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HINTS TO DISCRIMINATE THE CHOICE OF WET DEPOSITION MODELS APPLIED TO AN ACCIDENTAL RADIOACTIVE RELEASE

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Abstract: Atmospheric dispersion models are especially helpful for nuclear crisis management to anticipate the dispersion of the plume during the first hours following the release and later to analyze the transport and fate of radionuclides. In this process, the modelling of the wet deposition is a crucial point to correctly evaluate the ground contamination.
The wet deposition is generally divided in two main different mechanisms, the in-cloud scavenging (rainout) and the below-cloud scavenging (washout). For both of them a large number of models are proposed in the literature reflecting the uncertainties in our understanding of these phenomena. Currently there is no scientific consensus to discriminate between these models.
In order to improve our understanding on the magnitude of the modelling uncertainties a comprehensive sensitivity analysis was performed and it focused on the representation of the wet deposition fluxes.
A large number of model combinations are evaluated by comparison to the available observations of ground contamination. The hierarchies of models established from different statistical indicators show the lack of robustness of the wet deposition models. The aim is to establish a list of priority parameters for the ground contamination.

Key words: *Wet deposition, rainout, washout, Fukushima, parametrization choices*

INTRODUCTION

In order to model the transport of radionuclides bound to atmospheric particles and the ground contamination at the synoptic scale, the wet deposition is a crucial point. For example, Renaud et al (2004) show a very strong relationship between the rainfall and the ground contamination by 137-Cs over France consecutively to the Chernobyl accident. Unfortunately, the literature concerning wet deposition modelling is abundant but hardly consistent (Duhanyan and Roustan, 2011). This wide panel of models leads to a wide range of wet scavenging coefficient values, up to four decades and to a large uncertainty related to the choice of a wet deposition model to use for crisis and post-accidental simulations. This work is dedicated to the identification of the strength and the weakness of different modeling approaches of the wet deposition fluxes. There are two nuclear releases well documented and studied, the Chernobyl accident, and the Fukushima accident. But the data concerning the Fukushima accident are more numerous, and supposed to be more reliable. Furthermore, the Fukushima case shows important episodes of wet deposition, which dedicated particularly the event to this study. The case of the Fukushima accidental release of 137-Cs is simulated with the Eulerian model of the Polyphemus air quality modeling system (Mallet et al,, 2007, Quélo et al., 2007).

WET DEPOSITION

At the synoptic scale, the wet deposition is represented by a scavenging coefficient, which quantifies the share of the particle scavenged by unit of time. The two main mechanisms of collect occurring, the washout (below-cloud scavenging) and the rainout (in-cloud scavenging) are physically different, and can be modeled distinctly with their own scavenging coefficient. The below-cloud scavenging process is mainly due to the collect of the aerosol particles by the falling rain drops. For a detailed modeling, the particle size distribution, the drop size distribution, and the collection efficiency for each size of particles, drops and relative humidity should be known. Additionally, some other characteristics of the aerosol particles would be of interest (hygroscopicity, electrical charge), as for the drops (electrical charge). Due to the spatial resolution used for this kind of simulation and to the lack of corresponding observations, it is not possible to model carefully the below-cloud scavenging. However a large number of models are proposed in the literature to represent the below-cloud scavenging at the synoptic scale with reachable physical fields. The majority of these models quantify the scavenging coefficient as a function of the rainfall rate, but some others rely on another physical field, e.g. the relative humidity. For the in-cloud scavenging, the main process of particle collection by the cloud droplets is due to the droplets nucleation. But, again, the majority of the available models propose to determine the scavenging coefficient as a function of the rainfall rate. Some other introduces more complexity (liquid water content, relative humidity, cloud time-life …). Another issue about the wet scavenging is the determination of the cloud height (base and top). Some in-cloud scavenging models include already the cloud determination itself (Pudykiewicz, 1989), but for most of them, the determination of the cloud vertical extend is external.

METHODOLOGY

A sensitivity analysis is performed to try to discriminate between wet deposition models proposed in the literature. This analysis is based on the use of a large number of simulations, each simulation relying on a different combination of the models studied (models of wet deposition, models of dry deposition, sources…). All the simulations performed share a common platform and some common settings. An horizontal resolution of 0.05° is used, in a mesh of 120x120x15 cells. The vertical resolution is a non-linear scale from 0 m to 8000 m, (0, 40, 120, 280, …, 8000m). For these simulations, the horizontal diffusion is neglected. The meteorological fields used for all the simulations are provided by a WRF simulation, using a resolution of 0.05° with a one-hour time-step. A list of models and input data to be tested has been established. As the simulated wet deposition fluxes are sensitive to other components of the models, some parameterizations that impact the wet deposition have been also investigated in the analysis. These numerous simulations are compared to the observations of deposition of 137-Cs. The quality of each simulation is evaluated through the computation of some statistical indicators.

Choice of models and data

The choices of the “parameters” (model or data) have been done relying on some simple preliminary sensitivity tests. Thus, four important parameters are shown here. Two of them correspond directly to the purpose of the work, the in-cloud scavenging and the below-cloud scavenging. The below-cloud models used are of different complexity: those proposed by Laakso (2003) and Andronache (2003) provide scavenging coefficient that vary only with the rain intensity whereas the models of Slinn (1977) and Quérel (2014) propose a collection efficiency that take into account the size of the particles and of the raindrops and can be combined with different representation of the raindrop spectrum (Blanchard, 1953; Coutinho and Tomas, 1995; Sekhon and Srivastava, 1971). The scavenging coefficient of the in-cloud models can also be only rain dependent (Jylha, 1991; Scott, 1982; Ellenton, 1988), but it can integrate some other physical fields. The model of Roselle and Binkowski (1999) use the cloud liquid water content and an estimation of the clouds life time. The scavenging coefficient can also be completely rain independent (Pudykiewicz, 1989). Two other “parameters” are considered: the dry deposition and the source term. The use of a simple constant deposition velocity, set to 0.2 $cm.s^{-1}$, is compared to the use of a more detailed model able to take into account the size of the particles and the land cover (Zhang, 2001). The source terms considered in the study are those established in Mathieu et al. (2012), Saunier et al. (2013) and Winiarek et al. (2014). These sources are respectively a mixed-term, an inversion from the gamma dose rate, and an inversion from the deposition. A total of 240 simulations have been done, corresponding to the total number of possible combinations of the investigated “parameters”.

Comparison of the simulations and the observations

Following the Fukushima Daiichi NPP accident, we faced the lack of well formatted, usable and localized measurement data to compare with our models. It has led us to consider a database approach. We defined the main objectives that this database should reach: gathering the radiological measurement data of any type from multiple sources into a single place, ensuring the traceability of the measurement campaigns and simplifying the data mining to end users. The first objective has been achieved by developing a database schema taking into account the variety of the measurement data type. In this context, we favored user defined data types rather than a finite number of data types. This guarantees that any measurement data can be stored into the database, as far as the data are defined temporally and spatially. The second objective has required the storage of metadata, attached to the measurement campaigns. This ensures the traceability of the data and allows us to exclude or filter easily outliers on a metadata basis. The third objective is the most significant in terms of time saving and has been achieved by the use of a database with a spatial extension. Such an extension adds spatial functionalities to the database environment and authorizes data mining by spatial queries. In the context of the present work, the database was used to evaluate the mean deposition of Cs-137 on the computational meshes at a given date. This end-user need has been performed by querying all the Cs-137 deposition data on each computational cell of the mesh, deriving the value with radioactive decay and carry out an arithmetic mean. A wide area of 100 km near to the Fukushima nuclear power plant is excluded from the comparison. This exclusion allows keeping only three distinct interesting events: the north, south-west and south deposition pattern. Indeed, one of the difficulties in order to analysis the results with cumulated observations is the potential addition of different events.

Statistical indicators

Some statistical indicators are used to evaluate the quality of each simulation. The main ones are the correlation, the Normalized Mean Square Error (NMSE), the fractional bias, the factor 10, the factor 2 and the Bias-Corrected Root Mean Square Error (BCRMSE). These indicators are chosen to be representative of different aspects of the simulation's abilities (good bias, strong values, surface of deposition …)

RESULTS

The best configuration, a chimera

For each statistical indicator the "best configuration" is found to be different. Worst, none of the "parameters" investigated allows to reach a consensus. For the five best configurations for each statistical indicator, all the models tested appear at least one time, at the exception of the Laakso (2003) model, of the Coutinho and Tomas (1995) model of rain associated to the Slinn (1977) model of collection efficiency and the Blanchard (1953) model of rain associated to the Quérel (2014) model of collection efficiency. To illustrate this issue, the **Table 1** and the following figures show the configurations of the best factor 2 and the best BCRMSE and their corresponding deposition maps. The observations used to determine the statistical indicators are also shown.

Table 1. Best configuration example obtained for each parameter

Indicator looked	Dry Deposition	BCS	ICS	Source	BCRMSE	Fac2
BCRMSE	Zhang	monodispersed rain (Coutinho and Tomas, 1995) + Quérel, 2012	Pudykiewicz, 1989	Mathieu, 2012	**0,037**	0,129
Factor 2	constant	Function of rain intensity (Andronache, 2003)	Ellenton, 1988	Saunier, 2013	0,524	**0,589**

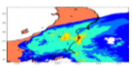

Figure 1. Deposition map obtained with the configuration providing the best factor 2.

Figure 2. Observations used to determine the statistical indicators.

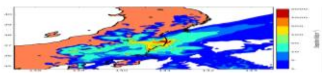

Figure 3. Deposition map obtained with the configuration providing the best BCRMSE.

Statistical impact of the choice of one "parameter"

An approach to estimate the impact of one "parameter" is to compare the results of a given choice for this "parameter" to the results of the other options investigated. Each member of a set of simulations performed with the same given choice is compared to the simulations sharing the same configuration except for the considered parameter. This comparison is made for each statistical indicator. At the end, a distribution of the differences is obtained for each parameter and statistical indicator.

With the help of these distributions, it is possible to determine the ratio of the simulation for which the use of one model rather than another one has improved the simulation, and how much the simulation was improved in mean. For example, if we consider the dry deposition and the correlation, 70% of the simulations give best results by using the constant deposition velocity rather than the Zhang (2001) deposition velocity. The best improvement in using a constant velocity is 30%, and the worst degradation is only 0.7%. The mean amelioration is equal to 12%. Thus, according to the correlation, the best dry deposition to use is the constant one.

This approach emphasizes the bad results obtained with the Andronache et al. (2003) below-cloud scavenging model. Indeed, in mean, the use of this model allows a significant degradation of the BCRMSE (+4 in mean), NMSE (+4 in mean), correlation (-4% in mean), fraction bias (-0.6 in mean). But, at the opposite, the use of this model strongly improves the value of the factor 2 and factor 10 (+8% in mean, + 30% in maximum for both). In fact, the value of the share of the below-cloud in using this model is surprisingly low (less than 5%).

Concerning the in-cloud scavenging, no significant influence in the model is observed, at the exception of the Pudykiewicz (1989) model for the factor 2 and 10. For these indicators, using the Pudykiewicz (1989) model decreases the factor 2 in mean of 4% (at worst the use of the Pudykiewicz decreases the factor 2 of 48%), and the factor 10 is decreased in mean of 14%, and for the worst case, the factor 10 falls of 64%. Finally, these factors are never improved by the use of the Pudykiewicz model. But, considering the other indicators, this model is not a degrading one. The *Figure 4* shows the distribution of the improvement of the factor 2 of each in-cloud models compared to the others.

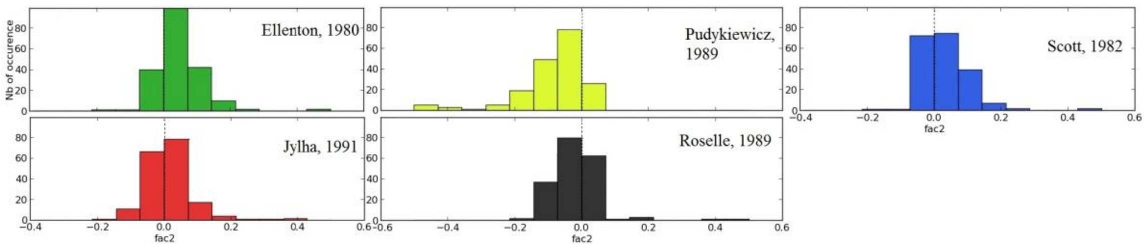

Figure 4. Relative improvement of the correlation of in-cloud scavenging ($Fac2_{tested\ model}$ - $Fac2_{other\ model}$)

DISCUSSION

The contradictions observed between the indicators lead to be careful with the results. For the cases where one model is statistically better than another, the improvement has to be evaluated by maps of differences. The evolution may be not significant for the modeled ground contamination. A "local" approach to determine the best model configuration is not suitable in these conditions. For both the below-cloud and the in-cloud scavenging, the use of more physically detailed models(e.g. estimating the drop size distribution) do not bring a systematic improvement in the simulations. The complex models are not over-represented in the established hierarchies of best combinations. For the next steps, the impact of models on the wet deposition will be analyzed for each particular deposition event. The aim is to identify some strong links between a combination of models and a particular meteorological situation. No particular model is coming with an universal improvement for the deposition modeling. However, the current analysis can be extended to groups of parameters (e.g. compare the statistics of Ellenton+ Laakso and Scott+Ulbrich&Slinn).

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