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Fractal aggregates in the atmosphere

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In this paper we consider the properties of fractal aggregates suspended in the Earth atmosphere. Fractal aggregates (FA) are now well established to appear in numerous natural and anthropogenic processes. Their role in the atmosphere is immense, for FA possess anomalous physico-chemical, mechanical and optical properties making them extremely effective atmospheric agents. The main goal of this paper is to overview the mechanisms of FA formation, their properties, to discuss sources and sinks of the atmospheric FA and their possible contribution into intra-atmospheric processes and influence on the public health.
KEYWORDS: Fractal aggregates; formation in the atmosphere; properties of fractals.

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1 Introduction

The presence of aggregated structures in the atmosphere was detected very long ago: forest fires and volcano eruptions are well known to produce tremendous amount of ash and other aggregated particles whose role in the formation of the Earth climate was attempted to be estimated by many authors. The transport and industrial aerosol exhausts are also often contain a considerable amount of aggregated particulate matter, not mentioning such intense anthropogenic sources like oil-gas fires. The specialists on Nuclear Winter did not pass this problem aside either.

Irregularly shaped particles were studied since rather long ago, but until fairly recently there was not found a unique and effective key idea for their characterization that would reflect common origin of irregular aggregates or would allow the explanation of their physico-chemical behavior from a unique position.

Therefore, the fractal ideas introduced into Physics (and other natural sciences) by *Mandelbrot* [1977] immediately attracted the attention of the aerosol scientists who applied them for characterization of atmospheric and laboratory-made aggregated aerosol particles.

So the fractal concept quickly found its way in the study of atmospheric aerosols. The success in its application to aerosols gave rise a splash of fractal activity at the end of 80-th – in the beginning of 90-th. The main efforts were directed on recording FA in the atmosphere, attempts to define their fractal dimension and return the Physics of FA into the scopes of former and so habitual notions like aerodynamical diameter, mobility, coagulation efficiencies etc. Although the successes along this route were doubtless – even the optical properties of Titan's hazes were explained by assuming they to consist of FA [*Cabane et al.*, 1993; *Chassefiere and Cabane*, 1995] – a slight coolness coming later resulted, perhaps, from the impression that there is almost nothing to investigate any further.

Of course, it is not so: newly discovered physical and chemical properties of aggregated particles are pertinent to be kept in mind in considering the aerosol processes.

This paper focusses on the properties of self-similar or, better, scaling invariant aggregates – so called fractals or fractal aggregates whose structure is repeated within a considerable range of spatial scales (from tens nm up to parts of cm or even cm). This very kind of order stipulates many unusual properties of FA.

The books ed. by *Avnir* [1989] and *Pietronero and Tosatti* [1986] contain sufficiently full information on the directions of the development of Fractal Physics and Chemistry. A regular account of fractal ideas the reader finds in the books of *Feder*, [1988] and *Smirnov*, [1990]. *Colbeck et al.* [1989] reviewed the fractal concept and its application to environmental aerosols. The fairly recent textbook by *Friedlander* [2000] also contains a chapter on fractals.

2 Phenomenology of Fractals

Typical FA consists of small spherules of the diameter of several tens nanometers united in the aggregate of the size of the order of microns. It is important to stress that the sizes of the spherules are much less than the characteristic parameters in the atmosphere like the mean free path

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of molecules or characteristic wavelength of Sun radiation, whereas the total aggregate sizes are either comparable with these parameters or even exceed them. It is also not surprising that the main attention in studying the atmospheric FA was given to soot aggregates.

In this Section the main concepts characterizing FA are introduced.

i. Mass of FA. Any FA is characterized with its total mass M that can be also measured in units of a spherule mass or, better, by number g of the spherules comprising FA.

ii. Size of FA. It is natural to introduce the gyration radius of FA as:

$$R^2 = \frac{1}{g(g-1)} \sum_{i \neq j} (\mathbf{r}_i - \mathbf{r}_j)^2,$$

where $\mathbf{r}_i$ is the position of the i-th spherule.

The maximal size of FA can also be of use:

$$d_{max} = \max |\mathbf{r}_i - \mathbf{r}_j|.$$

2.1 Fractal Dimension

Not each irregular aggregate is a fractal. The main point of the FA definition is the self-similarity at any scale that eventually leads to rather queer ramified structures of FA whose local mass distribution cannot be so easily measured.

The most straightforward way to measure fractal dimension D is to follow its definition: let FA (or other fractal object) be covered with boxes whose size ϵ goes to zero. If the number N of boxes filled with the elements of FA grows as $N \longrightarrow \epsilon^{-D}$, then D is identified with the fractal dimension of FA.

The simplest and still nontrivial example of the application of the fractal concept to real objects is the measurement of the length of a diffusion trajectory. The diffusion displacement $\Delta = \sqrt{2\mathcal{D}t_\Delta}$. If we represent Δ as the sum of smaller and smaller diffusion displacements $\epsilon = \sqrt{2\mathcal{D}t_\epsilon}$ then we find that the number $N(\epsilon)$ of the ϵ-displacements necessary to cover the diffusion route Δ is $N(\epsilon) = t_\Delta / t_\epsilon \propto \epsilon^{-2}$. The fractal dimension of the diffusion trajectory is thus $D = 2$.

FA are very loose objects. Their fractal dimension D characterizes the part of the space occupied by FA matter. This means that FA mass grows with its gyration radius R slower than R^3: $M \propto R^D$, where $D < 3$. Such the dependence assumes that FA density $\rho(r)$ changes with the distance r from its center as

$$\rho(r) \propto \rho_0 \left(\frac{r_0}{r} \right)^{3-D} \tag{1}$$

at $r < R$ and $\rho(r) = 0$ otherwise. Here ρ_0 is the density of the spherule and r_0 its radius. Equation (1) provides the R^D dependence of FA mass to hold:

$$g = k_D \left(\frac{R}{r_0} \right)^D, \tag{2}$$

where k_D is a fractal prefactor.

2.2 Correlation Function

Density-density correlation function also drops down as a power of distance r:

$$C(r) = \left\langle \sum_i m(\mathbf{r}_i) m(\mathbf{r}_i + \mathbf{r}) \right\rangle \propto r^{-(3-D)}, \tag{3}$$

where $m(\mathbf{r})$ is the density at the point $\mathbf{r}$, the sum goes over all centers of spherules and the triangle braces stand for averaging over all possible spatial configurations of spherules.

2.3 Distribution of Voids

FA thus mainly consists of "empty space" distributed among voids whose size spectrum is of great importance for the characterization of FA. This spectrum normalized to unity has the form:

$$n(a) = \frac{3-D}{R^{3-D}} a^{2-D}. \tag{4}$$

One immediately sees that the total volume occupied by the voids is exactly $4\pi R^3/3$ once the shape factor $\gamma = V/a^3$ defining the dependence of the average volume $V(a)$ of a void on its characteristic size a is $\gamma = 4\pi(6-D)/3(3-D)$ ($V = \gamma a^3$).

2.4 Phenomenology of Atmospheric FA

The measurements of D of atmospheric FA showed:

- Atmospheric FA (mainly soot aggregates) are not well developed fractal structure whose fractal dimensionality varies within 1.3–1.9 giving the indication that these FA are of coagulation origin.
- Such low fractal dimensions are explained by non-isotropy of observed FA which are mostly aligned in one direction. This anisotropy probably arises due to Coulomb or dipole-dipole interaction of FA.
- The fractal prefactor (Eq. (2)) for soot particles is $k_D \approx 27.46$ at $D = 1.75$.
- The structure of atmospheric (soot) fractals may change by condensation-evaporation cycles: the loose FA become more dense (D grows by 10–15%).

Katrinak et al. [1993] analyzed urban aggregates within the size range 0.21–2.61 μ and found that D varies within 1.35–1.38. Maximal value of D found by *Klinger and Roth* [1989] for diesel exhausts was $D = 1.2$, i.e. their particles were strongly aligned. Considerable attention to the processes of the transformation of FA in humid atmosphere was given by *Lesafre* [1989], *Huang et al.* [1994], *Nyeki and Colbeck* [1995a, 1995b] and *Ramachandran and Reist* [1995] to the process the transformation of FA in humid atmosphere. The chemical methods were applied by *Eltekova et al.* [1993] for determining D of soot FA. The value of fractal prefactor was discussed by *Nyeki and Colbeck* [1995a] who showed that k_f is close to 1.

3 Possible Sources of Fractal Particles

The sources of FA are subdivided into two groups: natural and anthropogenic ones.

3.1 Natural Sources

Volcanos. Volcanic eruptions produce a lot of volcanic ash consisting of aggregated oxide particles of the size from parts of μ up to mm. In addition, extreme volcanic conditions produce a lot of smaller aggregates.

Forest fires. They produce a huge amount of ash flakes whose sizes vary from parts of microns up to cm. No doubts that smaller aggregated particles accompany the combustion process (aggregated carbon + hydrocarbon particles or, better, soot). The chemical content of the ash flakes is known: they consists of mineral rests of the combustion process, resins, hydrocarbons and the products of their chemical interaction with atmospheric air.

Thunderstorms. High energy lightning processes are able to release carbon from carbon containing molecules and thus to produce small (nanometric) charged carbon particles (may be, in the fullerene form) which then aggregate forming FA and even aerogels.

Intra-atmospheric chemical processes. Intra-atmospheric chemical and photochemical processes are able to produce low volatile substances which then may produce solid nanoparticles. On colliding these objects form fractal structures.

3.2 Anthropogenic Sources

Industrial exhausts. They produce a lot of smoke particles, FA in that number. The chemical content of these aggregate corresponds to the average content of the smoke. Unfortunately, what share of these particles is aggregated is not yet established.

Transport exhausts. Transport produces aggregated aerosol particles consisting of nanometric soot particles. The sizes of these aggregates rarely exceed a micron.

Gas-oil fires. They produce aggregated soot particles (black smokes) consisting of nanometric units and reaching the sizes of the order of splits of cm.

There are many other less substantial sources of fractal aggregates.

4 Formation of Fractal Aggregates

One of the most important branch of the Fractal Science is the study of the growth kinetics of fractal objects. There exist two commonly accepted approaches to this problem:

- The direct modelling of the growth process. The elements of fractal construction (spherules or fractal fragments) are assumed to move on a lattice, collide, stick together and finally form a fractal structure. The whole process is modelled by computer from very beginning up to the end. This approach allows one to investigate the structure of a single fractal aggregate, to define its fractal dimension and other individual characteristics. In particular, it was shown that the fractal dimension D is totally stipulated with the type of the growth process. Namely, coagulation leads to the most loose structure ($D \approx 1.8$). Diffusion controlled condensation gives more dense particles with $D \approx 2$. Collision limited condensation process (low effective collisions do not permit the spherule to join to the aggregate right after the first collision) produces the most dense FA with $D \approx 2.4$.

- The growth process is considered within a kinetic scheme describing the time evolution of fractal mass spectra irrespective of the details of the fractal fragments motion, the latter ones being included via *kinetic coefficients* whose mass dependence alone defines the characteristic features of the mass spectra. In contrast to the direct modelling this approach accounts for the collective characteristics, the mass distribution of growing fractal aggregates first of all.

Below the second (kinetic) more traditional for the Aerosol Physics approach is used for studying the time evolution of the mass distribution of a collective of FA growing by condensation and coagulation. The collective is assumed to consist of aggregates whose fractal dimension D does not change during the growth process.

The initial stage of FA formation assumes the monomers (spherules) to form. We do not discuss this process, since it does not carry a specifics of the Fractal Physics.

4.1 Growth by Condensation

The latter includes joining monomeric units (spherules or monomers) of unit mass by one, with the condensation coefficients α_g being known functions of the fractal aggregate mass g.

It is not very difficult to see that the condensation coefficients should be proportional to the total number of spherules in FA in the free-molecular limit, and $\alpha_g \propto (FA\ size)$ in the continuous regime. Indeed, FA has a loose structure and the incident spherule readily reaches any point inside FA where it can be captured, unless the collisions with the molecules of the carrier gas make the incident spherule trajectory very lengthy and knotty. In this latter case FA becomes a "black absorber", i.e. the incident spherule randomly walks inside the FA long enough to be absorbed even if the absorption efficiency is not very high, and the average density of matter inside FA is negligibly low.

Hence

$$\alpha_g = \alpha_0 g \tag{5}$$

in the free-molecular regime and

$$\alpha_g = A\mathcal{D}nr_o g^{\frac{1}{D}} \quad (6)$$

in the opposite limit (the continuous regime).

The physical meaning of the constants entering Eqs (5) and (6) is apparent: α_0 (Eq. (5)) is the rate for an incident spherule capture by a vacancy incorporated into FA. The r.h.s. of Eq. (6) repeats that of the expression for the rate of condensational growth of a sphere in the continuous regime except the constant A be replaced by usual coefficient 4π specific for spherical geometry. Equation (6) thus describes the diffusion growth of FA whose radius is proportional to $g^{1/D}$. The values of α_0 and A cannot be found from theoretical considerations and should be thus considered as fitting parameters.

4.2 Growth by Coagulation

Coagulation seems to be the most efficient mechanism of FA growth. Sufficiently large fractal aggregates grown by coagulation have rather low fractal dimensionality $D \approx 1.8$. The rate of the coagulation process depends on the form of the coagulation kernel – the efficiency for two colliding particles to produce a new one whose mass is equal to the sum of the masses of the particles. The coagulation kernel is the collision cross section multiplied by the relative velocity of colliding fragments. The easiest way to estimate the coagulation kernels is just to extend well known expressions for the coagulation kernels for free molecular, continuous or transition regimes by substituting $R \propto g^{1/D}$ instead of $R \propto g^{1/3}$. So, one may expect that the following collection of coagulation kernels governs the time evolution of mass spectra of coagulating FA [*Wu and Friedlander*, 1993a, 1993b; *Wu et al.*, 1994]:

$$K(x,y) \propto (x^{\frac{1}{D}} + y^{\frac{1}{D}})^2 \sqrt{x^{-1} + y^{-1}}$$

(free molecular regime),

$$K(x,y) \propto (x^{\frac{1}{D}} + y^{\frac{1}{D}})(x^{-\frac{1}{D}} + y^{-\frac{1}{D}})$$

(continuous regime),

$$K(x,y) \propto \left(x^{\frac{1}{D}} + y^{\frac{1}{D}}\right)^3$$

(turbulent regime),

$$K(x,y) \propto x^{\frac{1}{D}} y^{\frac{1}{D}}$$

(coagulation of magnetic or electric dipoles),

$$K(x,y) \propto xy$$

(coagulating FA form linear chains). Here x and y stand for the masses of colliding particles. All above kernels are homogeneous functions of the variables of x and y: $K(ax, ay) = a^\lambda K(x,y)$. The homogeneity exponent $\lambda < 1$ for the first two kernels and may exceed unity otherwise. The latter fact means that the aerosol-aerogel transition is possible in the last three cases.

4.3 Aerosol-Aerogel Transition

This term refers to the formation of a macroscopic object (or objects) from initially microscopic aerosol particles. Everybody observed web-like structures or lengthy filaments suspended in the air or attached to the walls of cleaning devices. Sometimes such objects spontaneously arise in the carrier gas as a consequence of the coagulation process in the cases when the coagulation kernel grows sufficiently fast with the colliding particle masses ($\lambda > 1$). Aerosols consisting of fractal aggregates are the most probable candidates to form aerogels by coagulation.

The atmospheric aerogel objects may play crucial role in the formation of ball lightning. According to the model developed by *Smirnov* [1993] the ball lightning is a plasma ball spanned on an aerogel framework. This aerogel framework may form after a strike of a linear lightning which is able to produce the fractal aggregates by ablation or directly from carbon containing molecules in air. Although the dynamics of this process is not yet fully understood the aerogel model was shown to be perspective for explanation of many properties of ball lightnings.

Huge literature is devoted to the computer modelling of FA formation. It is summarized in the review article by *Meakin* [1991]. The mass spectrum of growing FA meets the set of kinetic equation describing FA condensational growth. These equations were analyzed and solved by *Lushnikov and Kulmala* [1995].

Coagulation of fractals in the free molecular regime was theoretically investigated by *Wu and Friedlander* [1993a, 1993b] who found considerable broadening of the particle mass spectra in decreasing the fractal dimensionality. Similar results were reported by *Vemury and Prastinis* [1994]. *Wu et al.* [1994] proposed a method for definition of D from the kinetics of coagulation. Rather simple introduction to Fractal Physics the reader will find in the review by *Smirnov* [1990], where considerable attention is given to the kinetics of FA formation.

The coagulation in the system with the kernel $K = xy$ was analyzed by *Lushnikov* [1978, 2004, 2005] who showed that a gel should form from coagulating sol after a finite interval of time. Experimentally this process was observed by *Lushnikov et al.* [1990, 1991] who supposed that the dipole-dipole interaction of FA is responsible for this phenomenon.

5 Optics of Fractals

Atmospheric fractals reveal very specific optical properties interacting intensely with sun light. This fact is linked closely with their structure: the geometrical size of the atmospheric FA lies in micron range, i.e. the particles are comparable with the wavelength of visible light and IR radiation. On the other hand, FA are composed of tiny nanosized units whose electrodynamic properties are often differ from those of macroscopic objects. This felicitous combination of micro- and macro-properties together with a kind of spatial order (scaling invariance) stipulate specific optical properties

of fractal aggregates.

Strong spatial correlations of the nanospherules (Eq. (3)) lead to the singularity in the differential elastic cross section at small angles:

$$\frac{d\sigma_e}{d\Omega} \propto \int d^3 r C(r) e^{i\mathbf{q}\cdot\mathbf{r}} \propto \frac{1}{q^D},$$

where $q = 2\pi\lambda^{-1}\sin(\theta/2)$ and θ is the scattering angle. This singular behavior serves in some cases for the experimental determination of the fractal dimension D.

Voids in FA (Eq. (4)) may create the conditions for surrendering light quants inside them. Sometimes (under special resonance conditions) FA consisting of weakly absorbed spherules are able to absorb the light.

In their comparison of a fractal smoke optical model with light extinction measurements *Dobbins et al.* [1994] used the following expressions for absorption and elastic scattering cross sections:

$$\sigma^{abs} = \frac{6\pi E(m)}{\lambda\rho_p}$$

$$\sigma_{sca} = \frac{4\pi\bar{n}^{(2)}F(m)}{\lambda\rho_p\bar{n}^{(1)}}\left[1 + \frac{4}{3D}k^2R_g^2\right]^{-D/2}$$

with $\bar{n}^{(1,2)}$ being the first and the second moments of FA size distribution function, $k = 2\pi/\lambda$, $x_p = 2\pi r_0/\lambda$, λ being the wavelength of the incident light and

$$E(m) = \mathrm{Im}\left(\frac{m^2-1}{m^2+2}\right), \qquad F(m) = \left|\frac{m^2-1}{m^2+2}\right|.$$

These rather simple expressions were applied for the analysis of the results on the light extinction of aggregated soot aerosols with $D = 1.75$ and found a reasonable agreement of predicted and measured values.

The paper by *Berry and Persival* [1986] gave the starting push to the studies of optics of FA. The computational analysis of the Rayleigh-Debye-Gans theory performed by *Farias et al.* [1996] (see also references therein) showed its applicability for soot FA. The authors concluded that this theory should replace other approximations for the description of soot optical properties, such as Rayleigh scattering and Mie scattering for an equivalent sphere. *Lushnikov and Maximenko* [1991] investigated the localization effects in FA and found that FA with $D < 3/2$ consisting of weakly absorbing materials may be, nevertheless, "black" owing to surrendering the incident light quants by voids inside FA. Other optical properties (hypercombinational scattering, scattering at small angles and photoabsorption) of FA were investigated by *Lushnikov et al.* [1989]. *Cabane et al.* [1993] and *Chassefiere and Cabane* [1995] explained the contradictions between the results of polarization and photometric measurements of upper layer of Titan's atmosphere by assuming that FA clouds are responsible for the light scattering effects.

6 Are the Atmospheric Fractals Longlivers?

The answer to this question depends on the mobility of FA which is expected to be much lower than that of compact particle of the same mass. The experimental and numerical studies of the mobilities of aggregated particles allow for some useful semiempirical relations to be established.

In the continuous regime *Rogak et al.* [1993] found that the mobility diameter of FA is:

$$d_{mc} = 2\beta R_g = \beta d_1\sqrt{\frac{D}{D+1}}g^{1/D}$$

with $\beta = 0.7$ – 1.0 and d_1 being the spherule diameter.

In the free molecular regime $d_{mk} \approx d_A$, where d_A is the radius of equivalent projected sphere.

Rather ancient "adjusted sphere" interpolation expression for the equvalent diameter by Dahnecke (cited from *Rogak et al.* [1993]) has the form:

$$\frac{d_m}{C_c(Kn_m)} = \frac{d_{mc}}{C_c(Kn_{ck})},$$

where d_m is the transition mobility diameter, d_{mc} and d_{mk} are the kinetic and continuum regime mobility diameters defined above. $Kn_m = 2\lambda/d_m$ and $Kn_{ck} = 2\lambda d_{mc}/d_{mk}^2$. The slip-correction factor is introduced as:

$$C_c(Kn) = 1 + Kn[A + B\exp(-C/Kn)]$$

with $A = 1.257$, $B = 0.4$ and $C = 1.1$. λ is the mean free path.

The sinks of FA in the atmosphere are:
Diffusion deposition is smaller by $g^{\frac{2}{D}-\frac{2}{3}}$ in the free molecular regime, and by $g^{\frac{1}{D}-\frac{1}{3}}$ in the continuous regime.
Sedimentation losses are smaller by $g^{\frac{1}{D}-\frac{1}{3}}$.
Other mechanisms:
Collapsing by humidification: Water condensation on atmospheric fractals may effectively enlarge their fractal dimensionality by 10–15% making them more and more compact.
Water capture by (even hydrophobic) FA.
Scavenging by rain- and snowfalls.
The latter two mechanisms are likely the most efficient.

Colbeck and Wu [1994] used the relation $d_m \propto d_V^{3(D-1)/2D}$ linking the mobility diameter d_m with the volume equivalent diameter d_V for determination of D of smoke FA. They found D to lie within the interval 1.40–1.96. A useful relations linking the particle mobility with optical diameter of soot aggregates ($d_{opt} \propto d_m^{1/3}$) is reported by *Schmidt-Ott and Wustenberg* [1995]. Expressions for thermal and sedimentation velocities in terms of a geometric particle diameter are given by *Magill* [1991]. *Huang et al.* [1994] performed experiments with diesel engines emit chain-agglomerate particles and found the changes in D from 1.56 to 1.76 and from 1.40 to 1.54 depending on the sulfur content. The "rigidity" of the chains was demonstrated to grow in increasing the sulfur content.

7 Concluding Remarks

- The fractal concept is undoubtedly fruitful for characterization the present day aerosol situation in the Earth atmosphere. At the same time it should be marked that the concept itself needs in a development, when applying to atmospheric aerosols. Still not numerous observations of atmospheric fractal aggregates show that their sizes (better the numbers of spherules comprising the aggregates) are not large enough to expose well developed fractal picture. Perhaps, distributions over D will be of use for their proper characterization.

- The fractal aggregates manifest anomalous physicochemical properties: their life times are much longer, than those of the compact particles of the same mass (by 10–100 times in the case of atmospheric fractals), their light scattering and absorption cross sections are higher by orders-of-magnitude than those of the equivalent collective non-aggregated spherules, their chemical and catalytic activities are also enhanced by the specifics of the fractal aggregate morphology. It is why even a small admixture of fractal aggregates may seriously change the existing estimates of the aerosol impact on the global radiation and chemical cycles in the atmosphere.

- The condensation of atmospheric moisture on fractal aggregates was shown to restructure them making the aggregates more compact. This process reduces their life times. The recognition of this fact helps to answer the question: where to seek them? The upper layers of the atmosphere and near cosmos are the most probable places for accumulating the fractal aggregates.

- The fractals of the lower troposphere are mainly of anthropogenic origin and hardly to be thought as very desirable guests. Being good absorbers they are able to accumulate harmful substances and radioactivity and transport them inside live organisms. Hence, the environmental aspects of the atmospheric fractal aggregates are of great importance.

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