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Pascal Rannou, Michel Cabane, Eric Chassefière. Growth of aerosols in Titan's atmosphere and related time scales: A stochastic approach. *Geophysical Research Letters*, 2012, 20 (10), pp.967-970. 10.1029/93GL00911 . insu-04748926

HAL Id: insu-04748926

<https://insu.hal.science/insu-04748926v1>

Submitted on 24 Oct 2024

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GROWTH OF AEROSOLS IN TITAN'S ATMOSPHERE AND RELATED TIME SCALES: A STOCHASTIC APPROACH

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Abstract. The evolution of Titan's aerosols is studied from their production altitude down to the ground using a stochastic approach. A background aerosol distribution is assumed, obtained from previous Eulerian modelling, and the evolution of a "tagged" particle, released near the formation altitude, is followed by simulating in a random way its growth through coagulation with particles of the background distribution. The two distinct growth stages, proposed by Cabane et al. (1992) to explain the formation of monomers and subsequent aggregates, are confirmed. The first stage may be divided into two parts. Firstly, within ≈ 1 terrestrial day, particles grow mainly through collisions with larger particles. They reach their size of monomer ($r_m \approx 0.09 \mu\text{m}$) through typically one to five such collisions. Secondly, within a few terrestrial days to ≈ 1 terrestrial month, particles evolve mainly by collisions with continuously created small particles and acquire their compact spherical structure. In the second stage, whose duration is ≈ 30 terrestrial years, or one Titan's seasonal cycle, particles grow by cluster-cluster aggregation during their fall through the atmosphere and reach, at low stratospheric levels, a typical radius of $0.4 - 0.5 \mu\text{m}$.

Introduction

The aim of this paper is to study the history of Titan's aerosol particles from their production level down to the planetary surface using a stochastic approach. From a previous Eulerian modelling (Cabane et al., 1992, denoted by C1 in the following), the size distribution of these particles has been obtained at any altitude between the production level and the ground. A specific growth scheme has been proposed which consists mainly of two separate stages.

The first stage corresponds to the initial growth of newly created aerosols near the formation altitude by accretion of very small elementary particles (synthesized organic molecules). Due to the large number of accreted particles, whose size is quite variable from one collision to the next, this leads to the formation of compact and nearly spherical particles, called "monomers" by West (1991) and West and Smith (1991). In the second stage these monomers slowly settle through the atmosphere and stick together to form aggregates. A microphysical treatment of the settling phase, including an analytical formulation of coagulation and settling laws adapted to the case of fractal particles, has already been developed to describe the second growth stage (Cabane et al.,

1993, denoted by C2 in the following).

The use of the Eulerian microphysical modelling does not allow the precise characterization of the temporal evolution of the particles from the moment they are created through photochemical processes. The goals of the present work are to present a quantitative approach and provide estimates of the relevant time scales.

In this spirit, we follow an individual "tagged" particle released near the formation altitude and descending through the atmosphere. Its interaction with particles of the stabilized distribution provided by the Eulerian model (C1, C2), used as a background environment, is treated using a stochastic approach. For this purpose, the evolution of this particle is studied by simulating its growth in a random way and memorizing both the times when collisions occur and the sizes of the collided particles. In addition to the basic information on time scales, this approach brings a more comprehensive understanding of the formation stage of monomers.

The stochastic model

The background aerosol distribution is fixed to its more likely state by consideration of the results obtained from the Eulerian modelling, either by assuming spherical particles over the whole height of the atmosphere (liquid drop model, see C1), or by using the liquid drop model above z_c and the fractal model (see C2) below this level. The average altitude of production, z_0 , is assumed to be 385 km. The mass production rate follows a gaussian law of standard deviation $\Delta z = 20 \text{ km}$. The integrated mass production rate is assumed to be $3.5 \cdot 10^{-13} \text{ kg m}^{-2} \text{ s}^{-1}$ and the radius of initial particles is fixed at $r_1 = 1.3 \text{ nm}$. An electrical charge of $30 e^-$ per micron of radius is chosen (C1). It is then possible to release an elementary particle of radius r_1 in the production region, its initial altitude having been drawn at random in the gaussian distribution ($z_0, \Delta z$), and to follow its evolution down to the ground. The only transport process considered in the present study is settling, since eddy diffusion equivalent velocities are smaller than settling velocities by an order of magnitude (C1).

The same grid as in our Eulerian model is used for radius and altitude. As far as the radius is concerned, bin number one corresponds to the size of the initial elementary particles ($r_1 = 1.3 \text{ nm}$). The other bins are defined recurrently by $r_{i+1} = 2^{1/3} r_i$, or $v_{i+1} = 2v_i$, where v_i is the volume of the particle. Let us denote by (i) the particles belonging to the bin number i. Coagulation reactions are of the type: (i)+(j) \rightarrow (i') and the corresponding coagulation coefficient is called $K_{i,j}$. Because the background distribution is stationary, the number of particles in a given two-dimensional bin at radius r and altitude z does not change with time. However, due to their motion along r and z , the particles present in this bin at time t and at time t' are not the same, outgoing particles being

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Paper number 93GL00911

0094-8534/93/93GL-00911\$03.00

replaced by incoming particles. It is then necessary to distinguish the number of particles present in the bin at any given time, that we shall denote by $[i]$, from the number of particles which were present at time t and are not present at time t' , which will be denoted by $[\bar{i}]$. By following the tagged particle (i) as a function of time, one may write:

$$\frac{d[\bar{i}]}{dt} = K_{i,1} [1] [\bar{i}] + K_{i,2} [2] [\bar{i}] + \dots + K_{i,i} [i] [\bar{i}] + \dots + K_{i,n} [n] [\bar{i}] \quad (1)$$

$$[\bar{i}] = [\bar{i}]_0 \exp \left\{ - \sum_{j=1}^n (K_{i,j} [j] [\bar{i}] t) \right\} = [\bar{i}]_0 \exp \left\{ - \frac{t}{\tau_{tot}} \right\} \quad (2)$$

where τ_{tot} is the characteristic time of disappearance through coagulation of the tagged particle (i) at the altitude z .

Choice of the time before coagulation

At any given time in the evolution process, it is necessary to draw at random the time t_r after which the tagged particle will collide with a particle belonging to the background distribution. From Eq. 2, the density of probability for the particle disappearance at time t_r may be written:

$$- \frac{1}{[\bar{i}]_0} \frac{d[\bar{i}]}{dt} = \frac{dP}{dt}(t_r) = \frac{1}{\tau_{tot}} \exp \left\{ - \frac{t_r}{\tau_{tot}} \right\} \quad (3)$$

The time t_r of disappearance may be drawn at random by using the classical property of the repartition function, that is to be uniformly distributed over the interval $[0,1]$. Knowing t_r and the settling velocity, the altitude reached by the particle at the time of coagulation may be determined.

Choice of the radius of the collided particle

From Eq. 2, the derivative with respect to time of the number of disappearing particles may be written:

$$\frac{d[\bar{i}]}{dt}(t) = [\bar{i}]_0 \frac{1}{\tau_{tot}} \exp \left\{ - \frac{t}{\tau_{tot}} \right\} \quad (4)$$

By setting $t = t_r + \theta$, we find:

$$\frac{d[\bar{i}]}{d\theta} = [\bar{i}]_{tr} \frac{1}{\tau_{tot}} \exp \left\{ - \frac{\theta}{\tau_{tot}} \right\} \quad (5)$$

which represents the rate of disappearance of the tagged particle (i) at the time $t=t_r$ chosen before. Considering only the coagulation path (i) + (k) → (k'), this rate is given by:

$$\left[\frac{d[\bar{i}]}{d\theta} \right]_k = [\bar{i}]_{tr} \frac{1}{\tau_k} \exp \left\{ - \frac{\theta}{\tau_k} \right\} \quad (6)$$

and the probability for (i) to disappear by colliding with (k) is:

$$P(k, \theta) = \frac{\left[\frac{d[\bar{i}]}{d\theta} \right]_k(\theta)}{\frac{d[\bar{i}]}{d\theta}(\theta)} \quad (7)$$

Consequently the branching ratio may be written:

$$P(k, \theta \rightarrow 0) = \frac{\tau_{tot}}{\tau_k} \quad (8)$$

$P(k)$ may be calculated for $k=1, \dots, j, \dots, n$, and $\sum P(k)=1$. The bin (k) is drawn at random by using the repartition function of P, which allows the path (i)+(j) → (i') to settle.

Thus, at time $t=t_r$, which is drawn at random, the tagged particle (i) collides with another particle (j), whose size is also drawn at random, at the altitude z . At this time, the size of the tagged particle increases and the process can start again.

Choice of the size of the particle after coagulation

Knowing the radius r_j of the collided particle, one needs to know the size of the final particle resulting from the collision-coagulation process. Due to the size discretization used in the present model, a particle needs to double its volume to climb one step in the size scale. If a particle of volume v_i collides with smaller particles, v_j , this will be performed after $n=v_i/v_j$ collisions. It may be assumed that, on average, one collision among n allows the particle to grow from (i) to (i+1).

For a tagged particle (i) colliding with a particle (j), the probability $p_{i,i'}$ to grow is given by:

- if $j < i$: $p_{i,i+1} = \frac{v_j}{v_i}$ and $p_{i,i} = 1 - \frac{v_j}{v_i}$
- if $j = i$: $p_{i,i+1} = 1$
- if $j > i$: $p_{i,j} = 1 - \frac{v_i}{v_j}$ and $p_{i,j+1} = \frac{v_i}{v_j}$

Results

Liquid drop model

The background aerosol distribution is chosen according to the results of the model by using the values of the parameters previously given. In order to make a statistical analysis of the possible evolutions of the tagged particle one thousand simulations are performed. For twenty simulations the radius and the altitude of the particle are shown as a function of time in Figure 1 and 2 respectively.

The release of the tagged particle corresponds to the beginning of the first stage of the growth process, denoted by (I). During stage (I), the tagged particle (i) collides with

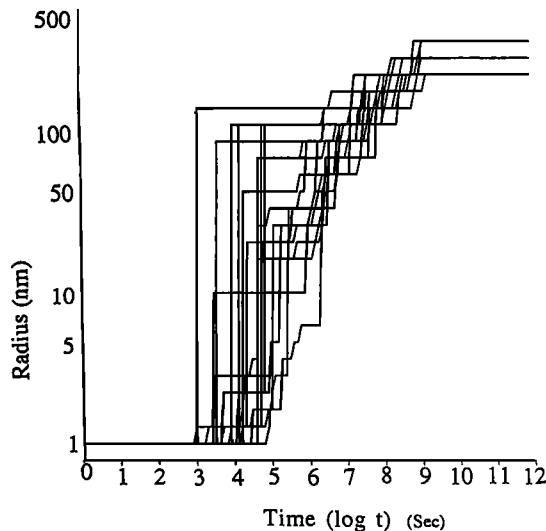


Fig. 1. Evolution of the radius of the tagged particle as a function of time for twenty simulations.

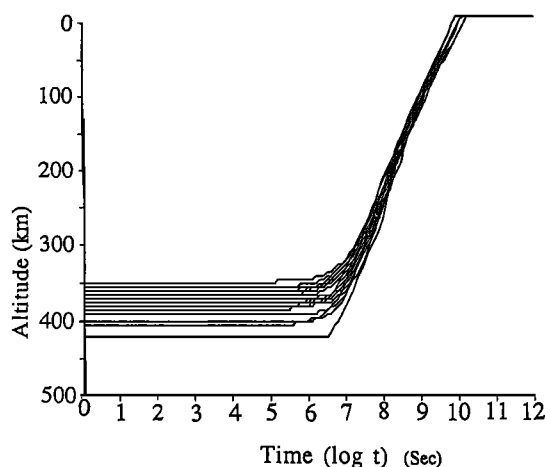


Fig. 2. Evolution of the altitude of the tagged particle as a function of time for twenty simulations.

particles characterized by a broad size spectrum and climbs in the size scale through a small number of collisions with larger particles, implying rather dispersed individual trajectories. The first collision occurs with such a large particle between 10^3 and 10^5 sec, with a corresponding large jump in the size scale, as can be seen in Figure 1. Then (i) becomes a large particle and, afterwards, it collides mainly with smaller particles. As outlined in C1 the coagulation coefficient is minimum for particles of the same radius. It is not surprising, therefore, to find that, when they are small, particles collide mainly with larger particles, whereas later, after having grown through a small number of such collisions, they undergo a lot of collisions with smaller particles. The growth of particles is driven by collisions with large particles, whereas collisions with small particles contribute to fill the gaps and tend to achieve the compact spherical structure of monomers. During this first stage of the growth process, extending from $\approx 10^4$ to 10^6 - 10^7 sec (Figure 1), the radius of the tagged particle increases from 1.3 nm to $0.09 \mu\text{m}$, within a factor of ≈ 2 on either side at the two sigma level. It must be emphasized that this value is in good agreement with the value of the monomer radius r_m proposed in C1. During the second stage, denoted by (II), for times greater than 10^6 - 10^7 sec, (i) grows mainly by collisions with particles of about the same size, with smaller jumps in the size scale of $\approx 2^{1/3}$ per collision (Figure 1). The dispersion of the radius distribution becomes small, with a nearly monodisperse distribution. The final radius, reached after $\approx 10^9$ sec (30 terrestrial years), is ≈ 0.2 - $0.3 \mu\text{m}$.

During its growth, and mainly during stage (II), (i) slowly settles through the atmosphere (Figure 2). This becomes noticeable at a time of $\approx 10^6$ sec, which roughly corresponds to the end of stage (I). Firstly there is a marked increase in the settling velocity of (i) due to the rapid growth of mass through coagulation, then (i) sinks more slowly in the dense atmosphere. The fall from the formation altitude down to the ground spreads over 10^{10} seconds (300 terrestrial years). In fact, below 80 km, reached towards 10^9 sec., ethane and methane are expected to condense in appreciable amounts on the particles, making them settle faster down to the ground.

The dispersion in size of particles accreted by the particle (i), and more precisely the average ratio between its radius and the radius of the collided particle, may be quantified as a

function of time and altitude using an index similar to the MSI (Mass Sharing Index) introduced in C1. Two quantities are defined, $\text{MSI}_{\text{st}}^{(1)}$ and $\text{MSI}_{\text{st}}^{(2)}$ (the "st" subscript refers to the stochastic model), which represent, respectively, the ratio of the radius r_s of the tagged particle to the radius r_c of smaller collided particles ($r_s > r_c$), and the converse ratio by consideration of larger collided particles ($r_s < r_c$):

$$\text{MSI}_{\text{st}}^{(1)} = \frac{\sum_{r_s > r_c} \frac{r_s}{r_c}}{n_{\text{coll}}^{(1)}}; \text{MSI}_{\text{st}}^{(2)} = \frac{\sum_{r_c > r_s} \frac{r_c}{r_s}}{n_{\text{coll}}^{(2)}} \quad (9)$$

where $n_{\text{coll}}^{(1)}$ and $n_{\text{coll}}^{(2)}$ are respectively the number per time unit of smaller and larger collided particles, the summation sign applying to the collisions occurring during the same lapse of time. The second definition of the MSI is close to the one introduced in C1. These quantities are calculated as an average over one thousand simulations. Results are presented in Tables 1 and 2. Several values of the MSI_{st} , corresponding to different characteristic times or positions of the tagged particle evolution, have been selected. Stage (I) may be divided roughly into two parts when considering the evolution of individual particles. In the first part ($t < 10^5$ s), the tagged particle collides mainly with larger particles ($\text{MSI}_{\text{st}}^{(2)} \approx 15$). After the first collision with a large particle ($10^5 \text{ s} < t < 10^6$ - 10^7 s), the collisions occur mainly with smaller particles ($\text{MSI}_{\text{st}}^{(2)} \rightarrow 1$; $\text{MSI}_{\text{st}}^{(1)} \approx 60$). In stage II, $\text{MSI}_{\text{st}}^{(1)}$ and $\text{MSI}_{\text{st}}^{(2)}$ both tend towards unity, which means that collisions occur between particles of about the same size.

Two distinct rates of increase of the mass of the tagged particle may be defined, due to collisions with, respectively, smaller ($dM^{(1)}/dt$) and larger ($dM^{(2)}/dt$) particles. These are plotted together in Figure 3, after averaging over one thousand simulations. The most important contribution in the first stage is due to collisions with larger particles, whose number is very small (typically between one and five, see Figure 1). They bring from $\approx 10^4$ to 10 times more matter than collisions with smaller particles between 10^3 and 10^5 sec. In the second part of stage (I), between 10^5 and 10^6 - 10^7 sec, smaller and larger particles bring a comparable contribution. The transition between stage (I) and stage (II) appears when the two curves merge, after 10^6 - 10^7 sec. At this moment there is a break in the history of the particles. At the end of stage (I) they have acquired a compact structure and a nearly spherical shape; moreover their size distribution is nearly monodisperse. Their shape and structure will not be maintained during the subsequent growth phase and fractal aggregates are expected to

Table 1. Evolution of $\text{MSI}_{\text{st}}^{(1)}$ and $\text{MSI}_{\text{st}}^{(2)}$ versus altitude.

z (km)	450	400	350	< 300
	← Stage I		← Stage II	
$\text{MSI}_{\text{st}}^{(1)}$	15	50	8	1.5
$\text{MSI}_{\text{st}}^{(2)}$	7	10	8	1.3

Table 2. Evolution of $\text{MSI}_{\text{st}}^{(1)}$ and $\text{MSI}_{\text{st}}^{(2)}$ versus time.

t (s)	< 10^4	10^5	10^6	> 10^7
	← Stage I		← Stage II	
$\text{MSI}_{\text{st}}^{(1)}$	70	65	65	1.5
$\text{MSI}_{\text{st}}^{(2)}$	15	15	2.5	1.3

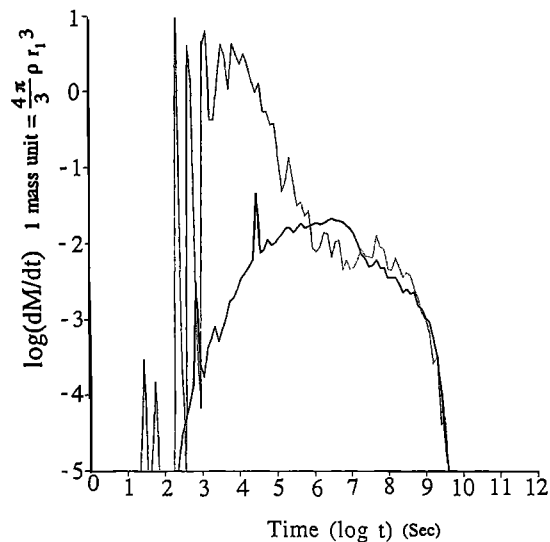


Fig. 3. Growth rates of the tagged particles through collisions with larger particles (dotted line) and smaller particles (solid line) as a function of time.

be formed (C2). Therefore, stage (II) has to be re-examined because the liquid drop model is no longer appropriate.

Fractal model

In order to refine our results as regards stage (II), the background distribution is now fixed according to the fractal model (C2). In this model, a new parameter is introduced in the microphysical laws: the fractal dimension of the aggregates which, in the case of cluster-cluster collisions is assumed to be $D_f=2$. This model is initialized by using the monomer distribution obtained from the liquid drop model at the altitude z_c of the transition between stages (I) and (II).

The tagged particle, of radius $r_m=0.09 \mu\text{m}$, is released at the altitude $z_c=350 \text{ km}$ where monomers are formed (C1) and the microphysical laws are calculated in the same way as in the fractal model. The great difference with the liquid drop model ($D_f=3$) is that a fractal particle falls slower than a compact particle of same mass, due to its larger cross-sectional area, but this allows it to collect more matter and its growth is enhanced. Calculations show that the average fall velocity of the particles at a given level has about the same value in the two models; consequently, although the mass of the particle is increased by a factor ≈ 3 , its fall time does not give significant differences in comparison with the results of the liquid drop model.

Conclusions

The stochastic model presented in this paper allows us to support the conclusions drawn from our Eulerian model (C1).

The existence of two distinct growth stages is confirmed. During the first stage, (I), the particles grow mainly through collisions with particles of very different sizes, leading to the formation of spherical monomers. The radius of these monomers, as deduced from the stochastic model, is a little smaller than $0.1 \mu\text{m}$, in agreement with (C1). The second stage, (II), consists of collisions between particles of about the same size and is expected to give rise to the formation of aggregates in the low stratosphere.

Stage (I) may be divided into two parts. Firstly, within ≈ 1 terrestrial day, particles grow mainly through collisions with larger particles and reach approximately their monomer size through one to five such collisions. Secondly, within a few terrestrial days to ≈ 1 terrestrial month, they undergo a multitude of collisions with smaller particles and acquire their compact spherical structure. In stage (II), whose duration is ≈ 30 terrestrial years, or one Titan's seasonal cycle, particles grow by cluster-cluster aggregation during their slow fall through the atmosphere. At low levels, their typical size is $0.2 - 0.3 \mu\text{m}$ in the liquid drop model, ($0.4 - 0.5 \mu\text{m}$ in the more realistic fractal model) The time to reach the surface is ≈ 300 terrestrial years, but condensation of main hydrocarbons and nitriles is expected to somewhat reduce this time.

Acknowledgments: This research was supported by the Programme National de Planétologie of the Institut National des Sciences de l'Univers (CNRS). Calculations were carried out using the Gould NP1 computer of the Centre de Calcul Recherche - Université Paris VI.

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(Received January 25, 1993;
revised: April 6, 1993;
accepted: April 6, 1993)