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A Mathematical Aspect for Liesegang Phenomena

– リーゼガング現象の数理的様相 –

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1 Liesegang Phenomena

Liesegang phenomenon is pattern formation appeared in a gel-containing system [1]. We can observe striped patterns like in Fig.1, 2, especially in the presence of concentration gradients in initial data. These striped patterns are called "the Liesegang band" and "the Liesegang ring" respectively, because they were discovered by R. E. Liesegang in 1896 for the first time. In this paper, we discuss about the mechanism of this kind of striped pattern formation.

The Liesegang band is obtained by, for example, the following procedure. A solution of one soluble electrolyte, for instance, lead nitorate ($Pb(NO_3)_2$), at relatively low concentration is placed in a test tube to which a gel-forming material is added. After a gel is formed, another electrolyte solution, such as the potasium iodide (KI), normally at substantially higher concentration, is poured on the top of the gel containing $Pb(NO_3)_2$. The iodine-ions (I^-) diffuse into the gel and react with lead ions (Pb^+) to form lead iodide (PbI_2) which is almost insoluble.

$$Pb^{2+} + 2I^- \longrightarrow PbI_2$$

After an interval of minutes there appear bands, so-called the Liesegang band like a Fig.1. The times after the start of the experiment at which pictures (a) to (c) were taken, are as follows: (a) 2 hours, (b) 8 hours, and (c) 48 hours.

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We can make the Liesegang ring similarly. A solution of KI is set up in the inner part of a petri dish whose outer part is occupied by $Pb(NO_3)_2$ contained in gel. Here, KI solution is much higher concentration than $Pb(NO_3)_2$. As I^- diffuse into an outer solution, the insoluble salt PbI_2 precipitates and rings, so-called the Liesegang ring appear like a Fig.2.

It is also well-known that these striped patterns satisfy three periodic laws, *spacing law*, *time law*, and *width law* in chemical experiments practically [5]. *spacing law* can be described as $X_{N+1} = pX_N$, where X_N is the distance of N-th band (ring) location from an original junction and p is a positive constant (Fig.3). *time law* and *width law* are expressed as $\sqrt{t_N} = qX_N$ and $w_N = rX_N$ respectively, where t_N, w_N, q and r are the interval from time when the experiment started to formation time of the N-th band (ring), width of the N-th band (ring) and positive constants.

There are a lot of mathematical models known, which describes the interesting phenomena. We adopt the reduced KR model which is reduced from the well-known Keller-Rubinow model. We can refer to the forthcoming paper [2] about the detail of the KR model and the reduction.

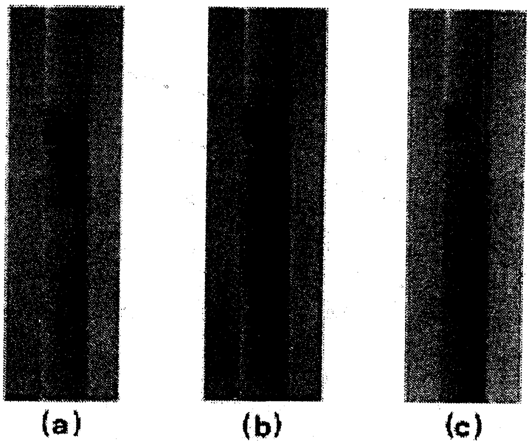


Fig. 1: the Liesegang band [3]

Fig. 2: the Liesegang ring [4]

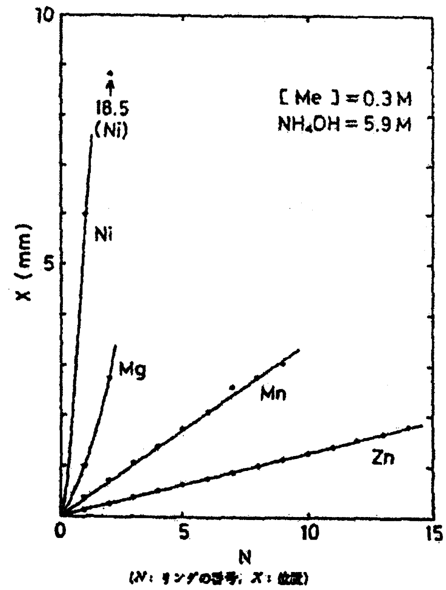
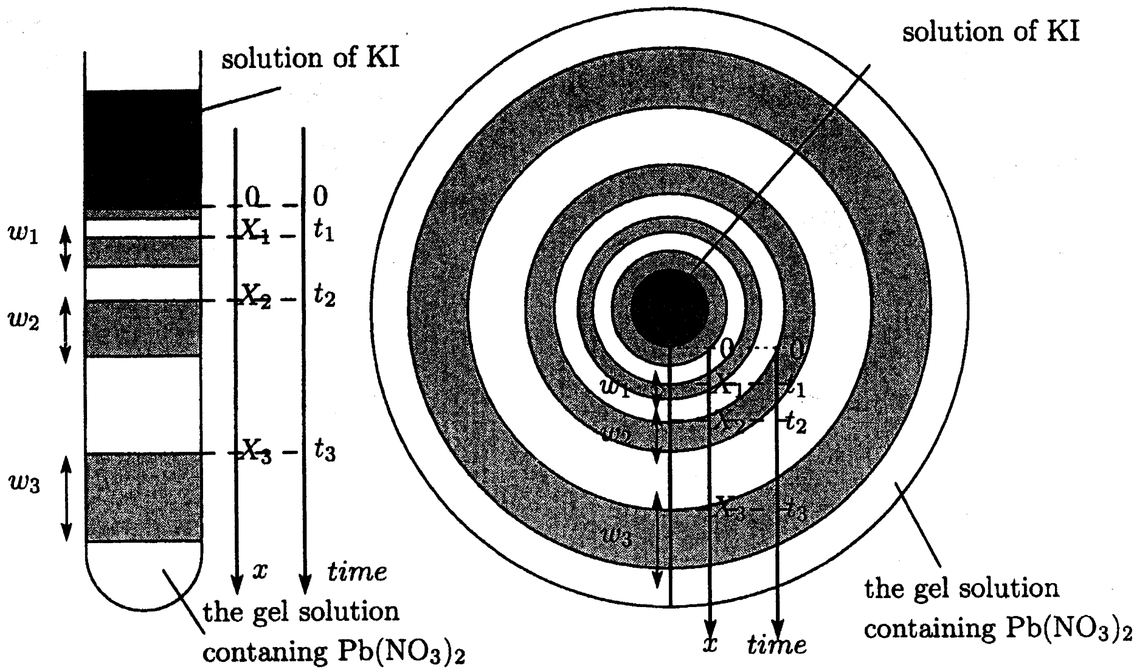


Fig. 3: *spacing law* (the courtesy of Kai) [6]



$$\frac{X_{n+1}}{X_n} = const. \qquad [spaceing\ law]$$

$$\frac{X_n}{\sqrt{t_n}} = const. \qquad [time\ law]$$

$$\frac{w_n}{X_n} = const. \qquad [width\ law]$$

Fig. 4: Pictures of experiments and the three laws

2 Mathematically rigorous Analysis for the reduced KR model

2.1 Existence of a time local weak solution

Without loss of generality, we make $D_c = 1$ as we change the reduced KR model to the dimension less form.

$$\begin{cases} c_t = \Delta c + b_0 S'(t)\delta\left(r - S(t)\right) - qP(c,d), & 0 < t < T,\ \mathbf{x} \in \mathbf{R}^n, \\ d_t = qP(c,d), & 0 < t < T,\ \mathbf{x} \in \mathbf{R}^n, \\ (B.C.)\ \lim\limits_{r\to\infty} c(t,\mathbf{x}) = 0, & 0 < t < T, \\ (\ I.C.)\ \ c(0,\mathbf{x}) = 0,\ \ d(0,\mathbf{x}) = 0, & \mathbf{x} \in \mathbf{R}^n, \end{cases} \tag{2.1}$$

where δ means the Dirac δ in one space dimension,

$$P(c,d) = \begin{cases} (c - C_a)_+, & \text{on}\ \ \{\mathbf{x} \in \mathbf{R}^n;\, c > C_s\ or\ d > 0\}, \\ 0, & \text{otherwise}, \end{cases}$$

$$\begin{aligned} q > 0,\ b_0 > 0,\ C_s \geq C_a \geq 0 \quad &: \ \text{given constants}, \\ S(t) = \alpha\sqrt{t}\ \ (\alpha > 0) \quad &: \ \text{given function}. \end{aligned}$$

r is defined by

$$r = |\mathbf{x}| = \sqrt{x_{n-1}^2 + x_2^2 + \cdots + x_n^2}\,, \quad \mathbf{x} = (x_1, x_2, \cdots, x_n) \in \mathbf{R}^n.$$

In this chapter we consider (2.1) in case of $C_a = 0$.

We first define a weak solution of (2.1). Let $c(\cdot,\cdot) \in L^1\left(0,T;\, W^{1,\infty}(\mathbf{R}^n)\right)$, $d(\cdot,\cdot) \in L^\infty\left((0,T) \times \mathbf{R}^n\right)$. If these satisfy

$$\begin{aligned} c(t,\mathbf{x}) \ &= \ \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\boldsymbol{\xi})^2}{4(t-s)}} b_0 S'(s)\delta(\lambda - S(s))\, d\boldsymbol{\xi}\, ds \\ &\quad -q\int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\boldsymbol{\xi})^2}{4(t-s)}} P(c,d)\, d\boldsymbol{\xi}\, ds, \\ d(t,\mathbf{x}) \ &= \ q\int_0^t P(c,d) ds, \end{aligned} \tag{2.2}$$

then we call a couple of them a weak solution of (2.1), where we define λ by

$$\lambda = |\boldsymbol{\xi}| = \sqrt{\xi_1^2 + \xi_2^2 + \cdots + \xi_n^2}\,,$$

for $\boldsymbol{\xi} = (\xi_1, \cdots, \xi_n) \in \mathbf{R}^n$.

We adopt the following form of the polar coordinate in $\mathbf{R}^n$ to rewrite (2.2):

$$\begin{cases} \xi_1 \ = \ \lambda \sin\beta_{n-1} \sin\beta_{n-2} \cdots \sin\beta_2 \cos\beta_1, \\ \xi_2 \ = \ \lambda \sin\beta_{n-1} \sin\beta_{n-2} \cdots \sin\beta_2 \sin\beta_1, \\ \quad \vdots \\ \xi_n \ = \ \lambda \cos\beta_{n-1}, \end{cases} \tag{2.3}$$

$$0 \le \lambda < \infty,$$
$$0 \le \beta_1 < 2\pi,$$
$$0 \le \beta_j < \pi \quad (j = 2, 3, \cdots, n-1).$$

The rewritten form of (2.2) is

$$\begin{aligned} c(t, \mathbf{x}) &= \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\boldsymbol{\xi}(\lambda))^2}{4(t-s)}} b_0 S'(s) \delta(\lambda - S(s)) J(\lambda, \boldsymbol{\beta})\, d\lambda\, d\boldsymbol{\beta}\, ds, \\ &\quad -q \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\boldsymbol{\xi}(\lambda))^2}{4(t-s)}} P(c, d) J(\lambda, \boldsymbol{\beta})\, d\lambda\, d\boldsymbol{\beta}\, ds, \end{aligned} \tag{2.4}$$

where $\boldsymbol{\beta} = (\beta_1, \cdots, \beta_{n-1})$, $d\boldsymbol{\beta} = d\beta_1 \cdots d\beta_{n-1}$, and

$$J(\lambda, \boldsymbol{\beta}) = \lambda^{n-1} \sin^{n-2} \beta_{n-1} \sin^{n-3} \beta_{n-2} \cdots \sin \beta_2$$

is the Jacobian of the polar coordinates, and $\boldsymbol{\xi}(\lambda)(= (\xi_1(\lambda), \xi_2(\lambda), \cdots, \xi_n(\lambda)))$ means the variables changed by use of (2.3). We emphasize the dependency only upon λ because there is the term of the Dirac δ on λ in the first term of the right-hand side of (2.4).

We remark that, if $n = 1$ and the boundary condition of c at $x = 0$ is the homogeneous Neumann, the corresponding integral equation of c is

$$\begin{aligned} c(t, x) &= \int_0^t \frac{b_0 S'(s)}{\sqrt{4\pi(t-s)}} \left(e^{-\frac{(x-S(s))^2}{4(t-s)}} + e^{-\frac{(x+S(s))^2}{4(t-s)}} \right) ds \\ &\quad -q \int_0^t \int_0^\infty \frac{1}{\sqrt{4\pi(t-s)}} \left(e^{-\frac{(x-\xi)^2}{4(t-s)}} + e^{-\frac{(x+\xi)^2}{4(t-s)}} \right) P(c, d)\, d\xi\, ds. \end{aligned}$$

Therefore we should also define the weak solution separately in one space deimension by use of the above expression. But the mathematical argument in this chapter is appricable to the case of one space dimension.

We define the operator G by

$$G(c) = (\text{the right-hand side of (2.4)})$$

and the space of functions K by

$$K = L^1\left(0, T; L^\infty(\mathbf{R}^n)\right).$$

Let us define the norm of K by

$$\|c\|_K = \int_0^T \|c(t, \cdot)\|_{L^\infty} dt,$$

and K is a Banach space. We note that G is a compact operator on K for any $d(\cdot, \cdot) \in L^\infty\left((0, T) \times \mathbf{R}^n\right)$, and note that let us

$$K_1 = \{c \in K;\ \|c\|_K \le 1\},$$

and K_1 is a bounded, convex, and closed set in K.

Theorem 2.1 (existence of a time local weak solution) *If $T > 0$ is small sufficiently, there exists a weak solution of (2.1) such that $c \in K$ and $d \in L^\infty((0,T) \times \mathbf{R}^n)$.*

pr.) We first note that, for any $c \in K$, $d(t,\mathbf{x})$ $(0 < t < T)$ satisfies

$$\begin{aligned} d(t,\mathbf{x}) &= q\int_0^t P(c,d)ds \\ &= \begin{cases} q\displaystyle\int_0^t c(s,\mathbf{x})\,ds, & \text{if } c > C_s \text{ or } d > 0, \\ 0, & \text{otherwise}, \end{cases} \end{aligned} \tag{2.5}$$

and $d(\cdot,\cdot) \in L^\infty((0,T) \times \mathbf{R}^n)$. Therefore we regard d as a function of c. If we put the function $d(c)$ into $P(c,d)$ of (2.2), then we consider of (2.2) as only c's equation. We will prove the existence of a solution $c \in K$ of (2.2). Let us decide d by use of (2.5) for c constructed already, and we can make a weak solution of (2.1) eventually.

Now we will estimete (2.4) for any $c \in K$.

[Estimate for the first term of the right-hand side of (2.4)]

Let us make the following change of variables to the first term of the right-hand side of (2.4),

$$\begin{cases} p^2 = \dfrac{s}{t}, \\ x_i = S(t)y_i \qquad (i = 1,2,\cdots,n), \end{cases} \tag{2.6}$$

and we get

$$\begin{aligned} &\int_0^t \int_{\mathbf{S}^{n-1}} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\boldsymbol{\xi}(S(s)))^2}{4(t-s)}} b_0 S'(s) J(S(s),\beta)\,d\beta\,ds \\ = &\int_0^1 \int_{\mathbf{S}^{n-1}} \frac{b_0\alpha^n}{(4\pi(1-p^2))^{\frac{n}{2}}} e^{-\frac{\alpha^2(\mathbf{y}-\boldsymbol{\xi}(p))^2}{4(1-p^2)}} J(p,\beta)\,d\beta\,dp, \end{aligned} \tag{2.7}$$

where $\mathbf{y} = (y_1, y_2, \cdots, y_n)$ and $\mathbf{S}^{n-1}$ means the unit sphere in $(n-1)$ space dimensions. The right-hand side of (2.7) is independent from t, and moreover it takes a bouded value at $\mathbf{y} = (0,0,\cdots,0)$ and converges to 0 as $|\mathbf{y}| \to \infty$. Therefore it takes a positive maximum in $\mathbf{R}^n$. There exists a positive constant M_1 independent of both t and $\mathbf{x}$ such that

$$|\text{the first term of the right-hand side of (2.4)}| \le M_1. \tag{2.8}$$

Thus we get

$$\int_0^T \|\text{the first term of the right-hand side of (2.4)}\|_{L^\infty}\,dt \le M_1 T.$$

[Estimate for the **x**-derivative of the first term of the right-hand side of (2.4)]

Let us note that

$$\frac{\partial}{\partial x_i} = \frac{1}{S(t)}\frac{\partial}{\partial y_i} \quad (i = 1, 2, \cdots, n),$$

and

$$\begin{aligned}&|x_1\text{-derivative of the first term of the right-hand side of (2.4)}|\\ =& \left|\frac{1}{S(t)}\frac{\partial}{\partial y_1}\left\{\int_0^1 \int_{\mathbf{S}^{n-1}} \frac{b_0 \alpha^n}{(4\pi(1-p^2))^{\frac{n}{2}}} e^{-\frac{\alpha^2(\mathbf{y}-\xi(p))^2}{4(1-p^2)}} J(p,\beta)\, d\beta dp\right\}\right|.\end{aligned}$$

Therefore there exists a positive constant M_2 such that

$$\begin{aligned}&|x_1\text{-derivative of the first term of the right-hand side of (2.4)}|\\ &\le M_2\sqrt{\frac{1}{t}}.\end{aligned}$$

For any $j = 2, 3, \cdots, n$, let us estimate x_j-derivative in the same manner, and there exists a positive constant M_3 independent of both t and $\mathbf{x}$ such that

$$|\text{the } \mathbf{x}\text{-derivative of the first term of the right-hand side of (2.4)}| \le nM_2\sqrt{\frac{1}{t}}.$$

Thus we get

$$\int_0^T \|\mathbf{x}\text{-derivative of the first term of the right-hand side of (2.4)}\|_{L^\infty}\, dt \le 2nM_2\sqrt{T}.$$

[Estimate for the second term of the right-hand side of (2.4)]

$$\begin{aligned}&\left|\int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\xi(\lambda))^2}{4(t-s)}} P(c,d)J(\lambda,\beta)\, d\lambda\, d\beta\, ds\right|\\ &\quad\le \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\xi)^2}{4(t-s)}}\, d\xi\, \|c(s,\cdot)\|_{L^\infty}\, ds.\end{aligned} \tag{2.9}$$

Let us remark that

$$\int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\xi)^2}{4(t-s)}}\, d\xi = 1,$$

and we get

$$(\text{the right-hand side of (2.9)}) = \int_0^t \|c(s,\cdot)\|_{L^\infty}\, ds \le \|c\|_K.$$

Therefore

$$\int_0^T (\text{the left-hand side of (2.9)})\, dt \le \|c\|_K \int_0^T dt = \|c\|_K T.$$

[Estimate for the **x**-derivative of the second term of the right-hand side of (2.4)]

$$
\begin{aligned}
&\left|\frac{\partial}{\partial x_1}\int_0^t\int_{\mathbf{R}^n}\frac{1}{(4\pi(t-s))^{\frac{n}{2}}}e^{-\frac{(x-\xi(\lambda))^2}{4(t-s)}}P(c,d)J(\lambda,\beta)\,d\lambda\,d\beta\,ds\right|\\
\leq\ &\int_0^t\left(\left|\int_{x_1}^{\infty}\frac{1}{2\sqrt{\pi(t-s)}}\left(-\frac{x_1-\xi_1}{4(t-s)}\right)e^{-\frac{(x_1-\xi_1)^2}{4(t-s)}}d\xi_1\right|\right.\\
&+\left.\left|\int_{-\infty}^{x_1}\frac{1}{2\sqrt{\pi(t-s)}}(-\frac{x_1-\xi_1}{4(t-s)})e^{-\frac{(x_1-\xi_1)^2}{4(t-s)}}d\xi_1\right|\right)\\
&\int_{\mathbf{R}^{n-1}}\frac{1}{(4\pi(t-s))^{\frac{n-1}{2}}}e^{-\frac{\sum_{j=2}^n(x_j-\xi_j)^2}{4(t-s)}}d\xi_2\cdots d\xi_n\,\|c(s,\cdot)\|_{L^\infty}\,ds.
\end{aligned}
\tag{2.10}
$$

Let us make the following chenge of variable

$$\theta_1=-\frac{x_1-\xi_1}{2\sqrt{t-s}},$$

and

$$
\begin{aligned}
\int_{x_1}^{\infty}\frac{-(x_1-\xi_1)}{8\sqrt{\pi}(t-s)^{\frac{3}{2}}}e^{-\frac{(x_1-\xi_1)^2}{4(t-s)}}d\xi_1 &= \frac{1}{2\sqrt{\pi(t-s)}}\int_0^{\infty}\theta_1e^{-\theta_1^2}d\theta_1\\
&= \frac{1}{4\sqrt{\pi(t-s)}}.
\end{aligned}
$$

Let us estimate the integral term on $(-\infty,x_1)$ in the same way as above, and note that

$$\int_{\mathbf{R}^{n-1}}\frac{1}{(4\pi(t-s))^{\frac{n-1}{2}}}e^{-\frac{\sum_{j=2}^n(x_j-\xi_j)^2}{4(t-s)}}d\xi_2\cdots d\xi_n=1,$$

and

$$(\text{the right-hand side of (2.10)})=\int_0^t\frac{1}{2\sqrt{\pi(t-s)}}\|c(s,\cdot)\|_{L^\infty}ds.$$

Therefore

$$
\begin{aligned}
\int_0^T(\text{the left-hand side of (2.10)})dt &\leq \int_0^T\int_0^t\frac{1}{2\sqrt{\pi(t-s)}}\|c(s,\cdot)\|_{L^\infty}dsdt\\
&= \int_0^T\int_s^T\frac{1}{2\sqrt{\pi(t-s)}}\|c(s,\cdot)\|_{L^\infty}dtds\\
&= \int_0^T\frac{1}{2\sqrt{\pi}}\|c(s,\cdot)\|_{L^\infty}\left(\int_s^T\frac{1}{\sqrt{t-s}}dt\right)ds\\
&= \frac{1}{\sqrt{\pi}}\int_0^T\|c(s,\cdot)\|_{L^\infty}(T-s)^{\frac{1}{2}}ds\\
&\leq \sqrt{\frac{T}{\pi}}\|c\|_K.
\end{aligned}
$$

For any $j=2,3,\cdots,n$, we estimate the x_j-derivative in the same manner.

Therefore, if $T > 0$ is small sufficiently, then for any $c \in K_1\ (\subset K)$

$$\begin{aligned} \|G(c)\|_K &\leq M_1 T + nM_2\sqrt{T} + \left(T + n\sqrt{\frac{T}{\pi}}\right)\|c\|_K \\ &\leq 1. \end{aligned}$$

Thus G is a compact operator from K_1 into K_1. By use of the Schauder's fixed point theorem, we conclude that there exists $c \in K_1$ such that $c = G(c)$. □

Remark 2.2 We see the solution c be bounded in $W^{1,\infty}(\mathbf{R}^n)$ uniformly in time except the time point $t = 0$ from the above proof.

2.2 Time global solution and its regularity

The weak solution satisfies the following:

- 'Gap' in time occurs at the moment when t becomes positive in the meaning of *sup*-norm because of the non-homogeneous term of the Dirac δ. But the solution is Lipschitz continuous in time if $t > 0$.
- We cannot expect that the solution on $r = S(t)$ is smoother than in $W^{1,\infty}(\mathbf{R}^n)$, although it is smooth in $r \neq S(t)$

In fact, let us calculate it directly by use of (2.4), and if the solution exists on a time interval $(0, T)$ for some positive constant T, it is Lipschitz continuous in $0 < t < T$ and is in $C^2(\mathbf{R}^n \setminus \partial D_{S(t)})$, and moreover

$$c \in L^1(0, T\,;\, W^{1,\infty}(\mathbf{R}^n) \cap L^p(\mathbf{R}^n)) \quad (1 \leq p \leq \infty).$$

Here $D_a = \{\mathbf{x} \in \mathbf{R}^n;\ |\mathbf{x}| < a\}$ for any $a > 0$.

Next, we will prove that a time global solution exists. If $c \equiv 0$, then the following differencial inequality holds:

$$\begin{cases} c_t \leq \Delta c + b_0 S'(t)\delta(t - S(t)) - qP(c, d) \\ \lim_{r \to \infty} c(t, \mathbf{x}) = 0 \\ c(0, \mathbf{x}) = 0. \end{cases}$$

Therefore, as long as it exists, the solution of (2.4) satisfies

$$c(t, \mathbf{x}) \geq 0.$$

We see precipitation occur continuously in space and time, if $C_s = C_a = 0$.

We estimate on the second term of the right-hand side of (2.4).

$$-q\int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(\mathbf{x}-\xi(\lambda))^2}{4(t-s)}} P(c, d) J(\lambda, \beta)\, d\lambda\, d\beta\, ds \leq 0.$$

As (2.7) and (2.8) are taken into account,

$$\begin{aligned} 0 \leq \quad c(t,\mathbf{x}) &\leq \int_0^1 \int_{\mathbf{S}^{n-1}} \frac{b_0 \alpha^n}{(4\pi(1-p^2))^{\frac{n}{2}}} e^{-\frac{\alpha^2(y-\xi(p))^2}{4(1-p^2)}} J(p,\beta)\, d\beta\, dp \\ &\leq M_1, \end{aligned} \tag{2.11}$$

uniformly for $\mathbf{x}$. Therefore for any $T > 0$,

$$\int_0^T \|c(t,\cdot)\|_{L^\infty} dt \leq M_1 T \tag{2.12}$$

We estimate on $c_{\mathbf{x}}(t,\mathbf{x})$ in the same manner as in the proof of Theorem 2.1. In fact, we make the same calculation as in [Estimate for the $\mathbf{x}$-derivative of the first term of the right-hand side of (2.4)] and [Estimate for the $\mathbf{x}$-derivative of the second term of the right-hand side of (2.4)], and we use (2.11) on the way. Therefore there exists a positive constant M_3 such that, for $i = 1, 2, \cdots, n$,

$$\int_0^T \|c_{x_i}(t,\mathbf{x})\|_{L^\infty} \leq M_3 \sqrt{T}. \tag{2.13}$$

Thus there exists a positive constant M_4 such that for any fixed $T > 0$

$$\|c\|_K \leq M_4 \left(\sqrt{T} + T\right), \tag{2.14}$$

by use of (2.12) and (2.13). From (2.14) we see the solution be in a bounded set for any $T > 0$. This means that a time global solution exists.

Remark 2.3 In the end of the section 2.2, we consider about radially symmetric solutions of (2.1). If it is assumed that the solution of (2.1) is radially symmetric, then it satisfies the equations of the radially symmetric problem:

$$\begin{cases} c_t = c_{rr} + \dfrac{n-1}{r} c_r + b_0 S'(t) \delta\left(r - S(t)\right) - qP(c,d), \\ d_t = qP(c,d), \\ (B.C.)\ \lim\limits_{r\to\infty} c(t,r) = 0, \\ (\ I.C.)\ \ c(0,r) = 0, d(0,r) = 0. \end{cases} \tag{2.15}$$

By putting $\mathbf{x} = (r, 0, 0, \cdots, 0)$ into (2.4), we naturally derive the integral equation of the weak formulation in the following:

$$\begin{aligned} c(t,r) &= \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{r^2 - 2r\xi_1(\lambda) + \lambda^2}{4(t-s)}} b_0 S'(s) \delta(\lambda - S(s)) J(\lambda,\beta)\, d\lambda\, d\beta\, ds, \\ &\quad -q \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{r^2 - 2r\xi_1(\lambda) + \lambda^2}{4(t-s)}} P(c,d) J(\lambda,\beta)\, d\lambda\, d\beta\, ds \\ d(t,r) &= q \int_0^t P(c,d) ds, \end{aligned} \tag{2.16}$$

We make the same argument as in the sections 2.1 and 2.2 to get the same kind of results about existence and smoothness of a radially symmetric time global solution. But, as we have no uniquness result of the solution of (2.1) because of the discoutinuity of $P(c,d)$, we do not conclude that the solution of (2.1) is only radially symmetric. Furthermore we note that, even if we assume that the solution is radially symmetric, we cannot immediately prove that the solution is unique, although in the following sections we focus on a radially symmetric solution to analyse the pattern formation of Liesegang phenomena.

2.3 Analysis to discontinuous precipitation

For (2.15), let us make a rescaling in the following:

$$\begin{cases} r = S(t)y, \\ t = e^{\tau}, \end{cases} \tag{2.17}$$

let $u(\tau,y) = c(t,r)$, and we get

$$\begin{cases} u_\tau = \dfrac{1}{\alpha^2}u_{yy} + \left(\dfrac{y}{2} + \dfrac{n-1}{\alpha^2 y}\right)u_y + \dfrac{b_0\delta(y-1)}{2} - e^{\tau}q\tilde{P}(u,\tilde{d}), & (-\infty, \log T)\times(0,\infty), \\ \tilde{d}_\tau = e^{\tau}q\tilde{P}(u,\tilde{d}), & (-\infty, \log T)\times(0,\infty), \\ \lim\limits_{y\to\infty} u(\tau,y) = 0,\ \dfrac{\partial u}{\partial y}(\tau,0) = 0, & -\infty < \tau < \log T, \\ u(-\infty,y) = 0,\ \tilde{d}(-\infty,y) = 0, & 0 < y < \infty, \end{cases} \tag{2.18}$$

Here we use

$$\delta(ax) = \frac{1}{a}\delta(x),$$

and we define

$$\tilde{P}(u,\tilde{d}) = P(c,d), \quad \tilde{d}(\tau,y) = d(t,r).$$

We have the corresponding existence and smoothness results to the ones in the original scale. Namely, (2.18) has a time global solution u, and for any $T > 0$ it is Lipschitz continuous in time τ and $C^2([0,1)\cup(1,\infty))$ in y. Moreover for any $1 \le p \le \infty$,

$$u \in L^1\left(0, \log T;\ W^{1,\infty}[0,\infty) \cap L^p(0,\infty)\right).$$

We now consider about the equation (2.15) without the term of $P(c,d)$ to focus only on c. If we define Ψ by

$$\Psi(t,r) = \int_0^t \int_{\mathbf{R}^n} \frac{b_0 S'(s)}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{r^2 - 2r\xi_1(S(s)) + S(s)^2}{4(t-s)}} J(S(s),\beta)\, d\beta\, ds, \tag{2.19}$$

from (2.16) we see this be a solution of (2.15) without the term of P. In (2.19), we make the following rescaling:

$$\begin{cases} r = S(t)y, \\ p^2 = \dfrac{s}{t}, \end{cases} \tag{2.20}$$

and we get

$$(\text{the right-hand side of (2.19)}) = \int_0^1 \int_{\mathbf{S}^{n-1}} \frac{b_0\alpha^n}{(4\pi(1-p^2))^{\frac{n}{2}}} e^{-\frac{\alpha^2(y^2 - 2y\xi_1(p) + p^2)}{4(1-p^2)}} J(p,\beta)\, d\beta\, dp.$$

This integral does not depend upon t (or τ). If $\Psi(y)$ is defined as the right-hand side of the above equality, then this is a stationary solution of (2.18) without the term of $\tilde{P}$. Namely, $\Psi(y)$ solves

$$\begin{cases} 0 = \frac{1}{\alpha^2} u_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y}) u_y + \frac{b_0}{2}\delta(y-1), \\ \lim_{y\to\infty} u(y) = 0,\ u_y(0) = 0. \end{cases} \tag{2.21}$$

We define C^* by

$$C^* = \Psi(1) = \int_0^1 \int_{\mathbf{S}^{n-1}} \frac{b_0 \alpha^n}{(4\pi(1-p^2))^{\frac{n}{2}}} e^{-\frac{\alpha^2(1-2\xi_1(p)+p^2)}{4(1-p^2)}} J(p,\beta)\, d\beta\, dp.$$

Lemma 2.4 (Estimate for Ψ)

$$\begin{aligned} &\Psi(y) = C^* && (0 \le y \le 1) \\ &\Psi(y) < \Psi(1) \ \ \mathit{and} \ \ \Psi_y(y) < 0 && (y > 1) \end{aligned}$$

pr.) If $0 \le y \le 1$, then (2.21) has a singularity apparently at $y \doteq 0$. Therefore we return to the original equation in n space dimensions. The solution c of (2.1) without $P(c,d)$ satisfies

$$c_t = \sum_{j=1}^n c_{x_j x_j} + b_0 S'(t)\, \delta\left(\sqrt{\sum_{j-1}^n x_j^2} - S(t)\right). \tag{2.22}$$

Let us make the following change of variables:

$$\begin{cases} x_j &= S(t) X_j \quad (j = 1, 2, \cdots, n), \\ t &= e^\tau, \end{cases} \tag{2.23}$$

and, if $u(X_1, X_2, \cdots, X_n, \tau) = c(x_1, x_2, \cdots, x_n, t)$, then u satisfies

$$u_\tau = \sum_{j=1}^n \left(\frac{1}{\alpha^2}(u_{X_j X_j}) + \frac{1}{2}(X_j u_{X_j})\right) + \frac{b_0}{2}\delta\left(\sqrt{\sum_{j=1}^n X_j^2} - 1\right), \tag{2.24}$$

and the extended function of $\Psi(y)$ constantly to the direction of β is a stationary solution of (2.24). Therefore this satisfies

$$0 = \sum_{j=1}^n \left(\frac{1}{\alpha^2}(u_{X_j X_j}) + \frac{1}{2}(X_j u_{X_j})\right) \quad \text{in } D_1^n = \left\{(X_1, \cdots, X_n); \sum_{j=1}^n X_j^2 < 1\right\}, \tag{2.25}$$

and this is equal to the constant C^* on ∂D_1^n. By use of the uniformly elliptic type of the strong maximum principle, this is equal to the constant C^* in $\overline{D_1^n}$. Thus we get

$$\Psi(y) \equiv C^* \quad (0 \le y \le 1).$$

If $y > 1$, then $\Psi(y)$ satisfies

$$\begin{cases} \frac{1}{\alpha^2}\Psi_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y})\Psi_y = 0, \\ \Psi(1) = C^*, \\ \lim_{y\to\infty} \Psi(y) = 0. \end{cases} \tag{2.26}$$

If there is $y_0 \in (1,\infty)$ such that $\Psi_y(y_0) = 0$, then the constant function $\Psi(y) \equiv \Psi(y_0)$ satisfies the first equation of (2.26). By use of the uniqueness of the solution of the initial value problem of the second order linear ordinary differencial equations, the solution of (2.26) must be the constant, which equals to $C^* > 0$ by the boundary condition at the origin. This is contradict to $\lim_{y\to\infty} \Psi(y) = 0$. Therefore $\Psi_y(y) \neq 0$ for any $y \in (1,\infty)$. As Ψ is smooth enough in $(1,\infty)$ and $C^* > 0$, it is seen that $\Psi_y(y) < 0 \quad (1 < y < \infty)$. □

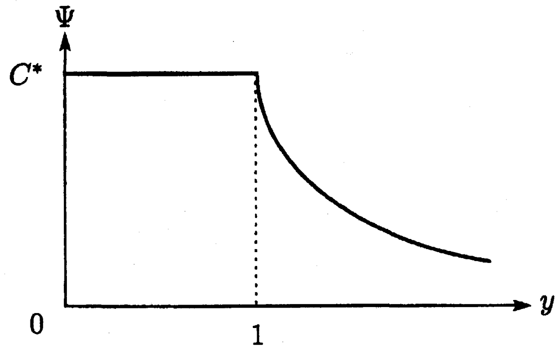


Fig. 5: Shape of $\Psi(y)$

Theorem 2.5 (The first precipitation)

- *If $C_s < C^*$, then there are $t^* > 0$ and $r^* > 0$ such that the first precipitation occurs in $\{(t,r) \in [0,\infty) \times [0,\infty)\,;\, 0 < t < t^*, 0 \le r < r^*\}$.*
- *If $C_s \ge C^*$, then the precipitation never occurs.*

pr.) For any $t > 0$, the solution of (2.15) without $P(c,d)$ is the following integral:

$$\int_0^t \int_{S^{n-1}} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{(r^2 - 2r\xi_1(S(s)) + S(s)^2)}{4(t-s)}} b_0 S'(s) J(S(s),\beta)\, d\beta\, ds.$$

Moreover, we remark that this integral is mapped to the stationary solution Ψ of (2.21) by the rescaling (2.17). This means that, if $C_s < C^*$, then there are $t^* > 0$ and $r^* > 0$ such that the first precipitation must occur in $\{(t,r) \in [0,\infty) \times [0,\infty)\,;\, 0 < t < t^*, 0 \le r < r^*\}$, and else if $C_s \ge C^*$, then the precipitation never occurs. □

In what follows, we assume that $(0 =)C_a < C_s < C^*$. We next prove that, if c becomes greater than C_s in some interval, then c must go down less than C_s after some finite time passes by.

Theorem 2.6 (discontinuous precipitation) *We define $\hat{c}(t,y) = c(t,r)$ by use of the rescaling $r = S(t)y$. It is assumed that there are $T_0 > 0$ and a subinterval $(y_1, y_2) \in [0,\infty)$ such that*

$$\begin{aligned} \hat{c}(T_0, y) &> C_s, \qquad &\forall y \in (y_1, y_2), \\ \hat{c}(T_0, y) &\le C_s, \qquad &\text{otherwise.} \end{aligned}$$

Then, there is a finite time $T^* > T_0$ such that

$$\hat{c}(T^*, y) \le C_s,$$

for all $y \in [0, \infty)$.

pr.) We first note that

$$\begin{aligned} c(t,r) &= \int_0^t \int_{\mathbf{S}^{n-1}} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{r^2 - 2r\xi_1(S(s)) + S(s)^2}{4(t-s)}} b_0 S'(s) J(S(s), \beta)\, d\beta\, ds, \\ &-q \int_0^t \int_{\mathbf{R}^n} \frac{1}{(4\pi(t-s))^{\frac{n}{2}}} e^{-\frac{r^2 - 2r\xi_1(\lambda) + \lambda^2}{4(t-s)}} P(c,d) J(\lambda, \beta)\, d\lambda\, d\beta\, ds. \end{aligned}$$

By use of (2.20), the first term of the right-hand side is changed to $\Psi(y)$, which is independent from t. By use of the following change of variables:

$$\begin{cases} r &= S(t)y \\ \lambda &= S(t)\eta \\ p &= \dfrac{s}{t} \end{cases} \tag{2.27}$$

to the second term, we get

$$-\frac{q\alpha^n t}{(4\pi)^{\frac{n}{2}}} \int_0^1 \int_{\mathbf{S}^{n-1}} \int_0^\infty \frac{1}{(1-p)^{\frac{n}{2}}} e^{-\frac{\alpha^2(y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} \hat{P}(\hat{c}, \hat{d})\, J(\eta, \beta) d\eta\, d\beta\, dp,$$

where

$$\begin{aligned} \hat{P}(\hat{c}, \hat{d}) &= P(c,d), \\ \hat{d}(t,y) &= d(t,r). \end{aligned}$$

Therefore

$$\begin{aligned} \hat{c}\,(t,y) &= \hat{c}\,(T_0, y) \\ &\quad -\frac{q\alpha^n (t - T_0)}{(4\pi)^{\frac{n}{2}}} \int_0^1 \int_{\mathbf{S}^{n-1}} \int_0^\infty \frac{e^{-\frac{\alpha^2(y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} \hat{P}(\hat{c}, \hat{d})\, J(\eta, \beta)}{(1-p)^{\frac{n}{2}}} d\eta\, d\beta\, dp. \end{aligned} \tag{2.28}$$

As $\hat{P}(\hat{c}, \hat{d}) = \hat{c}$ at least in the moving interval $\left(\sqrt{\frac{t_0}{t}} y_1, \sqrt{\frac{t_0}{t}} y_2\right)$ $(t > t_0)$,

$$\begin{aligned} &\int_0^\infty \frac{1}{(1-p)^{\frac{n}{2}}} e^{-\frac{\alpha^2(y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} \hat{P}(\hat{c}, \hat{d}) J(\eta, \beta) d\eta \\ \ge\; &\int_{\sqrt{\frac{T_0}{t}} y_1}^{\sqrt{\frac{T_0}{t}} y_2} \frac{1}{(1-p)^{\frac{n}{2}}} e^{-\frac{\alpha^2(y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} \hat{c}(\eta, t) J(\eta, \beta) d\eta \\ \ge\; &l \int_{\sqrt{\frac{T_0}{t}} y_1}^{\sqrt{\frac{T_0}{t}} y_2} \frac{1}{(1-p)^{\frac{n}{2}}} e^{-\frac{\alpha^2(y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} J(\eta, \beta) d\eta, \end{aligned}$$

as long as $\hat{c} > l$ for any fixed $l \in (0, C_s)$.

By use of Lemma 2.4, there is $y_0 > 0$ such that $\Psi(y) < C_s$ in $[y_0, \infty)$. Therefore, if $t > T_0$, then

$$\hat{c}(t, y) \le \Psi(y) < C_s,$$

in $[y_0, \infty)$. Let us take such a y_0 and fix it, and there exists $M_5 > 0$ such that, if $y \in [0, y_0]$, then

$$\int_0^1 \int_{S^{n-1}} \int_{\sqrt{\frac{T_0}{t}}y_1}^{\sqrt{\frac{T_0}{t}}y_2} \frac{1}{(1-p)^{\frac{n}{2}}} e^{-\frac{\alpha^2(y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} J(\eta, \beta)\, d\eta\, d\beta\, dp > M_5 |y_2 - y_1| \sqrt{\frac{T_0}{t}}.$$

Thus

$$\hat{c}\,(t, y) \le \hat{c}\,(T_0, y) - \frac{q\alpha^n l M_5 |y_2 - y_1| (t - T_0) \sqrt{\frac{T_0}{t}}}{(4\pi)^{\frac{n}{2}}}, \tag{2.29}$$

as long as $\hat{c} > l$.

On the other hand, there is a constant $M_6 > 0$ such that

$$\hat{c} \ge M_6 e^{-q(t - T_0)} \quad \text{in the moving interval } \left(\sqrt{\frac{T_0}{t}} y_1, \sqrt{\frac{T_0}{t}} y_2\right).$$

In fact, this is because the rescaling $r = S(t)y$ makes the moving interval pulled back to (y_1, y_2) and the solution satisfies the differential inequality:

$$c_t \ge c_{rr} + \frac{n-1}{r} c_r - q\, c, \qquad \text{in } (y_1, y_2).$$

Therefore, as we take l in (2.29) as $M_6 e^{-q(t-T_0)}$, there exists a constant $M_7 > 0$ such that

$$\hat{c}(t, y) \le \hat{c}(T_0, y) - \frac{q\alpha^n M_7 |y_2 - y_1| e^{-q(t-T_0)} (t - T_0) \sqrt{\frac{T_0}{t}}}{(4\pi)^{\frac{n}{2}}}.$$

Now we note that the function $e^{-q(t-T_0)}(t - T_0)\sqrt{\frac{T_0}{t}}$ attains the maximum M_q at a time point $T_1 > T_0$. Therefore, we conclude that

$$\hat{c}(T_1, y) \le \hat{c}(T_0, y) - \frac{q\alpha^n M_7 |y_2 - y_1| M_q}{(4\pi)^{\frac{n}{2}}},$$

where we define

$$M_q = \max_{t \ge T_0} \left(e^{-q(t-T_0)} (t - T_0) \sqrt{\frac{T_0}{t}} \right) > 0.$$

If $\hat{c}(T_0, y) - \frac{q\alpha^n M_7 |y_2 - y_1| M_q}{(4\pi)^{\frac{n}{2}}} \le C_s$, then the conclusion of the theorem holds. Otherwise, there is a subinterval $(y_3, y_4) \subset (y_1, y_2)$ such that $\hat{c}(T_1, y) > \frac{C_s}{2}$ for any $y \in (y_3, y_4)$ because of Lipschitz continuity of the solution $\hat{c}$. Therefore, let us replace T_1 to T_0 and (y_3, y_4) to (y_1, y_2), and let us continue to make the same argument as above finite times. Hence, there is a finite time $T^* > T_0$ such that

$$\hat{c}(T^*, y) \leq C_s,$$

for all $y \in [0, \infty)$, because $\hat{c}$ is bounded in $W^{1,\infty}([0, \infty))$ uniformly on time by Remark 2.2. Here we remark that there is a positive constant M_1^* such that $M_7 \geq M_1^* > 0$ in the above finite-time operation, and remark that there is a positive constant M_2^* such that $M_q \geq M_2^* > 0$ as T_0 is bigger and bigger. □

Remark 2.7 From Theorem 2.6, once c becomes bigger than C_s, c goes down and will become less than C_s after some finite time passes by. Therefore precipitation occurs discontinuously spatially and temporarily. Moreover, Theorem 2.6 tells us that the interval where precipitation occurs must be very small because c must go down at once if c exceeds C_s a little. But it is difficult that we estimate how small the interval is, because of the discontinuity of $P(c, d)$.

2.4 *time law & spacing law*

We first consider of the problem in the original scale. $(\underline{R_N}, \overline{R_N})$ is defined as the maximum open interval where the N'th precipitation happens, and $\overline{t_N}(> 0)$ is defined as the solution of the equation: $S(\overline{t_N}) = \overline{R_N}$. Especially, by Theorem 2.5, $\underline{R_1} = 0$. By Theorem 2.6 and the definition of $P(c, d)$, $(\underline{R_N}, \overline{R_N})$ must be a finite open interval.

We now think about the dynamics of the system after the N'th precipitation's being settled down and until $N + 1$'st precipitation's occuring. For this purpose, we separate the half line $[0, \infty)$ to $[0, \overline{R_N}]$ and $(\overline{R_N}, \infty)$. We prove that

(1) The $N + 1$'st precipitation will never occur in $[0, \overline{R_N}]$, and

(2) The $N+1$'st precipitation really occurs in $(\overline{R_N}, \infty)$, which satisfies *time law* rigorously and *spacing law* approximately.

We first prove (1).

Theorem 2.8 *The N+1'st precipitation will not occur in* $[0, \overline{R_N}]$.

pr.) We define $\overline{t_N^*}$ as T^* of Theorem 2.6 with $(y_1, y_2) = (\underline{R_N}, \overline{R_N})$. If $\overline{t_N^*} > \overline{t_N}$, then it is seen that the next precipitation does not occur in $(\overline{t_N}, \overline{t_N^*})$ by Theorem 2.6. Therefore we consider about the next precipitation only in $t > \overline{t_N^*}$.

Now, in $t > \overline{t_N^*}$ and $0 \leq r \leq \overline{R_N}$, c satisfies

$$\begin{array}{ll} c_t \le c_{rr} + \frac{n-1}{r} c_r, & 0 < r < \overline{R_N},\ t > \overline{t_N^*} \\ c(\overline{t_N^*}, r) < C_s, & 0 \le r \le \overline{R_N}. \end{array}$$

Therefore, by use of the maximum principle of parabolic type equation, the maximum value is taken either on $c(\overline{t_N^*}, r)$, on $c(t, 0)$ or on $c(t, \overline{R_N})$. Therefore, if c exceeds C_s in $[0, \overline{R_N}]$, it must be either $c(t, 0)$ or $c(t, \overline{R_N})$.

In what follows we prove that both $c(t, 0)$ and $c(t, \overline{R_N})$ do not exceed C_s actually. We show that if $c(t, r)|_{r=0,\ \overline{R_N}}$ takes the maximum, then it goes down. For the purpose, we use the integral expression in the rescaled system. β denotes either 0 or $\overline{R_N}$. For $\beta = 0$ or $\overline{R_N}$, we define $t_\beta^*(> \overline{t_N^*})$ as the time when $c(t, \beta)$ takes the maximum in $[0, \overline{R_N}]$. In the integral expression (2.28), let us substitute t_β^* for T_0, and we get

$$\begin{aligned} \hat{c}(t, y) &= \hat{c}(t_\beta^*, y) \qquad (2.30) \\ &\quad - \frac{q\alpha^n (t - t_\beta^*)}{(4\pi)^{\frac{n}{2}}} \int_0^1 \int_{\mathbf{S}^{n-1}} \int_0^\infty \frac{e^{-\frac{\alpha^2 (y^2 - 2y\xi_1(\eta) + \eta^2)}{4(1-p)}} \hat{P}(\hat{c}, \hat{d})\, J(\eta, \beta)}{(1-p)^{\frac{n}{2}}} d\eta\, d\beta\, dp. \end{aligned}$$

By the rescaling (2.17), the precipitation interval moves to the left-hand side. Therefore, we estimate the value of $\hat{c}\left(t, \sqrt{\frac{t_\beta^*}{t}} y\right)$. On the moving point $\sqrt{\frac{t_\beta^*}{t}} y$, it is seen that

$$\begin{aligned} \hat{c}\left(t, \sqrt{\frac{t_\beta^*}{t}} y\right) &= \hat{c}\left(t_\beta^*, \sqrt{\frac{t_\beta^*}{t}} y\right) \qquad (2.31) \\ &\quad - \frac{q\alpha^n (t - t_\beta^*)}{(4\pi)^{\frac{n}{2}}} \left(\frac{t_\beta^*}{t}\right)^{\frac{n}{2}} \int_0^1 \int_{\mathbf{S}^{n-1}} \int_0^\infty \frac{e^{-\frac{\frac{t_\beta^*}{t} \alpha^2 (y^2 - 2y\xi_1(\zeta) + \zeta^2)}{4(1-p)}} \hat{c}(t, \sqrt{\frac{t_\beta^*}{t}} \zeta)\, J(\zeta, \beta)}{(1-p)^{\frac{n}{2}}} d\zeta\, d\beta\, dp, \end{aligned}$$

where ζ is defined by

$$\eta = \sqrt{\frac{t_\beta^*}{t}} \zeta.$$

We take a constant $l \in (0, C_s)$ and a subinterval $[y_1', y_2'] \subset [0, \overline{R_N}]$ such that $\hat{c}\left(t, \sqrt{\frac{t_\beta^*}{t}} y\right) > l$ for any $y \in (y_1', y_2')$, and fix them. Therefore, there exists a constant $B^* > 0$, which is dependent on y_1', y_2' and is independent from y, t, t_β^*,

$$\text{(the second term of the right-hand side of (2.31))} < -\frac{q\alpha^n (t - t_\beta^*)}{(4\pi)^{\frac{n}{2}}} \left(\frac{t_\beta^*}{t}\right)^{\frac{n}{2}} l B^*$$

On the other hand, it is easily seen that there is a constant $\delta_1 > 0$ such that for any $y \in (t_\beta^*, t_\beta^* + \delta_1)$,

$$\hat{c}\left(t_\beta^*, \beta\right) \geq \hat{c}\left(t_\beta^*, \sqrt{\frac{t_\beta^*}{t}}\beta\right),$$

because $\beta = 0$ or because $\beta = \overline{R_N}$ and $\hat{c}$ takes the maximum value at $\overline{R_N}$. Moreover, let us define $M_{t_\beta^*}$ by

$$M_{t_\beta^*} = \sup_{t > t_\beta^*}\left((t - t_\beta^*)\left(\frac{t_\beta^*}{t}\right)^{\frac{n}{2}}\right) > 0.$$

(By simple calculation, it is seen that, if $n = 2$, then $M_{t_\beta^*} = t_\beta^*$ and, if $n \geq 3$, then $M_{t_\beta^*} = \frac{2t_\beta^*}{n-2}\left(\frac{n-2}{n}\right)^{\frac{n}{2}}$.) We note that there is a constant $\delta_2 > 0$ such that $(t - t_\beta^*)\left(\frac{t_\beta^*}{t}\right)^{\frac{n}{2}}$ is monotone increasing in $t \in [t_\beta^*, t_\beta^* + \delta_2]$, and also note that $M_{t_\beta^*}$ becomes bigger as t_β^* becomes bigger. It is, therefore, seen that there is a constant $\delta_3 > 0$ and $\delta_4 > 0$ such that for any $t \in (t_\beta^*, t_\beta^* + \delta_3)$

$$\hat{c}\left(t, \sqrt{\frac{t_\beta^*}{t}}\beta\right) < \hat{c}\left(t_\beta^*, \beta\right), \text{ and } \hat{c}\left(t_\beta^* + \delta_3, \sqrt{\frac{t_\beta^*}{t_\beta^* + \delta_3}}\beta\right) < \hat{c}\left(t_\beta^*, \beta\right) - \delta_4. \tag{2.32}$$

Therefore, both $c(t, 0)$ and $c(t, \overline{R_N})$ cannot exceed C_s. □

Next, we will prove (2). We remark that c has never reached C_s so far in $r > \overline{R_N}$, $t \leq \overline{t_N}$. We define functions $\varphi_N(t)$, $\eta_N(t)$, and $\psi_N(r)$ by

$$\begin{aligned}
\varphi_N(t) &= c(t, \overline{R_N}) \quad (t > \overline{t_N}), \\
\eta_N(t) &= c_r(t, \overline{R_N}) \quad (t > \overline{t_N}), \\
\psi_N(r) &= c(\overline{t_N}, r) \quad (0 < r < \infty),
\end{aligned}$$

for the solution c of the original equation (2.15). By Theorem 2.6 and the maximality of $(\underline{R_N}, \overline{R_N})$,

$$0 \leq \psi_N(r) < C_s \quad (\overline{R_N} < r < \infty). \tag{2.33}$$

c solves the following evolutionary equation:

$$\begin{cases}
c_t = c_{rr} + \frac{n-1}{r}c_r + b_0 S'(t)\delta(r - S(t)), & t > \overline{t_N},\ \overline{R_N} < r < \infty, \\
(B.C.)\ c(t, \overline{R_N}) = \varphi_N(t), & t > \overline{t_N}, \\
(B.C.)\ \lim\limits_{r \to \infty} c(t, r) = 0, & t > \overline{t_N}, \\
(\ I.C.)\ c(\overline{t_N}, r) = \psi_N(r), & \overline{R_N} < r < \infty,
\end{cases} \tag{2.34}$$

in $t > \overline{t_N}$ and $\overline{R_N} < r < \infty$.

One of our main tools is the comparison principle of the parabolic type equation with the problem with Homogeneous Dirichlet boundary condition at $r = 0$. Therefore, we must extend the equation (2.34) naturally into the interval $[0, \overline{R_N}]$. For this purpose, we consider the following evolutionary equation:

$$\begin{cases} \tilde{c}_t = \tilde{c}_{rr} + \frac{n-1}{r}\tilde{c}_r, & t > \overline{t_N},\ 0 < r < \overline{R_N}, \\ (B.C.)\ \tilde{c}(t, \overline{R_N}) = \varphi_N(t), & t > \overline{t_N}, \\ (B.C.)\ \tilde{c}_r(t, \overline{R_N}) = \eta_N(t), & t > \overline{t_N}, \\ (I.C.)\ \tilde{c}(\overline{t_N}, r) = \psi_N(r), & 0 < r < \overline{R_N}. \end{cases} \tag{2.35}$$

(2.35) has a unique solution $\tilde{c}$, and by use of the comparison principle,

$$\tilde{c}(t,r) > c(t,r) \geq 0$$

is satisfied in $t > \overline{t_N}$, $0 < r < \overline{R_N}$. Finally, let us consider the following evolution equation:

$$\begin{cases} v_t = v_{rr} + \frac{n-1}{r}v_r + b_0 S'(t)\delta(r - S(t)), & t > \overline{t_N},\ 0 < r < \infty, \\ (B.C.)\ v(t,0) = \eta_N(t), & t > \overline{t_N}, \\ (B.C.)\ \lim\limits_{r\to\infty} v(t,r) = 0, & t > \overline{t_N}, \\ (I.C.)\ v(\overline{t_N}, r) = \psi_N(r), & 0 < r < \infty, \end{cases} \tag{2.36}$$

and (2.36) has a unique solution v. Moreover, v satisfies

$$v(t,r) = \tilde{c}(t,r) > c(t,r)$$

in $t > \overline{t_N}$, $0 < r < \overline{R_N}$, and satisfies

$$v(t,r) = c(t,r)$$

in $t > \overline{t_N}$, $\overline{R_N} < r < \infty$.

Without loss of generality, we normalize $\overline{t_N} = 1$, as we fix $N \in \mathbf{N}$. In order to investigate behavior of the solution of (2.36), we study the following homogeneous problem:

$$\begin{cases} f_t = f_{rr} + \frac{n-1}{r}f_r + b_0 S'(t)\delta(r - S(t)), & t > 1,\ r > 0, \\ f(t,0) = 0, & t > 1, \\ \lim\limits_{r\to\infty} f(t,r) = 0, & t > 1, \\ f(1,r) = 0, & r > 0, \end{cases} \tag{2.37}$$

Furthermore, we need to consider the next problem to see properties of a solution of (2.37).

$$\begin{cases} g_t = g_{rr} + \frac{n-1}{r}g_r + b_0 S'(t)\delta(r - S(t)), & t > 0,\ r > 0, \\ g(t,0) = 0, & t > 0, \\ \lim\limits_{r\to\infty} g(t,r) = 0, & t > 0, \\ g(0,r) = 0, & r > 0 \end{cases} \tag{2.38}$$

A important difference between (2.37) and (2.38) is the time when the initial data is given. It is 1 in (2.37), although it is 0 in (2.38).

(2.38) has a unique time global solution. As $g(t,r)$ is transformed by the following change of variables:

$$\begin{cases} t' = \lambda^2 t, \\ r' = \lambda r, \end{cases} \tag{2.39}$$

$g(t', r')$ solves the quite same equation (2.38). Therefore it is satisfied that

$$g(t,r) = g(\lambda^2 t, \lambda r) \quad (\lambda > 0). \tag{2.40}$$

We let $\lambda = \frac{1}{\sqrt{t}}$, and we see

$$g(t,r) = g(1, \frac{r}{\sqrt{t}}). \tag{2.41}$$

Moreover we use the rescaling: $r = S(t)y$ to get

$$g(t,r) = g(1, \alpha y). \tag{2.42}$$

We remark that the right-hand side of (2.42) is independent from t. Let us define Ψ^D by

$$\Psi^D(y) = g(1, \alpha y),$$

and this is a stationary solution of the equation rescaled by $r = S(t)y$. Namely, Ψ^D solves

$$\begin{cases} 0 = \frac{1}{\alpha^2}\Psi_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y})\Psi_y + \frac{b_0}{2}\delta(y-1), & y > 0, \\ \Psi(0) = 0, \\ \lim\limits_{r\to\infty} \Psi(y) = 0. \end{cases} \tag{2.43}$$

This means that the solution of (2.38) has the "similar" shape to Ψ^D and its maximum point moves to the right-hand side.

On the other hand, we make the change of variables, $r = S(t)y$ and $t = e^\tau$, for the equation (2.37). If we define h by $h(\tau, y) = f(t, r)$, then the rescaled equation is

$$\begin{cases} h_\tau = \frac{1}{\alpha^2}h_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y})h_y + \frac{b_0}{2}\delta(y-1), & \tau > 0,\ y > 0, \\ h(\tau, 0) = 0, \\ \lim\limits_{y\to\infty} h(\tau, y) = 0, \\ h(0, y) = 0. \end{cases} \tag{2.44}$$

Now, let us consider the function $\Psi^D - h$, which satisfies the heat equation with homogeneous Dirichelet boundary condition and with the initial condition Ψ^D. Therefore, $\Psi^D - h$ converges to 0 uniformly in y, which means that f is monotone increasing and

$$f(t, S(t)y) \to \Psi^D(y) (= g(1, \alpha y)), \qquad \text{uniformly in } y,$$

as $t \to \infty$ (namely $\tau \to \infty$).

We next define C^{**} by $C^{**} := \Psi^D(1) > 0$, and study the shape of $\Psi^D(y)$ minutely.

Lemma 2.9 (Estimate for $\Psi^D(y)$)

$$\begin{aligned} \Psi^D(y) &> 0, \quad \textit{in } (0, \infty), \\ \Psi^D_y(y) &> 0, \quad \textit{in } (0, 1), \\ \Psi^D_y(y) &< 0, \quad \textit{in } (1, \infty), \end{aligned}$$

and $\Psi^D(y)$ attains its maximum C^{**} at $y = 1$.

pr.) $\Psi^D(y) > 0$ in $(0,\infty)$ is clear.

We will show that $\Psi^D_y(y) > 0$ in $(0,1)$ by contradiction. In $(0,1)$, Ψ^D solves

$$\begin{cases} 0 = \frac{1}{\alpha^2}\Psi^D_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y})\Psi^D_y, & \text{in } (0,1), \\ \Psi^D(0) = 0, \\ \Psi^D(1) = C^{**} > 0. \end{cases} \tag{2.45}$$

If there exists $y_0 \in (0,1)$ such that $\Psi^D_y(y_0) = 0$. Let us define $G(y)$ by

$$G(y) \equiv \Psi^D(y_0) \quad \text{(the constant function)},$$

and G solves

$$\begin{cases} 0 &= \frac{1}{\alpha^2}G_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y})G_y, \\ G_y(y_0) &= 0, \\ G(y_0) &= \Psi^D(y_0). \end{cases}$$

By use of the uniqueness theorem of the solution of the initial value problem of the second order linear partial differential equations, this does not have any solution more than G. Therefore the solution of (2.45) must correspond to G. Thus we see $\Psi(y) \equiv 0$ (for any $y \in (0,1)$) from $\Psi^D(0) = 0$. But it contradicys that $C^{**} > 0$, so that

$$\Psi^D_y(y) \neq 0 \text{ (for any } y \in (0,1)).$$

Taking $C^{**} > 0$ into account, we get $\Psi^D_y(y) > 0$ (for any $y \in (0,1)$). We can prove that $\Psi^D_y(y) < 0$, (for any $y \in (1,\infty)$) in the same manner, so we omit it. □

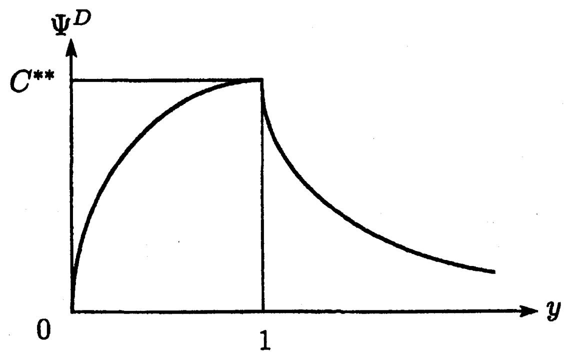


Fig. 6: $\Psi^D(y)$

For the non-homogeneous problem (2.36), we make a change of variables (2.17) and we define $w(\tau, y) = v(t,r)$ to get

$$\begin{cases} w_\tau = \frac{1}{\alpha^2}w_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y})w_y + \frac{b_0}{2}\delta(y-1) & \tau > 0,\ y > 0, \\ w(\tau, 0) = \eta_N(e^\tau), & \tau > 0, \\ \lim_{y\to\infty} w(\tau, y) = 0, & \tau > 0, \\ w(0,y) = \psi_N(\alpha y), & y > 0. \end{cases} \tag{2.46}$$

Lemma 2.10 (Estimate for (2.46)) *If $C_s < C^{**}$, then the solution of (2.46) continues to attain its maximum at $y = 1$ after some finite time passed by.*

pr.) The difference $w - h$ between solutions of (2.46) and (2.44) solves classically the following equation:

$$\begin{cases} z_\tau = \frac{1}{\alpha^2} z_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y}) z_y, & \tau > 0,\ y > 0, \\ z(\tau, 0) = \eta_N(e^\tau) > 0, & \tau > 0, \\ \lim_{y \to \infty} z(\tau, 0) = 0, & \tau > 0, \\ z(0, y) = \psi_N(\alpha y) > 0, & y > 0. \end{cases} \tag{2.47}$$

By use of preserving the positivity, we see

$$w > h, \tag{2.48}$$

for any $\tau > 0$, $y > 0$.

We now separate the interval where w defines to $(0,1)$ and $(1, \infty)$. In $(0,1)$, w solves

$$w_\tau = \frac{1}{\alpha^2} w_{yy} + (\frac{y}{2} + \frac{n-1}{\alpha^2 y}) w_y \quad \tau > 0,\ 0 < y < 1,$$

classically. We apply the parabolic type of strong maximum principle to see $w(\tau, y)$ attaining its maximum either at $\tau = 0$, $y = 0$ or $y = 1$. On the other hand, we have already known that the next precipitation does not occur in $[0, \overline{R_N}]$ by Theorem 2.8. Moreover, as taking (2.48), Lemma 2.9, and the fact that $h \to \Psi^D$ as $t \to \infty$ into account, we conclude that, if $C_s < C^{**}$, then w continues to attain its maximum at $y = 1$ after some finite time passes by.

In $(1, \infty)$, we take $R > 0$ large enough and fix it. We prove the same property in $[1, R]$. Finally we use the fact that $\lim_{y \to \infty} w(\tau, y) = 0$. We eventually see w attaining its maximum at $y = 1$ after some finite time. □

In what follows, we define τ'_{N+1} as the time when the solution w of (2.44) hits C_s, $N+1$'st time, and also define

$$t'_{N+1} = e^{\tau'_{N+1}}.$$

In original temporary and spatially scale, R'_{N+1} is defined as the spatial point where the solution c hits C_s, $N+1$'st time.

Theorem 2.11 (time law) *If $C_s < C^{**}$, then $R'_{N+1} = \alpha\sqrt{t'_{N+1}}$.*

pr.) By Lemma 2.10, $w(\tau, 1)$ continues to attain the maximum value and hits C_s in some finite time, if $C_s < C^{**}$. Therefore, in original scale, it means that

$$R'_{N+1} = \alpha\sqrt{t'_{N+1}},$$

which means *time law*. □

We define τ''_{N+1} as the time when the solution h of (2.46) hits C_s, $N+1$'st time, and also define $t''_{N+1} = e^{\tau''_{N+1}}$. Moreover, we define $R''_{N+1} = S(t''_{N+1})$ and $\overline{\tau_N} = \log \overline{t_N}$,.

Theorem 2.12 (spacing law) *It is assume that there exists a small constant $\varepsilon_1 > 0$ such that, for any $i, j \in \mathbf{N}$,*

$$\sup_{r>0} \left|\psi_i(r + \overline{R_i}) - \psi_j(r + \overline{R_j})\right| < \varepsilon_1. \tag{2.49}$$

Then there are constants $C^* > 0$ and $\delta_0 \geq 0$ such that

$$\frac{R'_{N+1}}{\overline{R_N}} = C^* + o(\varepsilon_1^{\delta_0}),$$

if $C^{**} > C_s$ and if $|C^{**} - C_s|$ is small enough.

pr.) In the interval $[\overline{R_N}, \infty)$, we can separate the solution v of (2.36) to the following three parts:

$$v(t,r) = f(t,r) + U(t,r) + V(t,r),$$

Here $f(t,r)$ solves (2.37), $U(t,r)$ solves the following:

$$\begin{cases} U_t = U_{rr} + \frac{n-1}{r} U_r, & t > \overline{t_N}, \quad r > \overline{R_N}, \\ U(t,0) = 0, & t > \overline{t_N}, \\ \lim\limits_{r\to\infty} U(t,r) = 0, & t > \overline{t_N}, \\ U(\overline{t_N}, r) = \psi_N(r), & r > \overline{R_N}, \end{cases} \tag{2.50}$$

and $v(t,r)$ solves the following:

$$\begin{cases} V_t = V_{rr} + \frac{n-1}{r} V_r, & t > \overline{t_N}, \quad r > \overline{R_N}, \\ V(t, \overline{R_N}) = \varphi_N(t), & t > \overline{t_N}, \\ \lim\limits_{r\to\infty} V(t,r) = 0, & t > \overline{t_N}, \\ V(\overline{t_N}, r) = 0, & r > \overline{R_N}. \end{cases} \tag{2.51}$$

For the solution U of (2.50), there exists a positive constant M_8 such that

$$\begin{aligned} \sup_{r\in[0,\infty)} |U(t,r)| &\leq \frac{1}{4\pi t} \int_0^{2\pi} \int_0^{\infty} \psi_N(r)\, r\, dr\, d\theta \\ &\leq \frac{M_8}{t} \to 0, \quad (t \to \infty). \end{aligned}$$

Taking the assumption of (2.49) into account, there exists a positive constant M_9 such that

$$\sup_{r>0,t>0} \left| U^{(i)}(t + t_i, r + \overline{R_i}) - U^{(j)}(t + t_j, r + \overline{R_j}) \right| \leq M_9 \varepsilon_1, \tag{2.52}$$

for any $i, j \in \mathbf{N}$. Here $U^{(i)}$ is the corresponding solution to (2.50) with $N = i$ for any $i \in \mathbf{N}$.

The solution of (2.51) satisfies that

$$\lim_{t\to\infty} |V(t,r)| = 0 \quad \text{(exponentially)}, \tag{2.53}$$

because of $\lim_{t\to\infty} \varphi_N(t) = 0$ (exponentially). Therefore f is only related to the $N+1$'st precipitation. We first consider about the solution f of (2.37). We have already made a rescale of (2.37) by (2.17) to get (2.44).

Let us remark that the right-hand side of (2.44) is independent from τ, and there exists a positive constant M_{10} independent of N such that, for any $N \in \mathbf{N}$, it holds that

$$\tau''_{N+1} - \overline{\tau_N} = M_{10}.$$

In the original scale of space anf time, it means that

$$\begin{aligned} \log t''_{N+1} - \log \overline{t_N} &= M_{10}, \\ \frac{t''_{N+1}}{\overline{t_N}} &= e^{M_{10}}. \end{aligned}$$

Furthermore, because $R''_{N+1} = S(t''_{N+1})$, we get

$$\begin{aligned} \frac{R''_{N+1}}{\overline{R_N}} &= \sqrt{\frac{t''_{N+1}}{\overline{t_N}}}, \\ &= e^{\frac{M_{10}}{2}}. \end{aligned}$$

We next think of the solution w of (2.46). By use of both (2.52) and (2.53), we see the difference between the solutions of (2.44) and (2.46) be at most in $O(\varepsilon_1)$ (ε_1 is small enough). Thus there exists $\delta_0 \geq 0$ such that

$$\frac{R'_{N+1}}{\overline{R_N}} = e^{\frac{M_{10}}{2}} + o(\varepsilon_1^{\delta_0}),$$

because of Theorem 2.11 $\square$

Remark 2.13 The assumption (2.49) means that small is the difference between the shape of the solution in $r > r_i$ at the moment when $t = t_i$ and the shape of the solution in $r > r_j$ at the moment when $t = t_j$ for any $i, j \in \mathbf{N}$. It apparently seems to be difficult that we prove this in mathematically rigorous manner, because of the hystericis happening. According to numerical simulations that we have already done, it seems that this is satisfied very well. We therefore think that we have made the essential mechanism by which Liesegang phenomena occurs clear.

As we state in Remark 2.7, we can consider of the interval $(\underline{R_N}, \overline{R_N})$ as very small. Therefore, as $R'_N \in (\underline{R_N}, \overline{R_N})$, we can regard the difference between R'_N and $\overline{R_N}$ as much smaller than the difference between $\overline{R_N}$ and $\overline{R_{N+1}}$. Hence we can regard Theorem 2.12 as *spacing law*. But it is difficult that we estimate how small the interval is because of the discontinuity of $P(c, d)$.

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