



# Universal Spectrum for Atmospheric Aerosol Size Distribution: Comparison with PCASP-B Observations of VOCALS 2008

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**Abstract:** Atmospheric flows exhibit scale-free fractal fluctuations. A general systems theory based on classical statistical physical concepts visualizes the fractal fluctuations to result from the coexistence of eddy fluctuations in an eddy continuum, the larger scale eddies being the integrated mean of enclosed smaller scale eddies. The model predicts (i) the eddy energy (variance) spectrum and corresponding eddy amplitude probability distribution are quantified by the same universal inverse power law distribution incorporating the golden mean. (ii) The steady state ordered hierarchical growth of atmospheric eddy continuum is associated with maximum entropy production. (iii) Atmospheric particulate size spectrum is derived in terms of the predicted universal inverse power law for atmospheric eddy energy spectrum. Model predictions are in agreement with observations. Universal inverse power law for power spectra of fractal fluctuations rules out linear secular trends in meteorological parameters. Global warming related climate change, if any, will be seen as intensification of fluctuations of all scales manifested immediately in high frequency fluctuations. The universal aerosol size spectrum may be computed for any location with two measured parameters, namely, the mean volume radius and the total number concentration and may be incorporated in climate models for computation of radiation budget of earth-atmosphere system.

**Keywords:** *complex systems and statistical physics; general systems theory; maximum entropy principle; universal inverse power law spectrum; universal spectrum for atmospheric suspended particulates; fractal fluctuations; chaos and nonlinear dynamics.*

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## 1 Introduction

Atmospheric flows exhibit self-similar fractal space-time fluctuations on all space-time scales associated with inverse power law distribution or  $1/\nu$  noise, where  $\nu$  is the frequency, for power spectra of meteorological parameters such as wind, temperature, etc [60]. Such  $1/\nu$  noise imply long-range correlations, identified as self-organized criticality generic to dynamical systems in nature and are independent of the exact physical, chemical, physiological and other properties of the dynamical system.

Mathematical modelling of Nonlinear dynamics is a multidisciplinary area of intensive study in recent years. Real world and mathematical models of dynamical systems are characterized by unpredictable or chaotic fractal fluctuations. Simple mathematical models of dynamical systems exhibit very complicated or chaotic dynamics [3]. A review of the several real world models was presented by Martunyuk [62] to illustrate the importance of the theory and applications of the general methods of stability analysis [5, 22, 23]. Slane and Tragesser [89] analysed a class of near-periodic systems in which the dynamics can be described by a set of nonlinear differential equations with no known equilibrium solution. Special solutions to rotating stratified Boussinesq equations were discussed by Desale and Sharma [24]. A fuzzy approach for the optimal design of robust control for uncertain systems was proposed by Chen [19]. Fuzzy theory was originally introduced to describe information (for example, the linguistic information) that is in lack of a sharp boundary with its environment [103].

The signature of fractals, namely, inverse power law form for power spectra of fluctuations was identified for isotropic homogeneous turbulence by Kolmogorov in the 1940s. The concept of fractals and its quantitative measure for space-time fluctuations of all scales was introduced by Mandelbrot in the late 1960s. The robust pattern of selfsimilar space-time fluctuations was identified by Bak, Tang and Wiesenfeld in the late 1980s as self-organised criticality (SOC) whereby the cooperative existence of fluctuations of all space-time scales maintains the dynamical equilibrium in dynamical systems. In this paper the author presents a general systems theory model applicable to all dynamical systems. The quantitative characteristics of the observed fractal space-time fluctuations and SOC are derived directly as a natural consequence of model concepts based on collective statistical probabilities of fluctuations such as in kinetic theory of gases as explained in the following. Visconti [97] states that according to Edward Lorenz the atmosphere may be intrinsically unpredictable. Today there is no theory that could predict the evolution of a cloud in the presence of updraft, wind, humidity advection, etc. In a completely different context, the kinetic theory of gases solves another impossible problem and avoids the question of how to describe the exact position of each molecule in a gas. Instead it gives their collective properties, describing their statistical behavior [97]. The most important problem of statistical mechanics is the Kinetic Theory of Gases. Notions like pressure, temperature and entropy were based on the statistical properties of a large number of molecules [20]. Recent work in dynamical systems theory has shown that many properties that are associated with irreversible processes in fluids can be understood in terms of the dynamical properties of reversible, Hamiltonian systems. That is, stochastic-like behavior is possible for these systems [27]. Maxwell's (1860s) and Boltzmann's (1870s) work on the kinetic theory of gases, and the creation of the more general theory of statistical mechanics persuaded many thinkers that certain very important large scale regularities – the various gas laws, and eventually the second law of thermodynamics – were indeed to be explained as the combined effect of the probability

distributions governing those systems parts [90].

A general systems theory developed by the author visualizes the fractal fluctuations to result from the coexistence of eddy fluctuations in an eddy continuum, the larger scale eddies being the integrated mean of enclosed smaller scale eddies. The model predicts that the probability distributions of component eddy amplitudes and the corresponding variances (power spectra) are quantified by the same universal inverse power law distribution which is a function of the golden mean. Atmospheric particulates are held in suspension by the vertical velocity distribution (spectrum). The atmospheric particulate size spectrum is derived in terms of the model predicted universal inverse power law characterizing atmospheric eddy energy spectrum.

Information on the size distribution of atmospheric suspended particulates (aerosols, cloud drops, raindrops) is important for the understanding of the physical processes relating to the studies in weather, climate, atmospheric electricity, air pollution and aerosol physics. Atmospheric suspended particulates affect the radiative balance of the Earth/atmosphere system via the direct effect whereby they scatter and absorb solar and terrestrial radiation, and via the indirect effect whereby they modify the microphysical properties of clouds thereby affecting the radiative properties and lifetime of clouds [36]. At present empirical models for the size distribution of atmospheric suspended particulates is used for quantitative estimation of earth-atmosphere radiation budget related to climate warming/cooling trends. The empirical models for different locations at different atmospheric conditions, however, exhibit similarity in shape implying a common universal physical mechanism governing the organization of the shape of the size spectrum. The pioneering studies during the last three decades by Lovejoy and his group [59, 60] show that the particulates are held in suspension in turbulent atmospheric flows which exhibit selfsimilar fractal fluctuations on all scales ranging from turbulence (mm-sec) to climate (kms-years). Lovejoy and Schertzer [59] have shown that the rain drop size distribution should show a universal scale invariant shape. The non-linear coupling between statistical mechanics, particle microphysics and atmospheric dynamics must be studied from scaling point of view [51].

In the present study a general systems theory for fractal space-time fluctuations developed by the author [78, 84-86] is applied to derive universal (scale independent) inverse power law distribution incorporating the golden mean for atmospheric eddy energy distribution. Atmospheric particulates are held in suspension by the spectrum of atmospheric eddy fluctuations (vertical). The suspended atmospheric particulate size distribution is expressed in terms of the atmospheric eddy energy spectrum and is expressed as a function of the golden mean  $\tau$  ( $\approx 1.618$ ), the total number concentration and the mean volume radius (or diameter) of the particulate size spectrum. A knowledge of the mean volume radius and total number concentration is sufficient to compute the total particulate size spectrum at any location. Model predicted atmospheric eddy energy spectrum is in agreement with earlier observational results [79, 81-83, 87]. Model predicted suspended particulate (aerosol) size spectrum is in agreement with observations using VOCALS 2008 PCASP-B data (Sections 8 and 9).

The paper is organized as follows. Section 2 contains the current state of knowledge of the size distribution of atmospheric suspended particulates. Section 3 contains a brief summary of the observed characteristics of selfsimilar fractal fluctuations in atmospheric flows. Section 4 summarizes the general systems theory for fractal space-time fluctuations in atmospheric flows. The normalized (scale independent) atmospheric eddy energy spectrum and the associated aerosol size spectrum are derived in Section 5. In

Section 6 it is shown that the General Systems Theory presented in this paper satisfies the Maximum Entropy Principle of classical Statistical Physics. Section 7 contains details of observational data sets used for validating the model predictions. Sections 8 and 9 contain results of analyses of the data sets and conclusions of the study respectively.

## 2 Atmospheric Suspended Particulates: Current State of Knowledge

### 2.1 Aerosol size distribution

As aerosol size is one of the most important parameters in describing aerosol properties and their interaction with the atmosphere, its determination and use is of fundamental importance. Aerosol size covers several decades in diameter and hence a variety of instruments are required for its determination. This necessitates several definitions of the diameter, the most common being the geometric diameter  $d$ . The size fraction with  $d > 1\text{-}2\ \mu\text{m}$  is usually referred to as the coarse mode, and the fraction  $d < 1\text{-}2\ \mu\text{m}$  is the fine mode. The latter mode can be further divided into the accumulation  $d \sim 0.1\text{-}1\ \mu\text{m}$ , Aitken  $d \sim 0.01\text{-}0.1\ \mu\text{m}$ , and nucleation  $d < 0.01\ \mu\text{m}$  modes. Due to the  $d^3$  dependence of aerosol volume (and mass), the coarse mode is typified by a maximum volume concentration and, similarly, the accumulation mode by the surface area concentration and the Aitken and nucleation modes by the number concentration. As the sources and sinks of the coarse and fine modes are different, there is only a weak association of particles in both modes [37]. The aerosol chemistry data organized first by Peter Mueller and subsequently analyzed by Friedlander and co-workers showed that the fine and coarse mass modes were chemically distinctly different [38]. Based on tedious and careful size distribution measurements performed over many different parts of the world, Junge and co-workers [43-46] have observed that there is a remarkable similarity in the gathered size distributions (number concentration  $N$  versus radius  $r_a$ ): they follow a power law function over a wide range from 0.1 to over  $20\ \mu\text{m}$  in particle radius [38]

$$\frac{dN}{d\log r_a} = c r_a^{-\alpha}.$$

The inverse power law exponent  $\alpha$  of the number distribution function ranged between 3 and 5 with a typical value of 4. This power-law form of the size distribution became known as the *Junge distribution* of atmospheric aerosols. In the 1960s the physical mechanisms that were responsible for maintaining the observed *quasi-stationary* size distribution of the size spectra were not known.

Whitby [99] introduced the concept of the multimodal nature of atmospheric aerosol and Jaenicke and Davies [41] added the mathematical formalism used today. Semi-quantitative explanation of the observed fine particle dynamics provided the scientific support for the bimodal concept and became the basis of regional dynamically coupled gas-aerosol models. Typically, the planetary boundary layer (PBL) aerosol is combination of three modes corresponding to Aitken nuclei, accumulation mode aerosols, and coarse aerosols, the shape of which is often modeled as the sum of lognormal modes [100, 18]. In a nutshell, the bimodal distribution concept states that the atmospheric aerosol mass is distributed in two distinct size ranges, fine and coarse and that each aerosol mode has a characteristic size distribution, chemical composition and optical properties [38].

### 3 Selfsimilar Fractal Fluctuations from Turbulence to Climate Scales in Atmospheric Flows

The Atmospheric particulates are suspended in the selfsimilar wind fluctuation pattern ranging from turbulence to climate scales manifested as inverse power law form for power spectra of temporal fluctuations of wind speed. A brief summary of observed long-range correlations on all space-time scales in atmospheric flows and implications for modeling atmospheric dynamical transport processes is given in the following.

Atmospheric flows exhibit self-similar fractal fluctuations generic to dynamical systems in nature. Self-similarity implies long-range space-time correlations identified as self-organized criticality [4]. The physics of self-organized criticality ubiquitous to dynamical systems in nature and in finite precision computer realizations of non-linear numerical models of dynamical systems is not yet identified. During the past three decades, Lovejoy and his group [60] have done extensive observational and theoretical studies of fractal nature of atmospheric flows and emphasized the urgent need to formulate and incorporate quantitative theoretical concepts of fractals in mainstream classical theory relating to Atmospheric Physics.

The empirical analyses summarized by Lovejoy and Schertzer [60], Bunde et al.[15], Bunde and Havlin [13, 14], Eichner et al. [28], Rybski et al. [73, 74], directly demonstrate the strong scale dependencies of many atmospheric fields, showing that they depend in a power law manner on the spacetime scales over which they are measured. In spite of intense efforts over more than 50 years, analytic approaches have been surprisingly ineffective at deducing the statistical properties of turbulence. Atmospheric Science labors under the misapprehension that its basic science issues have long been settled and that its task is limited to the application of known laws albeit helped by ever larger quantities of data themselves processed in evermore powerful computers and exploiting ever more sophisticated algorithms. Conclusions about anthropogenic influences on the atmosphere can only be drawn with respect to the null hypothesis, i.e. one requires a theory of the natural variability, including knowledge of the probabilities of the extremes at various resolutions. At present, the null hypotheses are classical so that they assume there are no long-range statistical dependencies and that the probabilities are thin-tailed (i.e. exponential). However observations show that cascades involve long-range dependencies and (typically) have fat tailed (algebraic) distributions in which extreme events occur much more frequently and can persist for much longer than classical theory would allow [60, 10, 11, 29, 12, 9].

A general systems theory for the observed fractal space-time fluctuations of dynamical systems developed by the author [78, 85] helps formulate a simple model to explain the observed vertical distribution of number concentration and size spectra of atmospheric aerosols. The atmospheric aerosol size spectrum is derived in terms of the universal inverse power law characterizing atmospheric eddy energy spectrum. The physical basis and the theory relating to the model are discussed in Section 4. The model predictions are (i) The fractal fluctuations can be resolved into an overall logarithmic spiral trajectory with the quasiperiodic Penrose tiling pattern for the internal structure. (ii) The probability distribution of fractal space-time fluctuations (amplitude) also represents the power (variance or square of amplitude) spectrum for fractal fluctuations and is quantified as universal inverse power law incorporating the golden mean. Such a result that the additive amplitudes of eddies when squared represent probability distribution is observed in the subatomic dynamics of quantum systems such as the electron or photon. Therefore

the irregular or unpredictable fractal fluctuations exhibit quantum-like chaos. (iii) Atmospheric aerosols are held in suspension by the vertical velocity fluctuation distribution (spectrum). The normalized (scale independent) atmospheric aerosol size spectrum is derived in terms of the universal inverse power law characterizing atmospheric eddy energy spectrum. Model predicted spectrum is in agreement (within two standard deviations on either side of the mean) with experimentally determined data sets for homogeneous aerosol size intervals (Sections 7 and 8).

#### 4 General Systems Theory for Fractal Space-Time Fluctuations in Atmospheric Flows

The study of the spontaneous, i.e., self-organized formation of structures in systems far from thermal equilibrium in open systems belongs to the multidisciplinary field of synergetics [35]. Formation of structure begins by aggregation of molecules in a turbulent fluid (gas or liquid) medium. Turbulent fluctuations are therefore not dissipative, but serve to assemble and form coherent structures [63, 69, 70, 39], for example, the formation of clouds in turbulent atmospheric flows. Traditionally, turbulence is considered dissipative and disorganized. Yet, coherent (organized) vortex roll circulations (vortices) are ubiquitous to turbulent fluid flows [93, 50, 32]. The exact physical mechanism for the formation and maintenance of coherent structures, namely vortices or large eddy circulations in turbulent fluid flows is not yet identified.

Turbulence, namely, seemingly random fluctuations of all scales, therefore, plays a key role in the formation of selfsimilar coherent structures in the atmosphere. Such a concept is contrary to the traditional view that turbulence is dissipative, i.e., ordered growth of coherent form is not possible in turbulent flows. The author [78, 85] has shown that turbulent fluctuations self-organize to form selfsimilar structures in fluid flows.

In summary, spatial integration of enclosed turbulent fluctuations give rise to large eddy circulations in fluid flows. Therefore, starting with turbulence scale fluctuations, progressively larger scale eddy fluctuations can be generated by integrating circulation structures at different scale ranges. Such a concept envisages only the magnitude (intensity) of the fluctuations and is independent of the properties of the medium in which the fluctuations are generated. Also, self-similar space-time growth structure is implicit to hierarchical growth process, i.e., the large scale structure is the envelope of enclosed smaller scale structures. Successively larger scale structures form a hierarchical network and function as a unified whole. Large eddy is the integrated mean of enclosed turbulent eddies and functions as a fuzzy logic network with two-way energy (information) flow between the scales. Fuzzy descriptions of system performance adds more insight to view the system performance [19].

The role of surface frictional turbulence in weather systems is discussed in Section 4.1. The common place occurrence of long-lived organized cloud patterns and their important contribution to the radiation budget of the earth's atmosphere is briefly discussed in Section 4.1.1.

##### 4.1 Frictional convergence induced weather

Roeloffzen et al. [72] discussed the importance of frictional convergence induced weather as follows. The coastline generally represents a marked discontinuity in surface roughness. The resulting mechanical forcing leads to a secondary circulation in the boundary layer, and consequently to a vertical motion field that may have a strong influence on the

weather in the coastal zone. In potentially unstable air masses, frictional convergence may cause a more-or-less stationary zone of heavy shower activity, for example. Of all meteorological phenomena typical for coastal regions, the fair weather sea-breeze circulation has probably been studied most extensively, e.g., Estoque [30], Walsh [98], Pielke [68], Pearson et al. [67]. In contrast to this thermally-driven circulation, the mechanical forcing due to the discontinuity in surface roughness may create circulation patterns of similar amplitude and scale. Frictional convergence is mentioned in some studies as the cause of increased precipitation in coastal zones under specific conditions, e.g., Bergeron [7], Timmerman [94], Oerlemans [64]; but its effect is generally underestimated [72].

Frictional convergence is analogous to Ekman pumping, namely, the process of inducing vertical motions by boundary layer friction [91].

Cotton and Anthes [21] emphasized the importance of the role of Ekman pumping on large scale weather systems. The strong control of cumulus convection by the larger scales of motion in tropical cyclones has been recognized for a long time. Syono et al. [92] showed that the rate of precipitation in typhoons was related to the updrafts produced by frictional convergence in the PBL (so-called Ekman pumping). Later observational and modeling studies have confirmed the cooperative interaction between cumulus convection and the tropical cyclones through frictionally induced moisture convergence and enhanced evaporation in the PBL (see review in Anthes [2]). Local winds such as Sea breezes actually are very important because they are determined mainly by the interaction of large scale motions with local topography [97].

New research suggests that rough areas of land, including city buildings and naturally jagged land cover like trees and forests can actually attract passing hurricanes. It was observed that storms traveling over river deltas hold together longer than those over dry ground. As a result, the city of New Orleans might feel a greater impact of hurricanes coming off the Gulf of Mexico than existing computer models predict [61].

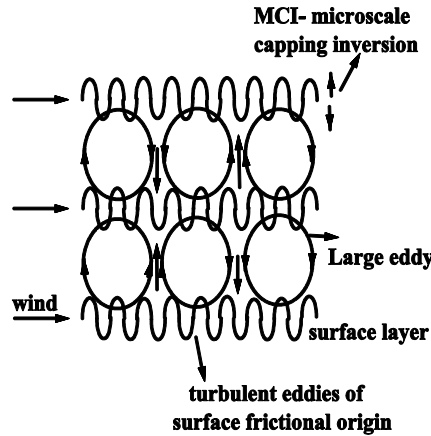
#### 4.1.1 Mesoscale cellular convection and radiation budget of the earth

Feingold et al. [31] discussed the importance of the observed large scale organized pattern of clouds in the radiation budget of the earth's atmosphere. Cloud fields adopt many different patterns that can have a profound effect on the amount of sunlight reflected back to space, with important implications for the Earth's climate. These cloud patterns can be observed in satellite images of the Earth and often exhibit distinct cell-like structures associated with organized convection at scales of tens of kilometers [49, 1, 33], i.e. mesoscale cellular convection. These clouds are important because they increase the reflectance of shortwave radiation and therefore exert a cooling effect on the climate system that is not compensated by appreciable changes in outgoing longwave radiation [96].

#### 4.2 Growth of macro-scale coherent structures from microscopic domain fluctuations in atmospheric flows

The non-deterministic model [78, 85, 86] incorporates the physics of the growth of macro-scale coherent structures from microscopic domain fluctuations in atmospheric flows. In summary, the mean flow at the planetary ABL possesses an inherent upward momentum flux of *frictional origin* at the planetary surface. This turbulence-scale upward momentum flux is progressively amplified by the exponential decrease of the atmospheric density with height coupled with the buoyant energy supply by micro-scale fractional

condensation on hygroscopic nuclei, even in an unsaturated environment [71]. The mean large-scale upward momentum flux generates helical vortex-roll (or large eddy) circulations in the planetary atmospheric boundary layer and under favourable conditions of moisture supply, is manifested as cloud rows and (or) streets, and mesoscale cloud clusters MCC in the global cloud cover pattern. A conceptual model of large and turbulent eddies in the planetary ABL is shown in Figures 1 and 2. The mean airflow at the planetary surface carries the signature of the fine scale features of the planetary surface topography as turbulent fluctuations with a net upward momentum flux. This persistent upward momentum flux of *surface frictional origin* generates large-eddy (or vortex-roll) circulations, which carry upward the turbulent eddies as internal circulations. Progressive upward growth of a large eddy occurs because of buoyant energy generation in turbulent fluctuations as a result of the latent heat of condensation of atmospheric water vapour on suspended hygroscopic nuclei such as common salt particles. The latent heat of condensation generated by the turbulent eddies forms a distinct warm envelope or a micro-scale capping inversion layer at the crest of the large-eddy circulations as shown in Figure 1.

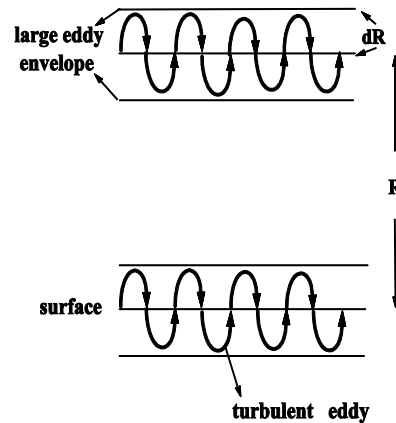


**Figure 1:** Micro-scale capping inversion (MCI) layer at the crest of the large-eddy circulations.

Progressive upward growth of the large eddy occurs from the turbulence scale at the planetary surface to a height  $R$  and is seen as the rising inversion of the daytime atmospheric boundary layer (Figure 2).

The turbulent fluctuations at the crest of the growing large-eddy mix overlying environmental air into the large-eddy volume, i.e. there is a two-stream flow of warm air upward and cold air downward analogous to superfluid turbulence in liquid helium [25, 26]. The convective growth of a large eddy in the atmospheric boundary layer therefore occurs by vigorous counter flow of air in turbulent fluctuations, which releases stored buoyant energy in the medium of propagation, e.g. latent heat of condensation of atmospheric water vapour. Such a picture of atmospheric convection is different from the traditional concept of atmospheric eddy growth by diffusion, i.e. analogous to the molecular level momentum transfer by collision. Molecules and turbulence eddies must





**Figure 2:** Progressive upward growth of the large eddy from the turbulence scale at the planetary surface.

have influence on atmospheric particles. However most theories on particle diffusional growth emphasize molecular effects, e.g., based on classical transport laws (Fick's first law for mass diffusion, Fourier law for heat diffusion) whereas the effects of turbulence are underestimated [51].

The generation of turbulent buoyant energy by the micro-scale fractional condensation is maximum at the crest of the large eddies and results in the warming of the large-eddy volume. The turbulent eddies at the crest of the large eddies are identifiable by a micro-scale capping inversion that rises upward with the convective growth of the large eddy during the course of the day. This is seen as the rising inversion of the daytime planetary boundary layer in echosonde and radiosonde records and has been identified as the entrainment zone [8, 34] where mixing with the environment occurs.

The general systems theory for eddy growth discussed so far for planetary atmospheric boundary layer (ABL) can be extended up to the upper atmospheric levels. In summary, a gravity wave feedback mechanism for the vertical mass exchange between the troposphere and the stratosphere is proposed. The vertical mass exchange takes place through a chain of eddy systems. The atmospheric boundary layer (ABL) contains large eddies (vortex rolls) which carry on their envelopes turbulent eddies of *surface frictional origin* [76, 78, 85]. The buoyant energy production by *microscale-fractional-condensation* (MFC) in turbulent eddies is responsible for the sustenance and growth of large eddies [77, 78, 85]. The buoyant energy production of turbulent eddies by the *microscale-fractional-condensation* (MFC) process is maximum at the crest of the large eddies and results in the warming of the large eddy volume. The turbulent eddies at the crest of the large eddies are identifiable by a *microscale-capping-inversion* (MCI) layer which rises upwards with the convective growth of the large eddy in the course of the day. The MCI layer is a region of enhanced aerosol concentrations. As the *microscale-fractional-condensation* (MFC) generated warm parcel of air corresponding to the large eddy rises in the stable environment of the *microscale-capping-inversion* (MCI), Brunt Vaisala oscillations are

generated [77, 78, 85]. The growth of the large eddy is associated with generation of a continuous spectrum of gravity (buoyancy) waves in the atmosphere. The atmosphere contains a stack of large eddies. Vertical mixing of overlying environmental air into the large eddy volume occurs by turbulent eddy fluctuations [76, 78, 85]. The circulation speed of the large eddy is related to that of the turbulent eddy according to the following expression [95, 78]

$$W^2 = \frac{2r}{\pi R} w^2. \quad (1)$$

In the above equation (1),  $W$  and  $w$  are respectively the r.m.s (root mean square) circulation speeds of the large and turbulent eddies and  $R$  and  $r$  are their respective radii.

The relationship between the time scales  $T$  and  $t$  respectively of the large and turbulent eddies can be derived in terms of the circulation speeds  $W$  and  $w$  and their respective length scales  $R$  and  $r$  from equation (1) [78] as follows

$$T = \frac{2\pi R}{W} \text{ and } t = \frac{2\pi r}{w},$$

$$\frac{T}{t} = \frac{Rw}{rW} = \frac{R}{r} \sqrt{\frac{\pi R}{2r}} = \left[ \frac{R}{r} \right]^{\frac{3}{2}} \sqrt{\frac{\pi}{2}}.$$

As seen from Figures 1 and 2 and from the concept of eddy growth, vigorous counter flow (mixing) characterizes the large-eddy volume. The total fractional volume dilution rate of the large eddy by vertical mixing across unit cross-section is derived from equation (1) [76, 78, 85] and is given as follows

$$k = \frac{wr}{dWR}. \quad (2)$$

In equation (2),  $w$  is the increase in vertical velocity per second of the turbulent eddy due to microscale fractional condensation (MFC) process and  $dW$  is the corresponding increase in vertical velocity of large eddy.

The fractional volume dilution rate  $k$  is equal to 0.4 for the scale ratio  $z$ , i.e.  $R/r = 10$ . Identifiable large eddies can exist in the atmosphere for scale ratios more than 10 only since, for smaller scale ratios the fractional volume dilution rate  $k$  becomes more than half. Thus atmospheric eddies of various scales, i.e., convective, meso-, synoptic and planetary scale eddies are generated by successive decadic scale range eddy mixing process starting from the basic turbulence scale [77, 78, 85]. From equation (2) the following logarithmic wind profile relationship for the ABL is obtained [76, 78, 85].

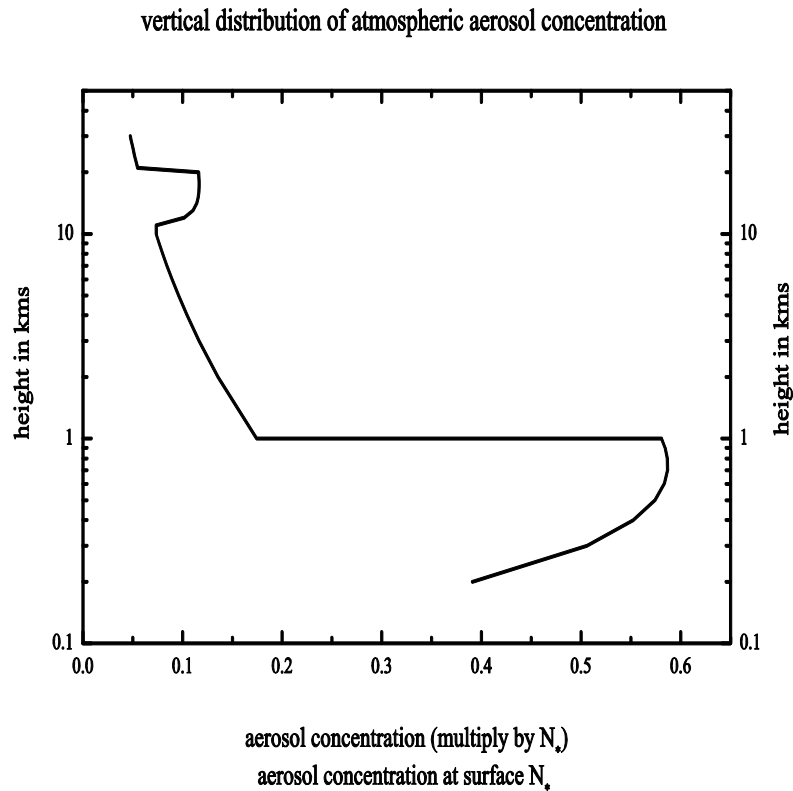
$$W = \frac{w}{k} \ln z. \quad (3)$$

The steady state fractional upward mass flux  $f$  of surface air at any height  $z$  can be derived using equation (3) and is given by the following expression [76, 78, 85].

$$f = \sqrt{\frac{2}{\pi z}}. \quad (4)$$

In equation (4)  $f$  represents the steady state fractional volume of surface air at any level  $z$ . Since atmospheric aerosols originate from surface, the vertical profile of mass and number concentration of aerosols follow the  $f$  distribution.

The magnitude of the steady state vertical aerosol mass flux is dependent on  $m_*$ , the aerosol mass concentration at the initial level (earth surface) and is equal to  $m_*f$  from (4), the non-zero values of  $f$  being given in terms of the non-dimensional length scale ratio  $z$ . Similarly the aerosol number concentration  $N$  at normalized height  $z$  is equal to  $N_*f$ , where  $N_*$  is the number concentration at the surface.



**Figure 3:** Model predicted aerosol vertical distribution.

The aerosol concentration vertical profile in Figure 3 is computed using equation (4) with appropriate length scale ratio  $z$  values corresponding to the associated steady state fractional volume dilution  $k$  values (2). The fractional volume dilution rate  $k$  is equal to 0.4 for the scale ratio  $z$ , i.e.,  $R/r = 10$ . Identifiable large eddies can exist in the atmosphere only for scale ratios more than 10 since, for smaller scale ratios the fractional volume dilution rate  $k$  becomes more than half. Thus atmospheric eddies of various scales, i.e., convective, meso-, synoptic and planetary scale eddies are generated by successive decadic scale range eddy mixing process starting from the basic turbulence scale [42, 76–87].

The peaks in the aerosol concentration at 1 km (lifting condensation level) and at about 10–15 km (stratosphere) identify the microscale capping inversion (MCI, Figure 1) at the crests of the convective and meso-scale eddies respectively, the appropriate

scale ratios for the convective and meso-scale eddies being 10 and 100 with respect to the turbulence scale. Thus for the turbulent eddy of radius 100m, the MCI's for the convective and meso-scale eddies occur at 1 km and 10 km respectively.

The model predicted profiles closely resemble the observed profiles associated with major quasi-permanent tropospheric inversion (temperature) layers reported by other investigators [46].

The vertical mass exchange mechanism predicts the  $f$  distribution for the steady state vertical transport of aerosols at higher levels. Thus aerosol injection into the stratosphere by volcanic eruptions gives rise to the enhanced peaks in the regions of microscale capping inversion (MCI) in the stratosphere and other higher levels determined by the radius of the dominant turbulent eddy at that level.

The time  $T$  taken for the steady state aerosol concentration  $f$  to be established at the normalised height  $z$  is equal to the time taken for the large eddy to grow to the height  $z$  and is computed using the following relation (see Section 4.3.2 below)

$$T = \frac{r}{w} \sqrt{\frac{\pi}{2}} \text{li} \sqrt{z}. \quad (5)$$

In equation (5),  $\text{li}$  is the logarithm integral.

The vertical dispersion rate of aerosols/pollutants from known sources (e.g., volcanic eruptions, industrial emissions) can be computed using the relation for  $f$  and  $T$  (equations 4 and 5).

### 4.3 Computations of model predictions and comparison with observations

#### 4.3.1 Vertical velocity profile

The microscale fractional condensation (MFC) generated values of vertical velocity have been calculated for different heights above the surface for clear-air conditions and above the cloud-base for in-cloud conditions for a representative tropical environment with favourable moisture supply. A representative cloud-base height is considered to be 1000m above sea level (a.s.l) and the corresponding meteorological parameters are, surface pressure 1000 mb, surface temperature 30 °C, relative humidity at the surface 80%, turbulent length scale 1 cm. The values of the latent heat of vapourisation  $L_v$  and the specific heat of air at constant pressure  $C_p$  are 600 cal  $\text{gm}^{-1}$  and 0.24 cal  $\text{gm}^{-1}$  respectively. The density of air at surface is 1.1495  $\text{Kg m}^{-3}$ . The ratio values of  $m_w/m_0$ , where  $m_0$  is the mass of the hygroscopic nuclei per unit volume of air and  $m_w$  is the mass of water condensed on  $m_0$ , at various relative humidities as given by Winkler and Junge [101, 102] have been adopted and the value of  $m_w/m_0$  is equal to about 3 for relative humidity 80%. For a representative value of  $m_0$  equal to 100g  $\text{m}^{-3}$  the temperature perturbation  $\theta'$  is equal to 0.00065°C and the corresponding vertical velocity perturbation (turbulent)  $w_*$  is computed and is equal to 21.1x10<sup>-4</sup> cm sec<sup>-1</sup> from the following relationship between the corresponding virtual potential temperature  $\theta_v$ , and the acceleration due to gravity  $g$  equal to 980.6 cm sec<sup>-2</sup>

$$w_* = \frac{g}{\theta_v} \theta'.$$

Heat generated by condensation of water equal to 300  $\mu\text{g}$  on 100  $\mu\text{g}$  of hygroscopic nuclei per metre<sup>3</sup> generates vertical velocity perturbation  $w_*$  (cm sec<sup>-2</sup>) equal to 21.1x10<sup>-4</sup> cm sec<sup>-2</sup> at surface levels. In the following it is shown that a value of  $w_*$  equal to 30x10<sup>-7</sup>

$\text{cm sec}^{-2}$ , i.e. about three orders of magnitude less than that shown in the above example is sufficient to generate clouds as observed in practice.

From the logarithmic wind profile relationship (3) and the steady state fractional upward mass flux  $f$  of surface air at any height  $z$  (4) the corresponding vertical velocity perturbation  $W$  can be expressed in terms of the primary vertical velocity perturbation  $w_*$  as

$$W = w_* f z. \quad (6)$$

$W$  may be expressed in terms of the scale ratio  $z$  as given below.

From equation (4)

$$f = \sqrt{\frac{2}{\pi z}} \ln z.$$

Therefore,

$$W = w_* z \sqrt{\frac{2}{\pi z}} \ln z = w_* \sqrt{\frac{2z}{\pi}} \ln z.$$

The steady state values of large eddy vertical velocity perturbation  $W$  equal to  $w_* f z$  at normalized heights  $z$  produced by the constant value of primary perturbation  $w_*$  generated by microscale fractional condensation at surface levels are computed from (6) and given in the Table 1.

Height $z$ above surface i.e., large eddy radius	Vertical velocity perturbation $W = w_* f z \text{ cm sec}^{-1}$
1 cm	$30 \times 10^{-7}$
100 cm	$1.10 \times 10^{-4}$
100 m	$2.20 \times 10^{-3}$
1000 m	$8.71 \times 10^{-3} \approx 0.01$

**Table 1:** The steady state values of large eddy vertical velocity perturbation  $W$ .

Microscale fractional condensation generated turbulent eddy perturbation speed increases progressively with height  $z$  from surface, the turbulent eddies being carried on the envelope of large eddy circulation of radius  $z$  and may be visualized as follows:

The 1 cm eddy at the surface generates perturbation speed  $w_*$  by microscale fractional condensation. At height  $z$  cm corresponding to the envelope of large eddy of  $z$  cm radius, the 1 cm eddy carried on the envelope of the large eddy has a perturbation speed of  $w_* f z$ . Thus at 1000m corresponding to large eddy radius 1000m, the 1cm eddy on the envelope has a perturbation speed of  $0.01 \text{ cm sec}^{-1}$ . The large eddy circulation time period at height  $z$  can be expressed in terms of the primary 1 cm radius eddy perturbation speed on its envelope as given in (5) (see Section 4.3.2 for the detailed derivation).

The above values of microscale fractional condensation related vertical velocities, although small in magnitude, are present for long enough periods in the lower levels and contribute for the formation and development of clouds as explained below.

### 4.3.2 Large eddy growth time

The time required for the large eddy of radius  $R$  to grow from the primary turbulence scale radius  $r_*$  is computed as follows

$$\text{The scale ratio } z = \frac{R}{r_*}.$$

Therefore for constant turbulence radius  $r_*$

$$dz = \frac{dR}{r_*}.$$

The incremental growth  $dR$  of large eddy radius is equal to

$$dR = r_* dz.$$

The time  $dt$  for the incremental cloud growth is expressed as follows

$$dt = \frac{dR}{W} = \frac{r_* dz}{W}.$$

$W$  is the increase in large eddy circulation speed resulting from enclosed turbulent eddy circulations of speed  $w_*$  and is given as  $W = w_* f z$  from (6). Therefore

$$\begin{aligned} dt &= \frac{r_* dz}{w_* f z} = \frac{r_* dz}{w_* z \sqrt{\frac{2}{\pi z} \ln z}}, \\ t &= \frac{r_*}{w_*} \sqrt{\frac{\pi}{2}} \int_2^z \frac{dz}{\sqrt{z} \ln z}. \end{aligned}$$

The above equation can be written in terms of  $\sqrt{z}$  as follows

$$\begin{aligned} \sqrt{z} &= \frac{dz}{2\sqrt{z}}, \\ dz &= 2\sqrt{z} d(\sqrt{z}). \end{aligned}$$

Therefore,

$$\begin{aligned} t &= \frac{r_*}{w_*} \sqrt{\frac{\pi}{2}} \int_{x_1}^{x_2} \frac{d\sqrt{z}}{\ln \sqrt{z}} = \frac{r_*}{w_*} \sqrt{\frac{\pi}{2}} \int_{x_1}^{x_2} \text{li}(\sqrt{z}), \\ x_1 &= \sqrt{z_1}, \quad x_2 = \sqrt{z_2}. \end{aligned}$$

In the above equation  $z_1$  and  $z_2$  refer respectively to lower and upper limits of integration and  $\text{li}$  is the Soldner's integral or the logarithm integral. The large eddy growth time  $t$  (equation 5 in Section 4.2) can be computed from the above equation in terms of the internal primary small eddy radius  $r_*$  (equal to 1 cm) and the corresponding eddy acceleration  $w_* f z$ .

Starting from surface, the time  $t$  seconds taken for the evolution of the 1000m ( $10^5$  cm) eddy from the 1 cm radius ( $r_*$ ) eddy energized by the microscale fractional condensation (MFC) induced primary perturbation  $w_* f z$  equal to  $0.01 \text{ cm sec}^{-2}$  can be

computed from the above equation by substituting for  $z_1 = 1\text{cm}$  and  $z_2 = 10^5\text{ cm}$  such that  $x_1 = \sqrt{1}=1$  and  $x_2=\sqrt{10^5} \approx 317$ ,

$$t = \frac{1}{.001} \sqrt{\frac{\pi}{2}} \int_1^{317} \text{li}\sqrt{z}.$$

The value of  $\int_1^{317} \text{li}\sqrt{z}$  is equal to 71.3. Hence  $t \approx 8938\text{ sec} \approx 2\text{ hrs } 30\text{ mins}$ .

Thus starting from the surface level cloud growth begins after a time period of 2 hrs 30 mins. This is consistent with the observations that convective cloud growth is visible in the afternoon hours.

## 5 Atmospheric Aerosol Size Spectrum

### 5.1 Vertical variation of aerosol number concentration

The atmospheric eddies hold in suspension the aerosols and thus the mass size spectrum of the atmospheric aerosols is dependent on the vertical velocity fluctuation spectrum of the atmospheric eddies as explained in the following. The distribution of atmospheric aerosols is not only determined by turbulence, but also by dry and wet chemistry, sedimentation, gas to particle conversion, coagulation, (fractal) variability at the surface, amongst others. However, at any instant, the mass (and therefore the radius for homogeneous aerosols) size distribution of atmospheric suspensions (aerosols) is directly related to the wind vertical velocity (eddy energy) spectrum which is shown to be universal (scale independent). The source for aerosols in the fine mode (diameter less than  $1\text{ }\mu\text{m}$ ) and coarse mode (diameter greater than  $1\text{ }\mu\text{m}$ ) are different [38] and may account for the differences in the departures of the observed from model predicted radius size spectrum for the fine and coarse aerosol modes (Section 6).

From the logarithmic wind profile relationship (3) and the steady state fractional upward mass flux  $f$  of surface air at any height  $z$  (4) the vertical velocity perturbation  $W$  is expressed in (6) as

$$W = w_* f z.$$

The corresponding moisture content  $q$  at height  $z$  is related to the moisture content  $q_*$  at the surface (or reference level) and is given as (6)

$$q = q_* f z. \quad (7)$$

The aerosols are held in suspension by the eddy vertical velocity perturbations. Thus the suspended aerosol mass concentration  $m$  at any level  $z$  will be directly related to the vertical velocity perturbation  $W$  at  $z$ , i.e.,  $W \approx mg$  where  $g$  is the acceleration due to gravity. Therefore

$$m = m_* f z. \quad (8)$$

In (8),  $m_*$  is the suspended aerosol (homogeneous) mass concentration in the surface layer. Let  $r_a$  and  $N$  represent the mean volume radius and number concentration of aerosols at level  $z$ . The variables  $r_{as}$  and  $N_*$  relate to corresponding parameters at the surface levels. Substituting for the average mass concentration in terms of mean radius  $r_a$  and number concentration  $N$  at normalized height  $z$  above surface

$$\frac{4}{3} \pi r_a^3 N = \frac{4}{3} \pi r_{as}^3 N_* f z. \quad (9)$$

The number concentration  $N$  of aerosol decreases with normalised height  $z$  according to the  $f$  distribution as shown earlier in Section 4.2 and is expressed as follows:

$$N = N_* f. \quad (10)$$

## 5.2 Vertical variation of aerosol mean volume radius

The mean volume radius of aerosol increases with height (eddy radius)  $z$  as shown in the following. At any height  $z$ , the fractal fluctuations (of wind, temperature, etc.) carry the signatures of eddy fluctuations of all size scales since the eddy of length scale  $z$  encloses smaller scale eddies and at the same time forms part of internal circulations of eddies larger than length scale  $z$ .

The wind velocity perturbation  $W$  is represented by an eddy continuum of corresponding size (length) scales  $z$ . The aerosol mass flux across unit cross-section per unit time is obtained by normalizing the velocity perturbation  $W$  with respect to the corresponding length scale  $z$  to give the volume flux of air equal to  $Wz$  and can be expressed as follows from (6):

$$Wz = (w_* f z) z. \quad (11)$$

The corresponding normalized moisture flux perturbation is equal to  $qz$  where  $q$  is the moisture content per unit volume at level  $z$ . Substituting for  $q$  from (7)

$$\text{normalised moisture flux at level } z = q_* f z^2. \quad (12)$$

The moisture flux increases with height resulting in increase of mean volume radius of CCN (cloud condensation nuclei) because of condensation of water vapour. The corresponding CCN (aerosol) mean volume radius  $r_a$  at height  $z$  is given in terms of the aerosol number concentration  $N$  at level  $z$  and mean volume radius  $r_{as}$  at the surface (or reference level) as follows from (12)

$$\frac{4}{3}\pi r_a^3 = \frac{4}{3}\pi r_{as}^3 N_* f z^2. \quad (13)$$

Substituting for  $N$  from (10) in terms of  $N_*$  and  $f$

$$\begin{aligned} r_a^3 &= r_{as}^3 z^2, \\ r_a &= r_{as} z^{\frac{2}{3}}. \end{aligned} \quad (14)$$

The mean aerosol size increases with height according to the cube root of  $z^2$  (14). As the large eddy grows in the vertical, the aerosol size spectrum extends towards larger sizes while the total number concentration decreases with height according to the  $f$  distribution. The atmospheric aerosol size spectrum is dependent on the eddy energy spectrum and may be expressed in terms of the recently identified universal characteristics of fractal fluctuations generic to atmospheric flows [86, 87] as shown in Section 5.3 below.

## 5.3 Probability distribution of fractal fluctuations in atmospheric flows

The atmospheric eddies hold in suspension the aerosols and thus the size spectrum of the atmospheric aerosols is dependent on the vertical velocity spectrum of the atmospheric eddies. Atmospheric air flow is turbulent, i.e., consists of irregular fluctuations of all space-time scales characterized by a broadband spectrum of eddies. The suspended



aerosols will also exhibit a broadband size spectrum closely related to the atmospheric eddy energy spectrum.

Atmospheric flows exhibit self-similar fractal fluctuations generic to dynamical systems in nature such as fluid flows, heart beat patterns, population dynamics, spread of forest fires, etc. Power spectra of fractal fluctuations exhibit inverse power law of form  $\nu^{-\alpha}$  where  $\alpha$  is a constant indicating long-range space-time correlations or persistence. Inverse power law for power spectrum indicates scale invariance, i.e., the eddy energies at two different scales (space-time) are related to each other by a scale factor ( $\alpha$  in this case) alone independent of the intrinsic properties such as physical, chemical, electrical etc of the dynamical system.

A general systems theory for turbulent fluid flows predicts that the eddy energy spectrum, i.e., the variance (square of eddy amplitude) spectrum is the same as the probability distribution  $P$  of the eddy amplitudes, i.e. the vertical velocity  $W$  values. Such a result that the additive amplitudes of eddies, when squared, represent the probabilities is exhibited by the subatomic dynamics of quantum systems such as the electron or photon. Therefore, the unpredictable or irregular fractal space-time fluctuations generic to dynamical systems in nature, such as atmospheric flows is a signature of quantum-like chaos. The general systems theory for turbulent fluid flows predicts [78, 84-87] that the atmospheric eddy energy spectrum follows inverse power law form incorporating the golden mean  $\tau$  [86, 87] and the normalized deviation for values of  $\sigma \geq 1$  and  $\sigma \leq -1$  as given below

$$P = \tau^{-4\sigma}. \quad (15)$$

The vertical velocity  $W$  spectrum will therefore be represented by the probability distribution  $P$  for values of  $\sigma \geq 1$  and  $\sigma \leq -1$  given in (15) since fractal fluctuations exhibit quantum-like chaos as explained above:

$$W = P = \tau^{-4\sigma}. \quad (16)$$

Values of the normalized deviation  $\sigma$  in the range  $-1 < \sigma < 1$  refer to regions of primary eddy growth where the fractional volume dilution  $k$  (2) by eddy mixing process has to be taken into account for determining the probability distribution  $P$  of fractal fluctuations (see Section 5.4 below).

#### 5.4 Primary eddy growth region fractal space-time fluctuation probability distribution

Normalized deviation  $\sigma$  ranging from -1 to +1 corresponds to the primary eddy growth region. In this region the probability  $P$  is shown to be equal to  $P = \tau^{-4k}$  (see below), where  $k$  is the fractional volume dilution by eddy mixing (2).

For the primary eddy growth region, the normalized deviation  $\sigma$  represents the length step growth number for growth stages more than one. The first stage of eddy growth is the primary eddy growth starting from unit length scale perturbation, the complete eddy forming at the tenth length scale growth, i.e.,  $R = 10r$  and scale ratio  $z$  equals to 10 [78, 85]. The steady state fractional volume dilution  $k$  of the growing primary eddy by internal smaller scale eddy mixing is given by (2) as

$$k = \frac{wr}{WR}. \quad (17)$$

The expression for  $k$  in terms of the length scale ratio  $z$  equal to  $\frac{R}{r}$  is obtained from (1) as

$$k = \sqrt{\frac{\pi}{2z}}. \quad (18)$$

A fully formed large eddy length  $R = 10r$  ( $z=10$ ) represents the average or mean level zero and corresponds to a maximum of 50% probability of occurrence of either positive or negative fluctuation peak at normalized deviation  $\sigma$  value equal to zero by convention. For intermediate eddy growth stages, i.e.,  $z$  less than 10, the probability of occurrence of the primary eddy fluctuation does not follow conventional statistics, but is computed as follows taking into consideration the fractional volume dilution of the primary eddy by internal turbulent eddy fluctuations. Starting from unit length scale fluctuation, the large eddy formation is completed after 10 unit length step growths, i.e., a total of 11 length steps including the initial unit perturbation. At the second step ( $z = 2$ ) of eddy growth the value of normalized deviation  $\sigma$  is equal to  $1.1 - 0.2 (= 0.9)$  since the complete primary eddy length plus the first length step is equal to 1.1. The probability of occurrence of the primary eddy perturbation at this  $\sigma$  value however, is determined by the fractional volume dilution  $k$  which quantifies the departure of the primary eddy from its undiluted average condition and therefore represents the normalized deviation  $\sigma$ . Therefore, the probability density  $P$  of fractal fluctuations of the primary eddy is given using the computed value of  $k$  as shown in the following equation

$$P = \tau^{-4k}. \quad (19)$$

The vertical velocity  $W$  spectrum will therefore be represented by the probability density distribution  $P$  for values of  $-1 \leq \sigma \leq 1$  given in (19) since fractal fluctuations exhibit quantum-like chaos as explained above (16):

$$W = P = \tau^{-4k}. \quad (20)$$

The probabilities of occurrence ( $P$ ) of the primary eddy for a complete eddy cycle either in the positive or negative direction starting from the peak value ( $\sigma = 0$ ) are given for progressive growth stages ( $\sigma$  values) in the following Table 2. The statistical normal probability density distribution corresponding to the normalized deviation  $\sigma$  values are also given in the Table 2.

The model predicted probability density distribution  $P$  along with the corresponding statistical normal distribution with probability values plotted on linear and logarithmic scales respectively on the left and right hand sides are shown in Fig.4. The model predicted probability distribution  $P$  for fractal space-time fluctuations is very close to the statistical normal distribution for normalized deviation  $\sigma$  values less than 2 as seen on the left hand side of Figure 4. The model predicts progressively higher values of probability  $P$  for values of  $\sigma$  greater than 2 as seen on a logarithmic plot on the right hand side of Figure 4.

### 5.5 Atmospheric wind spectrum and aerosol size spectrum

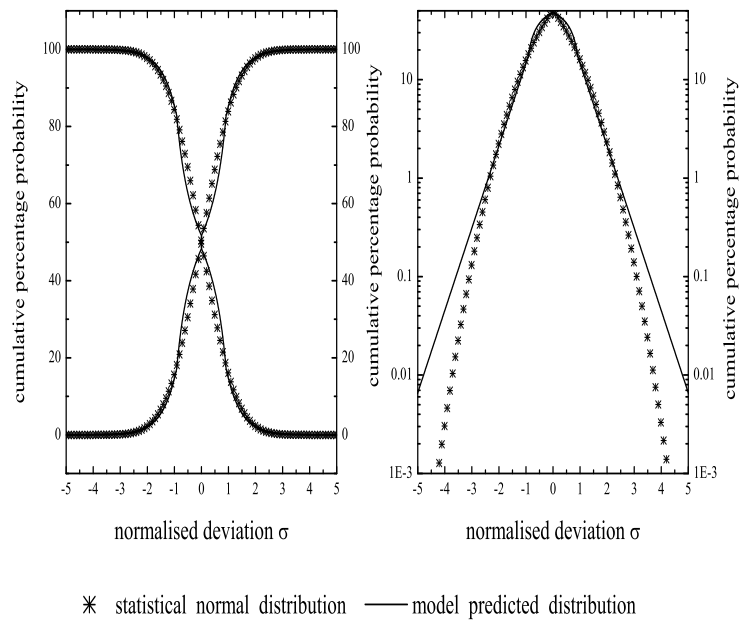
The steady state flux  $dN$  of cloud condensation nuclei (CCN) at level  $z$  in the normalized vertical velocity perturbation  $(dW)z$  is given as

$$dN = N(dW)z. \quad (21)$$

Growth step no	$\pm\sigma$	$k$	Probability(%) model predicted	Probability (%) Statistical normal
2	.9000	.8864	18.1555	18.4060
3	.8000	.7237	24.8304	21.1855
4	.7000	.6268	29.9254	24.1964
5	.6000	.5606	33.9904	27.4253
6	.5000	.5118	37.3412	30.8538
7	.4000	.4738	40.1720	34.4578
8	.3000	.4432	42.6093	38.2089
9	.2000	.4179	44.7397	42.0740
10	.1000	.3964	46.6250	46.0172
11	0	.3780	48.3104	50.0000

**Table 2:** Primary eddy growth.

fractal fluctuations probability distribution  
comparison with statistical normal distribution

**Figure 4:** Model predicted probability distribution  $P$  along with the corresponding statistical normal distribution with probability values plotted on linear and logarithmic scales respectively on the left and right hand sides.

The logarithmic wind profile relationship for  $W$  at (3) gives

$$dN = Nz \frac{w_*}{k} (d \ln z). \quad (22)$$

The general systems theory predicts universal logarithmic wind profile [78, 83] as manifested in the spiralling vortex air flows of tornadoes and the hurricane spiral cloud circulations.

Substituting for  $k$  from (2)

$$dN = Nz \frac{w_*}{w_*} W z (d \ln z) = NW z^2 (d \ln z). \quad (23)$$

The length scale  $z$  is related to the aerosol radius  $r_a$  (14). Therefore

$$\ln z = \frac{3}{2} \ln \left( \frac{r_a}{r_{as}} \right). \quad (24)$$

Defining a normalized radius  $r_{an}$  equal to  $r_a/r_{as}$ , i.e.,  $r_{an}$  represents the CCN mean volume radius  $r_a$  in terms of the CCN mean volume radius  $r_{as}$  at the surface (or reference level). Therefore,

$$\ln z = \frac{3}{2} \ln r_{an}, \quad (25)$$

$$d \ln z = \frac{3}{2} d \ln r_{an}. \quad (26)$$

Substituting for  $d \ln z$  in (23)

$$dN = NW z^2 \frac{3}{2} (d \ln r_{an}), \quad (27)$$

$$\frac{dN}{(d \ln r_{an})} = \frac{3}{2} NW z^2. \quad (28)$$

Substituting for  $W$  from equations (16) and (20) in terms of the universal probability density  $P$  for fractal fluctuations

$$\frac{dN}{(d \ln r_{an})} = \frac{3}{2} NP z^2. \quad (29)$$

The general systems theory predicts that fractal fluctuations may be resolved into an overall logarithmic spiral trajectory with the quasiperiodic Penrose tiling pattern for the internal structure such that the successive eddy lengths follow the Fibonacci mathematical series [78, 85]. The eddy length scale ratio  $z$  for length step  $\sigma$  is therefore a function of the golden mean  $\tau$  given as

$$z = \tau^\sigma. \quad (30)$$

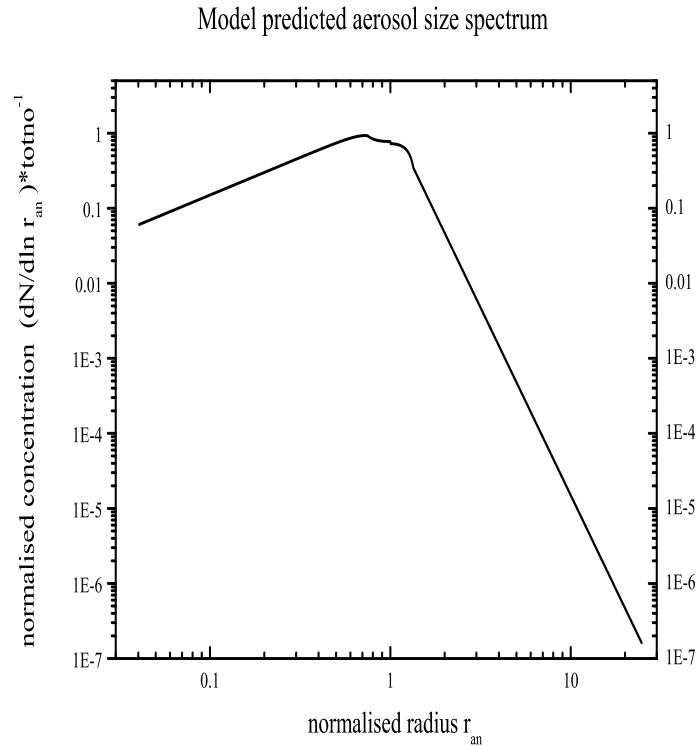
Expressing the scale length  $z$  in terms of the golden mean  $\tau$  in (29)

$$\frac{dN}{d(\ln r_{an})} = \frac{3}{2} NP \tau^{2\sigma}. \quad (31)$$

In (31)  $N$  is the steady state aerosol concentration at level  $z$ . The normalized aerosol concentration at any level  $z$  is given as

$$\frac{1}{N} \frac{dN}{d(\ln r_{an})} = \frac{3}{2} P \tau^{2\sigma}. \quad (32)$$

The fractal fluctuations probability density is  $P = \tau^{-4\sigma}$  (16) for values of the normalized deviation  $\sigma \geq 1$  and  $\sigma \leq -1$  on either side of  $\sigma = 0$  as explained earlier (Sections 5.3 and 5.4). Values of the normalized deviation  $-1 \leq \sigma \leq 1$  refer to regions of primary eddy growth where the fractional volume dilution  $k$  (2) by eddy mixing process has to be taken into account for determining the probability density  $P$  of fractal fluctuations. Therefore the probability density  $P$  in the primary eddy growth region ( $\sigma \geq 1$  and  $\sigma \leq -1$ ) is given using the computed value of  $k$  as  $P = \tau^{-4k}$  (20).



**Figure 5:** Model predicted universal (scale independent) aerosol size spectrum.

The normalized radius  $r_{an}$  is given in terms of  $\sigma$  and the golden mean  $\tau$  from equations (25) and (30) as follows

$$\begin{aligned} \ln z &= \frac{3}{2} \ln r_{an}, \\ r_{an} &= z^{\frac{2}{3}} = \tau^{\frac{2\sigma}{3}}. \end{aligned} \quad (33)$$

The normalized aerosol size spectrum is obtained by plotting a graph of normalized aerosol concentration  $\frac{1}{N} \frac{dN}{d(\ln r_{an})} = \frac{3}{2} P \tau^{2\sigma}$  (32) versus the normalized aerosol radius  $r_{an} = \tau^{\frac{2\sigma}{3}}$  (33). The normalized aerosol size spectrum is derived directly from the universal probability density  $P$  distribution characteristics of fractal fluctuations (equations 16 and 20) and is independent of the height  $z$  of measurement and is universal for aerosols in turbulent atmospheric flows. The aerosol size spectrum is computed starting from the minimum size, the corresponding probability density  $P$  (32) refers to the cumulative probability density starting from 1 and is computed as equal to  $P = 1 - \tau^{-4\sigma}$ . The universal normalized aerosol size spectrum represented by  $\frac{1}{N} \frac{dN}{d(\ln r_{an})}$  versus the normalized aerosol radius  $r_{an}$  is shown in Figure 5.

## 6 General Systems Theory and Maximum Entropy Principle of Classical Statistical Physics

Kaniadakis [47] states that the correctness of an analytic expression for a given power-law tailed distribution, used to describe a statistical system, is strongly related to the validity of the generating mechanism. In this sense the maximum entropy principle, the cornerstone of statistical physics, is a valid and powerful tool to explore new roots in searching for generalized statistical theories [47]. The concept of entropy is fundamental in the foundation of statistical physics. It first appeared in thermodynamics through the second law of thermodynamics. In statistical mechanics, we are interested in the disorder in the distribution of the system over the permissible microstates. The measure of disorder first provided by Boltzmann principle (known as Boltzmann entropy) is given by  $S = K_B \ln M$ , where  $K_B$  is the thermodynamic unit of measurement of entropy and is known as Boltzmann constant equal to  $1.38 \times 10^{-16}$  erg/ $^{\circ}$ C. The variable  $M$ , called thermodynamic probability or statistical weight, is the total number of microscopic complexions compatible with the macroscopic state of the system and corresponds to the degree of disorder or missing information [16].

The maximum entropy principle concept of classical statistical physics is applied to determine the fidelity of the inverse power law probability distribution  $P$  (15) for exact quantification of the observed space-time fractal fluctuations of dynamical systems ranging from the microscopic dynamics of quantum systems to macro-scale real world systems. The eddy energy probability distribution ( $P$ ) of fractal space-time fluctuations for each stage of hierarchical eddy growth is given by (15) derived earlier, namely

$$P = \tau^{-4t}.$$

The r.m.s circulation speed  $W$  of the large eddy follows a logarithmic relationship with respect to the length scale ratio  $z$  equal to  $R/r$  (3) as given below

$$W = \frac{w_*}{k} \log z.$$

In the above equation the variable  $k$  represents for each step of eddy growth, the fractional volume dilution of large eddy by turbulent eddy fluctuations carried on the large eddy envelope [78] and is given as (2)

$$k = \frac{w_* r}{WR}.$$

Substituting for  $k$  in (3), we have

$$W = w_* \frac{WR}{w_* r} \log z = \frac{WR}{r} \log z, \quad (34)$$

$$\text{and } \frac{r}{R} = \log z.$$

The ratio  $r/R$  represents the fractional probability  $P$  of occurrence of small-scale fluctuations ( $r$ ) in the large eddy ( $R$ ) environment. Since the scale ratio  $z$  is equal to  $r/R$ , (34) may be written in terms of the probability  $P$  as follows

$$\frac{r}{R} = \log z = \log \left( \frac{R}{r} \right) = \log \left( \frac{1}{(r/R)} \right), \quad (35)$$

$$P = \log \left( \frac{1}{P} \right) = -\log P.$$

For a probability distribution among a discrete set of states the generalized entropy for a system out of equilibrium is given as [75, 16, 6, 88]

$$S = - \sum_{j=1}^n P_j \ln P_j. \quad (36)$$

In (36)  $P_j$  is the probability for the  $j^{th}$  stage of eddy growth in the present study and the entropy  $S$  represents the missing information regarding the probabilities. Maximum entropy  $S$  signifies minimum preferred states associated with scale-free probabilities.

The validity of the probability distribution  $P$  (15) is now checked by applying the concept of maximum entropy principle [47]. Substituting for  $\log P_j$  (36) and for the probability  $P_j$  in terms of the golden mean  $\tau$  derived earlier (15) the entropy  $S$  is expressed as

$$S = - \sum_{j=1}^n P_j \ln P_j = \sum_{j=1}^n P_j^2 = \sum_{j=1}^n (\tau^{-4n})^2, \quad (37)$$

$$S = \sum_{j=1}^n \tau^{-8n} \approx 1 \text{ for large } n.$$

In (37)  $S$  is equal to the square of the cumulative probability density distribution and it increases with increase in  $n$ , i.e., the progressive growth of the eddy continuum and approaches 1 for large  $n$ . According to the second law of thermodynamics, increase in entropy signifies approach of dynamic equilibrium conditions with scale-free characteristic of fractal fluctuations and hence the probability distribution  $P$  (15) is the correct analytic expression quantifying the eddy growth processes visualized in the general systems theory. The ordered growth of the atmospheric eddy continuum is associated with maximum entropy production.

Paltridge [65] states that the principle of maximum entropy production (MEP) is the subject of considerable academic study, but is yet to become remarkable for its practical applications. The ability of a system to dissipate energy and to produce entropy "ought to be" some increasing function of the systems structural complexity. It would be nice if there were some general rule to the effect that, in any given complex system, the steady

state which produces entropy at the maximum rate would at the same time be the steady state of maximum order and minimum entropy [65].

Selvam [87] has shown that the eddy continuum energy distribution  $P$  (15) is the same as the *Boltzmann distribution* for molecular energies. The derivation of *Boltzmann's equation* from general systems theory concepts visualises the eddy energy distribution as follows: (i) The primary small-scale eddy represents the molecules whose eddy kinetic energy is equal to  $K_B T$  where  $K_B$  is the *Boltzmann's constant* and  $T$  is the temperature as in classical physics. (ii) The energy pumping from the primary small-scale eddy generates growth of progressive larger eddies [78]. The r.m.s circulation speeds  $W$  of larger eddies are smaller than that of the primary small-scale eddy (1). (iii) The space-time *fractal* fluctuations of molecules (atoms) in an ideal gas may be visualized to result from an eddy continuum with the eddy energy  $E$  per unit volume relative to primary molecular kinetic energy  $K_B T$  decreasing progressively with increase in eddy size.

The eddy energy probability distribution ( $P$ ) of fractal space-time fluctuations also represents the *Boltzmann distribution* for each stage of hierarchical eddy growth and is given by (15) derived earlier, namely

$$P = \tau^{-4t}.$$

The general systems theory concepts are applicable to all space-time scales ranging from microscopic scale quantum systems to macroscale real world systems such as atmospheric flows.

A systems theory approach based on maximum entropy principle has been applied earlier in cloud physics to obtain useful information on droplet size distributions without regard to the details of individual droplets [51-57]. Liu, Daum et al. [57] conclude that a combination of the systems idea with multiscale approaches seems to be a promising avenue. Checa and Tapiador [17] have presented a maximum entropy approach to Rain Drop Size Distribution (RDSD) modelling. Liu, Liu and Wang [58] have given a review of the concept of entropy and its relevant principles, on the organization of atmospheric systems and the principle of the Second Law of thermodynamics, as well as their applications to atmospheric sciences. The Maximum Entropy Production Principle (MEPP), at least as used in climate science, was first hypothesised by Paltridge [66].

## 7 Data

VOCALS PCASP-B data sets were used for comparison of observed with model predicted suspended particle size spectrum in turbulent atmospheric flows.

During October and November, 2008, Brookhaven National Laboratory (BNL) participated in VOCALS (VAMOS Ocean-Cloud-Atmosphere Land Study), a multi agency, multi-national atmospheric sampling field campaign conducted over the Pacific Ocean off the coast of Arica, Chile. Support for BNL came from DOE's Atmospheric Science Program (ASP) which is now part of the Atmospheric System Research (ASR) program following a merger with DOE's Atmospheric Radiation (ARM) program. A description of the VOCALS field campaign can be found at <http://www.eol.ucar.edu/projects/vocals/>

Measurements made from the DOE G-1 aircraft are being used to assess the effects of anthropogenic and biogenic aerosol on the microphysics of marine stratus. Aerosols affect the size and lifetime of cloud droplets thereby influencing the earth climate by making clouds more or less reflective and more or less long-lived. Climatic impacts resulting from interactions between aerosols and clouds have been identified by the IPCC (2007)



as being highly uncertain and it is toward the improved representation of these processes in climate models that BNL's efforts are directed.

The parent data set from which the Excel spreadsheet has been derived is archived at the BNL anonymous ftp site:

ftp://ftp.asd.bnl.gov/pub/ASP%20Field%20Programs/2008VOCALS/Processed\_Data/PCASP\_BPart/.

Data are archived as ASCII files.

### 7.1 VOCALS 2008 PCASP-B aerosol size spectrum

Data from the DOE G-1 Research Aircraft Facility operating during the 2008 VAMOS Ocean Cloud Atmosphere Land Study (VOCALS) 2008 based in part at Chacalluta Airport (ARI) north of Arica, CHILE.

PCASP\_BPart - Contains detailed size-binned (30 bins, 0.1 - 3  $\mu\text{m}$  diameter) data obtained from the PCASP (Passive Cavity Aerosol Spectrometer Probe, Unit B). This probe was on the isokinetic inlet in the cabin before 10/29/08. On flight 081029a it was moved to the nose pylon of the plane.

The following 17 data sets were used for the study, the file names giving Flight Designation (yymmddflight of day letter), 081014a\_10.txt, 081017a\_10.txt, 081018a\_10.txt, 081022a\_10.txt, 081023a\_10.txt, 081025a\_10.txt, 081026a\_10.txt, 081028a\_10.txt, 081029a\_10.txt, 081101a\_10.txt, 081103a\_10.txt, 081104a\_10.txt, 081106a\_10.txt, 081108a\_10.txt, 081110a\_10.txt, 081112a\_10.txt, 081113a\_10.txt. The letter is a for first flight. Max.Data Frequency is 10s-1 indicated as \_10 in the file name.

## 8 Analysis and Discussion of Results

The atmospheric suspended particulate size spectrum is closely related to the vertical velocity spectrum (Section 5). The mean volume radius of suspended aerosol particulates increases with height (or reference level  $z$ ) in association with decrease in number concentration. At any height (or reference level)  $z$ , the fractal fluctuations (of wind, temperature, etc.) carry the signatures of eddy fluctuations of all size scales since the eddy of length scale  $z$  encloses smaller scale eddies and at the same time forms part of internal circulations of eddies larger than length scale  $z$  (Section 5.2). The observed atmospheric suspended particulate size spectrum also exhibits a decrease in number concentration with increase in particulate radius. At any reference level  $z$  of measurement the mean volume radius  $r_{as}$  will serve to calculate the normalized radius  $r_{an}$  for the different radius class intervals as explained below.

The general systems theory for fractal space-time fluctuations in dynamical systems predicts universal mass size spectrum for atmospheric suspended particulates (Section 5). For homogeneous atmospheric suspended particulates, i.e. with the same particulate substance density, the atmospheric suspended particulate mass and radius size spectrum is the same and is given as (Section 5.5) the normalized aerosol number concentration equal to  $\frac{1}{N} \frac{dN}{d(\ln r_{an})}$  versus the normalized aerosol radius  $r_{an}$ , where (i)  $r_{an}$  is equal to  $\frac{r_a}{r_{as}}$ ,  $r_a$  being the mean class interval radius and  $r_{as}$  being the mean volume radius for the total aerosol size spectrum (ii)  $N$  is the total aerosol number concentration and  $dN$  is the aerosol number concentration in the aerosol radius class interval  $dr_a$  (iii)  $d(\ln r_{an})$  is equal to  $\frac{dr_a}{r_a}$  for the aerosol radius class interval  $r_a$  to  $r_a + dr_a$ .

### 8.1 Analysis results, VOCALS PCASP-B aerosol size spectrum

A total of 17 data sets between 14 October and 13 November 2008 are available for the study. The data used in this study for each of the 17 flights are (i) average and standard deviation for particle number concentration per cc in 29 class intervals ranging from .1 to 3  $\mu\text{m}$  for the particle diameter (ii) average and standard deviation for total particle number concentration per cc (bins 2 to 30) (iii) average and standard deviation for total volume (cc).

Details of data sets used for the study are shown in Figure 6(a - d) as follows. (i) Figure 6a: lower and upper radius size limits for bin numbers 2 to 30 (ii) Figure 6b: average and standard deviation for total particle number concentration per cc for the 17 flights (iii) Figure 6c: average and standard deviation for total volume (cc) for the 17 flights (bins 2 to 30) (iv) Figure 6d: average and standard deviation for mean volume radius ( $\mu\text{m}$ ) for the 17 flights. The dispersion (equal to standard deviation/mean) expressed as percentage gives a statistical measure of variability of measured particle number concentration. Computed dispersion (%) values are plotted for the two size ranges (i) less than 1  $\mu\text{m}$  diameter (bins 2 to 20) and (ii) 1 to 3  $\mu\text{m}$  diameter (bins 21 to 30) in Figure 7a and Figure 7b respectively.

The average total number concentration exhibits a variability of about  $\pm 100 \text{ cc}^{-1}$  around a mean value of about  $200 \text{ cc}^{-1}$  except for the first three flights which show larger variability (Figure 6b). The total volume is one order of magnitude larger for flight numbers 10 onwards compared to earlier flights (Figure 6c) consistent with larger median volume radii for flight numbers 10 onwards (Figure 6d) and exhibit large variability, particularly for size ranges more than 1  $\mu\text{m}$  (Figure 6d).

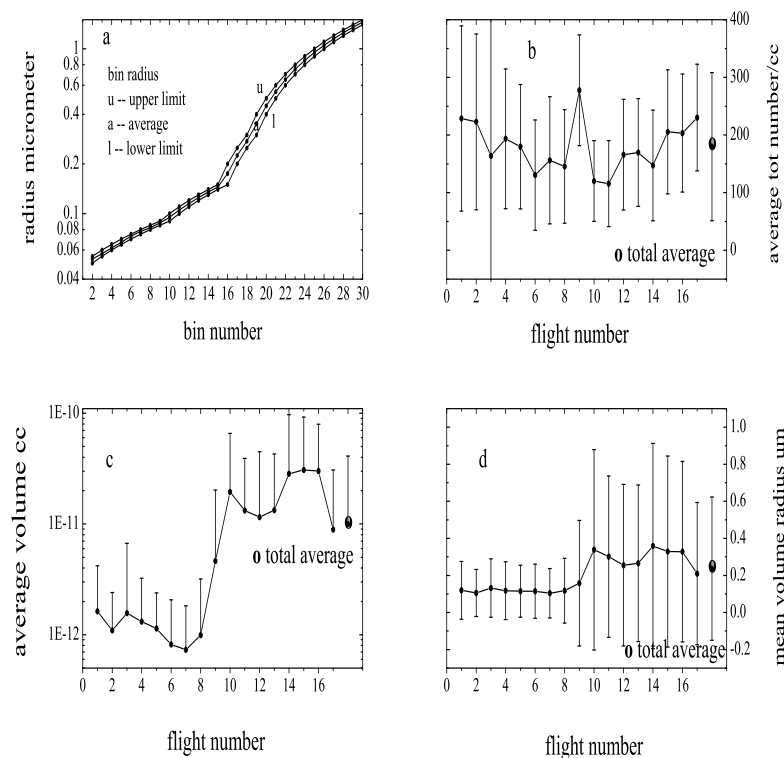
For particle diameter range less than 1  $\mu\text{m}$  (bins 2 to 20) the computed dispersion (%) for particle number concentration is within 100% for bins 2 to 14 size range and thereafter increases rapidly to a maximum of 500%. The computed dispersion (%) for particle number concentration for bins 21 to 30 (1 to 3  $\mu\text{m}$  diameter) increases steeply from 500% to nearly 5000% with increase in particle size.

The mid-point diameter of the class interval was used to compute the corresponding value of  $d(\ln r_{an})$ . The average aerosol size spectra for each of the 17 data sets are plotted on the left hand side and the total average spectrum for the 17 data sets is plotted on the right and side in Figure 8 along with the model predicted scale independent aerosol size spectrum. The corresponding standard deviations for the average spectra are shown as error bars in Figure 8.

The total average aerosol size spectrum (right hand side of Figure 8a) for size (radius) range less than about 0.5  $\mu\text{m}$  (accumulation mode) is closer to the model predicted spectrum while for particle size range greater than 0.5  $\mu\text{m}$  (coarse) the spectrum shows appreciable departure from model predicted size spectrum possibly attributed to different aerosol substance densities in the accumulation and coarse modes. The aerosol size spectra for the two different homogeneous aerosol substance densities corresponding to the two size (radius) ranges, namely (i) 0.1 to about 0.5  $\mu\text{m}$  (accumulation mode) and (ii) 0.5 to 1.5  $\mu\text{m}$  (coarse mode) were computed separately and shown in Figures 9 and 10 respectively. The observed aerosol size distribution for the two size categories now follow closely the model predicted universal size spectrum for homogeneous atmospheric suspended particulates. Earlier studies [38] have shown that the source for submicron (diameter) size accumulation mode aerosols is different from the larger (greater than 1  $\mu\text{m}$  diameter) coarse mode particles in the atmosphere and therefore may form two different homogeneous aerosol size groups.

The amount and longitudinal gradient of aerosol sulfate, and a consideration of the locations of Cu smelters and power plants in Chile, strongly suggest that the sub micron aerosol is dominated by anthropogenic emissions [48].

details of data sets (averages) for 17 flights, oct - nov 2008



error bars indicate one standard deviation on either side of mean

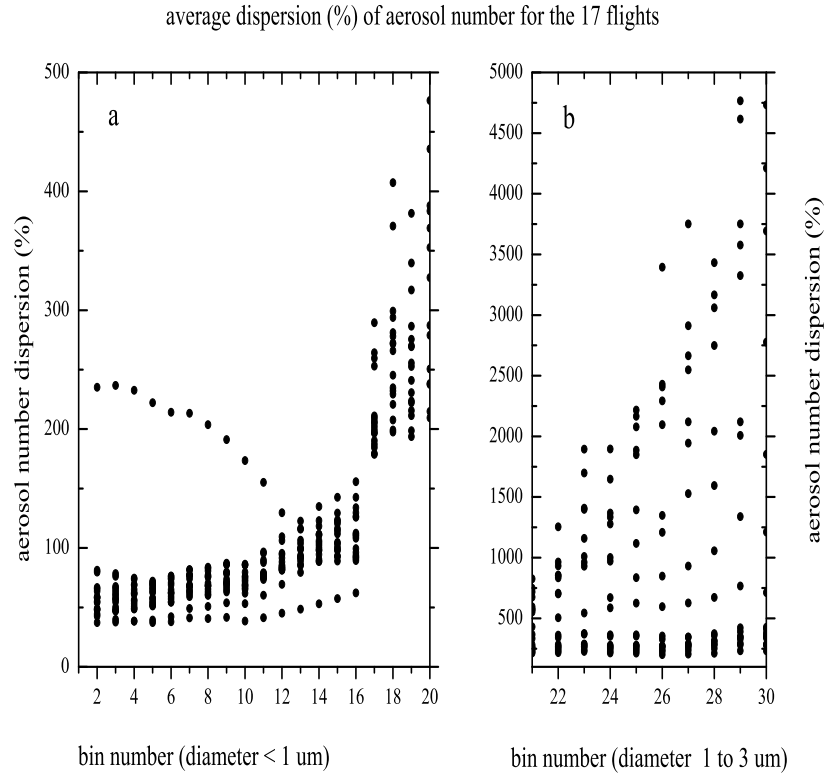
**Figure 6:** (a): lower and upper radius size limits for bin numbers 2 to 30 (b) average and standard deviation for total particle number concentration per cc for the 17 flights (c) average and standard deviation for total volume (cc) for the 17 flights (bins 2 to 30) (d) average and standard deviation for mean volume radius ( $\mu m$ ) for the 17 flights.

## 9 Conclusions

The apparently irregular (turbulent) atmospheric flows exhibit selfsimilar fractal fluctuations associated with inverse power law distribution for power spectra of meteorological parameters on all time scales signifying an eddy continuum underlying the fluctuations. A general systems theory [78] visualizes each large eddy as the envelope (average) of enclosed smaller-scale eddies, thereby generating the eddy continuum, a concept analogous to Kinetic Theory of Gases in Classical Statistical Physics. It is shown that the

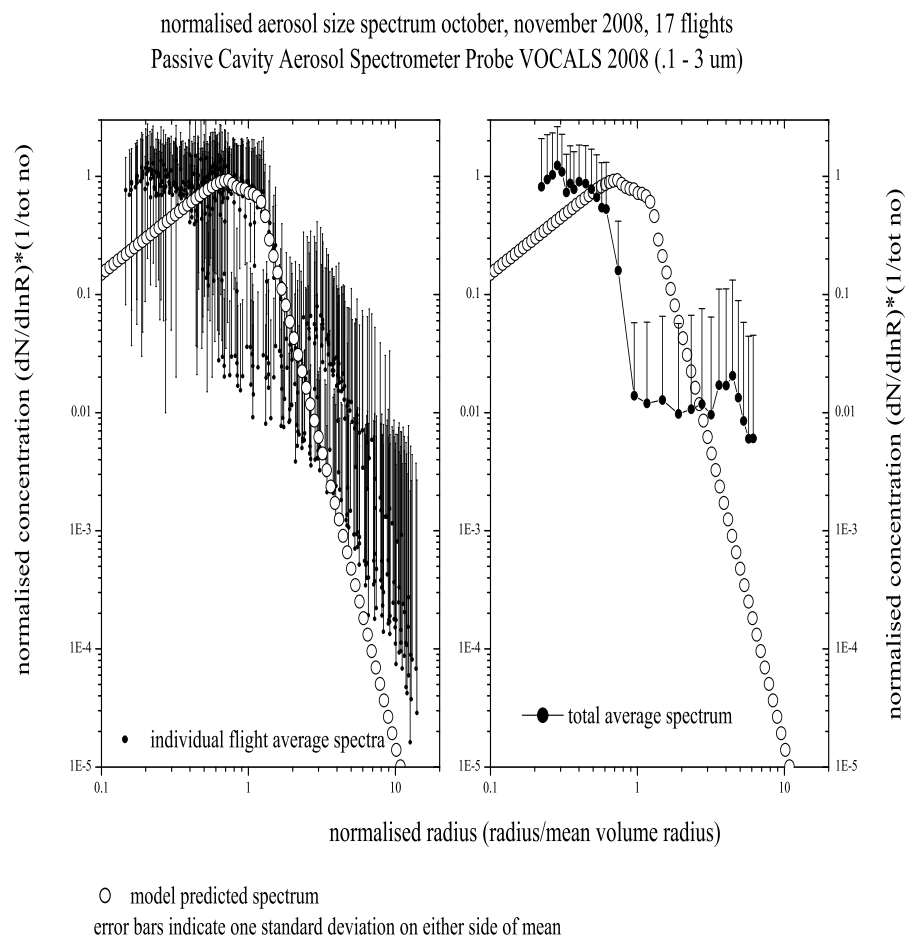
ordered growth of atmospheric eddy continuum in dynamical equilibrium is associated with Maximum Entropy Production.

Two important model predictions of the general systems theory for turbulent atmospheric flows and their applications are given in the following:

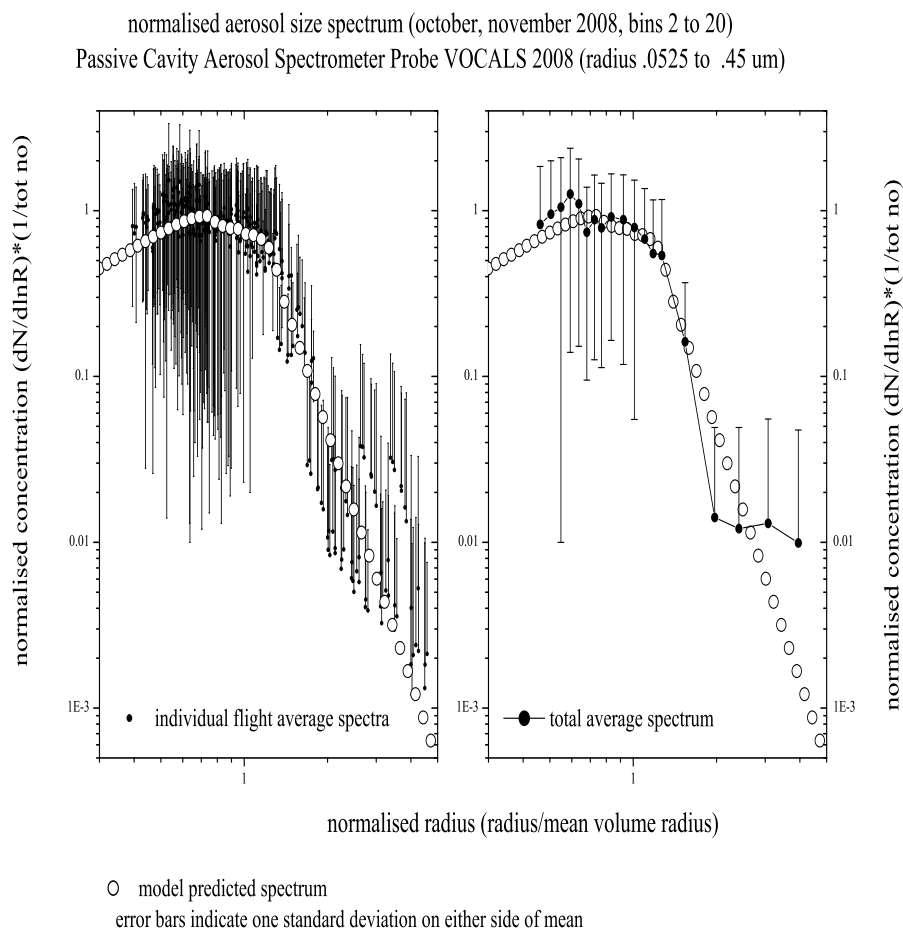


**Figure 7:** Computed dispersion (%) values for size range (a) less than 1  $\mu\text{m}$  diameter (bins 2 to 20) (b) 1 to 3  $\mu\text{m}$  diameter (bins 21 to 30).

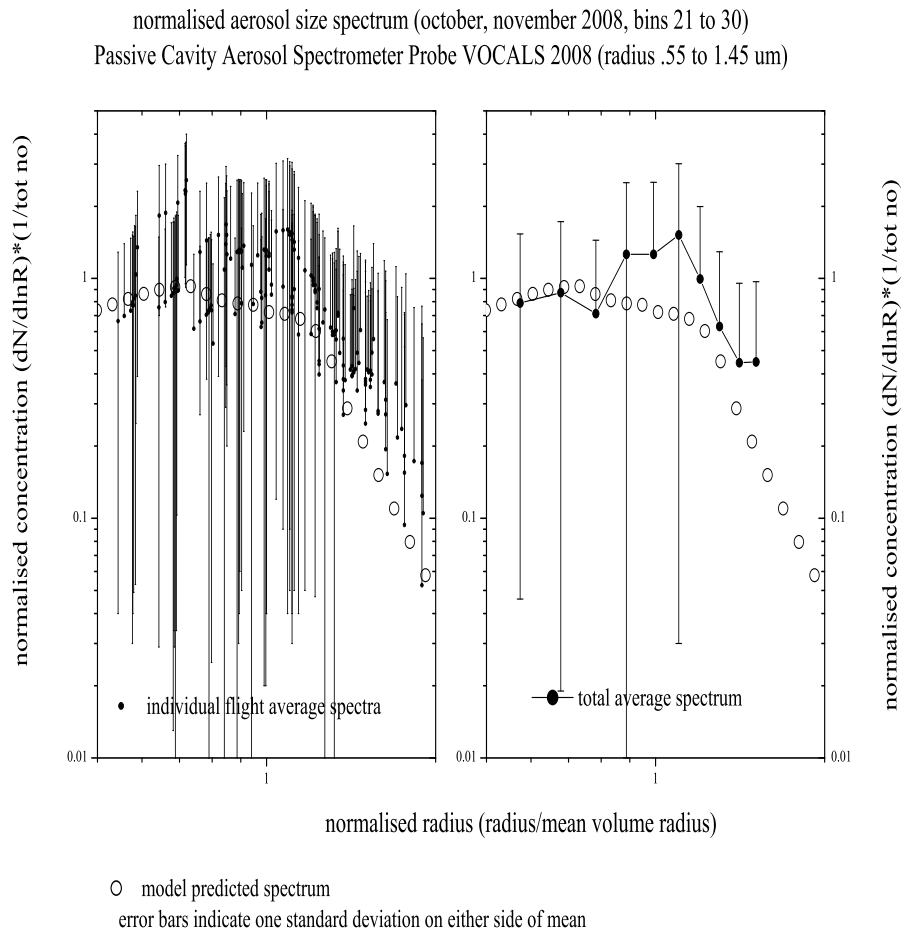
- The probability distributions of amplitude and variance (square of amplitude) of fractal fluctuations are quantified by the same universal inverse power law incorporating the golden mean. Universal inverse power law for power spectra of fractal fluctuations rules out linear secular trends in meteorological parameters. Global warming related climate change, if any, will be seen as intensification of fluctuations of all scales manifested immediately in high frequency fluctuations [79, 87].
- The mass or radius (size) distribution for homogeneous suspended atmospheric particulates is expressed as a universal scale-independent function of the golden mean  $\tau$ , the total number concentration and the mean volume radius. Model predicted aerosol size spectrum is in agreement (within two standard deviations on either side of the mean) with total averaged radius size spectra for the VOCALS



**Figure 8:** The average aerosol size spectra (bins 2 to 30) for each of the 17 data sets are plotted on the left hand side and the total average spectrum for the 17 data sets is plotted on the right along with the model predicted scale independent aerosol size spectrum.



**Figure 9:** The aerosol size spectra for homogeneous aerosol substance density in the accumulation mode corresponding to the size (radius) range 0.1 to about 0.5  $\mu\text{m}$  (bins 2 to 20). The average aerosol size spectra for each of the 17 data sets are plotted on the left hand side and the total average spectrum for the 17 data sets is plotted on the right along with the model predicted scale independent aerosol size spectrum



**Figure 10:** The aerosol size spectra for homogeneous aerosol substance density in the coarse mode corresponding to the size (radius) range 0.5 to 1.5  $\mu\text{m}$  (bins 21 to 30). The average aerosol size spectra for each of the 17 data sets are plotted on the left hand side and the total average spectrum for the 17 data sets is plotted on the right along with the model predicted scale independent aerosol size spectrum

2008 PCASP-B data sets. SAFARI 2000 aerosol size distributions reported by Haywood et al. [36] also show similar shape for the distributions. Specification of cloud droplet size distributions is essential for the calculation of radiation transfer in clouds and cloud-climate interactions, and for remote sensing of cloud properties. The general systems theory model for aerosol size distribution is scale free and is derived directly from atmospheric eddy dynamical concepts. At present empirical models such as the log normal distribution with arbitrary constants for the size distribution of atmospheric suspended particulates is used for quantitative estimation of earth-atmosphere radiation budget related to climate warming/cooling trends (Section 2.1). The universal aerosol size spectrum presented in this paper may be computed for any location with two measured parameters, namely, the mean volume radius and the total number concentration and may be incorporated in climate models for computation of radiation budget of earth-atmosphere system.

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### Appendix I: List of frequently used Symbols:

- $\nu$ : frequency,
- $d$ : aerosol diameter,
- $N$ : aerosol number concentration,
- $N_*$ : surface (or initial level) aerosol number concentration,
- $r_a$ : aerosol radius,
- $\alpha$ : exponent of inverse power law,
- $W$ : circulation speed (root mean square) of large eddy,
- $w$ : circulation speed (root mean square) of turbulent eddy,
- $R$ : radius of the large eddy,
- $r$ : radius of the turbulent eddy,
- $w_*$ : primary (initial stage) turbulent eddy circulation speed,
- $r_*$ : primary (initial stage) turbulent eddy radius,
- $T$ : time period of large eddy circulation,
- $t$ : time period of turbulent eddy circulation,
- $k$ : fractional volume dilution rate of large eddy by turbulent eddy fluctuations,
- $z$ : eddy length scale ratio equal to it  $R/ r$ ,
- $f$ : steady state fractional upward mass flux of surface (or initial level) air,



$q$ : moisture content at height  $z$ ,

$q_*$ : moisture content at primary (initial stage) level,

$m$ : suspended aerosol mass concentration at any level  $z$ ,

$m_*$ : suspended aerosol mass concentration at primary (initial stage) level,

$r_a$ : mean volume radius of aerosols at level  $z$ ,

$r_{as}$ : mean volume radius of aerosols at primary (initial stage) level,

$r_{an}$ : normalized mean volume radius equal to  $r_a/r_{as}$ ,

$P$ : probability density distribution of fractal fluctuations,

$\sigma$ : normalized deviation.

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