Theses of the PhD dissertation

INVESTIGATION OF THE DISPERSION OF AIR POLLUTANTS BY THE RePLaT MODEL

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1 Introduction

In the last decades the demand for precise tracking and forecasting of atmospheric pollutants has increased due to the growing interest in environmental problems and consequently requirements for detailed prediction of health and economic hazards. Pollutants from different sources may be advected far away from their initial position and may cause air pollution episodes at distant locations. The effects of ash clouds from volcano eruptions (e.g. Mount St. Helens (1980), Pinatubo (1991), Eyjafjallajökull (2010)) and of pollutants from industrial accidents (e.g. Fukushima (2011)) underline the need for investigating pollutant dispersion in the atmosphere. According to the scale of the dispersion phenomenon, various models can be used to compute the dispersion of the pollutant. From regional to global scale the dispersion can be simulated by Eulerian models or by Lagrangian models.

Lagrangian models are useful for simulating the trajectories of pollutants from single sources, since they track individual particles or puffs. Their advantage against the Eulerian ones is that for the dispersion calculation no grid is needed. A class of the Lagrangian particle-tracking models tracks "ghost particles" (computational particles): any of these particles carry an artificial mass which is determined by the emitted mass and the number of particles used in the simulation. The properties of these particles usually do not coincide with those of any real pollutant particle. The mass attributed to the ghost particle decreases exponentially due to dry and wet deposition. In this approach a ghost particle is considered to be the center of mass of a large amount of adjacent pollutants. As the trajectory of these pollutants is represented by a single trajectory, the method of ghost particles tacitly supposes that these neighboring particles remain together forever. However, the chaotic nature of the advection dynamics implies that an initially small, compact ball of particles in the atmosphere becomes rapidly deformed into a complicated, filamentary shape of large extent. Therefore, the physical reality of ghost particle models remains questionable. Another class of the Lagrangian models follows "real particles": the particles in these models have fixed, realistic size and density. However, the recently available models of this type do not take into account the deposition and the scavenging of particles by precipitation.

In three-dimensional flows, as it is the case in the atmosphere, the advection of pollutants is chaotic. It is important to emphasize that this feature is independent of the turbulent nature of the atmosphere (a behavior, which is more complex than chaotic and characteristic to systems with high degrees of freedom). The advection dynamics of pollutants shows typical characteristics of the chaos, such as sensitivity to the initial conditions (the motion is unpredictable on long time range, the trajectories of initially nearby particles diverge within short time), irregular motion, and complex but regular (fractal) structures. An initially compact pollutant cloud becomes soon strongly stretched, its length grows rapidly, while becoming tortuous and folded. Although, the trajectory of a single particle is unpredictable, the investigation of a pollutant cloud containing

several particles gives insight to the spread of the contaminants.

The meteorological forecasts used for dispersion modeling are produced by the numerical solution of the atmospheric hydro-thermodynamic equations. Due to inaccuracies in the measurements and approximations used, the initial conditions of the meteorological model cannot be determined precisely. Initial errors are amplified due to the sensitivity to initial conditions. Therefore, there is uncertainty in the meteorological forecasts which can be quantified by the so-called ensemble technique based on the execution of multiple meteorological forecasts. Dispersion models are usually run by a single forecast which is considered to be the best one. However, it can be useful to perform simulations using the whole ensemble forecast, i.e. producing an ensemble dispersion prediction in order to get a detailed and more reliable overview of the uncertainties and possible hazards related to the dispersion event.

2   Scientific aims

The research focuses on the large-scale dispersion of pollutants in the atmosphere with the combination of two main topics: the simulation of air pollutant dispersion is related to the investigation and characterization of chaotic behavior in the approach of dynamical systems theory.

The aim of the work was to develop a relatively simple Lagrangian dispersion model which is able to describe real physical processes reasonably well. The model tracks individual aerosol particles with realistic size and density, and takes into account more processes than other models which also follow real particles. The model was to be evaluated by simulating documented events and comparing the results to measurement data.

Since our model describes the motion of realistic aerosol particles, it is suitable for the investigation of the chaotic behavior in dispersion and deposition processes from a dynamical systems point of view. Another aim of us was to study the phenomena mentioned in Introduction by means of the model developed, as:

- Investigation of the impact of the uncertainty of the meteorological forecast on the dispersion calculation.
- Investigation and characterization of the chaotic dynamics of the particles by means of some useful quantities, well-known from dynamical systems theory, that have not been applied for the atmosphere yet. These measures are:
    - the so-called *topological entropy* which characterizes the stretching rate of pollutant clouds, and
    - the so-called *escape rate* which describes the deposition rate of particles falling out from the atmosphere.

3 Applied methods

The goal of the development of our Lagrangian dispersion model was to create a model which is simple and easy to understand, and at the same time considers the atmospheric physical processes in a realistic manner. The simulated pollutant clouds contain several particles having realistic radius and density. The equation of motion of the particles is derived from Newton's equation. The drag force depends on the Reynolds number that quantifies the relation of hydrodynamical and viscous acceleration. Scale analysis reveals that the horizontal velocity of a particle takes over the actual local wind speed within a fraction of a minute, while in the vertical direction the terminal velocity also has to be taken into account besides the vertical velocity component of air. Therefore a particle is advected by the wind components in the horizontal direction and its vertical motion is, in addition, influenced by its terminal velocity which depends on the size and density of the particle and the density and viscosity of the air at the location of the particle.

Since the meteorological data utilized by the dispersion model have coarse resolution without resolving turbulent diffusion, the effect of small-scale turbulent diffusion on the particles is built into the model as a stochastic term. In the planetary boundary layer the computation of the vertical turbulent diffusivity is based on the Monin–Obukhov similarity theory, while the horizontal turbulent diffusivity is assumed to be constant in both this layer and the free atmosphere. In addition, the model also takes into account the effect of precipitation (for more detail see Thesis No. 1).

Far from the surface, in the free atmosphere, a particle has much smaller chance to be captured by a raindrop than particles traveling lower in the atmosphere. An order of magnitude estimate indicates that the role of turbulent diffusion in large-scale dispersion can be neglected. Therefore in simulations conducted solely in the free atmosphere, the motion of particles was determined only by the advection and by the gravitational settling. As the terminal velocity depends hardly on the altitude, in these simulations a constant terminal velocity was used in this layer.

For the dispersion simulations the forecasts and ERA-Interim reanalysis fields of the European Centre for Medium-Range Weather Forecasts (ECMWF) were used.

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 used. The model solves these differential equations for the subsequent position of the particles by using the explicit Euler method.

The applied meteorological data are available on a given latitude–longitude grid on different pressure levels or in a hybrid coordinate system with a given time resolution. The meteorological variables at the actual location of a particle are calculated using bicubic spline interpolation in the horizontal direction and linear interpolation in the vertical direction and in time. The only

exception is the boundary layer height which is determined by linear interpolation with the exception of the period between 12 and 18 o'clock (in local time) when the value at 12 o'clock is used in order to avoid the underestimation of the boundary layer height.

When the pollution was emitted into the free atmosphere (e.g. volcano eruption) the pollutant cloud was tracked in the simulations using the meteorological data on pressure levels. The reason behind this consideration was the fact that these computations are faster and easier than using data of the levels of a hybrid coordinate system. When the pollution was released close to the surface (e.g. nuclear power plant accident) calculations were carried out with data on hybrid levels, since the hybrid coordinate system has much more levels close to the surface that implies more accurate trajectory computations there.

In order to get comparable results (to measurements and to other model results) the concentration based on the number of the particles per volume or per area were calculated .

4 Theses

1. We developed the Real Particle Lagrangian Trajectory (RePLaT) model, which is a Lagrangian dispersion model tracking aerosol particles. The RePLaT model includes the effect of turbulent diffusion on the particles and the scavenging of particles by precipitation. The description of the model can be found in [4].

   - A novel feature of the model is the application of the impact of precipitation on individual particles by a random process which depends on precipitation intensity. The parametrization of wet deposition is based on the general Eulerian approach. We consider wet deposition as a random process that results in a particle being captured by a raindrop with certain probability (depending on precipitation intensity). Thereby the radius and density of the particle change suddenly to that of the raindrop. The trajectory of the "new" particle (a particle that turned into a raindrop) is computed using the terminal velocity based on the new properties of the particle. The particle does not leave the atmosphere instantaneously, but as a raindrop falling through the air.
   - Since the dynamics of aerosol particles is a kind of dynamical system, concepts from chaos theory can be applied and studied by using the RePLaT model.

2. In order to validate the RePLaT model we simulated the dispersion of volcanic ash from the Eyjafjallajökull's eruptions and the dispersion and deposition of the radioactive particles released during the accident of the Fukushima Nuclear Power Plant. The simulations were carried out, on the one hand, only within free atmosphere and, on the other hand, by taking into consideration the processes of the boundary layer, like precipitation, and turbulent diffusion depending on the height.

- In the case of the eruptions of the Eyjafjallajökull volcano we found reasonable agreement between the distribution of volcanic ash in the simulation and that indicated by satellite measurements. [1]

- In the case of the radioactive material released during the Fukushima Nuclear Power Plant accident the arrival times of the pollution at different locations coincided with the observations. In addition, the simulations were able to reproduce the measured concentrations of the noble gas ^{133}Xe appropriately. However, the RePLaT model overestimated the concentration of ^{137}Cs in some time periods mainly for European locations.

3. We also investigated the sensitivity of the dispersion characteristics with respect to the uncertainties coming from the input meteorological fields and, additionally, its dependence on the particle size. The dispersion of aerosol particles of different sizes was studied utilizing the members of a meteorological ensemble forecast using the simplifications applicable in the free atmosphere. [2]

- Considerable differences were found regarding the horizontal and vertical extension of the pollutant clouds of the different ensemble members. These clouds covered an area 5–10 times larger than the cloud of the deterministic forecast. We found that the smaller the particles are, the larger is the variance between the ensemble dispersion members.

- The differences in the ensemble of dispersion prediction were quantified by means of different statistical measures. The difference in the center of mass and in the standard deviation of each pollutant cloud of the ensemble members were calculated. We introduced a new quantity, the root-mean-square distance of each particle from its own clones in the different ensemble members. The relative variance of the Lagrangian ensemble statistics based on the standard deviation of the particles was found to be 2–3 times larger than the variability of the meteorological wind speed forecasts, therefore the uncertainties from the meteorological characteristics are growing in the dispersion model.

4. We also studied the dispersion of aerosol particles focusing on the chaotic motion of particles using the topological entropy, a quantity, well-known from dynamical systems theory, but not yet applied for the atmosphere. [1, 3]

- We illustrated in a case study that an initially small, one-dimensional pollutant cloud becomes strongly stretched and folded, and within short time the material line with its stretched and folded shape covers a significant fraction of a hemisphere. The length of the filament increases typically in an exponential manner in time. The topological entropy is the rate of the exponential increase of the filament length.

5. The seasonal distribution of the average topological entropy computed from 10-day time spans for ideal particles is determined for several geographical locations. [3]

- The largest topological entropies appear in the mid- and high latitudes, especially in winter (0.8–0.9 day^{-1}), while the smallest values can be found in the tropical belt (0.2–0.3 day^{-1}).

- The average topological entropy depends only slightly on the initial altitude of the particles. Based on our preliminary studies the topological entropy seems to hardly change in the presence of turbulent diffusion.

- It turned out that the local value of the topological entropy is a measure of the chaoticity of the state of the atmosphere and it can provide information about the speed of pollutant spread from a given location.

6. We illustrated that escape rate, a concept well-known from the theory of transient chaos, can be successfully applied to describe the deposition rate of aerosol particles. These investigations provide a Lagrangian foundation for the concept of deposition rates. [1, 4]

- In order to study the deposition dynamics on global scale, several particles were distributed uniformly over the globe at given altitudes. The number of non-escaped particles was found to decrease exponentially in time. We observed a time-scale separation with two different exponential decays. The short-term decay and long-term decay are described by the so-called short-term and long-term escape rate, respectively: they characterize the deposition of typical particles and that of exceptional ones that remain in the atmosphere for an extremely long time, respectively.

- The long-term escape rate — due to long-lived particles which become well-mixed in the atmosphere — does not depend significantly on the initial altitude of the particles, while the short-term escape rate is larger for particles initialized lower in the atmosphere.

7. We studied the influence of advection, turbulent diffusion and precipitation on the deposition dynamics of the particles, and the dependence of the escape rates on the particle size. [1, 4]

- Regarding the dependence of the escape rate on the particle radius the best approximate fit appears to be exponential both for the short-term escape rate and the long-term one in each of the three simulation setups.

- The impacts of wet deposition and turbulent diffusion are independent of particle size, but the role of precipitation was found to be stronger. The weakest dependence on the radius is found in the setup that takes into account the role of precipitation, while the

strongest dependence appears in the setup that neglects the impact of precipitation and turbulent diffusion. In contrast to precipitation that enhances the deposition for each particles size, turbulent diffusion increases the number of deposited particles for small ($r < 5$ µm) particles, and decreases it for larger ($r \approx 5$–10 µm) ones.

- Besides the escape rates we also determined the average lifetime of the chaotic motion of the particles, i.e. the average atmospheric residence time for different initial altitudes and particle sizes. The higher is the initial position of the particles, the larger is their average residence time. This quantity (as well as the escape rate) is found to depend exponentially on the particle radius. The reciprocal value of the short-term escape rate provides an estimate of the average residence time of typical particles, while that of the long-term escape rate characterizes the average residence time of small particles initialized in the free atmosphere.

5 Conclusions

In our research we developed the RePLaT Lagrangian dispersion model. In order to refine the simulations, in the future the model should be improved by additional factors related mostly to a more detailed description of the wet deposition. The separation of the in-cloud and below-cloud scavenging and the distinction between scavenging by rain and scavenging by precipitation can be some of the main aspects of the improvement.

In conclusion, we can state that the simulations carried out by the RePLaT model agree reasonably well with observations, and additionally, the model provides opportunity for a novel approach to the study of chaotic behavior in the atmosphere.

Publications related to the theses

[1] Haszpra, T. and Tél, T.: Volcanic ash in the free atmosphere: A dynamical systems approach. *Journal of Physics: Conference Series*, **333**, 012008, 2011. doi:10.1088/1742-6596/333/1/012008.

[2] Haszpra, T., Lagzi, I. and Tél, T.: Dispersion of aerosol particles in the free atmosphere using ensemble forecasts. *Nonlinear Processes in Geophysics*, **20** (5), 759–770, 2013. doi:10.5194/npg-20-759-2013.

[3] Haszpra, T. and Tél, T.: Topological entropy: a Lagrangian measure of the state of the free atmosphere. *Journal of the Atmospheric Sciences*, **70** (12), 4030–4040, 2013. doi:10.1175/JAS-D-13-069.1.

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Other publications related to the dissertation

[5] Haszpra, T., Kiss, P., Tél, T. and Jánosi, I. M.: Advection of passive tracers in the atmosphere: Batchelor scaling. *International Journal of Bifurcation and Chaos*, **22**, 1250241, 2012. doi:10.1142/S0218127412502410.

[6] Haszpra, T.: Világjáró részecskék a légkörben – Az Eyjafjallajökull vulkán kitörésének és a fukushimai balesetnek a tanulságai. *Természet Világa*, **144** (Káosz, környezet, komplexitás különszám), 67–72, 2013.

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[12] Haszpra, T., Lagzi, I. and Tél, T.: Dispersion of aerosol particles in the atmosphere: Fukushima. In: *Geophysical Research Abstracts*, vol. 15, p. 11028 (European Geosciences Union (EGU) General Assembly 2013, Wien, Austria), 2013.04.07–2013.04.12. (abstract and short talk).

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