

# RePLaT–Chaos

Illustration of the chaotic behavior of atmospheric pollutant spreading

Theoretical background and user guide

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Budapest, 2018

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# 1. Introduction

The main aim of RePLaT-Chaos is to make the simulation of global-scale atmospheric spreading of pollutant clouds and the studying of the so-called chaotic features of the advection easy. The pollutant clouds consist of aerosol particles or gas particles. The software is a simpler version of the previously developed RePLaT (Real Particle Lagrangian Trajectory) model [1, 2]. RePLaT-Chaos determines the motion of the particles by taking into account the advection by the wind components in the horizontal direction and its vertical motion is, in addition, influenced by its terminal velocity due the gravity.<sup>1</sup>

In three-dimensional flows, as it is the case in the atmosphere, the advection of pollutants is chaotic. The advection dynamics of pollutants shows typical characteristics of the chaos, such as sensitivity to the initial conditions (the trajectories of initially nearby particles diverge within short time), irregular motion, and complex but regular (fractal) structures. It can be easily observed in the RePLaT-Chaos simulations that pollutant clouds do not disperse in the atmosphere like dye blobs on clothes, rather an initially compact pollutant cloud becomes soon strongly stretched, its length grows rapidly, while becoming filamentary, tortuous and folded. The software can also calculate two quantities which describe the chaoticity of the advection. The stretching rate (topological entropy) characterizes the rate of the exponential stretching of pollutant clouds, while the escape rate quantifies the deposition rate of particles falling out from the atmosphere to the surface [3].

## 2. User guide

### 2.1. Short description

RePLaT-Chaos simulates the atmospheric spreading of pollutant clouds in time interval and with different simulation setups given by the user. Pollutant clouds consist of individual particles the number of which is determined by the user. For the spreading calculations meteorological files containing the appropriate meteorological data overlapping the defined time interval are required. The initial position, size and other properties of the pollutant cloud (and its particles) can be given in the user interface, as well as pre-generated pollutant clouds can also be read for the simulations. RePLaT-Chaos determines the new position of each particle of the pollutant cloud in each time step based on the meteorological data and writes the particle data to file. Furthermore, it computes the length of the pollutant cloud and the ratio of the particles not deposited from the atmosphere. These data are needed for the calculation of two quantities describing the chaoticity of the spreading: one of them is the topological entropy characterizing the stretching rate of the length of the pollutant clouds and the other one is the escape rate quantifying the rapidity of the particle deposition. RePLaT-Chaos provides an opportunity to replay simulations saved in files and to determine the above-mentioned two chaotic measures.

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<sup>1</sup> Neglecting the impact of other processes (e.g., turbulent diffusivity and the scavenging of particles by precipitation).

## 2.2. Launch RePLaT-Chaos

Depending on the operation system the software can be downloaded and installed in different ways described in detail on the webpage <http://hatimi.web.elte.hu/RePLaT/index.html>. The menu items and buttons with underlined letter/number can be reached not only by mouse clicks but also by keyboard shortcut **Alt + <letter>/<number>**.

## 2.3. Meteorological data

For the simulation of the spreading of pollutant clouds meteorological data in NetCDF (Network Common Data Form) format files are needed. Such data can be freely downloaded in different temporal and spatial resolution, e.g., from the ECMWF (European Center for Medium-range Weather Forecasts) databases (ERA-40, ERA-Interim, etc. [7]) after registration. Data should be downloaded for the entire globe and at least 6 hours of time resolution. The properties of the files required by RePLaT-Chaos are:

- Meteorological data on regular longitude–latitude grid ( $[0^\circ : dx : 360^\circ - dx] \times [-90^\circ : dy : 90^\circ]$ ) at different pressure levels, where  $dx$  and  $dy$  are grid sizes. The software considers the lowest level as Earth’s surface and the highest one as the “top” of the atmosphere.
- Required meteorological variables: zonal [m/s] and meridional [m/s] wind components, vertical velocity component [Pa/s] and temperature [K].
- One meteorological file contains values of one of the meteorological variables on the above-mentioned grid for a single time instant.
- File name convention: **<variable name><yyyyMMddhhmmss>.nc**.<sup>2</sup>

For the sum of the choosen time instants a single compressed NetCDF file can be downloaded from the ECMWF database. Single meteorological files for the series of subsequent time instants can be created, e.g., by the software NCO [8] on Linux operation systems as:

- **ncpdq -P upk input.nc output1.nc**: unpacking the downloaded NetCDF file,
- **ncks -d time,\$t,\$t -v \$v output1.nc output2.nc**: applying in loop it writes the fields of variable \$v at time instant \$t from output1.nc to outout2.nc.

## 2.4. Output data

RePLaT-Chaos writes the data of the particles of the pollutant clouds to command separated text files with CSV file extension, therefore these files can be easily read or analyzed with other softwares, too. One output file represents again single time instant and contains one line for each particle. File name convention: **<file name pattern><yyyyMMddhhmmss>.csv**. The comma separated data in a line are:  $\lambda$ ,  $\varphi$ ,  $z$ ,  $r$ ,  $\rho_p$ ,  $in$ , where

- $\lambda$ : longitude coordinate of the particle [radian]  $[0; 2\pi)$ ,
- $\varphi$ : latitude coordinate of the particle [radian]  $[-\pi; \pi]$ ,

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<sup>2</sup> The format <yyyyMMddhhmmss> denotes the following: year (yyyy), month (MM), day (dd), hour (hh), minute (mm), second (ss) given in 4, 2, 2, 2, 2, 2 digits, respectively.

- $z$ : vertical coordinate of the particle [m] calculated from its pressure coordinate  $p$  based on the equations of the standard atmosphere,
- $r$ : particle radius [ $\mu\text{m}$ ]  $[0; \infty)$ ,
- $\rho_p$ : particle density [ $\text{kg}/\text{m}^3$ ]  $[0; \infty)$ ,
- $in$ : is the particle in the atmosphere yet [1] or not [0].

The software computes the length of the pollutant cloud [km] and/or the ratio of the non-escaped particles if the user chooses the option(s). It writes the natural logarithms of these quantities to file at the time instants given by the user. The file of the length and the ratio of the non-escaped particles contain lines of format of `<yyyyMMddhhmmss><tab>ln(value of the quantity)`.

## 2.5. Start simulation

After strtaing the software different setup options for a new simulation appear on the screen. These include the settings of parameters both for the simulation and for the pollutant cloud. This screen is also accessible via **File** menu clicking menu item **New simulation – set parameters**. Via the menu an other option for starting new simulation is also accessible: in **New simulation – read parameters** pollutant clouds are not initialized according to user-given parameters rather its particles are read from a file.

### 2.5.1. Setting the simulation parameters

The user choose either the **New simulation – set parameters** or the **New simulation – read parameters** menu item, in both cases the user should give the simulation parameters at first (left panel of the screen in Figure 1 and Figure 2):

- **Start**: start date and time of the simulation (format: `<yyyy.MM.dd.hh:mm:ss>`).
- **End**: end date and time of the simulation (format: `<yyyy.MM.dd.hh:mm:ss>`).
- **Time step**: if it is **constant** then the textfield represent the value of the constant time step [s] (format: integer); if it is **variable** then the textfield corresponds to  $\Delta t_{\min}$  defined in section 3.1.
- **Input folder**: folder of the meteorological files. Clicking the **Choose input folder** button it can be chosen or it can be written directly to the textfield.
- **File name pattern for the met. fields**: the part of the file name before date and time for the meteorological files of the zonal (**u**), meridional (**v**), vertical (**w**) velocity component of air and the temperature (**T**).
- **Reflect from the surface?**: if the box is checked each particle bounces back from the lowest meteorological level according to the **Reflection coefficient for the surface** (format: real). For topological entropy (length) calculation, the box is worth checking and write 1 for reflection coefficient.
- **Reflect from the top?**: if the box is checked each particle bounces back from the highest meteorological level according to the **Reflection coefficient for the top** (format: real). Generally, the box is worth checking unless the user wants to especially study how many particles leave the meteorological region at the highest level.

- **Save particle data?:** should the data of the particles of the pollutant cloud (and the length data and the ratio of the non-escaped particles) be written to file. You should not check the box if you carry out test calculations and would like to see the spreading of pollutant cloud only once, i.e., you do not need the data later.
- **Output folder:** the folder of the files for particle data, length data and ratio of the non-escaped particles. Clicking the **Choose output folder** button it can be chosen or it can be written directly to the textfield.
- **File name pattern for the output:** the part of the particle data file name before date and time.
- **Calculate length? (filename):** should the length of the pollutant cloud be calculated, and if yes, what should be the name of the length file to which the data are written. The length of the pollutant cloud is computed as the sum of the distances between the subsequent particles. Therefore, the result of the calculation equals to the real length only if the particles are initiated as a one-dimensional “pollutant cloud”, i.e, a line segment.
- **Calculate ratio of non-escaped particles? (filename):** should the ratio of the non-escaped particles be calculated, and if yes, what should be the name of the escape file to which the data are written.
- **Time interval for the output:** the time interval [s] for writing the data of the pollutant cloud, the length and the ratio of the non-escaped particles to file (format: integer)
- **Insert new particles if the distance [km] of two particles:** should new particles be inserted in the pollutant cloud if the distance of two subsequent particles is greater than the given distance (format: real). The box is worth checking for length calculation; otherwise subsequent particles may travel far away from each other which results in the “cut-off” and underestimation of the length of the pollutant cloud. It implies that the end of the length curve shows smaller values than the exponential function fitted to the beginning of the graph.
- **Max. number of particles:** The number of the particles, including the inserted particles, does not exceed the given number (format: integer).

By default, the fields are filled with the default setting parameters loaded from **default/default\_simulation\_setup.txt** (the data in the file can be changed). After overwriting any field, the user can reload the default values by clicking the **Default** button, new data can be loaded in the fields from a chosen file by clicking the **Load** button, or the values of the fields can be saved in a new file by clicking the **Save** button. If there is a wrong or empty textfield, data could not be saved: the problem is indicated by an alert window and the wrong fields are marked by red.

At first, for starting a simulation the generation of the simulation setup is required: the user can generate it by clicking the **Generate simulation setup** button. If there is a wrong or empty textfield, similarly to saving, the problem is indicated by an alert window and the wrong fields are marked by red. If there is no wrong or empty textfield, a pop-up window indicates that the „Simulation setup is generated.” Then the disabled right panel (the parameter settings of the pollutant cloud (Figure 1) or the data for reading particles of a pollutant cloud (Figure 2)) becomes enabled.

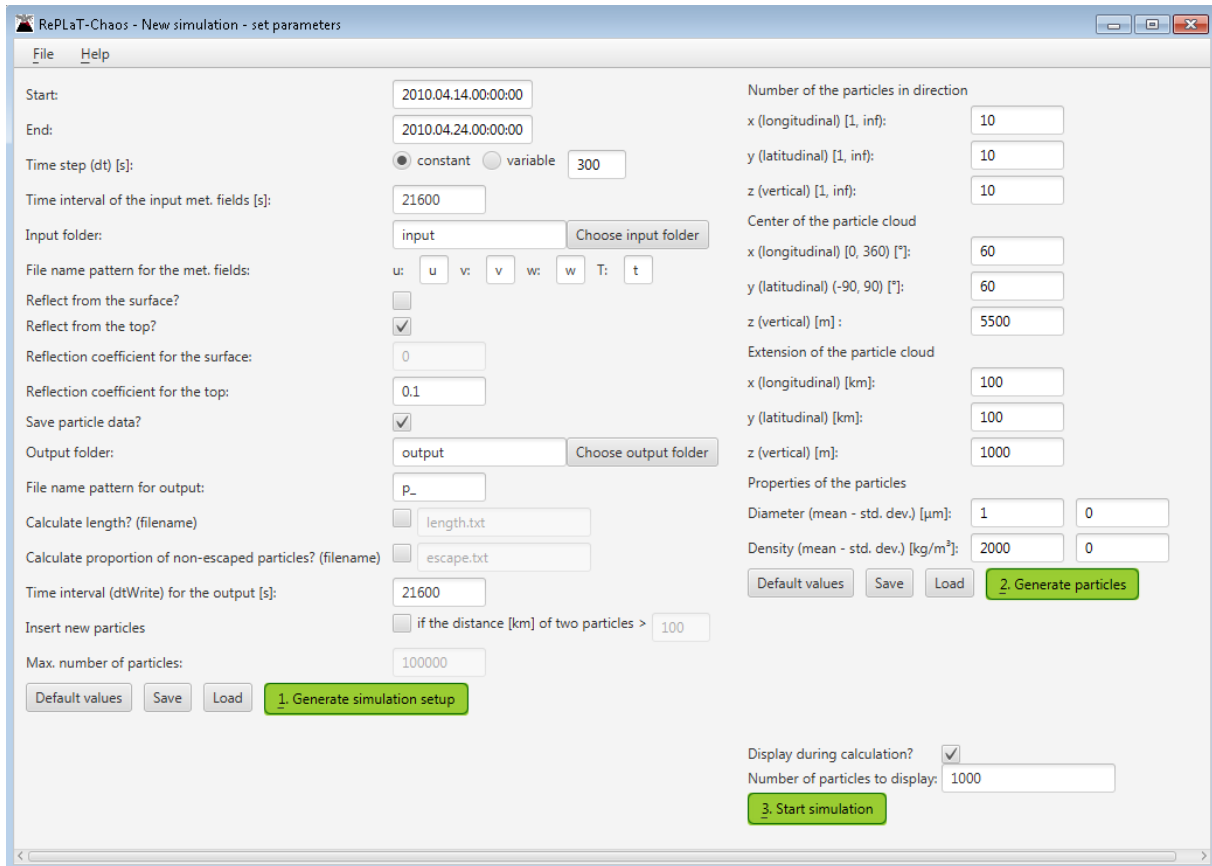


Figure 1. New simulation - set parameters. Starting a new simulation by setting the parameters of the simulation and the pollutant cloud.

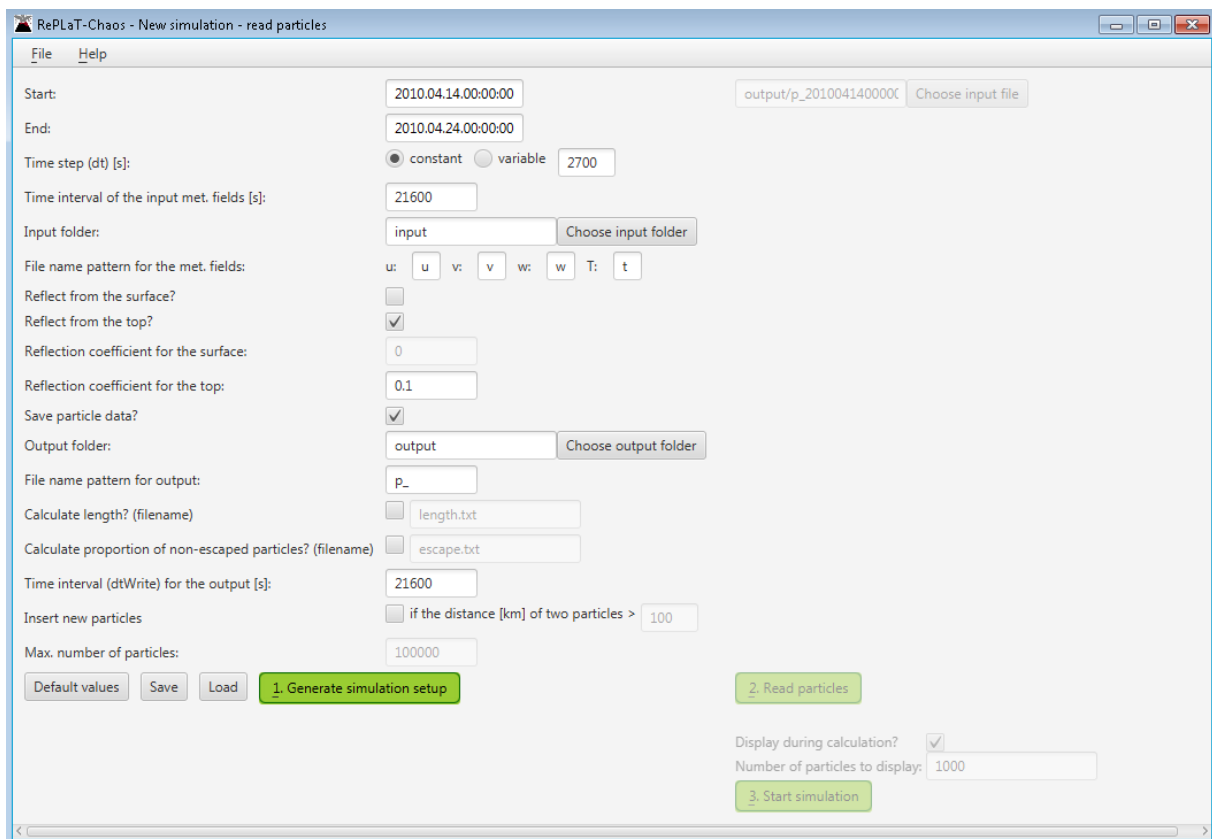
## 2.5.2. Setting the parameters of the pollutant cloud

In the case of setting the parameters of a pollutant cloud the user should set the following parameters of the particles filling a rectangular cuboid:

- **Number of particles in direction x / y / z:** number of the particles in zonal, meridional and vertical direction (format: integers, greater or equal than 1).
- **Center of the particle cloud x / y / z:** zonal [°], meridional [°] and vertical [m] coordinate of the center of the pollutant cloud (format: real numbers in the given intervals)
- **Extension of the particle cloud x / y / z:** length of the rectangular cuboid representing the pollutant cloud in zonal [°], meridional [°] and vertical [m] direction (format: non-negative real)
- **Diameter (mean – std.dev.):** the diameter of the particles is log-normally distributed with the user given mean and standard deviation [ $\mu\text{m}$ ] (format: non-negative real). The diameter 0  $\mu\text{m}$  corresponds to gas particles which are advected by the instantaneous velocity of the atmospheric flow at each time instant (their terminal velocity is 0).
- **Density (mean – std.dev.):** the density of the particles is log-normally distributed with the user given mean and standard deviation [ $\text{kg}/\text{m}^3$ ] (format: non-negative real).

By default, the fields are filled with the default setting parameters loaded from **default/default\_particle\_parameter\_setup.txt** (the data in the file can be changed). The usage of the **Default**, **Save** and **Load** buttons are similar as in the simulation setup panel.

For starting a simulation the generation of the pollutant cloud is also required: the user can generate it by clicking the **Generate particles** button. If there is a wrong or empty textfield, the problem is indicated by an alert window and the wrong fields are marked by red. If there is no wrong or empty textfield, a pop-up window indicates that the pollutant cloud is generated („Number of particles: <particle number>.”). Then the disabled bottom right panel (for setting the display properties of the simulation and starting the simulation calculation (Figure 1)) becomes enabled.



**Figure 2. New simulation - read particles. Starting a new simulation by setting the parameters of the simulation and reading the particles of the pollutant clouds from file.**

### 2.5.3. Reading the particles of a pollutant cloud

In the case of reading the particles of the pollutant cloud (Figure 2), the user choose the file containing the initial conditions of its particles by clicking the **Choose input file** button. The default file path is in the file **default/default\_particle\_file\_setup.txt**. The formats and the values of data in the file should meet the requirements listed in section 2.4. The particle data are read from the file by clicking the **Read particles** button. In the case of wrong values an alert window indicates the problem: „There were invalid data while reading from file <file> <wrong lines>”. If every line is correct, a pop-up window indicates that the pollutant cloud is generated („Number of particles: <particle number>.”). Then the disabled



bottom right panel (for setting the display properties of the simulation and starting the simulation calculation (Figure 1, Figure 2)) becomes enabled.

This way of generating a pollutant cloud is worth applying when the user does not want to initialize the particles filling a rectangular cuboid (mentioned in the previous section) rather than arbitrary positions. E.g., in this way several different particle groups (i.e., different pollutant clouds) initialized at different locations can be tracked simultaneously.

#### 2.5.4. Starting a new simulation

In the bottom right panel (Figure 1 and Figure 2) the user should check whether if she/he wants to watch the spreading of the pollutant cloud during the simulation calculation (**Display during calculation?**) and if yes, how many particles of the pollutant cloud should be drawn (**Number of particles to display**, format: integer). By clicking the **Start** button the simulation calculation starts and the progress (and the particle cloud if chosen) can be tracked in a new window (Figure 3).

#### 2.5.5. Setting the display properties of the simulation

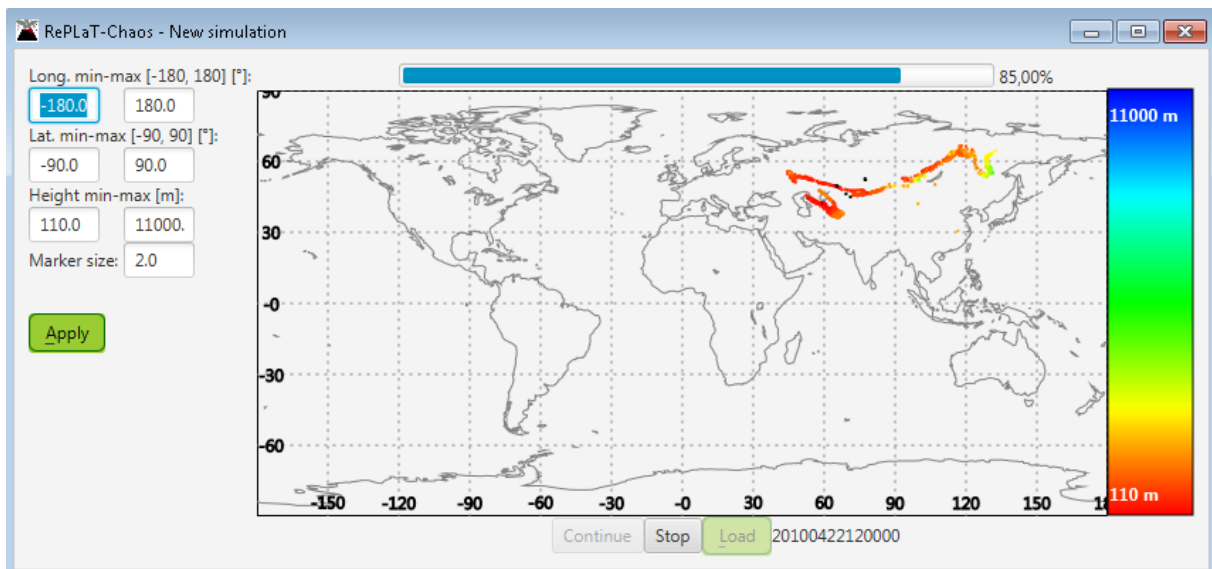


Figure 3: Tracking the calculation of the simulation with displaying the particle positions.

Depending on whether the user has chosen this option or not, during the simulation calculation the current position of the particles of the pollutant cloud is displayed colored according to the vertical coordinate of the particles on a map or only a progressbar is visible to show the percentage of the progress of the calculation and the corresponding date and time. The user can stop and continue the calculation by clicking the **Stop** or **Continue** buttons, respectively, and she/he can load the particle data saved in files for replay by clicking the **Load** button. The longitudinal and latitudinal boundaries of the map, the vertical boundaries of the coloring and the marker size of the particles can be changed on the left side (formats: real numbers in the given intervals). The settings are applied by clicking the **Apply** button. If there is a wrong or empty textfield, the problem is indicated by an alert window and the wrong fields are marked by red.

## 2.6. Replaying a saved simulation

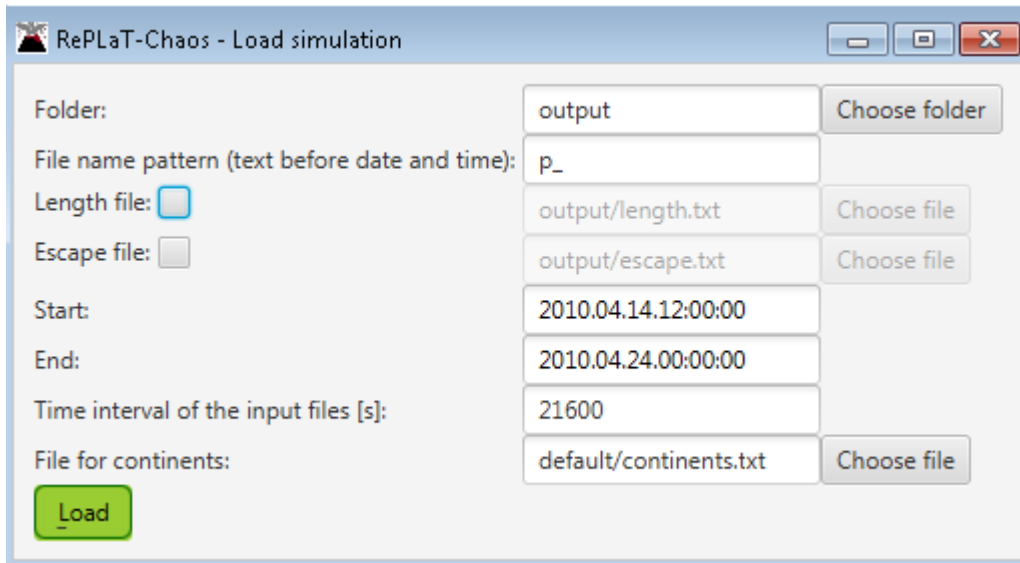
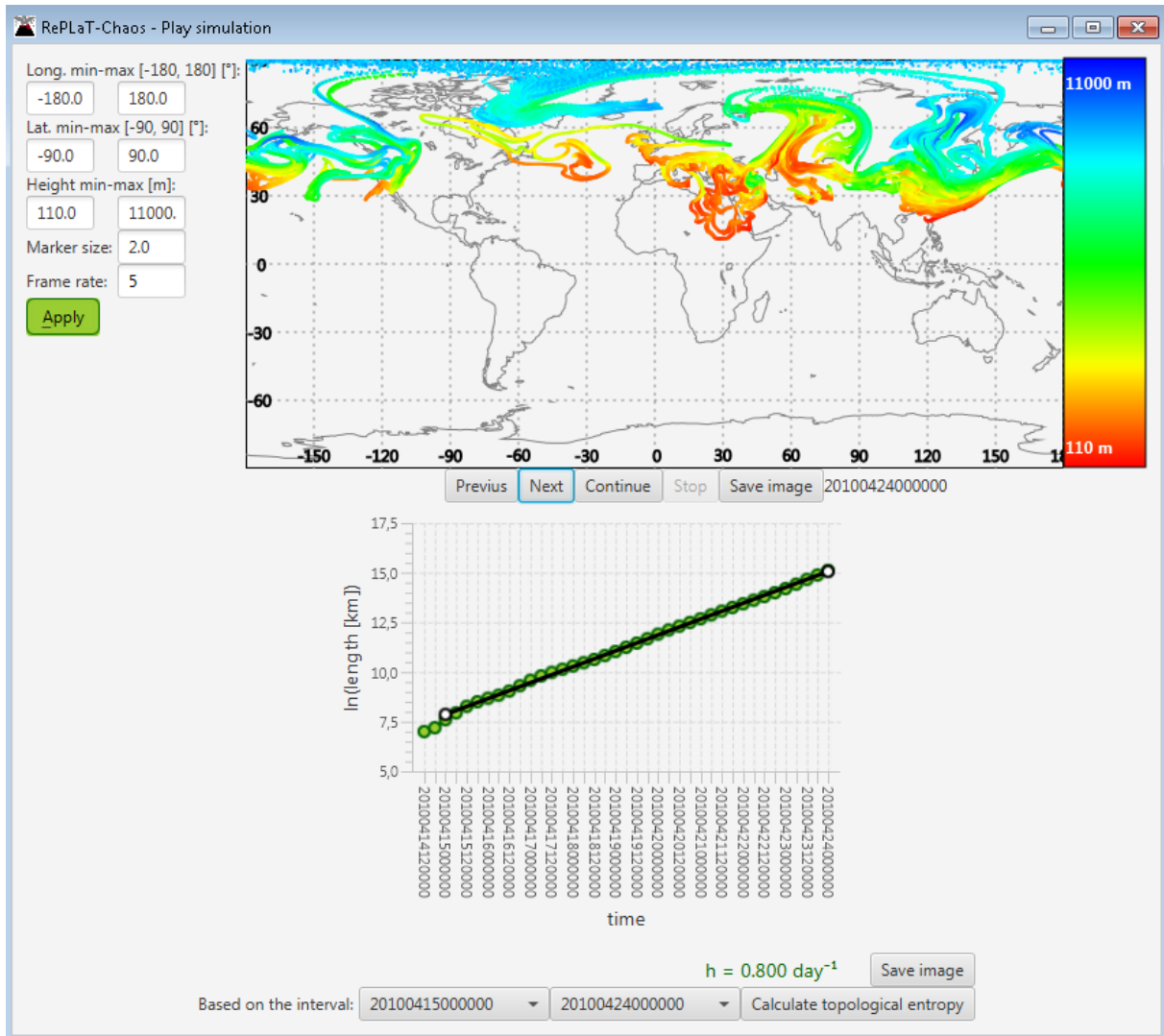


Figure 4: Simulation data setting to load saved simulation.

Saved simulation can be loaded by clicking the **Load simulation** menu item in **File** menu on the new simulation screen or by clicking the **Load** button in the case of an ongoing simulation. Then the parameters defining the saved simulation appear in a new window (Figure 4). For ongoing simulation textfields are filled with the parameters of the simulation, otherwise they are filled with the values loaded from a **default/default\_load\_setup.txt**. Parameters:

- **Folder:** folder of the files to be loaded. Clicking the **Choose folder** button it can be chosen or it can be written directly to the textfield.
- **File name pattern:** the part of the name of the particle files before date and time.
- **Length file:** should length data be loaded and if yes from which file. Clicking the **Choose file** button it can be chosen or it can be written directly to the textfield.
- **Escape file:** should data of the ratio of non-escaped particles be loaded and if yes from which file. Clicking the **Choose file** button it can be chosen or it can be written directly to the textfield.
- **Start:** start of the display (format: <yyyy.MM.dd.hh:mm:ss>)
- **End:** end of the display (format: <yyyy.MM.dd.hh:mm:ss>)
- **Time interval of the input files:** time interval [s] between subsequent files to be displayed (format: integer)
- **File for continents:** the file containing the coordinates ( $[-\pi; \pi] \times [-\pi/2; \pi/2]$  [rad]) of the curves of continents. Clicking the **Choose file** button it can be chosen or it can be written directly to the textfield.



**Figure 5. Displaying saved simulation and the time dependence of the length of the pollutant cloud.**

The selected simulation is loaded by clicking the **Load** button. If the **Length file / Escape file** is checked the time dependence of the natural logarithm of the length of the pollutant cloud / the time dependence of the natural logarithm of the ratio of the non-escaped particles is also appear on the panel (Figure 5, Figure 6). By default, the forward loop of the spreading of pollutant cloud is displayed according to the given **Frame rate**. The user can stop and continue the replay by clicking the **Stop** and **Continue** buttons, respectively, and can move the replay one by one frame forward/backward by clicking the **Previous/Next** buttons. The instantaneous position of the pollutant cloud can be saved by clicking the **Save image** button. The properties of the display can be changed similarly as described in section 2.5.5 by clicking the **Apply** button. Beyond those options the speed of the animation (frame rate) can be modified, too.

If the user loaded length data / data for the ratio of the non-escaped particles from file, she/he can select a start and end date and time from the two lists in the bottom of the panel. By clicking the **Calculate topological entropy / Calculate escape rate** button a line is fitted to the graph between the given time instants using the least squares approach and its slope

(i.e. the topological entropy (Figure 5)) / (-1)×slope (i.e., the escape rate (Figure 6)) is also presented. The graphs can be saved by clicking the **Save image** button.

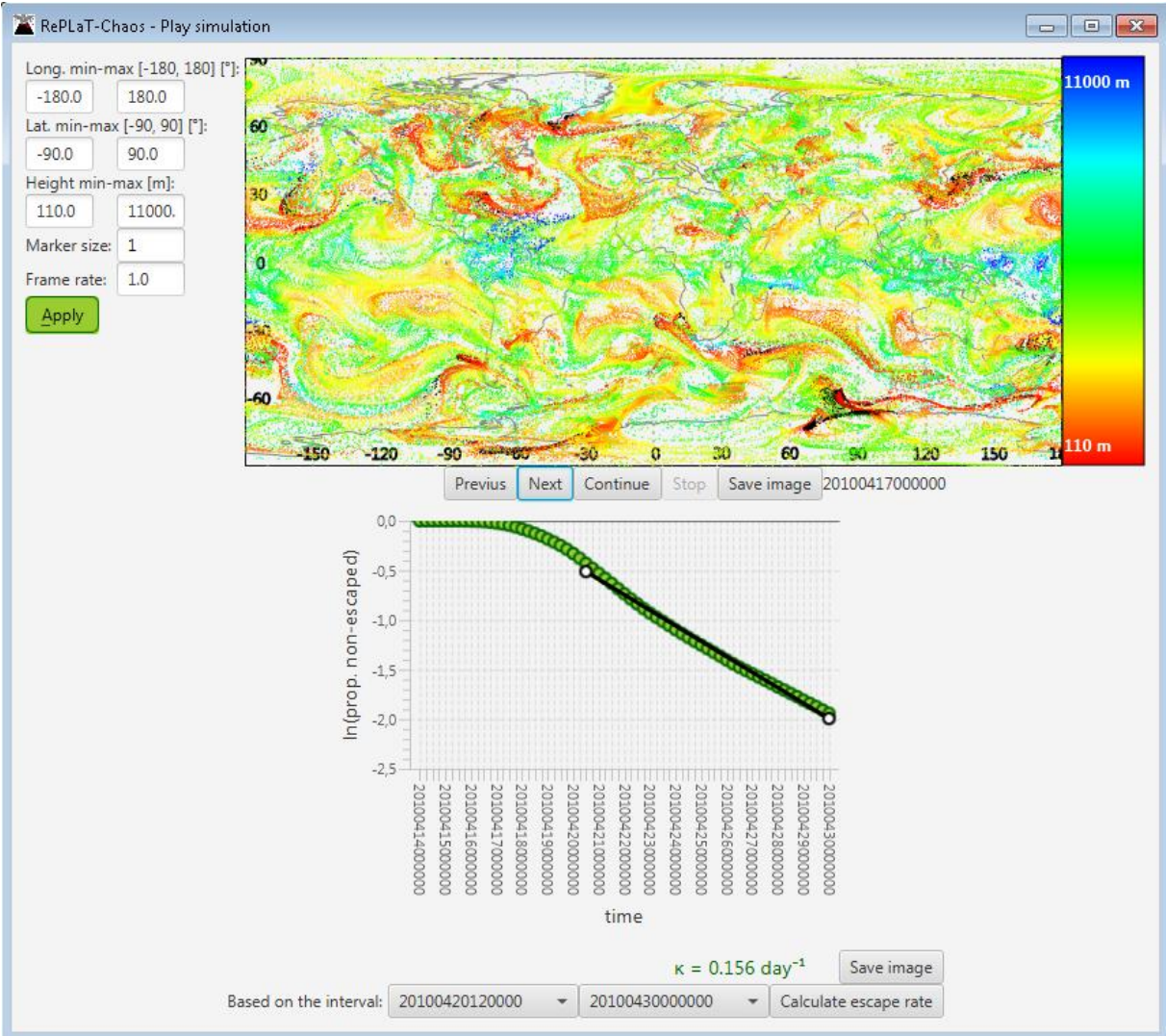


Figure 6. Displaying saved simulation and the time dependence of the ratio of non-escaped particles.

### 3. Theoretical background

#### 3.1. Calculation of particle trajectories

For small aerosol particles it can be shown that a particle is advected by the wind components in the horizontal direction and its vertical motion is influenced by its terminal velocity<sup>3</sup> and the vertical velocity component of air [2]. RePLaT-Chaos utilizes meteorological data given on regular longitudinal–latitudinal grid in the horizontal direction and at different pressure levels in the vertical direction. Therefore, the equations of motion of the particles are written in spherical coordinates in the horizontal and in pressure coordinates in the vertical direction in agreement with the structure of the meteorological data:

$$\begin{aligned}\frac{d\lambda(t)}{dt} &= u_{\text{rad}}(\lambda(t), \varphi(t), p(t), t) = \frac{u(\lambda(t), \varphi(t), p(t), t)}{R \cos(\varphi(t))}, \\ \frac{d\varphi(t)}{dt} &= v_{\text{rad}}(\lambda(t), \varphi(t), p(t), t) = \frac{v(\lambda(t), \varphi(t), p(t), t)}{R}, \\ \frac{dp(t)}{dt} &= \omega(\lambda(t), \varphi(t), p(t), t) + \omega_{\text{term}}(\lambda(t), \varphi(t), p(t), t),\end{aligned}$$

where  $\lambda(t)$ ,  $\varphi(t)$ ,  $p(t)$  denotes the longitude, latitude [rad] and pressure [Pa] coordinates of the particle at time instant  $t$ ,  $u$ ,  $v$ ,  $\omega$  are the zonal, meridional [m/s] and vertical [Pa/s] velocity components of air,  $u_{\text{rad}}$ ,  $v_{\text{rad}}$  are the zonal and meridional wind components measures in [rad/s],  $R = 6371$  km is the radius of the Earth, and  $\omega_{\text{term}}$  is the terminal velocity of the particle. The terminal velocity is

$$\omega_{\text{term}} = \begin{cases} \frac{2}{9} \frac{\rho_p r^2 g^2}{\nu} & \text{if } \text{Re} \ll 1, \\ \sqrt{\frac{8}{3} \frac{\rho_p \rho r}{C_D} g^3} & \text{if } \text{Re} \gg 1, \end{cases}$$

where  $\rho_p$  is the density of the particle,  $\rho$  is the density of air,  $r$  is the radius of the particle,  $g$  is the gravitational acceleration,  $\nu$  is kinematic viscosity,  $C_D$  is the drag coefficient (for particles assumed to be spheres  $C_D = 0.4$ ),  $\text{Re} = 2rV/\nu$  is the Reynolds number (where  $V$  denotes a characteristic velocity about which it can be shown that converges to the terminal velocity). The limit of  $r = 0$   $\mu\text{m}$  can be considered as gas „particles”, the terminal velocity of which is  $\omega_{\text{term}} = 0$ .

The required meteorological data for the above equations are the velocity components  $u$ ,  $v$ ,  $\omega$  and the density  $\rho$  and kinematic viscosity  $\nu$  of air. Since the latter two variables are not available in meteorological databases, we calculate them by means of temperature  $T$ . The density can be derived from the ideal gas law as follows:

$$\rho = \frac{p}{R_d T},$$

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<sup>3</sup> A particle falls through a motionless air with its terminal velocity if the sum of the drag force and the buoyancy is equal to the downward force of gravity.

where  $R_d = 287 \text{ J/kg/K}$  is the specific gas constant of dry air. The kinematic viscosity can be expressed by means of the dynamic viscosity  $\mu$ , where a  $\mu$  depends again on the temperature  $T$  represented by Sutherland's law [4]:

$$\nu = \frac{\mu}{\rho}, \text{ where}$$

$$\mu = \beta \frac{T^{3/2}}{T + T_S}, \text{ that is}$$

$$\nu = \frac{\mu}{\rho} = \beta \frac{T^{5/2}}{T + T_S} \frac{R_d}{p},$$

where  $\beta = 1.458 \times 10^{-6} \text{ kg m}^{-1} \text{ s}^{-1} \text{ K}^{-0.5}$  is Sutherland's constant and  $T_S = 110.4 \text{ K}$  is a reference temperature.

Now each quantity on the right-hand side of ordinary differential equation system describing the motion of the particles is known. RePLaT-Chaos solves the differential equations by an explicit second-order Runge–Kutta method (applied often also in other dispersion models). Hence the position  $\mathbf{r}(t + \Delta t) = [\lambda(t + \Delta t), \varphi(t + \Delta t), p(t + \Delta t)]$  of a particle at time instant  $t + \Delta t$  using the velocity  $\mathbf{v}(\mathbf{r}(t), t) = [u_{\text{rad}}(\mathbf{r}(t), t), v_{\text{rad}}(\mathbf{r}(t), t), \omega(\mathbf{r}(t), t) + \omega_{\text{term}}(\mathbf{r}(t), t)]$  at time  $t$  can be written as:

$$\mathbf{r}_0(t + \Delta t) = \mathbf{r}(t) + \mathbf{v}(\mathbf{r}(t), t) \Delta t \quad \text{and}$$

$$\mathbf{r}(t + \Delta t) = \mathbf{r}(t) + \frac{1}{2} [\mathbf{v}(\mathbf{r}(t), t) + \mathbf{v}(\mathbf{r}_0(t + \Delta t), t + \Delta t)] \Delta t.$$

The utilized meteorological data are available on a regular latitude–longitude grid on different pressure levels with a given (e.g., 3 or 6 hours) time resolution. Therefore, in order to solve the equations of motion of the particles and to calculate the particle trajectories, the quantities  $u$ ,  $v$ ,  $\omega$ ,  $T$  are interpolated to the location of the particles in each time step. RePLaT-Chaos applies linear interpolation in each of the three directions and in time.

Users have the option to choose between variable time step and constant time step for the trajectory calculation. In the former option, each time step for each particle is determined by the RePLaT-Chaos model based on the grid size and the current atmospheric velocity components using the Courant–Friedrichs–Lewy condition:

$$\Delta t_{\text{CFL}} = \text{CFL} \cdot \min \left\{ \frac{\Delta \lambda_g}{|u_{\text{rad}}(\mathbf{r}(t), t)|}; \frac{\Delta \varphi_g}{|v_{\text{rad}}(\mathbf{r}(t), t)|}; \frac{\Delta p_g}{|\omega(\mathbf{r}(t), t) + \omega_{\text{term}}(\mathbf{r}(t), t)|} \right\},$$

where  $\Delta \lambda_g$ ,  $\Delta \varphi_g$ ,  $\Delta p_g$  are the grid size in zonal, meridional [rad] and vertical [Pa] direction,  $\text{CFL} < 1$  is the Courant number. The minimal time step  $\Delta t_{\text{min}}$  is determined by the user. If the obtained  $\Delta t$  would be larger than the time interval  $(t_{\text{next}} - t)$  up to the next writing of particle data to file or up to the next reading of new meteorological fields then the time step is modified as:

$$\Delta t = \min \{ t_{\text{next}} - t; \max \{ \Delta t_{\text{CFL}}; \Delta t_{\text{min}} \} \}.$$

### 3.2. Topological entropy

The so-called topological entropy, a well-known quantity from dynamical systems theory, quantifies the complexity, irregularity of the advection of pollutant clouds in the atmospheric context. According to a definition of chaos, a system is chaotic if its topological entropy is positive [3]. It can be shown that an initially small pollutant cloud in the atmosphere becomes strongly stretched and folded; its length grows rapidly, while becoming filamentary and tortuous. The length  $L$  of the filament increases typically in an exponential manner in time  $t$ :  $L(t) \sim \exp(h t)$  (Figure 7). The exponent  $h$  is called the topological entropy and it is the rate of the exponential increase of the filament length. The larger the topological entropy is, the faster the length grows and the more complex, tortuous shape the pollutant cloud forms [5].

For one-dimensional pollutant clouds (line segments or filaments) the length of the pollutant cloud is calculated by the sum of the distances between its particles:

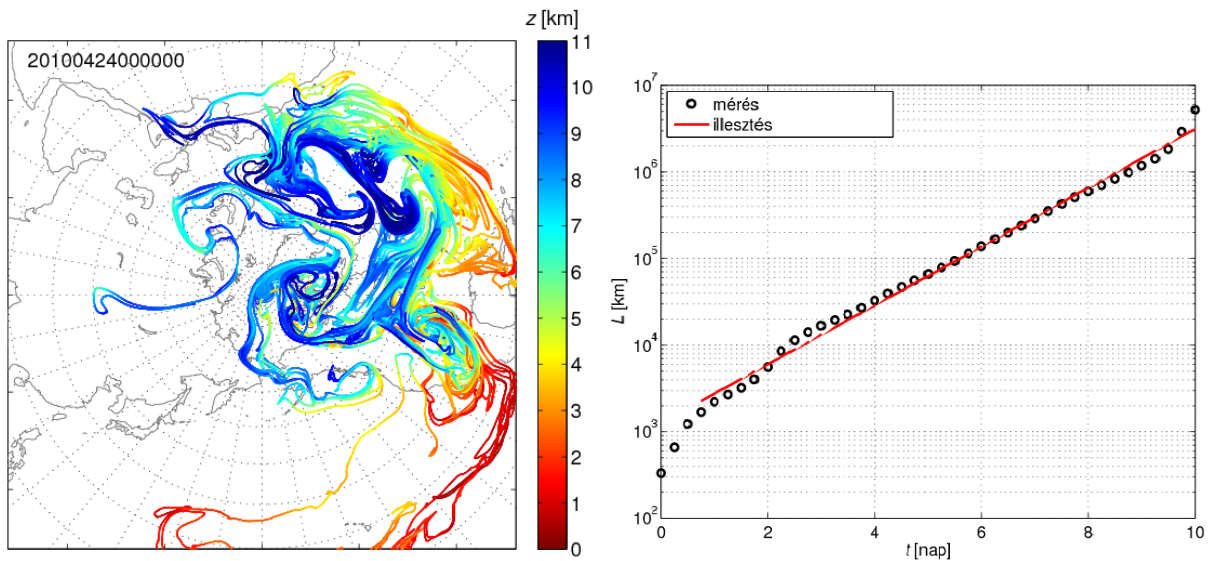
$$L(t) = \sum_{i=1}^{n-1} |\mathbf{r}_{i+1}(t) - \mathbf{r}_i(t)|,$$

where  $\mathbf{r}_i(t)$  is the position of the  $i$ th particle. Since vertical stretching proved to be  $10^{-2}$ – $10^{-3}$  times smaller than the horizontal one [3], it can be neglected. Thus, the length of a filament is obtained from the horizontal distances between particle pairs, and the distance [km] is calculated along great circles by

$$|\mathbf{r}_{i+1}(t) - \mathbf{r}_i(t)|_{\text{hor}} = \arccos [\sin \varphi_i \sin \varphi_{i+1} + \cos \varphi_i \cos \varphi_{i+1} \cos(\lambda_i - \lambda_{i+1})] \times \frac{180}{\pi} \times 111.1$$

where  $\lambda_i$  and  $\varphi_i$  are the longitudinal and latitudinal coordinate of the  $i$ th particle, respectively, and  $(180/\pi) \times 111.1$  converts the unit from radians to kilometers using the fact that the spherical distance of  $1^\circ$  along a great circle corresponds to a length of 111.1 km along the surface.

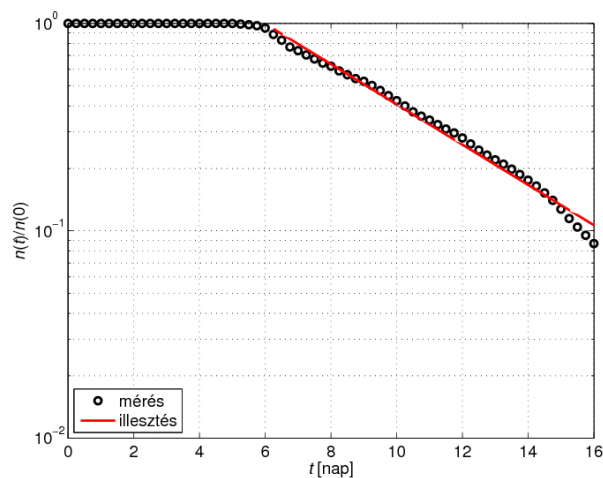
Since subsequent particles may travel far away from each other in time, the length of a pollutant cloud that consists of finite number of particles may be underestimated compared to a pollutant cloud with same initial conditions that consists of infinite number of particles (i.e. the pollutant cloud is continuous). This implies that after certain time when there are several “cut-off” segments among the particles the computed length differs from an exponential function, its values are lower than the expected ones. Hence to reduce the underestimation of the length if the distance of two neighboring particles becomes larger than a critical distance, a new particle is inserted between them.



**Figure 7.** The length of pollutant clouds grows exponentially in the atmosphere. Left: A filament with an initial length of 300 km covers a large part of the Northern Hemisphere within 10 days. Right: The time-dependence of the length of the filament.

### 3.3. Escape rate

The particles of the pollutant clouds carry out the chaotic motion for finite duration because, e.g., reaching the surface of the Earth they may deposit on the ground. This type of chaos is called transient chaos [3]. Nevertheless, there also exists a null measure set, the so-called chaotic saddle, initialized on which the particles would never leave it and carry out chaotic motion for an infinite amount of time. Being a null measure set the probability for the particles to be initially located on the chaotic saddle is zero and the particles sooner or later leave (i.e. “escape”) any arbitrary preselected regions of the saddle. The rapidity of this leaving is characterized by the escape rate. After a sufficiently long time  $t_0$ , the decay in the ratio  $n(t)/n(0)$  of survivors in the regions (in atmospheric spreading simulations in the region defined by the meteorological data) is generally exponential (similar to the law of radioactive decay):  $n(t)/n(0) \sim \exp(-\kappa t)$ , for  $t > t_0$  (Figure 8). Coefficient  $\kappa$  is called the escape rate [1].



**Figure 8.** The ratio of non-deposited particles of the pollutant cloud decreases exponentially in time.



## Acknowledgement

The project is supported by the János Bolyai Research Scholarship of the Hungarian Academy of Sciences and by the National Research, Development and Innovation Office (NKFIH) under Grants PD-121305, FK-124256.

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