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SENSITIVITIES IN WET DEPOSITION MODELLING APPLIED TO THE FUKUSHIMA NUCLEAR ACCIDENT

Stijn Van Leuven[1,2,3], Pieter De Meutter[1,3], Johan Camps[1], Andy Delcloo[2,3] and Piet Termonia[2,3]

[1]SCK CEN, Belgian Nuclear Research Centre, Mol, Belgium
[2]Department of Physics & Astronomy, Ghent University, Ghent, Belgium
[3]Royal Meteorological Institute of Belgium, Brussels, Belgium

Abstract: Atmospheric transport and dispersion modelling can provide crucial information to evaluate the impact of regular or accidental releases of radionuclides. One of the processes that depletes radionuclides from the atmosphere is deposition, which causes ground contamination thereby potentially impacting the food chain. Wet deposition in particular plays an often dominant role in the total deposition of radioactive material following a release of radioactive particulates in the atmosphere. In this work, the sensitivities of wet deposition contributions will be quantified with atmospheric dispersion model FLEXPART. The transport and deposition of ^{137}Cs as a result of the Fukushima Daiichi Nuclear power plant accident is used as a test case for this sensitivity study. A novel method is developed which extracts the individual scavenging contributions from the FLEXPART simulations and optimises them by comparing to measurements.

Key words: *ATM, wet deposition, sensitivity study*

INTRODUCTION

The simulation of wet deposition in atmospheric transport modelling (ATM) remains difficult in large part due to uncertainties in the parameterisation schemes used in simulations (Draxler et al., 2015, Solazzo and Galmarini, 2015, Quérel et al., 2021, Fang et al., 2022). In general, the wet scavenging coefficients range between 10^{-5} and 10^{-2} s^{-1} (Baklanov and Sørensen, 2001, Sportisse, 2007). This variation is mostly caused by the dependence of wet scavenging on the particle size distribution and rain intensity. The values of scavenging coefficients and their dependencies are difficult to measure, leading to large uncertainties in their implementation in ATM's. With the atmospheric dispersion model FLEXPART (Stohl et al., 1998, Stohl and Thomson, 1999, Stohl et al., 2005), the user is able provide a scaling factor for the wet scavenging coefficients as an input to the simulations. A revised wet deposition scheme was introduced in FLEXPART with its latest version v10.4, introducing four parameters that need to be specified for wet deposition of aerosols (Grythe et al., 2017, Pisso et al., 2019). The appropriate model parameter values are to be chosen by the user, and may differ from case to case (Grythe et al., 2017).

It is however not straightforward to find the optimal parameter values, and any such method can be computationally expensive. Here we explore a new method to optimise the scavenging coefficients which should allow for more efficient prescribing of the wet deposition parameters in FLEXPART. This is demonstrated by applying the proposed methodology to the transport and deposition of ^{137}Cs following the Fukushima Daiichi Nuclear power plant (FDNPP) accident.

METHODOLOGY

The wet deposition scheme of FLEXPART v10.4 contains up to four different scavenging processes for a given particle species, each with a corresponding scavenging coefficient Λ. The scavenging processes depend on the physical conditions of the atmosphere where the particle is located. A distinction between gases and aerosol particles is made (see Table 1).

Table 1. Model parameters for the scavenging coefficients used in FLEXPART 10.4 wet deposition scheme.

	Gases	**Particles**	
		$T < 0°C$	$T > 0°C$
In-cloud	A', B'	IN	CCN
Below-cloud	A, B	C_{snow}	C_{rain}

This new scheme was developed to better take into account the scavenging efficiencies in different conditions (Grythe et al. 2017). For gases the common parameterisation $\Lambda = AI^B$ is used (I being the rain intensity). The parameterisation of aerosol particles is more elaborate. It consists of in-cloud scavenging (nucleation) and below-cloud scavenging (impaction). The nucleation occurs by activated particles forming either cloud droplet condensation nuclei or ice nuclei, and is parameterised by the column cloud water and surface rain intensity. The overall strength of nucleation can be manually scaled by CCN and IN respectively. Below-cloud scavenging is driven by rain or snow, and is parameterised by the precipitation intensity and aerosol size. It can be manually scaled by C_{rain} and C_{snow}.

To improve the process of selecting the appropriate parameter values in ATM's, we develop a method which consists of two steps: 1) quantify individual scavenging contributions and 2) rescale the obtained contributions in an optimal way. These steps are described in more detail below.

1. Quantifying scavenging contributions

The airborne concentration at a given receptor is reduced by the scavenging that has taken place in the plume during its trajectory from the source to the receptor. The idea is to quantify how much the concentration has been reduced due to the individual scavenging processes in Table 1. Extracting these from the simulations is however not as straightforward as simply disabling some scavenging contributions, as this method will introduce certain 'compensation effects': decreasing one scavenging process causes the contribution of the other processes to simultaneously increase since there is now more concentration available to deplete. To avoid this effect, the individual scavenging contributions are directly extracted from the FLEXPART simulations.

2. Optimisation scheme

The concentration c that remains after scavenging is given by

$$c = c_0 - \sum_i \Delta c_i \,, \tag{1}$$

where c_0 is the concentration if there were no scavenging, and Δc_i are the scavenging contributions for each process. The goal is to scale the Δc_i's to find an optimal fit of c to the observations. However, directly scaling the different contributions Δc_i can quickly lead to negative concentrations. Therefore a more involved scaling scheme is proposed. With every scavenging process i, a scavenging factor A_i will be associated which acts on the concentration field through $\Delta c_i = A_i\left(c_0 - \sum_{j\neq i} \Delta c_j\right)$. In other words, the scavenging factor A_i acts on the part of the concentration field that is not affected by the other scavenging processes. From this definition, it can be shown that the individual Δc_i's scale as follows:

$$\Delta c_i = c_0 \frac{A_i}{1 - A_i}\left(1 + \sum_j \frac{A_j}{1 - A_j}\right)^{-1}. \tag{2}$$

This formulation prevents the concentrations from becoming negative and also reproduces the physical compensation effect when altering the strength of one of the scavenging processes. New Δc_i's are then generated by scaling the A_i's, so that an optimised fit can be obtained between the remaining concentrations c and the observations.

The above methodology is applied to ^{137}Cs transport and deposition following the FDNPP accident. For the FLEXPART simulations, we use meteorological data from European Centre for Medium-Range Weather

Forecasts (ECMWF) and the FDNPP source term of Terada et al., 2020. The ^{137}Cs observations are provided by the Comprehensive Nuclear Test-Ban-Treaty Organisation, which were made using the radionuclide stations from the International Monitoring System (IMS).

RESULTS

The different scavenging contributions as obtained by FLEXPART for IMS station RN71 (as an example) are shown in Figure 1. The left panel shows the contributions for the default FLEPXART scavenging parameters. These contributions are scaled by c_0 so that the sum $c/c_0 + \sum_i \Delta c_i/c_0$ is equal to 1, in accordance with Equation (1). Overall the impaction by snow (C_{snow}) has the greatest contribution for this station. Optimising the Δc_i's gives the results on the right panel. By fitting the scaling to the observations, impaction by rain (C_{rain}) and scavenging by cloud condensation nucleation (CCN) substantially increase.

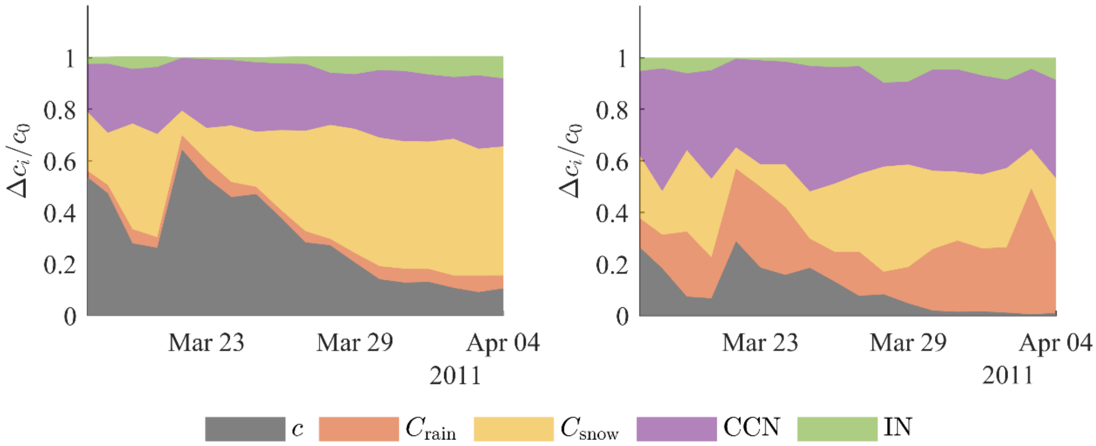

Figure 1. Scavenging contributions to simulated ^{137}Cs concentration at IMS station 71. Left: default FLEXPART deposition, Right: optimised scavenging contributions.

The distributions of the observational and model concentrations are shown on Figure 2 and Figure 3. Figure 2 shows the result for default FLEXPART parameters, while Figure 3 shows the concentrations as a result of optimising the scavenging contributions as seen above. For both Figures the distributions are shown in histogram form (left panels) and on a scatter plot (right panels). The default FLEXPART parameter values lead to an overestimation of the concentrations by around one to two orders of magnitude in this case, i.e. there is too little deposition (Figure 2). The optimisation scheme reduces this bias significantly by increasing the scavenging (Figure 3).

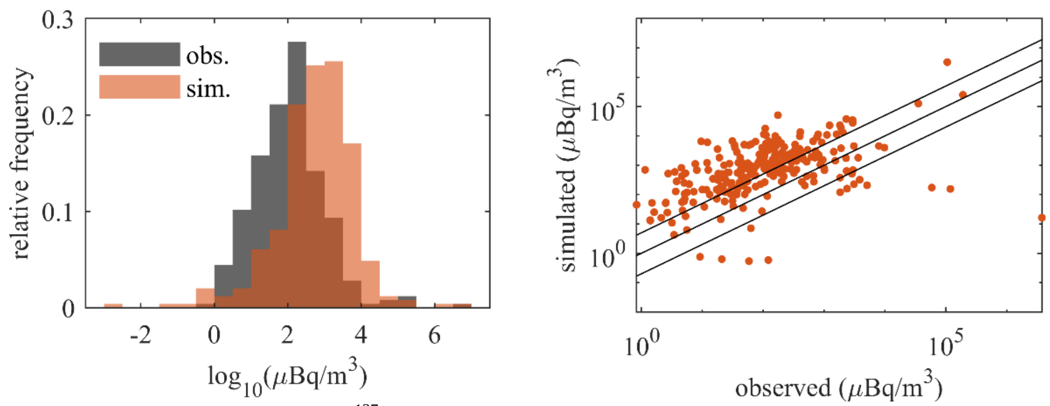

Figure 2. Concentrations with default ^{137}Cs deposition parameters compared with observational data. Black lines on the left panel show an offset of factors (1/5, 1, 5).

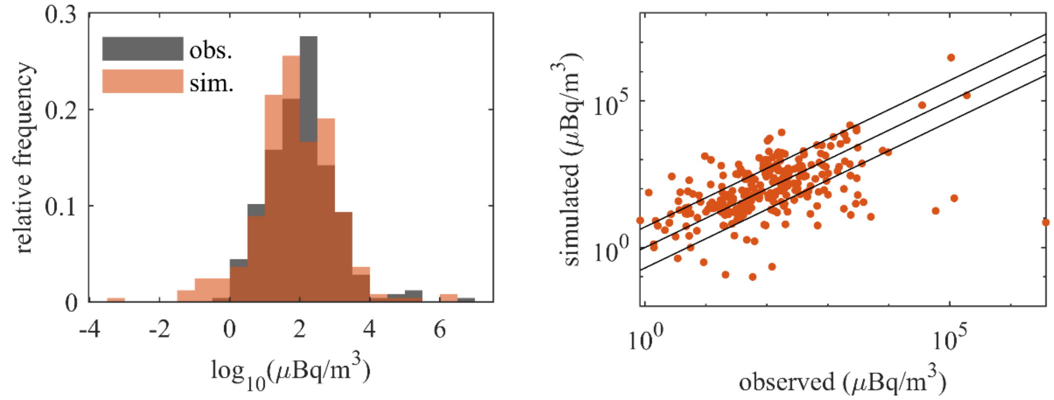

Figure 3. Optimised concentrations compared with observational data. Black lines on the left panel show an offset of factors (1/5, 1, 5).

CONCLUSIONS

Modelling of wet deposition is a crucial aspect of atmospheric transport modelling, yet large uncertainties remain in its parameterisations due to limited measurements of some scavenging processes. Sensitivity experiments could be conducted with ATM's which involve perturbing the scavenging coefficients and running the model thousands of times. Here, we have introduced a new method to scale and fit the deposition to concentration measurements using FLEXPART v10. The scaling is fitted to IMS observations, which shows a drastic improvement over the default FLEXPART deposition parameters. In principle this method only requires a single simulation, thereby greatly increasing the efficiency of prescribing the appropriate deposition parameters in FLEXPART. In the future, we intend to include the contribution of dry deposition as well.

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