 75 °C, extrapolation is used. Moreover, for refractive index $n$, Lorenz-Lorentz equation can also be applied. Lorenz-Lorentz equation for liquids is shown by [16, pp. 183],

$$\frac{n^2 - 1}{n^2 + 2 \rho} = \text{const.}$$  \hspace{1cm} (Eq. 3.30)

where $\rho$ is density of the liquid. The comparison of refractive index $n$ from extrapolation and Lorenz-Lorentz equation as well as absorption index $k$ from
extrapolation are shown in Table 3.2. Although at some wavelengths, two refractive indices are not close, but when using these two sets of optical constants in Monte Carlo simulation, transmissivity yields almost the same results. Consequently, to make it simple, optical constants from extrapolation are adopted in the simulation.

Table 3.2 Comparison of optical constants from extrapolation and Lorenz-Lorentz equation.

<table>
<thead>
<tr>
<th>$\tilde{\nu}$ [cm$^{-1}$]</th>
<th>$\lambda$ [\mu m]</th>
<th>$n$ (extrapolation)</th>
<th>$n$ (Lorenz-Lorentz)</th>
<th>$k$ (extrapolation)</th>
</tr>
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<tbody>
<tr>
<td>3300</td>
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<tr>
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<tr>
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<td>5.000</td>
<td>1.3026</td>
<td>1.3133</td>
<td>0.0070</td>
</tr>
</tbody>
</table>

Table 3.3 Coefficients of polynomial curve-fitting for $H/B$, by Wang [20] in Eq. 3.29 at temperature 75 °C.

<table>
<thead>
<tr>
<th>$C_0$</th>
<th>$C_1$</th>
<th>$C_2$</th>
<th>$C_3$</th>
<th>$C_4$</th>
<th>$C_5$</th>
<th>$C_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$3.39 \times 10^{-3}$</td>
<td>$-1.04 \times 10^{-1}$</td>
<td>$-2.06 \times 10^{-1}$</td>
<td>$3.45 \times 10^1$</td>
<td>$-3.97 \times 10^2$</td>
<td>$1.58 \times 10^3$</td>
<td>$-1.53 \times 10^3$</td>
</tr>
</tbody>
</table>
In Eq. 3.8 – Eq. 3.10, a complicated ratio $H/B_v$ is presented. Therefore, a six-order polynomial curve-fitting by Wang [20] at 3-8 $\mu m$ is used to provide an analytical expression,

$$\frac{H}{B_v} = \sum_{i=0}^{6} C_i \lambda^i. \quad \text{(Eq. 3.31)}$$

Resulting coefficients for 75 °C are displayed in Table 3.3.

### 3.2 Monte Carlo Simulation

Monte Carlo method on radiative heat transfer is a statistical method to capture photons’ behaviors in a given volume [16, pp. 196-215, pp. 470-490]. Energy that passes the volume is treated as individual energy bundles, and by tracking each bundle whether it is emitted, scattered, or transmitted, radiative properties of the volume such as transmissivity, reflectivity, and absorptivity can be calculated. The algorithm for Monte Carlo radiative transfer simulation is:

1. Assign values for energy bundle in the beginning.
2. Choose a location for emitting an energy bundle.
3. Choose a wavelength for the bundle.
4. Choose a propagation direction of the bundle.
5. Choose a path length of the bundle to determine the next action of the bundle.
6. Compute new location of the bundle.
7. Decide whether the bundle is absorbed or scattered at the new location.
8. Jump to step 1 or 3 if the bundle is absorbed or scattered respectively.

9. Repeat step 2 to 8 for all energy bundles.

**Assign values for energy bundle in the beginning**

Assigning values for energy bundle is arbitrary to some extent. Brewster [16] suggested starting from net radiant energy balance in an elemental volume,

\[ Q_{\text{net}} = \text{emission} - \text{absorption} \]  \hspace{1cm} (Eq. 3.32)

The emission energy can be manipulated into,

\[ \text{emission} = wN \]  \hspace{1cm} (Eq. 3.33)

where \( w \) is the energy per bundle, and \( N \) is the number of emitted bundles. Assuming the energy per bundle is constant for all bundles, absorption energy is,

\[ \text{absorption} = wS \]  \hspace{1cm} (Eq. 3.34)

where \( S \) is the number of absorbed bundles. The number of emitted bundles \( N \) is selected by user, which decides the accuracy of simulation. Typically, larger \( N \) would have more accurate results but at the same time larger \( N \) yields long computational time. A proper number of \( N \) is chosen to fit user’s needs, and in this work \( N \) is chosen at 8000. The energy per bundle \( w \) in the simulation is utilizing blackbody intensity of IR source at 4.95 \( \mu \text{m} \) divided by \( N \) and using this \( w \) value to get number of emitted bundles at different wavelengths.
Choose a location for emitting an energy bundle

The location of the bundle is decided by its probability distribution function and its cumulative density function. The probability of emission in a differential volume $dV$ into a small solid angle $d\Omega = \sin\theta d\theta d\phi$ at a small wavelength region $d\lambda$ is the ratio of the emitted power in $dV$, $d\Omega$, $d\lambda$ interval to total emitted power,

$$P(\lambda, \theta, \phi, r) d\theta d\phi d\lambda dr = \frac{K_{a_i} I_{b_i} \cos \theta \sin \theta d\theta d\lambda dV}{\int_{\lambda} \int_{\theta} \int_{\phi} K_{a_i} I_{b_i} \cos \theta \sin \theta d\theta d\lambda dV} \quad \text{(Eq. 3.35)}$$

where $P(\lambda, \theta, \phi, r)$ is probability distribution function, $K_{a_i}$ is spectral absorption coefficient, and $I_{b_i}$ is spectral blackbody intensity. To calculate the probability of emission from $dV$, integration of Eq. 3.35 along directional and wavelength dependence with assumptions of uniform temperature and uniform spectral absorption coefficient is performed,

$$P(r) dr = \frac{dV}{V} \quad \text{(Eq. 3.36)}$$

The cumulative density function $R$ is obtained by integrating Eq. 3.36 over the subset volume $\partial V$,

$$R(r) = \int_{\partial V} P(r) dr = \frac{\partial V}{V} \quad \text{(Eq. 3.37)}$$

Since the cumulative density function $R$ is at the range 0 to 1, it is reasonable to pick a value between 0 and 1 randomly to decide the location of emission.
Choose a wavelength for the bundle

The idea of choosing a wavelength is the similar as choosing a location. The probability of emitting the bundle in \( d\lambda \) is by integrating Eq. 3.35 over volume and directions,

\[
P(\lambda) d\lambda = \frac{K_{a_1} e_b}{K_{em_p} e_b} d\lambda \quad \text{(Eq. 3.38)}
\]

where \( e_b \) is spectral blackbody radiation flux, and \( K_{em_p} \) is the Planck mean emission coefficient defined as,

\[
K_{em_p} = \int_0^1 K_{a_1} df(\lambda T) \quad \text{(Eq. 3.39)}
\]

where \( df(\lambda T) \) is external fractional function. The cumulative density function for wavelength is,

\[
R(\lambda) = \int_0^\lambda P(\lambda') d\lambda' = \frac{1}{K_{em_p}} \int_0^{f(\lambda T)} K_{a_1} df(\lambda T) . \quad \text{(Eq. 3.40)}
\]

In this work, wavelength region 3.1-4.95 \( \mu m \) has to be included; therefore, an interval 0.01 divides this wavelength region uniformly. Spectral transmissivity is calculated at each wavelength and integrated to give a total transmissivity at the working wavelength range.

Choose a propagation direction of the bundle

The probability distribution function of bundle propagation direction can be written by assuming azimuthally symmetric scattering,
where $\theta_p$ is out-scattering direction, and $p(\theta_p)$ is phase function. The cumulative density function of angular angle is expressed as,

$$R(\theta_p) = \int_0^{\theta_p} P(\theta_p')d\theta_p'$$  \hspace{1cm} \text{(Eq. 3.42)}$$

The cumulative density function of azimuthal angle with the assumption of azimuthally symmetric scattering is,

$$R(\phi_p) = \frac{\phi_p}{2\pi}$$  \hspace{1cm} \text{(Eq. 3.43)}$$

By generating random number from 0 to 1, propagation direction can be attained. As for $\theta_p$, this simulation uses a random number between 0 to $\pi/2$.

**Choose a path length of the bundle to determine the next action of the bundle**

Assuming extinction coefficient $K_e$ in Eq. 3.25 is not a function of path length $s$, the probability function of path length is,

$$P(s)ds = K_e e^{-K_es} ds$$  \hspace{1cm} \text{(Eq. 3.44)}$$

and cumulative density function is,

$$R(s) = \int_0^s P(s')ds' = 1 - e^{-K_es}$$  \hspace{1cm} \text{(Eq. 3.45)}$$

Once a random number is brought about, path length can be solved from Eq. 3.45,
\[ s = -\frac{1}{K_e} \ln (1 - R) = -\frac{1}{K_e} \ln (R) \] 

(Eq. 3.46)

while \( R \) is at the range 0 to 1, \( 1 - R \) can be replaced by \( R \).

**Decide whether the bundle is absorbed or scattered at the new location**

To decide the bundle is absorbed or scattered, albedo \( w_0 \) in Eq. 3.27 is compared with random number,

\[ R > w_0 \quad \text{(absorbed)} \] 

(Eq. 3.47)

\[ R > w_0 \quad \text{(scattered)} \] 

(Eq. 3.48)

and if the bundle is scattered, a new direction is obtained by comparing cumulative density function of propagation direction in Eq. 3.42 with a random number. Angle of direction at 0 to \( \pi \) is divided by a grid size at 0.008, and the comparisons starts from angle at 0. Once the cumulative density function is larger than the random number, the angle of propagation is determined.

**Other Simulation Condition Settings**

According to the shape the cloud chamber, a cylindrical coordinate system is made in Monte Carlo simulation. Energy bundles are emitted from the area of optical shutter and an imaginary area is set in the simulation to serve as IR camera collecting photons. Due high spectral reflectivity of Al (>0.98) in this working spectral range [26-27], the chamber wall is regarded as a place having all reflection without absorption. In addition, the chamber wall is optical thin, which enhances the rationality of this assumption. The
transmissivity is defined by following the same principle in Eq. 2.5. By counting number of energy bundles \( S \) gathered by IR camera, transmissivity is computed by integrating each number count along the working wavelength range and taking ratio of two difference values, the difference of opened and closed shutter number counts when water clouds exist and the difference of opened and closed shutter number counts when no water clouds in the chamber,

\[
\tau_{MC} = \frac{S_{w,o} - S_{w,c}}{S_{wo,o} - S_{wo,c}} \quad (\text{Eq. 3.49})
\]

A Matlab code for this Monte Carlo simulation on transmissivity is shown in Appendix. A which developed by Wang [20] and modified to fit the conditions in this work. A flowchart for this Monte Carlo simulation is shown in Fig. 3.4.
Input number of bundles $N$ and albedo $w_0$
Initialize transmission counter $S$

Emit all energy bundles $i=1$ to $N$

Initialize emitted bundle location

Choose new direction for diffuse emitter:
$\theta = R_\pi / 2$, $\phi = 2\pi R$, $\phi = -\ln(R) / K$

Decide new location

Hit the wall?

Y

Update location after reflection

N

In the chamber?

N

Detected by IR camera?

N

Absorbed $w_0 < R$

Y

$S = S + 1$

N

Choose new direction after scattering:
$\theta_r = \theta_r (R)$, $\phi = 2\pi R$, $\phi = -\ln(R) / K$

Fig. 3.4 Monte Carlo flowchart for cloud chamber transmissivity simulation. [20]
3.3 Result Discussion

Results of Monte Carlo transmissivity simulation are presented to compare with experimental results at 75 °C. The comparison of transmissivity data at 60 °C in [20] and 75 °C in this work and the prediction of transmissivity at 50 °C are also revealed to show temperature effect of phase transition radiation. The working wavelength range in this work, 3.1-4.95 μm, is applied in many IR remote sensors, which makes research in this spectral range substantial. Assumption of mono-dispersed droplets is made throughout the simulation.

Table 3.4 CCN numbers for different $d^*$'s while reference volume fraction $f_v$ is set at $1.6 \times 10^{-4}$.

<table>
<thead>
<tr>
<th>Reference $f_v$</th>
<th>$d^*$ (μm)</th>
<th>CCN (cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1.6 \times 10^{-4}$</td>
<td>15</td>
<td>9.05×10$^4$</td>
</tr>
<tr>
<td></td>
<td>16</td>
<td>7.46×10$^4$</td>
</tr>
<tr>
<td></td>
<td>17</td>
<td>6.22×10$^4$</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>5.24×10$^4$</td>
</tr>
<tr>
<td></td>
<td>19</td>
<td>4.46×10$^4$</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>3.82×10$^4$</td>
</tr>
<tr>
<td></td>
<td>21</td>
<td>3.30×10$^4$</td>
</tr>
</tbody>
</table>

Experimental and Simulation Results Discussion at 75 °C

The best simulation fit is shown in Fig. 3.5 with the conditions, CCN at $6.22 \times 10^4$ cm$^{-3}$ and probability constant $a_{21}$ at $3 \times 10^{-8}$. To get CCN value in this work, the first step is to choose a reference $f_v$ value at $1.6 \times 10^{-4}$ and then diameter of droplets $d^*$ at this volume fraction is assigned at 17 μm. With $f_v$ and $d^*$, together using Eq. 3.5,
CCN can be deduced, as shown in Table 3.4. The probability constant $a_{si}$ is determined by fitting experimental data at 60 °C shown in next section. The CCN values listed in Table 3.4 are at the order of $10^4 \, cm^{-3}$, which agree with typical CCN values in an environment without industries. [28] Due to the difficulties of measurement of aerosol size composition, an empirical relation between CCN and supersaturation $s\%$ was introduced by Seinfeld and Pandis [28, pp. 792-794] providing a qualitative expression on CCN,

$$CCN = cs^k$$  \hspace{1cm} (Eq. 3.50)

where $c$ and $k$ are the parameters varying from different environments, which can be found in [28, table 17.4]. A comparison of observed CCN in real environment and predicted CCN from the empirical relation was shown in [28, Fig. 17.17]. When supersaturation $s$ was at 0.1%, CCN values in continental environment both from observation and prediction are between $10^1$ and $10^5 \, cm^{-3}$. Therefore, the calculated CCN values in Table 3.4 are acceptable speculations. Besides, CCN is assumed a constant in the cloud chamber during simulations. In each experiment CCN may be different in the lab environment, which influences the size of droplet; however, highly repetitive experimental data suggests that the fluctuation of CCN in the experiments is minor. Consequently, it is reasonable to assume a constant CCN value in Monte Carlo simulation.
Fig. 3.5 Best curve fit of Monte Carlo transmissivity simulation for 75 °C experiment.

Fig. 3.6 Monte Carlo 75 °C transmissivity simulation for different $d^*$ values while fixing $a_{21}$ at $3 \times 10^{-8}$.
Fig. 3.7 Effects from volumetric absorption, scattering and extinction on 75 °C transmissivity results when surface absorption is neglected.

For the same $a_{21}$ value at $3\times10^{-8}$, simulation results at different $d^*$ (CCN) values are shown in Fig. 3.6. It is clear that when $d^*$ increases (CCN number decreases), transmissivity increases. The curve for $d^*$ at 17 $\mu m$ (CCN at $6.22\times10^4$ $cm^{-3}$) comprises two transitions while $f_v$ is at $0.1\times10^{-4}$ and $0.5\times10^{-4}$. When $d^*$ equals to 15 and 16 $\mu m$, the condition of two transitions cannot be maintained. While $d^*$ is equal to and larger than 19 $\mu m$, the parabola-like trend in the range of $f_v=0.1\times10^{-4}$ and $f_v=0.5\times10^{-4}$ is broken. Although the curve of $d^*$ at 18 $\mu m$ shows the characteristics of two transitions and parabola-like trend inside the transition, the simulation result does not match experimental data. Two energy extinction mechanisms of volumetric
contributions, absorption and scattering, are able to check this phenomenon. For absorption, larger CCN would lead to a bigger absorption coefficient resulting in a smaller transmissivity. On the other hand, for scattering, at a fixed $f_v$, larger CCN means smaller droplet radius in Eq. 3.5 contributing to a strong forward scattering. Therefore, higher transmitted energy accompanying a higher transmissivity can be anticipated. The diameter of radius in Fig. 3.5 is $17 \mu m$ and it can be seen in Fig. 3.6 that there is a restriction in assigning $d'$. Simulation results using $d'$ values other than the values in Fig. 3.5 do not maintain the agreement with experimental data.

Two volumetric contributions of energy extinction, absorption and scattering, need to be discussed further. Since the aluminum chamber wall provides a high reflectivity boundary, the path length of a photon from IR source has a high probability of becoming optical thick. Accordingly, multiple scattering is expected to happen in the cloud chamber, while the assumption of single scattering for optical thin condition is not suitable in this work. To know more about the effect of volumetric scattering, volumetric absorption efficiency $Q_{av}$ and surface absorption efficiency $Q_{as}$ are moved out in Eq. 3.24 to let $Q_v = Q_{sv}$. Similarly the condition, $Q_v = Q_{av}$, is achieved to investigate the effect of volumetric absorption. Another condition while surface absorption $Q_{as}$ is neglected, $Q_v = Q_{sv} + Q_{sv}$, is also calculated using the same Monte Carlo transmissivity code. The simulation results of these cases are shown in Fig. 3.7. It can be seen that scattering is the major contribution to energy extinction when $f_v$ is less than $0.2 \times 10^{-4}$, while contribution from absorption is comparing weak due to small
droplet size causing small droplet size parameter $x$. When $f_v$ increases, energy extinction from scattering becomes less because larger droplet has a tendency to contain a behavior of strong forward in-scattering. Absorption contribution increases gradually with respect to increasing droplet size. Even though scattering effect is weaker when $f_v$ increases, contribution from scattering is still not negligible. The curve of the condition, $Q_x = Q_{sv} + Q_{sv}$, seems to contain two transitions but simulation results when $f_v$ is larger than $0.5 \times 10^{-4}$ is a little far away from experimental data.

![Graph](image)

Fig. 3.8 Effects of probability constant $a_{z1}$ in surface absorption on 75 °C transmissivity results.
Table 3.5 droplet diameter $d$, relaxation time $A_{21}^{-1}$, and binary collision rate $Z_{\text{coll}}$ for CCN=$5.24\times10^4$ $cm^{-3}$ and $a_{21}=$$3\times10^{-8}$ at $f_i=1.6\times10^{-4}$.

<table>
<thead>
<tr>
<th>$f_i$($\times10^{-4}$)</th>
<th>$d$($\mu m$)</th>
<th>$A_{21}^{-1}$ (sec)</th>
<th>$Z_{\text{coll}}$ ($cm^{-3}$ sec$^{-1}$)</th>
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</thead>
<tbody>
<tr>
<td>0.01</td>
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</tr>
<tr>
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</tr>
<tr>
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<tr>
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</tr>
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<td>$3.37\times10^{22}$</td>
</tr>
<tr>
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<td>6404</td>
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</tr>
<tr>
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<td>18.00</td>
<td>3291</td>
<td>$8.20\times10^{22}$</td>
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<tr>
<td>2.0</td>
<td>18.31</td>
<td>3181</td>
<td>$8.49\times10^{22}$</td>
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</table>
Fig. 3.9 Monte Carlo transmissivity simulation results when excluding surface absorption ($a_{21} = 0$).

Fig. 3.10 Comparison of $Q_{av}$ and $Q_{tot}$ at $f_v = 0.1 \times 10^{-4}$ in the working spectral region, 3.1-4.95 $\mu m$. 
Fig. 3.11 Comparison of $Q_{av}$ and $Q_{as}$ at $f_i = 2 \times 10^{-4}$ in the working spectral region, 3.1-4.95 $\mu$m.

Surface absorption due to phase transition is another key contribution to energy extinction affecting transmissivity. While Einstein coefficient for spontaneous emission $A_{21}$ [s$^{-1}$] describes the probability of spontaneous emission from excited state, the inverse of $A_{21}$ [s] represents the relaxation time of excited state (water vapor in this work). For a fixed CCN value, of transmissivity simulation at different probability constant $a_{21}$ are shown in Fig. 3.8. It can be detected that energy absorption is strengthened by this surface absorption. Comparing the result with zero surface absorption and curves with none-zero surface absorption, it is clear that surface absorption is stronger as $f_i$ increases. The condition, $a_{21} = 3 \times 10^{-8}$, is able to capture two transitions of transmissivity data at $f_i = 0.1 \times 10^{-4}$ and $f_i = 0.5 \times 10^{-4}$. When $a_{21}$ is set
to be larger than $1 \times 10^{-7}$, the physical meaning of $Q_{sa}$ cannot be maintained. In Eq. 3.14, surface absorptivity $f_s$ is approximated by single surface absorption efficiency $Q_{sa}$ and $a_{21}$ is proportional to $Q_{sa}$. Therefore, too large $a_{21}$ leads to a high efficiency $Q_{sa}$, which destroys its physical meaning. As CCN is chosen at $6.22 \times 10^4$ while $f_s$ is at $1.6 \times 10^{-8}$ and $a_{21}$ is at $3 \times 10^{-8}$ in this study, the corresponding diameter of droplet $d$, relaxation time $A_{21}^{-1}$, and binary collision rate $Z_{coll}$, for different $f_v$ are presented in Table 3.5. Comparing binary collision rate with number density of water vapor molecules $n_v = 8.10 \times 10^{18}$ at 75 °C saturation condition, binary collision rate $Z_{coll}$ is $6.04 \times 10^{22}$ when $f_v$ is at $1.2 \times 10^{-4}$. Lower temperature or vapor pressure leads to decreasing binary collision rate if the condition of water condensation is maintained. The condition $a_{21} = 0$ symbolizes the condition of neglecting phase transition absorption and the traditional theory cannot yield satisfying results by only changing CCN or assigning $d^*$ values. Fig. 3.9 shows various transmissivity curves for different $d^*$s when $a_{21} = 0$. It is clear that when surface absorption is excluded in Monte Carlo simulation, none simulation results can agree with experimental data no matter what CCN ($d^*$) is chosen. This analysis is a proof of the existence of phase transition radiation of water. The importance of surface absorption is emphasizing by comparing surface absorption to volumetric absorption. For two limiting volume fractions, $f_v = 0.1 \times 10^{-4}$ and $f_v = 2 \times 10^{-4}$, the comparisons of volumetric absorption efficiency $Q_{av}$ and surface absorption efficiency $Q_{sa}$ are shown in Fig. 3.10 and Fig. 3.11. At the
wavelength $\lambda = 4 \ \mu m$, the absorption index $k$ is small leading to a relative small $Q_{av}$ where $Q_{av}$ is significant. On the other hand, at the wavelength $\lambda = 3.1 \ \mu m$, the effect $Q_{av}$ of is shadowed by $Q_{av}$ where $Q_{av}$ is one order magnitude larger than $Q_{av}$.

**Comparison of Transmissivity Data at 60 °C and 75 °C**

Two experimental transmissivity measurements for 60 °C [20] and 75 °C are shown in Fig. 3.12. In 60 °C data, there are two transitions at volume fraction $0.15 \times 10^{-4}$ and $0.5 \times 10^{-4}$. The first transition stands for the occurrence of water cloud inside the chamber. When volume fraction is at the range, $0.15 \times 10^{-4} \leq f_v \leq 0.5 \times 10^{-4}$, transmissivity maintains nearly a constant. For $f_v \geq 0.5 \times 10^{-4}$, transmissivity decreases deliberately. In 75 °C data, two transitions can be observed at volume fraction $0.1 \times 10^{-4}$ and $0.5 \times 10^{-4}$, but the behavior of 75 °C data is different at the range, $0.1 \times 10^{-4} \leq f_v \leq 0.5 \times 10^{-4}$. Instead of a constant transmissivity, a parabola-like transmissivity change is shown at this range. For $f_v \geq 0.5 \times 10^{-4}$, $\tau$ is not as sensible as in the range, $0.1 \times 10^{-4} \leq f_v \leq 0.5 \times 10^{-4}$, and gradual decrease is found in 75 °C data as in 60 °C data. Multiple surface absorption effect was not included in 60 °C simulation. Therefore, in order to demonstrate the consistency of multi-surface effect, simulation is performed by changing some simulated conditions. It is discovered that slightly decrease on $a_z$ to $3 \times 10^{-8}$ while other parameters were remained unchanged can support a fit as good as in [20], shown in Fig. 3.13.
Fig. 3.12 Experimental results for transmissivity measurement at 60 °C [20] and 75 °C

Fig. 3.13 Best simulation fit for 60 °C [20] transmissivity experiment while multiple surface absorption is added.
Another clue to understand the difference of transmissivity measurement between 60 °C and 75 °C is to investigate the two mechanisms of energy extinction. Extinction and absorption coefficients from volumetric and surface contributions are integrated along 3.1-4.95 μm wavelength to provide total-sense coefficients at each $f_i$. Surface absorption coefficient $K_{as}$, volumetric absorption coefficient $K_{av}$, volumetric extinction coefficient $K_{ev}$, and combined extinction coefficient $K_{e}$ from volumetric and surface contributions for 60 °C and 75 °C at different $f_i$ are shown in Fig. 3.14. It
is found that coefficients of 75 °C are always larger than coefficients of 60 °C. Since CCN at 75 °C is bigger than CCN at 60 °C, the droplet diameter at 75 °C is smaller than at 60 °C for a fixed $f_v$. In Eq. 3.4, both larger CCN and smaller droplet diameter make coefficients increase. Fig. 3.15 shows efficiencies, $Q_{as}$, $Q_{av}$, $Q_{ev}$, and $Q_e$ at each $f_v$ in this work. Efficiencies of 75 °C are not always larger than efficiencies of 60 °C due to the effects of optical constants and droplet size parameter. However, it can be identified that CCN and droplet diameter play key roles in explaining transmissivity variation at different temperatures.

Fig. 3.15 Integration of absorption and extinction efficiencies along wavelength 3.1-4.95 μm at each $f_v$. (a) Surface absorption efficiency $Q_{as}$. (b) Volumetric absorption efficiency $Q_{av}$. (c) Volumetric extinction efficiency $Q_{ev}$. (d) Combined extinction efficiency $Q_e$. 

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Fig. 3.16 Transmissivity Results of Monte Carlo simulation at 50 °C, 60 °C, and 75 °C with CCN at $2.22 \times 10^4$, $3.82 \times 10^4$, and $6.22 \times 10^4$ $cm^{-3}$ respectively.

**Predictions of Transmissivity Data at 50 °C**

Utilizing the ideas and information in this work, transmissivity at 50 °C can be predicted. Although the actual CCN value at 50 °C is unknown, the analysis of CCN at 60 °C and 75 °C still offers a concept to CCN at 50 °C. While probability constant $a_{21}$ remains the same, CCN at 50 °C is $2.22 \times 10^4$ using linear extrapolation from 60 °C and 75 °C. The prediction of transmissivity at 50 °C along with simulation results at 60 °C and 75 °C are shown in Fig. 3.16. Fig 3.16 also includes transmissivity at
different temperatures while surface absorption is neglected \(a_{21} = 0\). It can be found generally surface absorption has greater effect at higher temperature and at higher volume fraction of liquid water. Transmissivity at 50 \(^\circ\)C seems to have two transitions at 0.1\(\times\)10\(^{-4}\) and 0.3\(\times\)10\(^{-4}\). Inside the transition, transmissivity approaches a constant as 60 \(^\circ\)C transmissivity. When \(f_v\) is larger than 0.3\(\times\)10\(^{-4}\), transmissivity decreases faster than data at 60 \(^\circ\)C and 75 \(^\circ\)C. It can be concluded that at low temperature, the transition range shrinks and transmissivity goes up mainly due to lower CCN values. The major temperature effect comes from different CCN values while probability constant \(a_{21}\) is regarded as an independent constant. Once precise CCN values are given at different temperatures, the extinction coefficient and other knowledge in this work are expected to apply well in a wide temperature range.
Chapter 4 Conclusion and Future Work

Transmissivity measurement of IR radiation passing through a 75 °C cloud chamber is performed. Monte Carlo simulation of the same transmissivity test is made to compare with experimental data and understand the temperature effect of radiation from water phase transition. Homogeneous water cloud is generated inside the chamber steadily to provide a condition of liquid-vapor mixture. By comparing measured photon counts from the IR camera, it is found that photon counts with cloud inside the chamber are less than photon counts without water cloud in the chamber. This phenomenon suggests that energy extinction happens when there is water cloud in the chamber.

Experimental transmissivity data at 75 °C shows two transitions when \( f_v = 0.1 \times 10^{-4} \) and \( f_v = 5 \times 10^{-4} \). To make the knowledge of phase transition radiation of water more complete, multiple surface absorption of a droplet is introduced. Utilizing approximations of radiative scattering theory, Monte Carlo transmissivity simulation matches experimental data well with the conditions, \( CCN = 6.22 \times 10^4 \text{ cm}^{-3} \) and \( a_{21} \) at \( 3 \times 10^{-8} \) while in thermodynamic equilibrium. CCN value in simulations is assumed a constant at the order of \( 10^4 \text{ cm}^{-3} \), which agrees with the observed CCN in urban environment without industrial pollution. Two energy extinction mechanisms, volumetric and surface extinction, are examined individually. The transmissivity simulation of two volumetric components, absorption and scattering, are shown at each \( f_v \). The effect of the only surface component, absorption, is inspected to demonstrate
the existence of phase transition of water. Comparison of experimental data at 60 °C and 75 °C recommends the CCN or droplet diameter at a specific $f_v$ is the key factor to the transmissivity difference. Coefficients and efficiency of extinction and absorption are analyzed to obtain an understanding of temperature effect of phase transition radiation of water. Predictions of transmissivity at 50 °C is presented to support the importance of temperature effect of phase transition radiation of water.

Although several knowledge on phase transition radiation of water has been revealed, much work can be done to acquire detailed comprehension of this issue.

1. Transmissivity experiment at other spectral regions or a monochromatic wavelength.
2. Measurements of CCN and volume fraction of water liquid $f_v$.
3. IR emission tests of water cloud.

It is useful to obtain transmissivity data at different spectral ranges such as visible range or microwave range where many remote sensors are applied. Experiment at a monochromatic wavelength produce a profound knowledge, since measurement in 3.1-4.95 $\mu m$ is an integrated data acquirement in this work. CCN and $f_v$ measurements can offer a precise simulation condition and data processing during experiments. Direct emission test of water cloud will demonstrate the existence of phase transition radiation of water by comparing the emission intensity to blackbody intensity. It is anticipated that emission intensity is bigger than blackbody intensity whenever phase transitions take place. While this kind of phase transition radiation is firm and accepted by
scientists and engineers, a better weather prediction with such radiation including can be expected in the future.
References


Appendix A Monte Carlo Simulation Code

**MCsim.m**

clear % need to convert the units of wavelength and droplet size
delete('MCtransmissivity.dat') % from [micron m] to [cm]
format short e

% reading bulk water optical constants
load wavelength.dat
load abscoef75C.dat
load refcoef75C.dat

% reading the pre-calculated reference value for intensity difference
load MC_B12_diff75C.dat

% reading the pre-calculated poly-fit coefficients for H/B at 75 degrees C
load CoeffH_B75C.dat

% reading droplet size [micron m] at fv0, fv~d^3
fv0=1.6E-4;
d0=17;
m0=0; %m0=input('enter initial fv [m0*1E-5] in terms of m0 (such as 0 or 10):');
n_nc=fv0/(pi*(d0*1E-4)^3/6); % number density of nucleation sites, #/cm^3
reflonoff=1; %reflonoff=input('reflection calculation on/off (off= 0; on= non-zero)');
refltrace=0; %refltrace=input('reflection tracing on/off (off= 0; on= non-0)');

% the number of energy bundles at T=T1 and wavelength=3.1 micron
N0=8000; %N0=input('enter number of bundles at T=T1 and wavelength=3.1 micron (reference value of m is 4000)');

% path length=41cm=s=xa, lens diameter=2.54cm
% object diameter=0.95cm, distance btn chamber and lens=20cm=xb
% units in [cm]
s=41;
xa=41;
xb=20;
dch=10.16;
dshutter=3.175;
dlens=2.54;
% dobj=0.95;
rch=dch/2;
rshutter=dshutter/2;
rlens=d1ens/2;
% robj=dobj/2;
l=[3.1:0.01:4.95]; % working wavelength of IR camera: 3.1~4.95 micron
i_l=(4.95-3.1)/0.01+1; % grid size=0.01 micron
T1=300; % IR source T when shutter is closed
T2=378; % IR source T when shutter is open
Tg=273.15+75; % medium temperature @ 75 C
ng=8.10e18; % the number density of vapor molecules [#/cm^3] @ 75 C
h=6.626*10^-34; % Planck's constant [J-s]
c=3*10^10; % [cm/s]
kb=1.38E-16; % [erg/K]
mh2o=18/(6.02E23); % water molecular mass [g/molecule]
a21=3e-8; % trial value for a21 [-/-]
%a21=0; % limiting case
n1=1.0003; % for air
k1=0;

% data record
aaa=sprintf('%s%d', 'd', d0, 'N', N0, 'a', a21, '.txt'); % new
fid = fopen(aaa, 'w'); % new

% theta for phase function calculation
pi=3.1416;
theta=[0:0.0008:pi];
i_theta=(pi-0)/0.0008+1; % grid size=0.0008 rad
i_theta=round(i_theta);

% intensity calculations for the blackbody-like IR signal source
C1=37413;
C2=14388;
B1=C1./(l.^5.*(exp(C2./(l*T1)))-1))/pi; % B1=B(T1), optical shutter closed
B2=C1./(l.^5.*(exp(C2./(l*T2)))-1))/pi; % B2=B(T2), optical shutter open

% converting the unit of spectral intensity [W/cm^2-micron]
% to [# of photons/cm^3-s]
B1=B1./(h*c./l);
B2=B2./(h*c./l);
e0=B1(i_l)/N0; % photonic energy per bundle
N01=B1/e0; % #'s of bundles for different wavelengths
N01=round(N01);

n=zeros(i_l, i_l); % bulk water refractive coefficient
n=zeros(i_l, i_l); % bulk water refractive coefficient

for i=1:i_l
    k(i)=interp1(wavelength,abscoef75C,l(i));
    n(i)=interp1(wavelength,refcoef75C,l(i));

    % droplet surface reflection
    thetar=0:pi/1000:pi/2; % incident angle
\[
u = \sqrt{0.5 \cdot ((n(i)^2 - k(i)^2) - n1^2 \cdot (\sin(\theta_r))^2) + \sqrt{(n(i)^2 - k(i)^2) - n1^2 \cdot (\sin(\theta_r))^2 + 4 \cdot n(i)^2 \cdot k(i)^2)}}; \\
v = \sqrt{0.5 \cdot ((-1) \cdot (n(i)^2 - k(i)^2) - n1^2 \cdot (\sin(\theta_r))^2) + \sqrt{(n(i)^2 - k(i)^2) - n1^2 \cdot (\sin(\theta_r))^2 + 4 \cdot n(i)^2 \cdot k(i)^2)}}; \\
R_{perp} = ((n1 \cdot \cos(\theta_r) - u)^2 + v^2) / ((n1 \cdot \cos(\theta_r) + u)^2 + v^2); \\
R_{para} = (((n(i)^2 - k(i)^2) \cdot \cos(\theta_r) - n1 \cdot u)^2 + (2 \cdot n(i) \cdot k(i) \cdot \cos(\theta_r) + n1 \cdot v)^2) / ((n(i)^2 - k(i)^2) \cdot \cos(\theta_r) + n1 \cdot u)^2 + (2 \cdot n(i) \cdot k(i) \cdot \cos(\theta_r) + n1 \cdot v)^2; \\
R_{dir\_spec} = (R_{perp} + R_{para}) / 2; \\
R_{spec}(i) = 2 \cdot \text{trapz}(\theta_r, R_{dir\_spec}); \\
\text{\% integrate along wavelength}
\]

end

phb=CoeffH_B75C; % Poly-fit coefficients for H/B

% Monte Carlo transmissivity calculation--------------------------

% Monte Carlo transmissivity calculation--------------------------

temprans=MC_B12_diff75C; % pre-calculated relative radiation strength
at m=0;
temp1=phb(1)*l.^6+phb(2)*l.^5+phb(3)*l.^4;
temp2=phb(4)*l.^3+phb(5)*l.^2+phb(6)*l+phb(7);
fitH_B=temp1+temp2;
Ka_cond0=(a21/(4*pi))*(fitH_B.*(1E4*h*c./l))*ng ... 
* n_nc*sqrt((8*pi*kb*Tg/mh2o) 
% converting B[W/cm^2] to H/B to (() photons/sec)/cm^2]; T=75 C 
% Ka_cond=Ka_cond0*(d/2)^2 to appear in later calculations 
f=((a21/4*pi))*(fitH_B.*(1E4*h*c./l))*ng ... %used for multiple 
surface 
*sqrt((8*pi*kb*Tg/mh2o) 

clear temp1 temp2 fitH_B 

for m=m0:20 
fv=m*1E-5+1E-8; 

% assuming n_nucleation_site=const: fv~d^3, 
d=d0*(fv/fv0)^(1/3); 

% extinction and absorption efficiencies, Qe and Qa, from anomalous 
diffraction 
% Rayleigh-Gans scattering for phase function 
% calculating Qe,a,s and Ke,a,s 
% size parameter x 

x=pi*d./l; 
ro=2*x.*(n-1); 

% parameters for the modified A.D. 

omega=2*x.*k+j.*ro; 
m_complex=n-j.*k;
alpha_mdf=0.5+((n-1)-k.^0.5*2/3-k/2)+((n-1)+(k.^0.5-5*k)*2/3).^2;
gamma_mdf=(3/5-(n-1).^0.5*3/4+3*(n-1).^4)+5./(6/5+(n-1)./k);
mu=alpha_mdf+gamma_mdf./x;

% parameters for A.D.
beta=atan(k./(n-1));
u=cos(beta)./ro;
v=4*x.*k;
Qe_adiff=2+4*u.^2.*cos(2*beta)-4*exp(-ro.*tan(beta)).*(u.*sin(ro ...
-Q beta)+u.^2.*cos(ro-2*beta));
Qa_adiff=1+(2./v).*exp(-v)-(2./(v.^2)).*(1-exp(-v));

% modified anomalous diffraction
Qv=real(2+4.*exp(-omega)./omega+4*(exp(-omega)-1)./(omega.^2));
T=2-exp(-x.^(-2/3)); % correction to x>>1, G.O. limit
Qext=T.*Qv;
Qe_adiff=Qe_adiff; % using modified A.D. for Qe while keeping Qa unchanged

% absorption test
%    Qe_adiff=Qa_adiff;
Qs_adiff=Qe_adiff-Qa_adiff;

% scattering test
%    Qe_adiff=Qs_adiff;
Ke_adiff=1.5*Qe_adiff.*f_v/(d*1E-4);
Ka_cond=Ka_cond0*(d*1E-4/2).^2;

% consider second surface
R=R_spec;
Qa_cond=fs.*(1+(1-R-fs).*(1-Qa_adiff));
Ka_cond=Ka_cond.*(1+(1-R-fs).*(1-Qa_adiff)./(1-(1-Qa_adiff).*(R)));%

% evaporative absorption shadowing effect test
%    Qe_adiff
%    pause
%    Qa_cond=Ka_cond/n_nc/(pi*(d*1E-4/2).^2);
%    Shadow_e=Qa_cond./Qe_adiff;
%    Shadow_e
%    pause

A21_value=a21*n_nc*(d*1E-4/2).^2*sqrt(8*pi*kb*Tg/mh2o);
% bi_coll_rate=A21_value/a21*ng; % binary collision rate
% bi_coll_rate
% condensation radiation test
Ke_adiff=Ke_adiff+Ka_cond;
Ks_adiff=1.5*Qs_adiff.*fv/(d*1E-4);
clear ro beta u v Qe_adiff Qa_adiff Qs_adiff

% surface absorption test
% Ke_adiff=Ka_cond;
% Ks_adiff=0*Ks_adiff;

% calculating phase function, $p(\theta)$
arg=zeros(i_theta,i_l);
g_arg=zeros(i_theta,i_l);
phase=zeros(i_theta,i_l);
temp_=zeros(i_theta,i_l);
for i=1:i_l
    arg(:,i)=2*x(i).*sin(theta/2);
    g_arg(:,i)=3*(sin(arg(:,i))-arg(:,i).*cos(arg(:,i)))+1E-8./...
    (arg(:,i).^3+3E-8);
    phase(:,i)=g_arg(:,i).^2.*(1+(cos(theta)).^2);
end

% replacing the numerical singularity of phase function at $\theta=0$
% by an adjacent value
    phase(1,:)=phase(2,:);

% updating phase function
const=zeros(1,i_l);
R_theta=zeros(i_theta,i_l);
for i=1:i_l
    const(i)=2./trapz(theta,temp_(:,i));
    phase(:,i)=phase(:,i)*const(i);
end

% updating cumulative probability function for phase function, $R_\theta$, % in terms of polar angle, $\theta$, and wavelength, $l$
temp__=0;
for j0=1:i_theta
    temp__=temp__+(0.5*phase(j0,i)*sin(theta(j0))*(pi/(i_theta-1)));
end
R_theta(j0,i)=temp__;
clear temp__;
end
clear const temp_arg g_arg phase
\texttt{N02=\texttt{zeros}(1,i\_l);}
\texttt{S\_t1=\texttt{zeros}(1,i\_l);}
\texttt{S\_t2=\texttt{zeros}(1,i\_l);}

\texttt{\textbf{for }i=1:i\_l}

\% optical depth of cloud chamber at wavelength lambda, \( t_c \)
\texttt{\textit{t\_c=Ke\_adiff(i)\*xa;}}

\% albedo at wavelength lambda, \( W_0 \)
\texttt{\textit{W0=Ks\_adiff(i)/Ke\_adiff(i);}}

\% initialization of bundle counters
\texttt{S\_t=0;}
\texttt{S\_tJ0=0;}
\texttt{S\_tJ1=0;}
\texttt{S\_tJ2=0;}
\texttt{S\_e=0;}
\texttt{S\_eNeg=0;}
\texttt{S\_eJ0=0;}
\texttt{S\_eJ1=0;}
\texttt{S\_eJ2=0;}
\texttt{S\_a=0;}
\texttt{S\_eRefl=0;}
\texttt{badno=0;}

\texttt{\textbf{for }ii=1:N01(i)}
\texttt{jj=0;}
\texttt{t\_l=0;}
\texttt{tempi=rand;}
\texttt{\textit{thetai=tempi\*pi/2;}}
\texttt{\% thetai=\pi/180/10; \% pseudo-parallel light}
\texttt{\textit{oldmu=cos(thetai);}}
\texttt{\% old mu: not accounting for reflection yet}
\texttt{mu=cos(thetai); \% new mu: for reflection calculation}
\texttt{temprl1=rand;}
\texttt{rl1=temprl1\*rshutter;}
\texttt{ph1=0;}
\texttt{x1=0;}
\texttt{thetal1=0;}
\texttt{tempt\_l=rand;}
\texttt{\% temporary \texttt{O1(rl1,ph1,x1)} for ray tracing: \texttt{O1} -> \texttt{O2}}
\texttt{temprl1=rl1;}
\texttt{tempphi1=ph1;}
\texttt{tempx1=x1;}

\texttt{\% letting the photon leave the cloud chamber for \( m=0 \)}
\texttt{\textbf{if }m==0}
\texttt{\textit{tempt\_l=exp(-2\*t\_c/oldmu); \% old mu: not including refl yet}}
clear tempi temp_r1
% re-setting reflection counter for the new energy bundle
reflcount=0;

% tracing an energy bundle
if refltrace~=0
    [X Y Z]=pol2cart(phi1,r1/rch,x1/rch);
    xyz=[X Y Z phi1 r1/rch];
    save MCrefl.dat xyz -ascii -append
    clear xyz X Y Z
end

% the initial polar angle and azimuthal angle
theta1=thetai;
S12=-log(tempt_l)/Ke_adiff(i);
x2=x1+S12*oldmu; % old mu: not accounting for
reflection yet
temp=rand;
phi1_=2*pi*temp;
oldphi1_=phi1_;

% the coordinates of photon where the first scattering takes place
if oldphi1_>pi
    phi1_=2*pi-phi1_;
end
r2=(r1^2+S12^2*(sin(theta1))^2
    -2*r1*S12*sin(theta1)*cos(phi1_))^.5;
tempr2=r2;
alpha=acos((r1^2+r2^2
    -(S12*sin(theta1))^2)/(2*r1*r2));
if oldphi1_<pi
    alpha=abs(alpha);
else
    alpha=-abs(alpha);
end
phi2=phi1+alpha;
clear thetai alpha

% new coordinates if reflection occurs before the first scattering
if reflonoff~=0
if r2>rch
    if oldphi1_<pi
        reflcount=1;
    else
        reflcount=-1;
    end
    clear oldphi1_
    beta_=asin(r1*sin(phi1_)/rch);
end
alpha_ = pi - phi1_ - beta;
dra = rch * sin(alpha_) / sin(phi1_);
dsa = dra / sin(theta1);
dsb = S12 - dsa;

% tracing an energy bundle
reflx = x1 + (S12 - dsb) * cos(theta1);

if reflx >= xa % leaving the cloud chamber
    r2 = 0.1 * rch; % passing the while loop for
reflection iteration
else
    drb = dsb * sin(theta1);
    r2 = (drb^2 + rch^2 - 2 * drb * rch * cos(beta_))^0.5;
    temp2r = r2;
    alpha0 = acos((r2^2 + rch^2 - drb^2)/(2 * r2 * rch));

    if reflcount < 0
        alpha_ = -alpha_;
        alpha0 = -alpha0;
    end
    phi2 = phi1 + alpha_ + alpha0;
    reflr = rch;
    reflphi = phi2 - alpha0;
    tempri = reflr; % updating temporary O1 if
reflection occurs
    tempphi = reflphi;
    tempx1 = reflx;

    if (isreal(r2) == 0) || (isnan(r2) == 1)
        badno = badno + 1;
        S_e = S_e + 1;
        clear beta_ alpha_ dra dsa dsb drb
        clear alpha_ alpha0
        r2 = 0.1 * rch; % passing the while loop for
reflection iteration
    end
    t_l = 1.1 * t_c; % passing the while loop of
    t_l to the next ii
end
end

while r2 > rch
    if refltrace == 0
        [X Y Z] = pol2cart(reflphi, reflr/rch, reflx/rch);
        xyz = [X Y Z reflphi reflr/rch];
        save MCrefl.dat xyz -ascii -append
        clear xyz X Y Z
    end
    clear reflr reflphi reflx
end

phil = beta;
alpha_ = pi - phi1 - beta
  dra = rch * sin(alpha_) / sin(phi1_)
  dsa = dra / sin(theta1);
  dsb = dsb - dsa;

% tracing an energy bundle
  reflx = x1 + (S12 - dsb) * cos(theta1);

  if reflx >= xa % outside the cloud chamber
    r2 = 0.1 * rch; % jumping out of the while loop
  else
    drb = dsb * sin(theta1);
    r2 = (drb^2 + rch^2 - 2 * drb * rch * cos(betah))^0.5;
    tempr2 = r2;
    phi2 = phi2 - alpha0;
    alpha0 = acos((r2^2 + rch^2 - drb^2) / (2 * r2 * rch));

    if reflcount < 0
      alpha_ = -alpha_;
      alpha0 = -alpha0;
      reflcount = reflcount - 1;
    else
      reflcount = reflcount + 1;
    end

    if abs(reflcount) > 10000
      S_e = S_e + 1;
      badno = badno + 1;
      t_l = 1.1 * t_c; % passing the while loop of t_l to the next i
    break
  end

  phi2 = phi2 + alpha_ + alpha0;
  reflr = rch;
  reflphi = phi2 - alpha0;
  tempr1 = reflr; % updating temporary O1 if reflection occurs
  tempphi1 = reflphi;
  tempx1 = reflx;
end

r2 = tempr2; % retrieving r2
clear beta_ alpha_ dra dsa dsb drb alpha0 tempr2
clear reflr reflphi reflx

% updating S12 and the corresponding polar angle for scattering after reflection
S12 = ((r1 * cos(phi1) - r2 * cos(phi2))^2 + (r1 * sin(phi1) - r2 * sin(phi2))^2 + (x1 - x2)^2)^0.5;
theta1 = acos((x2 - x1) / S12);
\( \mu = \cos(\theta_1); \) \% new \( \mu \): reflection already accounted for

end

else
    \( S_e = S_e + 1; \)
    \( S_{eRefl} = S_{eRefl} + 1; \)
end

% passing the while loop of \( t_l \) to the next ii

end

\% tracing an energy bundle
if \( \text{refltrace} = 0 \)
    \[ X \ Y \ Z \] = pol2cart(\( \phi_2, r_2/\text{rch}, x_2/\text{rch} \));
    \text{xyz} = [\( X \ Y \ Z \ \phi_2 \ r_2/\text{rch} \]);
    save \text{MCrefl.dat} \text{xyz} -ascii -append
    clear \text{xyz} \text{X} \text{Y} \text{Z}
end

% iteration loops for energy bundles in the cloud chamber
while \( t_l < t_c \)
    \( t_l = t_l - \mu \log(\text{tempt}_l); \) \% old \( \mu \): as if no reflection
end

% leaving the cloud chamber, on the IR source side
if \( t_l < 0 \)
    \( S_e = S_e + 1; \)
    \( S_{eNeg} = S_{eNeg} + 1; \)
break

% leaving the cloud chamber, on the IR camera side
elseif \( t_l \geq t_c \)

% using temporary O1 for ray tracing purpose
\% vector \( O_{02} = O_{12} \times (x_2-x_1) \)
\% vector \( O_{b2} = O_{12} \times (x_2-(x_a+x_b))/(x_2-x_1) \)
\( r_0 \cos \phi_0 = r_2 \cos \phi_2 - (r_2 \cos \phi_2 - \text{tempr1} \ldots \) *cos(tempphi1)) \times x_2/(x_2-\text{tempxl});
\( r_0 \sin \phi_0 = r_2 \sin \phi_2 - (r_2 \sin \phi_2 - \text{tempr1} \ldots \) *sin(tempphi1)) \times x_2/(x_2-\text{tempxl});
\( r_0 = (r_0 \cos \phi_0^2 + r_0 \sin \phi_0^2)^{0.5}; \)
\( r_{bcosphi} = r_2 \cos \phi_2 - (r_2 \cos \phi_2 - \text{tempr1} \ldots \) *cos(tempphi1)) \times (x_2-(x_a+x_b))/(x_2-\text{tempxl});
\( r_{bsinphi} = r_2 \sin \phi_2 - (r_2 \sin \phi_2 - \text{tempr1} \ldots \) *sin(tempphi1)) \times (x_2-(x_a+x_b))/(x_2-\text{tempxl});
\( r_b = (r_{bcosphi}^2 + r_{bsinphi}^2)^{0.5}; \)
clear \text{r0cosphil} \text{r0sinphil} \text{rbcosphib} \text{rbsinphib}
clear \text{tempr1 tempphi1 tempxl}

% being detected
if (rb<=rlens) % energy bundles collected by the lens
clear r0 rb
S_t=S_t+1;
if jj==0
    S_tJ0=S_tJ0+1;
elseif jj==1
    S_tJ1=S_tJ1+1;
else
    S_tJ2=S_tJ2+1;
end
break
% leaving the cloud chamber but not entering the collecting region
else
clear r0 rb
S_e=S_e+1;
if jj==0
    S_eJ0=S_eJ0+1;
elseif jj==1
    S_eJ1=S_eJ1+1;
else
    S_eJ2=S_eJ2+1;
end
break
end
% still inside the cloud chamber, i.e. t_l<t_c
else
    temp=rand;
end

% the probability of an energy bundle being absorbed in the cloud chamber
if W0<temp
    S_e=S_e+1;
    S_a=S_a+1;
    break
end
jj=jj+1;

% updating the after-scattering polar angle wrt ray direction
temp_theta=rand;
for j0=1:i_theta
    if R_theta(j0,i)>=temp_theta
        theta2=theta(j0);
        break
    end
end
clear temp_theta
tempt_l=rand;
S23=-log(tempt_l)/Ke_adiff(i);

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% calculation after-scattering coordinates as if no reflection

\begin{verbatim}
temp = rand;
phi2_ = 2*pi*temp;

% updating 0c
rcsphic = r2*sin(phi2) + (r2*sin(phi2) - r1 ... *sin(phi1))*(S23*cos(theta2)/S12);
rccosphic = r2*cos(phi2) + (r2*cos(phi2) - r1 ... *cos(phi1))*(S23*cos(theta2)/S12);
xc = x2 + (x2-x1)*(S23*cos(theta2)/S12);

% updating 03_
rc3sphic = r2*sin(phi2) + (S23*sin(theta2)) ... ^2/((r2*cos(phi2) - r1*cos(phi1)) ... )^2))^0.5;
r3coshphic = r2*cos(phi2) - r1*cos(phi1); 

% updating 03
x3 = xc + S23*sin(theta2)*sin(phi2_)*sin(theta1);
a1 = rccosphic - r2*cos(phi2);
b1 = rcsphic - r2*sin(phi2);
c1 = -(x3-xc)*(xc-x2);
a2 = r3_coshphic - rccosphic;
b2 = r3_sphic - rcsphic;
c2 = (S23*sin(theta2))^2*cos(phi2_);
AA = [a1 b1 a2 b2]; 
CC = [c1; c2];
XX = AA\CC;
r3coshphic = rccosphic + XX(1,1);
r3sinphic = rcsphic + XX(2,1);
r3r = (r3sinphic^2 + r3coshphic^2)^0.5;
tempr3 = r3;
phi3 = asin(r3sinphic/r3);
\end{verbatim}

\begin{verbatim}
if (cos(phi3)*r3coshphic)<0
phi3 = pi - phi3;
end

% updating temporary 01 for ray tracing
	tempr1 = r2;
	tempphi1 = phi2;

tempx1 = x2;

% calculating the after-scattering polar angle wrt axial direction
theta1_ = acos((x3-x2)/S23);
oldmu = cos(theta1_); % old mu: not including reflection yet

\end{verbatim}
if (isreal(r3)==0) || (isnan(r3)==1)
    badno=badno+1;
    S_e=S_e+1;
    break
end

% new coordinates for the previous scattering trip if reflection occurs
if r3>rch

    if reflonoff==0
        S_e=S_e+1;
        S_eRefl=S_eRefl+1;
        break
    end

    reflcount=1;
    alpha_=acos((r2^2+(S23*sin(theta1_))^2 - r3^2)/(2*r2*S23*sin(theta1_)));
    beta_=asin(r2*sin(alpha_)/rch);
    dra=r2*sin(alpha_+beta_)/sin(beta_);
    dsa=dra/sin(theta1_);
    dsb=S23-dsa;

    % tracing an energy bundle
    reflx=x2+(S23-dsb)*cos(theta1_);

    if reflx>=xa % outside the cloud chamber
        r3=0.1*rch; % passing the while loop
    else
        drb=dsb*sin(theta1_);
        r3=(drb^2+rch^2-2*drb*rch*cos(beta_))^(0.5);
        temp_r3=r3;
        alpha0=pi-beta_alpha;
        alpha1=acos((r3^2+rch^2-2*drb^2)/(2*r3*rch));

        if (phi3-phi2)<0
            reflcount=-1;
            alpha1=-alpha1;
            alpha0=-alpha0;
        end
        phi3=phi2+alpha0+alpha1;
        reflr=rch;
        reflphi=phi3-alpha1;
        temp_reflphi=reflphi;
        temp_x1=reflx;

        if (isreal(r3)==0) || (isnan(r3)==1)
            badno=badno+1;
            S_e=S_e+1;
            clear beta_alpha dra dsa dsb drb
        end

end
clear alpha1 alpha0
break
end
end

while r3>rch

if refltrace==0
[X Y Z]=pol2cart(reflphi,reflr/rch,refl/rch);
    xyz=[X Y Z reflphi reflr/rch];
    save MCrefl.dat xyz -ascii -append
    clear xyz X Y Z
end

clear reflr reflphi reflx

if reflcount>0
    reflcount=reflcount+1;
else
    reflcount=reflcount-1;
end

if abs(reflcount)>10000
    S_e=S_e+1;
    badno=badno+1;
    break
end

alpha_=beta_;
dra=rch*sin(alpha_+beta_)/sin(beta_);
dsa=dra/sin(theta1_);
dsb=dsb-dsa;

% tracing an energy bundle
reflx=x2+(S23-dsb)*cos(theta1_);

if reflx>=xa % outside the cloud chamber
    r3=0.1*rch; % jumping out of the while loop
else
    drb=dsb*sin(theta1_);
    r3=(drb^2+rch^2-2*drb*rch ... *cos(beta_))^0.5;
    tempr3=r3;
    phi3=phi3-alpha1;
    alpha0=pi-alpha1-beta_;
    alpha1=acos((r3^2+rch^2-drb^2) ... /
        (2*r3*rch));

    if reflcount<0
        alpha1=-alpha1;
        alpha0=-alpha0;
    end

end

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\[
\phi_3 = \phi_3 + \alpha_0 + \alpha_1; \\
\text{refl} = r_{ch}; \\
\text{reflphi} = \phi_3 - \alpha_1; \\
\text{tempr1} = \text{refl}; \quad \% \text{ updating temporary O1}
\]

if reflection occurs
\[
\text{tempphi1} = \text{reflphi}; \\
\text{tempx1} = \text{refl};
\]
end
end
\[
r_3 = \text{tempr3}; \quad \% \text{ retrieving r3}
\]
clear \[\beta_\alpha_\text{dra} \alpha_0 \alpha_1 \text{tempr3} \]
clear \[\text{refl} \text{reflphi} \text{reflx} \]
\[
\text{if abs(reflcount)} > 10000 \\
\text{break}
\]
end

% updating S23 and associated numbers after reflection
\[
S23 = ((r_3 \cos(\phi_3) - r_2 \cos(\phi_2))^2 + (r_3 \sin(\phi_3) - r_2 \sin(\phi_2))^2 + (x_3 - x_2)^2)^{0.5}; \\
\theta_1_\text{=} \text{acos}((x_3 - x_2)/S23); \\
\mu = \cos(\theta_1_\text{;}) \quad \% \text{ new mu: reflection already accounted for}
\]
end

% tracing an energy bundle
\[
\text{if refltrace} = 0 \\
[X \ Y \ Z] = \text{pol2cart}(\phi_3, r_3/r_{ch}, x_3/r_{ch}); \\
xyz = [X \ Y \ Z \ \phi_3 \ r_3/r_{ch}]; \\
\text{save MCrefl.dat xyz -ascii -append}
\]
clear xyz X Y Z
end

% updating coordinates for scattering calculations
\[
\theta_1 = \theta_1_\text{;} \\
S_{12} = S_{23}; \\
r_1 = r_2; \\
\phi_1 = \phi_2; \\
x_1 = x_2; \\
r_2 = r_3; \\
\phi_2 = \phi_3; \\
x_2 = x_3; \\
clear \[\theta_1 \ S_{23} \ r_3 \ phi_3 \ x_3 \]
\]
end
end
clear \[r_1 \ phi_1 \ x_1 \ r_2 \ phi_2 \ x_2 \ r_3 \ phi_3 \ x_3 \ phi_1_\phi_2_\text{;}
\]
clear \[\theta_1 \ \text{theta2} \ \theta_1_\text{;} \ S_{12} \ S_{23} \ \text{tempt_l} \ \text{temp mu oldmu} \]
t_l
if refltrace~=0
    load MCrefl.dat;
    [X,Y,Z]=cylinder(1,50);

    subplot(1,2,1)
    tempcrh=ones(i_theta,1);
    [X1,Y1]=pol2cart(2*theta,tempcrh);
    [X2,Y2]=pol2cart(2*theta,tempcrh*rshutter/rch);
    Z2=tempcrh-1;
    [X3,Y3]=pol2cart(2*theta,tempcrh*r lens/rch);
    Z3=(xa+xb)/rch+Z2;
    plot (MCrefl(:,1),MCrefl(:,2),'-or',X1,Y1,'b', ... 
         X2,Y2,'g',X3,Y3,'m')
    xlim([-1 1]); ylim([-1 1]);

    subplot(1,2,2)
    temprch=ones(i_theta,1);
    [X1,Y1]=pol2cart(2*theta,temprch);
    [X2,Y2]=pol2cart(2*theta,temprch*rshutter/rch);
    Z2=temprch-1;
    [X3,Y3]=pol2cart(2*theta,temprch*r lens/rch);
    Z3=(xa+xb)/rch+Z2;
    plot (MCrefl(:,1),MCrefl(:,2),'-or',X1,Y1,'b', ... 
         X2,Y2,'g',X3,Y3,'m')
    xlim([-1 1]); ylim([-1 1]);

pause
    clear MCrefl X Y Z temprch X1 Y1 X2 Y2 X3 Y3
    delete MCrefl.dat
end

S_t1(i)=S_t;
N02(i)=N01(i)*B2(i)/B1(i);
S_t2(i)=S_t1(i)*N02(i)/N01(i);
clear t_c W0 S_t S_tJ0 S_tJ1 S_tJ2
clear S_e S_eNeg S_eJ0 S_eJ1 S_eJ2 S_eRef1 badno
end

clear i
i=[1:i_l];

if m==0
    temptrans=(trapz(i,S_t2)-trapz(i,S_t1));
    save MC_B12_diff.dat temptrans -ascii
else

% saving transmissivity data
    transmissivity=(trapz(i,S_t2)-trapz(i,S_t1))/temptrans;
    temp1=[fv transmissivity];
    save MCtransmissivity.dat temp1 -ascii -append
    fprintf(fid, '%d %d
', temp1); %data record
    clear i fv temp1 transmissivity
end
end

fclose(fid); %data record
load MCtransmissivity.dat
fv=MCtransmissivity(:,1);
transmissivity=MCtransmissivity(:,2);

plot (fv,transmissivity), xlabel('volume fraction')...
    ,ylabel('transmissivity')
legend('Monte Carlo Simulation')
xlim ([0 2e-4]); ylim ([0 1]);