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(54) **TECHNIQUES FOR CHARACTERIZING CLOUD CONDENSATION NUCLEL**

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(58) **Field of Search** 356/37, 335, 336; 250/335, 573, 576, 222.2; 73/25.01, 28.01; 374/15

(56) **References Cited**

U.S. PATENT DOCUMENTS

2,915,939 A * 12/1959 Van Luck 73/29.05 X
2,953,686 A * 9/1960 Garrison 356/37 X
3,351,759 A * 11/1967 Rich 73/28.01 X

(List continued on next page.)

FOREIGN PATENT DOCUMENTS

JP 57-42839 * 3/1982 73/865.8

OTHER PUBLICATIONS

Fukuta et al, "The principle of a new horizontal thermal gradient cloud condensation nucleus spectrometer", *Journal de Recherches Atmospheriques*, vol. 13, No. 3, pp. 169-188, Jul.-Sep. 1979.

Fukuta et al, "A Horizontal Thermal Gradient Cloud Condensation Nucleus Spectrometer", *Journal of Applied Meteorology*, vol. 18, No. 10, pp. 1352-1362, Oct. 1979.

Hoppel et al, "Errata", *Journal of Aerosol Science*, vol. 11, No. 4, pp. 421-422, 1980.

Hoppel et al, "A segmented thermal diffusion chamber for continuous measurements of CN", *Journal of Aerosol Science*, vol. 10, No. 4, pp. 369-373, 1979.

Hudson, "An Instantaneous CCN Spectrometer", *Journal of Atmospheric and Oceanic Technology*, vol. 6, No. 6, pp. 1055-1065, Dec. 1989.

Radke et al, "A cloud condensation nucleus spectrometer designed for airborne measurements", *Journal de Recherches Atmospheriques*, vol. 15, No. 3-4, pp. 225-229, Jul.-Dec. 1981.

Hudson et al, "Performance of the continuous flow diffusion chambers", *Journal de Recherches Atmospheriques*, vol. 15, No. 3-4, pp. 321-331, Jul.-Dec. 1981.

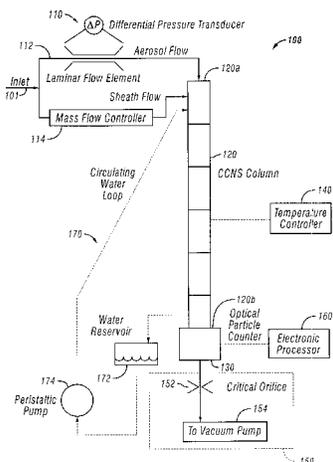
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(57) **ABSTRACT**

A cloud condensation nucleus spectrometer having a stream-wise segmented condensation nucleus growth column. The condensation nucleus growth column includes alternating hot and cold temperature-maintaining segments arranged next to one another. The temperature difference between adjacent hot and cold temperature-maintaining segments increases from the input opening to an output opening of the condensation nucleus growth column to produce a supersaturation distribution that increases from the input opening to the output opening.

4 Claims, 4 Drawing Sheets



U.S. PATENT DOCUMENTS

3,462,609	A	*	8/1969	Beattie	356/37	X	4,967,187	A	*	10/1990	Dumas et al.	73/863.01	X
3,589,169	A	*	6/1971	La Fitte et al.	73/25.01		5,026,155	A	*	6/1991	Ockovin et al.	356/37	
3,597,084	A	*	8/1971	Pagano	73/23.2	X	5,098,657	A	*	3/1992	Blackford et al.	356/37	X
3,651,686	A	*	3/1972	Dizio	73/25.01		5,239,356	A	*	8/1993	Höllander et al.	356/37	
4,790,650	A	*	12/1988	Keady	73/28.01	X	5,278,626	A	*	1/1994	Poole et al.	356/37	X
4,902,311	A	*	2/1990	Dingfors et al.	55/60		5,659,388	A	*	8/1997	Scheer et al.	356/37	
4,967,095	A	*	10/1990	Berger et al.	250/222.2	X	5,903,338	A	*	5/1999	Mavliev et al.	356/37	X

* cited by examiner

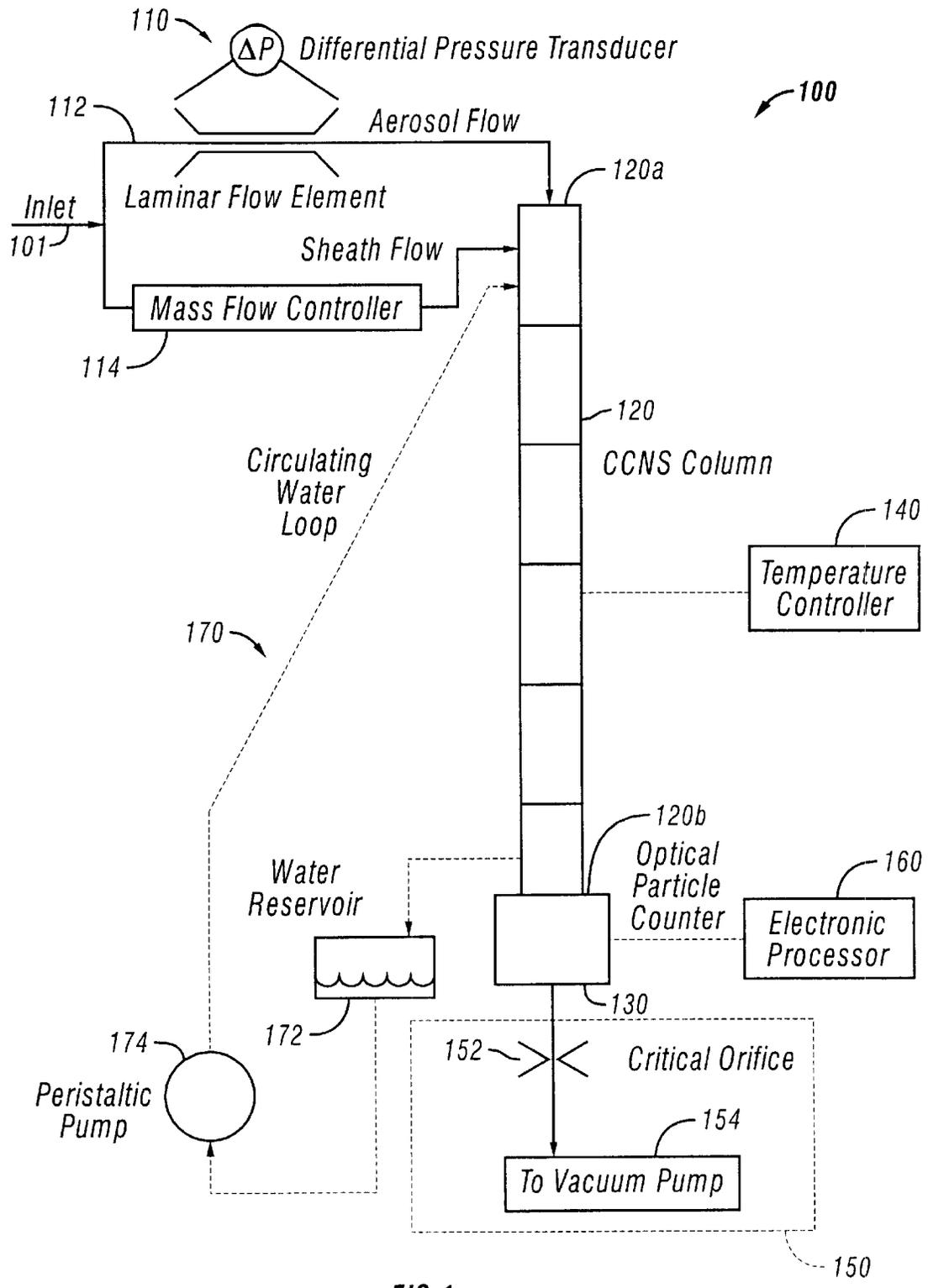


FIG. 1

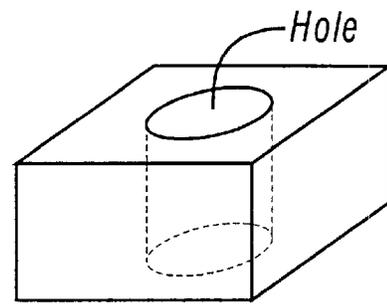
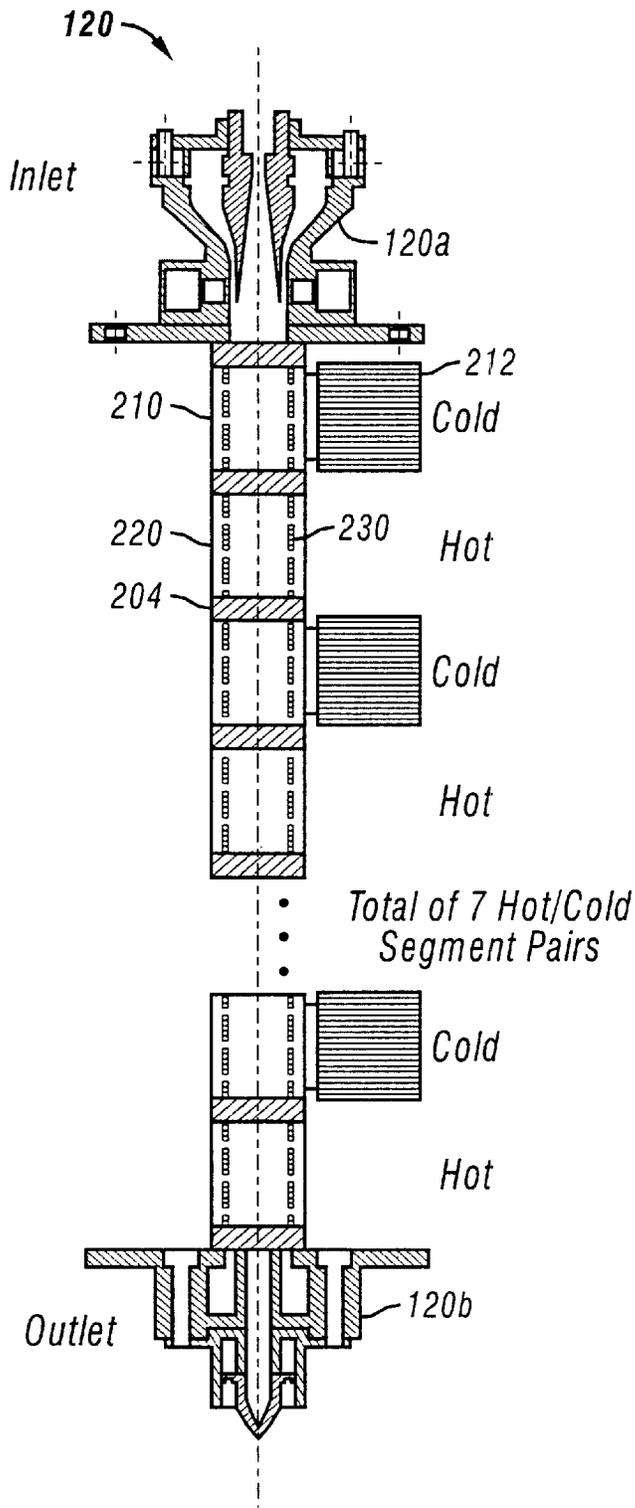


FIG.2A

FIG.2

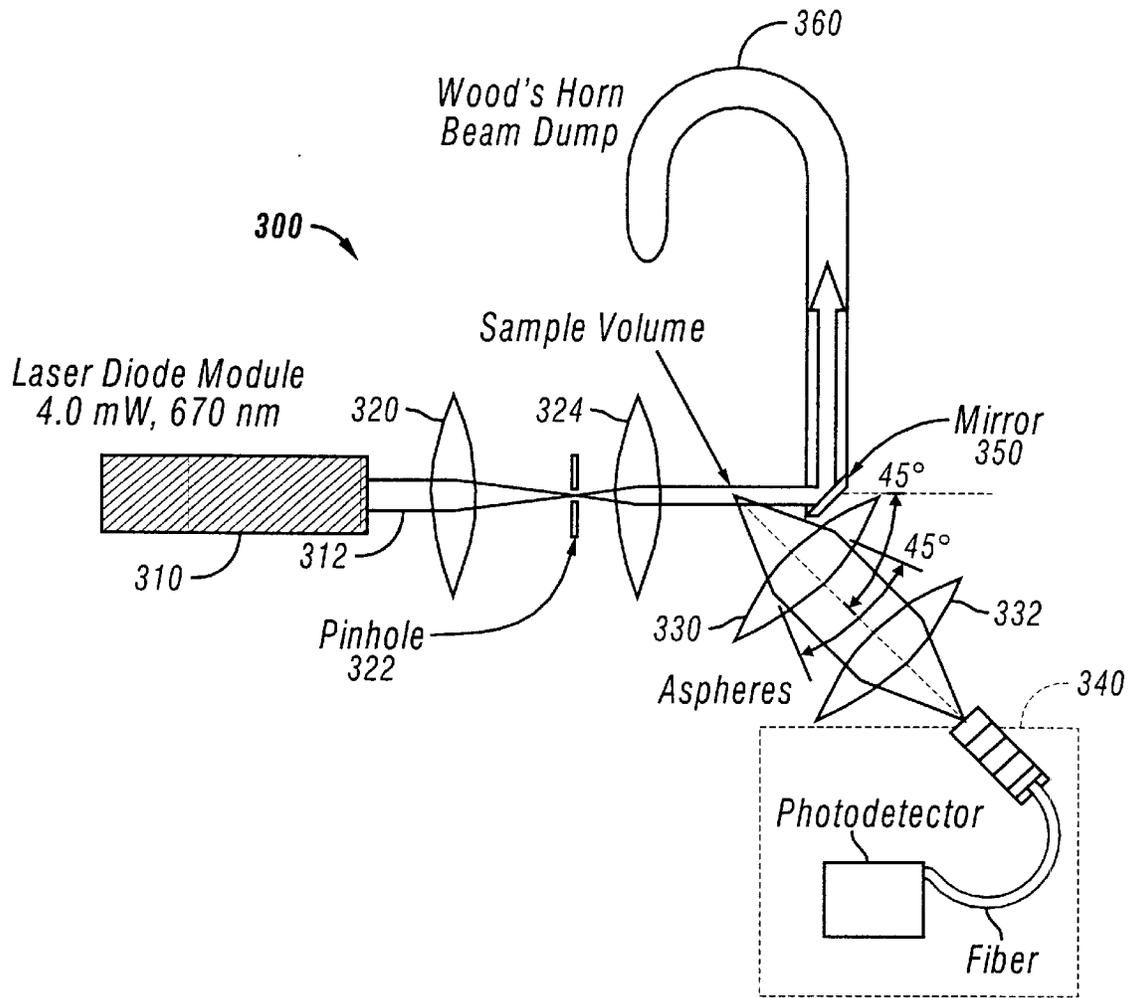


FIG. 3

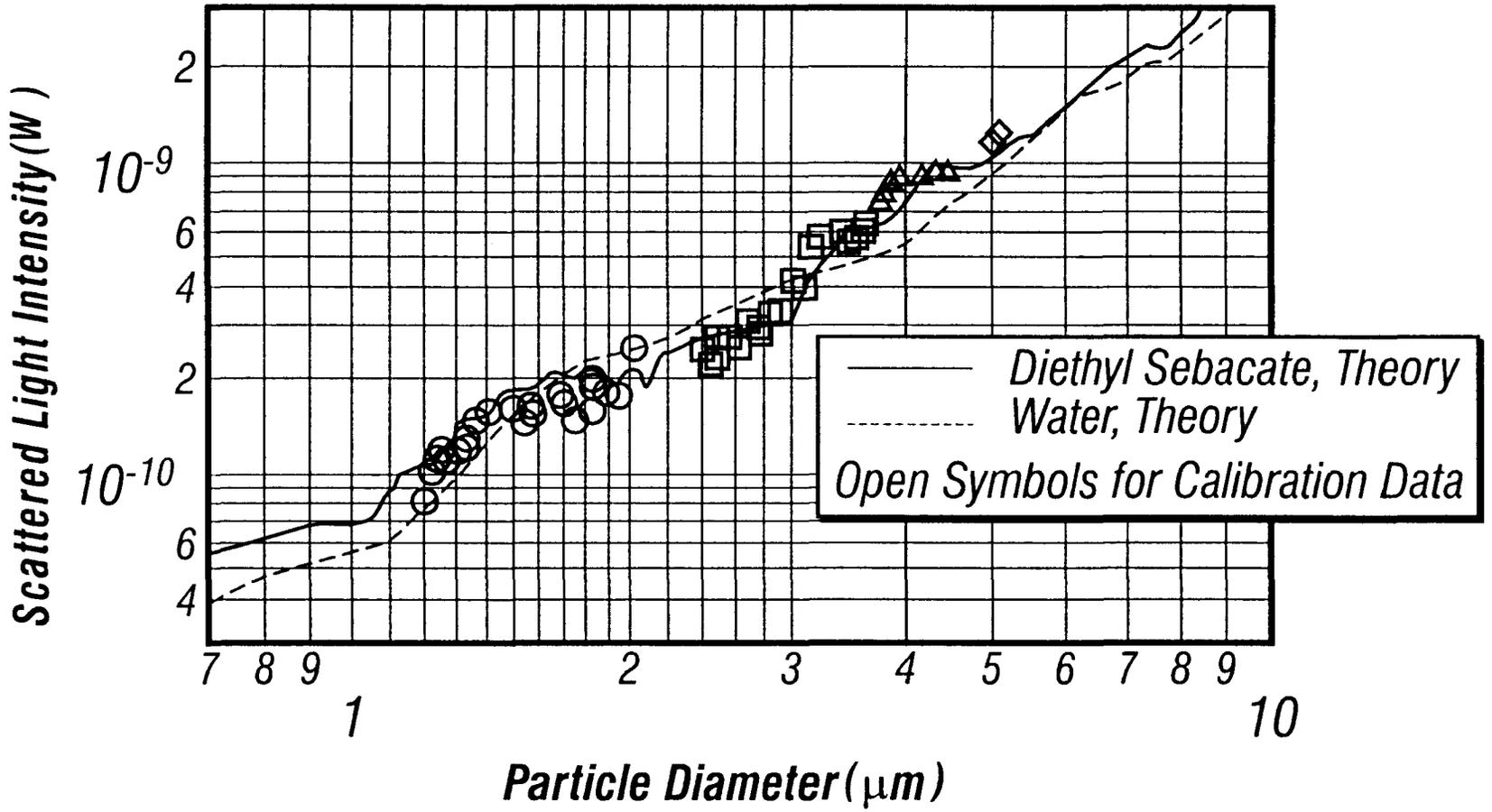


FIG.4

TECHNIQUES FOR CHARACTERIZING CLOUD CONDENSATION NUCLEI

This application is a divisional application (and claims the benefit of priority under 35 USC 120) of U.S. application Ser. No. 09/169,274, filed Oct. 9, 1998 which is now U.S. Pat. No. 6,330,060 and claims the benefit of U.S. Provisional Application No. 60/062,013, filed Oct. 10, 1997.

ORIGIN OF THE INVENTION

The U.S. Government has certain rights in this invention pursuant to Grant No. N00014-96-1-0119 awarded by the Navy.

FIELD OF THE INVENTION

The present invention relates to aerosol measurements, and more particularly, to instruments and techniques for characterizing cloud condensation nuclei.

BACKGROUND

Atmospheric particles influence the climate system, radiative transfer, visibility, and air quality. Hence, aerosol measurements of concentration, sizes, and chemistry of atmospheric particles are important in many applications, including monitoring air pollution and predicting climate change.

One aspect of aerosol measurements is characterization of cloud condensation nuclei ("CCN"). Under proper humidity conditions, certain aerosol particles are able to nucleate to form cloud droplets. Properties of cloud condensation nuclei provide important information on cloud formation and cloud properties. For example, cloud condensation nuclei can influence the droplet number and size distribution in a cloud, which ultimately affect a variety of processes including cloud lifetime and precipitation rate.

The ability of a particle to nucleate is at least in part determined by the saturation level of the environment, the size of the particle, and the chemical composition of the particle. For example, water vapor is more likely to condense on salt particles such as NaCl than on organic particles. When the relative humidity exceeds the saturation level where the vapor phase and the liquid phase are in equilibrium, a supersaturation state establishes and vapor begins to condense on surfaces and some particles to form droplets or condensation nuclei. At a certain critical supersaturation, when the diameter of a condensation nucleus of a given chemical composition exceeds a critical diameter, the nucleus is said to be "activated", that is, vapor will condense spontaneously on that nucleus and cause the nucleus to grow to a very large size which is limited only by the kinetics of condensational growth and the amount of vapor available for the condensational growth.

The critical diameter at a given supersaturation usually changes with the chemical composition of the particles. Hence, particles of different chemical compositions can become activated at different sizes.

One way to characterize condensation nuclei is to measure the critical supersaturation at which a particle activates. Instruments for such measurements are generally referred to as cloud condensation nucleus counters. Cloud condensation nucleus spectrometers are such counters capable of producing and measuring supersaturations in a desired range. See, for example, Hudson, "An Instantaneous CCN Spectrometer," *Journal of Atmospheric and Oceanic Technology*, Vol. 6, p. 1055, December, 1989, and Hoppel et

al., "A Segmented Thermal Diffusion Chamber for Continuous Measurements of CN," *Journal of Aerosol Science*, Vol. 10, p. 369, 1979, which are incorporated herein by reference.

The atmospheric environment is usually dynamic. The activation and subsequent growth of cloud condensation nuclei originated from a subset of atmospheric aerosols are essential to formation of cloud droplets. Therefore, it is desirable to perform in situ measurements in order to accurately measure aerosol samples in real time and monitor the changing climate at a target location. A compact airborne cloud condensation nucleus spectrometer can be used to meet such demand. However, many conventional condensation nucleus spectrometers are ill-suited for small aircraft platforms due to limitations in various factors such as weight, size, time resolution, range of measurable supersaturation.

SUMMARY

The present invention provides a novel CCN spectrometer which has been designed specifically for use on a remotely piloted aircraft for long periods of unattended operation, and which can measure CCN spectra over a wide range of supersaturation at high frequency (one spectrum per minute or faster). The instrument is also designed to be light and consume minimum power in order to conserve the limited resources available on small aircraft.

One embodiment of the CCN spectrometer implements a segmented cloud condensation nucleus growth column. A gas flow channel is formed within the column to receive and transfer a gas flow from an input opening to an output opening and having an inner wall which is wetted by a liquid. The segmented column has a plurality of alternating hot and cold temperature-maintaining segments arranged next to one another relative to the gas flow channel to control and maintain a temperature distribution along the gas flow channel. Each hot temperature-maintaining segment is maintained at a temperature higher than a cold temperature-maintaining segment. The temperatures produce a varying supersaturation environment within the gas flow channel.

In particular, a temperature difference between adjacent hot and cold temperature-maintaining segments increases from the input opening to the output opening to produce a supersaturation distribution that also increases from said input opening to said output opening.

A special optical particle counter is implemented to produce an optical probe beam to illuminate the gas flow in a close proximity to the output opening and to determine presence and dimension of particles in the gas flow.

These and other aspects and advantages of the present invention will become more apparent in light of the accompanying drawings, the detailed description, and the appended claims.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a diagram showing one embodiment of a cloud condensation nucleus spectrometer.

FIG. 2 shows a segmented cloud condensation nucleus column.

FIG. 2A shows one embodiment of a segment for the column shown in FIG. 2.

FIG. 3 shows the layout of an optical particle counter integrated to the column of FIG. 2.

FIG. 4 shows intensity of the scattered light from output aerosol flow as a function of particle size measured by the optical particle counter shown in FIG. 3, where a calculated calibration by using a Diethyl Sebacate flow is also shown.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

FIG. 1 shows one embodiment **100** of a cloud condensation nucleus spectrometer. The spectrometer **100** includes an input flow module **110**, a CCN growth column **120**, a temperature controller **140** for the CCN growth column **120**, and a particle counter **130**, an electronic processor **160**, and an output flow module **150**. The CCN growth column **120** is configured to produce an increasing supersaturation profile from an input end **120A** to an output end **120B** along the aerosol flow. The aerosol particles having critical supersaturation within the supersaturation range produced by the CCN growth column **120** are activated and exit the CCN growth column **120** with increased sizes.

The particle counter **130** is located at the output end **120B** of the condensation column **120** and measures the number of the activated particles in the aerosol flow. Examples of such particle counter includes an optical particle counter which infers particle size from intensity of light scattered from individual particles, and an aerodynamic time-of-flight counter which measures particle size by the particle velocity acquired through rapid acceleration of the gas flow. The electronic processor **160** receives and processes the output signal from the particle counter **130** to produce the respective number of activated particles as a function of the critical supersaturation.

The spectrometer **100** also includes a water supply module **170** having a reservoir **172** to provide water to the CCN growth column **120**. Preferably, the CCN growth column **120** may be positioned vertically so that water can be recirculated through the CCN growth column **120** from the top to the bottom by using a single water pump **174**. This also minimizes buoyancy induced secondary flows and loss of particles to the column walls by gravitational settlement. The flow rate of the water may be maintained at a constant low flow rate (e.g., less than 0.5 ml/min).

The condensation column **120** is preferably divided into a plurality of column segments at different temperatures. FIG. 2 shows the preferred structure of the condensation column **120**. Each column segment may be a metal block with a central through hole as shown in FIG. 2A. For example, aluminum block of about 28 mm (H)×25 mm (W)×25 mm (L) with a central hole of about 20 mm in diameter can be used. A thin-walled thermal conductive tube **230** (e.g., formed of stainless steel) may be placed in the center of the condensation column **120** through all column segments to conduct the aerosol flow. Alternatively, the aerosol flow may be conducted by directly using a flow channel formed by the through holes of the column segments. However implemented, the side wall of the flow channel is wetted running the water through the side wall with the water supply module **170**. One preferred way of wetting is to line the side wall with filter paper (e.g., Whatman 1 Chr).

The column segments are alternatively maintained at different high and low temperatures which are respectively referred to as "hot" column segments **220** and "cold" column segments **210**. Each hot column segment **220** has an electrical heating element (e.g., one or more power resistors) connected to the temperature controller **140** and is maintained at a desired elevated temperature for that segment. A thermal control loop may be implemented to actively control the temperature of each segment. This is well known in the art. Each cold column segment **210** is attached to a cooling element **212**, e.g., a thermoelectric cooler, and is maintained at a desired low temperature. A heat sink may be attached to the cooling element **212** to increase the cooling efficiency. At

least one thermal sensor (e.g., a thermistor) is disposed in each column segment and connected to the temperature controller **140** to measure the temperature. Adjacent hot and cold column segments **220** and **210** are thermally insulated from each other by a thermal insulation layer **204**.

One feature of the condensation column **120** is that the temperature difference between two successive hot and cold column segments increases. One implementation maintains the cold column segments **210** at different temperatures that decrease from the input end **120A** to the output end **120B** while keeping all hot column segments **220** at a common elevated temperature. Alternatively, the cold column segments **210** may be maintained at a common low temperature and the temperatures of the hot column segments **220** are higher than that low temperature and increase from the input end **120A** to the output end **120B**. In another variation, neither the cold column segments **210** nor the hot column segments **220** are maintained at a common temperature. However implemented, the temperature profile along the condensation column **120** not only changes in an alternating manner between high and low temperatures from one segment to another but the temperature difference also increases in the hot column segments **220** from the input end **120A** to the output end **120B**. In the embodiment shown in FIG. 2, the condensation column has a total of seven pairs of cold and hot column segments. The temperature difference between the two segments in each pair can be set at 2° at the beginning and increases 1° per pair. The temperature difference in the last pair at the end **120B** is 8°.

This special temperature profile can produce a monotonically increasing supersaturation profile along the center line of the condensation column **120** and can maintain a desired high spatial rate throughout the condensation column **120** without a significant decay near the output end **120B**.

The flow rate of the aerosol flow in the condensation column **120** also has a significant impact on the supersaturation profile along the center line of the condensation column **120**. When the flow rate is not controlled within a proper range, the supersaturation profile may not be monotonic but may have the same supersaturation at two different positions. The output flow module **150** is used to adjust the flow rate for a given temperature profile in the condensation column **120** to achieve a desired monotonically increasing supersaturation profile.

The input flow module **110** includes an aerosol flow path **112** and a sheath flow **114**. The sheath flow **114** is produced by filtering out aerosol particles with a mass flow controller. The relative amount of aerosol versus sheath flow can be controlled by using a mass flow controller **114** (e.g., Sierra Instruments 840). The aerosol flow to the condensation column **120** is the difference between the total input flow **101** and sheath flow **114** and is monitored by measuring the pressure drop across a laminar flow tube in the aerosol flow path **112**. The total flow **101** may be controlled by using a critical orifice **152** in the output flow module **150** (e.g., with a nominally flow rate of 0.75 lpm). The output flow module **150** also includes a vacuum pump **154** to induce the aerosol flow.

It should be noted that in this configuration, the instrument is not maintained at a constant pressure. Since the diffusivity of water vapor in air is pressure dependent, this must be accounted for in interpreting the data. Also, since the mass flow controller does not maintain constant volumetric flow rate, rather, the sheath volumetric flow rate increases with decreasing pressure, the relative amounts of aerosol and sheath flow vary with pressure, which also must

be accounted for. These flow rates are fed into the electronic processor **160** for data processing.

FIG. **3** shows an optical implementation **300** of the particle counter **130**. The aerosol flow is perpendicular to the paper. The optical particle counter **300** is specially designed to improve accuracy in particle counting and to reduce the instrument weight. The optical particle counter **300** is designed for measuring water droplets within a range from about 1 μm to about 20 μm in size. The optical particle counter **300** includes a diode laser **310**, a beam collimator formed of lenses **312**, **324** and a pinhole **322**, an optical collector formed of lenses **330**, **332** and a photodetection module **340**. The diode laser **310** may be an industrially-packaged laser diode module (e.g., ThorLabs 98002-005) with a line output nominally 1 mm \times 5 mm wide. The pinhole may be of about 50 μm .

The collimated laser beam from lens **324** is directed to the aerosol flow from the output end **120B** of the condensation column **120**. The laser beam is preferably perpendicular to the aerosol flow. The dimension of the illuminated region in the aerosol flow is less than the average spacing between two aerosol particles so that, on the average, only a single particle is illuminated by the laser beam. This substantially reduces the probability of two droplets passing through the beam simultaneously while maximizing the uniformity of the intensity seen by particles passing through slightly different parts of the beam. The beam is spatially-filtered by the pinhole **322** because spurious uncollimated light can introduce an excessively high background light level. A particle is counted when a strong optical signal caused by scattering from a particle is detected by the photodetection module **340**.

In general, the output nozzle of the output end **120B** is as small as possible so that the CCN droplets all pass through the same part of the laser beam. However, it has been found that tip diameters significantly smaller than 1 mm may cause droplet impaction. Preferably, a nozzle with a half angle of about 15° and an output diameter of about 1 mm may be used. Other means of directing the grown particles exiting the growth column to the view volume may also be used. A mirror **350** and a beam dump **360** may be implemented to guide the main laser beam away from the lens **330** to reduce the background noise.

Collection lenses **330** and **332** are positioned relative to the intersection point **326** of the laser beam and the aerosol flow so that the scattered light is collected at a direction of about 45° off the propagation direction of the laser beam. The lens **330** has a solid collection angle of about 45°. Two aspheric lenses may be used as the lenses **330** and **332**. The direction of detection and the solid collection angle of the lens **330** should be properly selected to ensure a monotonic dependence of the intensity of the collected light and the particle size.

The photodetection module **340** may include a fibre optic patch cord (e.g., 1 mm core) to receive and transmit the light to a photodetector such as a photomultiplier tube or a photodiode (e.g., an avalanche photodiode detector). A multichannel analyzer (e.g., EG&G Ortec Trump-2k) card may be included in the electronic processor **160** to measure the peak height of the pulses, each of which represents one droplet.

FIG. **4** shows a calculated response of the optical particle counter and measured calibration data using droplets of a clear, non-volatile organic liquid, diethyl sebacate

($n=1.436$), generated from a Berglund-Liu vibrating orifice aerosol generator.

The optical particle counter **300** of FIG. **3** can be integrated to the condensation column **120** so that the laser probe beam intersects with the output aerosol flow at a location close to the output end **120B**. Since water droplets are volatile and water condensed on an aerosol particle can vaporize after leaving the condensation column **120**, it is critical to measure the particles at a location as close to the output end **120B** as possible. Many conventional particle counters including optical counters often introduce the output aerosol flow into a flow path before measurements and can significantly degrade the accuracy of the measurements. Implementation of the optical particle counter **300** can essentially eliminate the additional flow path outside the output end **120B** and improves the accuracy of measurements.

The above design can be used to significantly reduce the weight (e.g., less than 35 pounds) and size of a CCN spectrometer and thereby reduce the power consumption (e.g., about 100 W) and requirements of the power supply (e.g., a 28-V source). The on-board electronic processor further allows for automatic operation without human intervention.

Although the present invention has been described in detail with reference to the preferred embodiments, various modifications and enhancements may be made without departing from the appended claims.

What is claimed is:

1. A method, comprising:

directing a gas flow into a gas flow channel from an input opening to an output opening;

vaporizing a liquid to produce a vapor from the liquid to fill the gas flow channel;

producing a spatial temperature distribution in the gas flow channel that changes with position from the input opening to the output opening, and that includes a plurality of alternating hot and cold regions arranged next to one another where a hot region has a temperature higher than an adjacent cold region;

selecting temperatures of the hot and cold regions to spatially increase a temperature difference between adjacent hot and cold segments from the input opening to the output opening in the gas flow channel; and

determining presence and dimension of particles in the gas flow which are condensed with the vapor.

2. The method as in claim 1, wherein the hot regions in the gas flow channel are maintained at a constant elevated temperature and the cold regions are maintained at different temperatures which monotonically decrease from the input opening to the output opening and are lower than the elevated temperature.

3. The method as in claim 1, wherein the cold regions are maintained at a constant low temperature and the hot regions are maintained at different temperatures which increase with position from the input opening to the output opening and are higher than the low temperature.

4. The method as in claim 1, wherein the presence and dimension of particles in the gas flow are measured optically by illuminating an optical beam to the gas flow at the output opening, without introducing another flow into the gas flow.