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## NoTice

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(HASA-Came-NPO-13606-2) FREQUENCI-SCANNIHC
particle sig Application (Hasa) 21 p HC A02/ HF a01
csc 14B
63/35


Uncles

IENCY-SCANNING PARTICLE
SIZE SPECTROMETER
The invention relates to appanatus and method for
remotely reconstructing the effective spectrum of sizes of
scattering particulates of unknown chemical composition in
equivalent spheres of lower radius value approximatelv one
micrometer and unbounded upper radius value. Conventional
direct sampling methods, where particles are thermally precin
itated, impacted or otherwise mechanically collected, present
several problems, such as: disturbing of samrile, low samplin
rate, discontinuous sampling, collection efficiency problems,
long sampling time and painstaking analvsis.

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## 



Where $\rho_{k}$ is a depolarization factor of the $k^{\text {th }}$ gas defined as the ratio of scattered intensities in directions parallel and perpendicular to the plane of scatterinp, for ar incident bean of natural light. The preferred scattering angle $\theta$ is 150 ming but may be should be smaller than 0,2 inverse picrometers ( $\mu^{-1}$ ) over a range of approximately $0.20 \mu \mathrm{~m}^{-1}$ to $4.0 \mu \mathrm{~m}^{-1}$. The novelty resides in determining particle size distribution independently of refractive indices of particles, including the accurate location of the mode radius of particles forming a homogeneous cloud or medium between the light source and the detector, using remote and passive instrumentation. Even the light source may be regarded as passive since it may be natural light. This determination is provided without constraining the size distribution to any analytical model. Use of such models would necessarily bias the distribution.
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THT



## $20 \Gamma 4$



$\angle 1,4$


## SPECIEICATIO:

TO ALL UUN IT PAY CO:BERR:

BE IT KiON: THAT ALAIM L. FMMis, a citizen of Morocco, residing at 1080 South El Molino Avenue, Fasadera, Caliform 9110 , in the County of Los Angeles, State of Californte, has invented a new and useful

FREマJEiCY-SCA:MI!G PAPTICLE SIZE SEECTROMETER
of which the folioning is a specification:
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## ABSERAOT OE T: D DSCTOEDEX

A particie size spectrometer having a fined fiald of view within the forward light scatterine cone at an anEle $\theta_{s}$ between approximately 100 and 200 minutes of arc (preEerably at 150 minutes), a spectral range extending approximately fron 0.2
 between about 0.1 and $0.2 \mu \mathrm{~m}^{-1}$ (preferably toward the lower and of this range of spectral resolution), is employed to determine the distrizution ce particle sizes, indepencently of the chemical composition of the zarticles, from measurements of incident iight, $I_{0}$, at each fresuency, $\sigma(=1 / \lambda)$, and scattered lift.t, I( $\sigma$ ), according to the equation:

$$
f_{n}(r)=-\frac{\ell^{3}}{2 \pi r^{2}} \int_{0}^{\infty} J_{1}(y) Y_{1}(y) y \frac{d}{d \sigma}\left(I I_{\sigma}\right) d \sigma, y=2 r_{\sigma},
$$

where $\left\{=2 \pi \sin \theta\right.$, 3 being the fixec $\because$ iewine ancle $\theta_{S}$ at which scattered linht is reasured, $r$ is particle size, $\sigma$ as the

kind and order unity, $Y_{1}$ is a Bessal function of second kind and order urity. The cuantity, $I_{c}$, is the ratio of scattered light to incident light at each frequency interval. The apparatus is a passive remote sensor that can be used in laboratories, field stations, flying aircrafts and airships, and on board an orbiting satellite.

## ORIGI:. OF THE IMVE:TIO::

The invention dascribed herein ::as mace in the performance of work under a JASA contract and is subject to the provisions of Section 305 of the Hational Aeronautics and Space Act of $195 \varepsilon$, Pubiic Law $85-5 € 8$ (72 Stat. 435 ; L2 USC 2457).

## EACKGROU:D OF ST:E IMYE:OIO:

This invention relates to a method and apparatus for determining the distribution of particie sizes of a scattering medium of unknown chenical composition. The medium may consist of a composite mixture of arbitrary numbers of chemically different particles emberdec in various gases.

There is a continuing need to determine the size distribution of any arbitrary number of particle species which may present a distribution in their refractive indices, and which may be embedced in a scattering medium of unknown chemical composition, for such irdustrial purposes as environmental pollution monitoring and control, chenical analyses, aeronautical engineering and rocket engineering as well as for such other purnoses as meteorological research, geophysical researzh, anc bioredical anaivses. The composite mixture os
particles and gases may be contained in an experimental chamber, suspended freely in the air, or suspended in fluids such as medical plasmas, or in any other similar situation where particulate sizes must be determined without disturbing the sample.

Conventional direct sampling methods where particles are thermally precipitated, impacted or otherwise mechanically collected, as well as in situ imaging methods, present several problems. Some of these problems are: disturbing of sample, low sampling rate, discontinuous sampling, collection efficiency problems, long sampling time and painstaking analyses. Additionally, they cannot be used as remote sensors for real-


To avoid these problems of direct sampling, and at the same time provide a remote sensing capability, methods have been developed which exploit the forward light scattering properties of particles at frequencies within the range 0.193 to 4.070 inverse micrometers ( $\mu \mathrm{m}^{-1}$ ) or within any larger range extending down to $0.02 \mu \mathrm{~m}^{-1}$ and up to $9.27 \mu \mathrm{~m}^{-1}$. Incident light, $I_{0}$, possessing a spectrum of frequencies in the ranges above specified, is characterized only by its wavenumber, $\sigma_{i}$, the reciprocal of the wavelength, $\lambda_{i}$. Its direction of propagation, $\theta_{i}$, is the direction of reference, $\theta_{i}=0$. The subscript, $i$, here stands for "incidence." Particles are characterized by their nature and micro-structure, respectively represented by a refractive index, $m$, a complex number, and a size distribution, $* n(r)$, representing, a partial concentration per unit volume and per unit increment of the radius $r$. Only $\Omega(r)$ is ${ }^{\text {亚 }}$ independent
of the wavenumber. The various paritcles, assumed to be spherical in shape, may also present a distribution in mitself. Gases will be similarly characterized by their number density, i.e. number of atoms or molecules jer unit volume, refractive index and geometrical shape. Scattered light, on the other hand, is characterized by its wavenumber, $\sigma_{s}$, and direction, $\theta_{8}$, of propagatirn. Here the subscript, $s$, stands fior "scattering." 7.e direction, $\theta_{s}$, is th s the scattering angle Qwith reference to the incident light direction $\theta_{i}$.

For a given incident ilght, i.e., for a situation where $\sigma_{i}$ and $\theta_{i}$ are given, and for a given assembly of scatterers, i.e., for an assembly of particles of given refractive indices, $m\left(\sigma_{s}\right)$, and size distribution, $n(r)$, Mie's theory of scattering dy a conducting sphere enables us to determine the scattered light for any arbitrary $\sigma_{s}$ and $\theta_{s}$. Conversely if the incident light is known for any $\sigma_{i}$ and $\theta_{i}$ to be prescribed, and the scattered light is measured for various $\sigma_{s}$ and $\theta_{s}$ to be also prescribed, we should be able in principle to infer the properties of the particles, that is their size distribution, and refractive index values at the various us. In other words, Mie's theory enables us to compute the output, $I_{s}$, in all its detailed variations if we know the input, $I_{0}$, and the properties of the particles $m\left(\sigma_{s}\right)$ and $n(r)$. However, what is of interest is a determination of the scatterers from a knowledge of the input, $I_{0}$, and the output, $I_{s}$. But from an inversion of lie's theory it would be possible to determine only the combined relation between the refractive indices, $m\left(\sigma_{s}\right)$, and size distribution, $n(r)$, and nop the size distribution separatel".

It has been discovered that the Fraunhofer theory of angular diffraction of light of fixed single frequency at an aperture in a plane screen can be extender to a range of multiple frequencies. Under these conditions, the light scattered by a particle is essentially independent of the refractive index, $m\left(\sigma_{8}\right)$, and depends only on its size as though the particle Fere an aperture in a screen of the same radius. It has also peen discovered that, working within the restricted domain of applicability of the theory thus exterided ( $\theta_{s}$ between 100 and 200 minutes of arc, approximately, preferably $\theta_{s}=150$ minutes; and $0.2 \mu^{-1} \leq \sigma_{s} \leq 4.0 \mu \mathrm{~m}^{-1}$, approximately), it is possible to determine size distribution, $n(r)$, independently of refractive indices $\mathrm{m}\left(\sigma_{s}\right)$, i.e., to determine size distribution of particles of radius larger than approximately one micrometer ( $\mu \mathrm{m}$ ) without knowing anything of their refractive indices, from known incident light, $I_{0}$, and measured scattered light, $I_{s}$.

If the assembly of scatterers under study consists of particles of $j$ different species and $k$ different gases, the identical determination of $n(r)$ can still be made provided only $j, k$, and the so-called depolarization factor of each gas (this is defined as the ratio of scattered intensities $1 n$ directions parallel and perpendicular to the plane of scattering for an incident beam of natural light), $\rho_{k}$, are known.

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In accordance with the present invention, a natural or artificial light source is passed through a volune of scattering particles the size distribution of which is to be determined. The intensity of light scattered by the particles,
at a fixed angle between approximately 100 and 200 minutes of arc from the direction of incident light, preferably 150 minutes of arc, is recorded as a function of frequency, $I(\sigma)$. Records are made across the approximate frequency interval $0.20 \mu \mathrm{~m}^{-1}$ to $4.0 \mu \mathrm{~m}^{-1}$, at frequency steps smaller than about $0.2 \mathrm{um}^{-1}$. Simultaneously, the intensity of the incident light at each ffrequency, $I_{0}$, is recorded. The ratio $I_{\sigma}$ of scattered light, $f(\sigma)$, to incident light, $I_{0}$, is then obtained. In order to determine the size distribution, $n(r)$, from the ratio $I_{0}$, the following equation has been derived and investigated with regard to its correctness, the uriqueness of the solution obtained from it, and the stability of this solution with regard to inherent experimental and analytical noises :

$$
n(r)=-\frac{\ell^{3}}{2 \pi r^{2}} \int_{0}^{\infty} J_{1}(y) Y_{1}(y) y \frac{d}{d \sigma}\left(\sigma I_{\sigma}\right) d \sigma, y=\ell r \sigma
$$

where $J_{1}$ is a- Bessel function of first kind and order unity, $Y_{I}$ is a Bessel function of second kind and order unity, $\ell=2 \pi \sin \theta, \theta$ being the fixed viewing angle at which scattered light is measured, $r$ is particle radius, and $\sigma$ is the reciprocal of wavelength. In the case of $j$ particle species embedded in $k$ gas species, the former equation remains valid provided only $I_{\sigma}$ is changed to the following expression:

$$
I_{\sigma} \rightarrow G_{\varepsilon}=-\frac{1}{j}\left[4 \cos \theta I_{\sigma}-\frac{3}{2} \sum_{k} \frac{2-\rho_{k} \cos 2 \theta}{2+\rho_{k}}\right],
$$

where $\rho_{k}$ is a so-called depolarization factor of the $k^{\text {th }}$ gas defined as the ratio of scattered intensities in directions parallel and perpendicular to the plane of scattering for an incident beam of natural light.

The novel features of the invention are set forth with particularity in the appended claims. The invention will best be understood from the following description when read in conjunction with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWIMGS

The FIG. l illustrates schematically an example of apparatus for the method of the present invention, where the method steps operating on measured quantities are represented by functional blocks. angle, $\theta_{s}$, on determining particle size distribution from light scattered by the particles according to the present invention.

FIG. 3 is a graph showing the effect of spectral range $\theta_{\min }$ to $\theta_{\max }$, in scanning the frequency of light incident on particles for determining particle size distribution.

FIG. 4 is a graph showing the effect of spectral resolution, $\Delta \sigma$, in the process of determining particle size distribution according to the present invention.

Referring to FIG. 1, a natural or artifitial light source 10 is directed to a cell 11 filled with a medium of unknown chemical composition through a chopper 12 which cyclically deflects the incident light, $I_{0}$, to a reference multiple-filter detector 13 for the purpose of measuring the incident light intensity as the light source is scanned in frequency. The filter of the detector is preferably changed as the light source is scanned in order that the reference measurement $I_{0}$ be accurate for each frequency of incident light. An object multiple-filter detector 14 is likewise controlled to detect scattered light at the different prescribed frequencies as the source is scanned. That is represented by dotted lines 15 and 16 for synchronized control of the detector and light source. The reference and object multiple-filter detectors can be detector arrays in which each detector element is slaved to a specific frequency. $R_{L: S}$
OR:
 field of view at a fixed angle $\theta_{s}$ of less than $6^{\circ}$ within the forward scattering cone of the cell 11 . For best results, the scattering angle $\theta_{s}$ is made about 150 minutes of arc, but it may be any angle between 100 and 200 minutes of arc, approximately. It is also designed to have a spectral resolution, i.e., a frequency step, $\Delta \sigma$, as coarse as $0.2 \mu \mathrm{~m}^{-1}$ or as fine as $0.1 \mu \mathrm{~m}^{-1}$, and to operate within the approximate spectral frequency Iimits of $\sigma_{\min }=0.2 \mu \mathrm{~m}^{-1}$ and $\sigma_{\max }=4.0 \mu \mathrm{~m}^{-1}$.

Although a cell is indicated, the scattering particles forming a homogeneous cloud or medium between the light source and the detector may be contained in a laboratory chamber, suspended freely in the air, contained in fluids such as plasmas and blood, emitted from any type of industrial or engine nozzle, or in any other similar situation where particulate sizes must be determined without disturbing the sample in any manner. Also, although a variable light source is indicated, radiance from particles that exhibit significant thermal emission e.g., radiance from atmospheric particles which emerges from the atmosphere, can also be used. On the other hand, a light source may be employed that emits all of the frequencies to be scanned. In the latter case, the control for frequency scanning may be effected at only the detectors 13 and 14 , while in the former case it is effected only at detector 14.

If the foregoing restrictions on the fixed angle, $\theta_{s}$, spectral frequency range, $\sigma_{\min }$ to $\sigma_{\max }$, and spectral resolution, $\Delta \sigma$, of the reference and object detectors are observed, the measurements of scattered light $I(\sigma)$ can provide the complete spectrum of particulate sizes, i.e., particle size distribution, $n(r)$, in radii of equivalent spheres, from approximately $1 \mu \mathrm{~m}$ to any arbitrarily larger value, including the accurate location of the mode radius (corresponding to the largest concentration of particles). The technique will now be described. Consider an inciden: beam of radiation of wavenumber, $\sigma=1 / \lambda$, ( $\lambda=$ wavelength), and intensity, $I_{o}$, illuminating a single particle of radius, $r$. The singly-scattered light intensity, $I(\sigma)$, at angle, $\theta$, in units of $I_{0}$, is expressed by the formula:

$$
I_{\sigma} \equiv \frac{I(\sigma)}{I_{0}} \cong\left[\frac{r J_{1}(k r \sin \theta)}{\sin \theta}\right]^{2}, \theta=0 s,
$$

where $k=2 \pi \sigma$ and $J_{1}$ is a Bessel function of first kind and order unity. For a distributio of sizes, $n(r)$, the generaliza-
? tion of this expression will provide che total scattered light intensity:

$$
I_{\sigma}=\frac{1}{\sin ^{2} \theta} \int_{0}^{\infty} n(r) r^{2} J_{1}^{2}(x) d r, x=k r \sin \theta .
$$

It may be noted that equations (1) and (2) depend only on $r$. In order to determine $n(r)$ from measurements of $I_{\sigma}$, it is necessary to invert this equation. The analytical inverse of equation (2) is obtained in the form:

$$
n(r)=-\frac{\ell^{3}}{2 \pi r^{2}} \int_{0}^{\infty} J_{1}(y) Y_{1}(y) y \frac{d}{d \sigma}\left(\sigma I_{\sigma}\right) d \sigma, y=\ell r \sigma,
$$

where $\ell=2 \pi \sin \theta$ and $Y_{1}$ is $a$. Bessel function of second kind and order unity.

If the scattering medium being illuminated consists of $j$ different species of particles embedded in $k$ different species of gases, equations (2) and (3) remain valid provided only we replace in these equations the quantity $I_{o}$ by the following quantity:

$$
\begin{equation*}
I_{\sigma} \rightarrow G_{\sigma}=\frac{I}{j}\left[4 \cos \theta I_{\sigma}-\frac{3}{2} \sum_{k} \frac{2-\rho_{k} \cos 2 \theta}{2+\rho_{k}}\right] \tag{1i}
\end{equation*}
$$

where $\rho_{k}$ is the depolarization factor of the $k^{\text {th }}$ gas.

It must be stressed that Equation (3) utilizing either $I_{\sigma}$ or $G_{\sigma}$ is a closed form expression for $n(r)$ which makes no assumption on any analytical form by which this function may be modeled. It is exactly true for the function $I_{\sigma}$ defined by

Equation (2) or the function $G_{\sigma}$ defined by Equation (4). It may be noted, however, that $\sigma$ will not vary over an infinite range as indicated by Equation (3).
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For example, if it is wished to use this instrument as a remote sensor on board a planetary orbiter, $\sigma$ has a lower bound which will vary according to the brightness temperature of the planetary atmosphere of interest. This value of $\sigma$ is that at which the planetary radiation dominates the visible solar radiation reflected by the planet. This is because the thermal radiation from the planet surface and atmosphere system, which is emitted due to the temperature of this surface and atmosphere, is not the absolute zero. Thus, on Earth, this cutoff value is approximately $0.37 \mu \mathrm{~m}^{-1}$. However, although the integration indicated in Equation (3) cannot be carried out completely, and it is conceivable that in some extreme cases the distribution $n(r)$ may either not be reproduced exactly or not at all, it has been discovered that an object detector designed to operate within the limits set forth hereinbefore will permit the integration to be carried out to provide the detailed spectrum of sizes (particle size distribution) in equivalent spheres of the scattering particles. These limits provide a unique method which has been submitted to detailed scrutiny as to stability with regard to both inherent experimental and analytical noises.

The first computational step of forming the ratio indicated by Equation (2) is represented by a functional block 17, and the second computational step indicated by Equation (3) is represented by a functional block l8. The ratio can be formed using analog circuits, or upon converting the detector signals to digital form, by digital techniques. In either case, the result of the first computational step is preferably obtained in digital form for storage and subsequent retrisval for solui. on of Equation (3). The values of this ratio at the several prescribed frequencies are then inserted in Equation (3) which is subsequently solved for various values of $r$ larger than approximately lum. For example, for $r=r_{1}$, the equation yields $n_{1}=n\left(r=r_{1}\right)$; for $r=r_{2}$, the solution yields $n_{2}=n\left(r=r_{2}\right)$ and so forth. The set of values $n_{1}, n_{2}, \ldots$ is the desired particle distribution $n(r)$. A general purpose computer can be used for solving Equation (3). The computer can be used on a time-sharing basis or a special-purpose computer could be slaved to ti:ニ apraratus. The output from such a computer is converted to an electrical signal which is then displayed visually on a cathode ray tube (CRT) display 19. Thus, a real-time visualization of the size distribution is provided, such as for the purpose of monitoring the changes in particle sizes that may take place as is the case, for example, with environmental particulate pollution.

The effect of viewing angle, $\theta_{s}$, can be appreciated from the graph of FIG. 2 which shows a true plot of particle size distribution by a bell-shaped curved labeled "n(true)" for comparison with particle size distribution curves determined
at viewing angles of 50 minutes, 150 minutes and 350 minutes. The distribution curve labeled $" \theta_{s}=150 \mathrm{~min}$ " is found to be very close to the true distribution curve. Consequently, it is concluded that a viewing angle of about 150 min is optimum. At viewing angles between approximately 100 min and 200 min , the results are still very good, although small negative tails develop close to these two values below $1.5 \mu \mathrm{~m}$ radius; but beyond that range there is not only further degradation of the results from the true curve, but totally incorrect results over much of the range of particle sizes. For example, at a viewing angle as small as 50 rin , the results indicate a negative distribution between 14 and $17 \mu \mathrm{~m}$ radius, and although it seems to follow the true distribution curve as particle radius decreases, it becomes clear at below $8 \mu \mathrm{~m}$ radius that the distribution curve is totally in error (meaningless) since size distribution increases, apparently without limit, as the particle radius decreases. Deviation from the optimum viewing angle in the other direction will not produce such absurd results, as shown by the distribution curve labeled " $\theta_{s}=350 \mathrm{~min}$ ". But still the results have so departed from the true distribution as to not be useful except perhaps in locating the peak position. Note the negative distribution above llum and below $2.7 \mu \mathrm{~m}$ radius. The true distribution cannot, of course, go below zero. This demonstrates the limited range of viewing angles that can be effectively used in this technique.

The effect of spectral range can be appreciated from the graph of FIG. 3. In the range of about 0.2 to $4.0 \mu \mathrm{~m}^{-1}$, the result is very nearly identical to the true distribution curve as shown by a curve labeled "0.198 $\leqslant \sigma \leqslant 4.070 \mu \mathrm{~m}^{-1}$."

This result remains substantially the same if the starting value of 0.198 were to be lowered down to $0.02 \mu \mu^{-1}$. At the lower part of the range, namely at 0.198 to $2.860 \mu \mathrm{~m}^{-1}$, the form of the distribution curve remains about the same but becomes negative at less than $2 \mu \mathrm{~m}$ radius. Increasing the upper end of the range to approximately $9.00 \mu \mathrm{~m}^{-1}$ produces about the same results, but the distribution curve becomes more negative at less than $2 \mu \mathrm{~m}$ radius. Working in a subinterval of the lower part of the optimum range, namely in the range from 0.44 to $2.86 \mu \mathrm{~m}^{-1}$, the result is about the same as working at just the lower part of that oftimum range, except that the distribution is found to be regative below $3 \mu \mathrm{~m}$ radius and above $12 \mu \mathrm{~m}$ radius, with decrease of approximately $18 \%$ in the peak. Operating at only a narrow central part of the optimum spectral range, namely between about 0.3 to $1.2 \mu \mathrm{~m}^{-1}$ produces the absurd result indicated by the curve labeled " $0.286 \leq \sigma \leq 1.190 \mu \mathrm{~m}^{-1}$." Operating below the optimum range, such as in a range from 0.03 to $0.2 \mu \mathrm{~m}^{-1}$ also produces an absurd result, as shown by the curve labeled " $0.031 \leq \sigma \leq 0.235 \mu \mathrm{~m}^{-1}$." Lastly, extending the optimum range, for example from 1.0 to $10.0 \mu \mathrm{~m}^{-1}$ also produces the absurd resultis shown by the curve labeled $1.0 \leq \sigma \leq 10.0 \mu \mathrm{~m}^{-1}$."吅

The effect of spectral resolution is illustrated in FIG. 4. Again the curve labeled $n$ (true) is the theoretical or true particle distribution. The optimum is shown to be a spectral resolution of about $0.06 \mu \mathrm{~m}^{-1}$, as shown by the curve labeled $" \Delta \sigma=0.061 \mu \mathrm{~m}^{-1} "$. However, spectral resolution is less critical than spectral range or viewing angle. A smaller resolution would have no deleterious effect on the result.

A larger resolution can be tolerated to about three times the optimum. There, at the resolution of $0.18 \mu \mathrm{~m}^{-1}$, the results begin to degrade, and at about $0.80 \mu \mathrm{~m}^{-1}$, the results become absurd as shown by the curve labeled $" \Delta \sigma=0.774 \mu \mathrm{~m}^{-1} . "$

The necessary spectral range with the optimum spectral resolution may be achieved by scanning frequency at the detector using a detector-array in which each detector is slaved to a specific frequency, as noted hereinbefore. Alternatively, one or more voltage variable photodetectors are used, and frequency scanning is achieved by varying the bias voltage to the detectors. For most laboratory applications, there will be a specific light source, either variable in frequency or containing all frequencies of interest. In the latter case, scanning the frequencies in the desired range could be accomplishe by the use of filters. Such an arrangement would also be used outside the laboratory from a ground-based station such as for determining the size distribution of particles in the Earth's atmosphere, where light from the sun is filtered for scanning the frequency of light passed through a volume of scattering particles. In the case of an orbiting satellite, on the other han it is the thermal emission from the atmosphere-surface system that would be filtered.

Although particular embodiments of the invention have been described and illustrated herein, it is recognized that modifications and equivalents may readily occur to those skilled in the art and consequently it is intended that the claims be interpreted to cover such modifications and equivalents.

