18th International Conference on
Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
9-12 October 2017, Bologna, Italy

A PARTICLE SIZE DEPENDENT WET DEPOSITION SCHEME FOR THE LAGRANGIAN DISPERSION MODEL, NAME

Helen N. Webster[1] *and David J. Thomson*[1]

[1]Met Office, FitzRoy Road, Exeter, EX1 3PB, UK

Abstract: A particle size dependent wet deposition scheme for the Lagrangian dispersion model NAME is described, The scheme calculates a scavenging coefficient which varies with particle diameter and solubility and accounts for known variations in the efficiency of different sized particles to act as cloud condensation (or ice) nuclei in clouds or to be captured by falling precipitation elements below clouds. For a range of aerosols, comparisons are made between the predicted wet deposition and air concentration fields obtained using the particle size dependent wet deposition scheme and the traditional bulk wet scavenging parametrization used by default in NAME.

Key words: *wet deposition, particle diameter, aerosols, scavenging, Lagrangian model.*

INTRODUCTION

In regions of precipitation, wet deposition processes are highly efficient in scavenging atmospheric aerosols. Aerosols can enter precipitation elements in different ways. Within cloud they can act as cloud condensation (or ice) nuclei in the formation of cloud droplets (or cloud ice crystals). Below cloud, aerosols can be swept out by impaction with falling precipitation elements.

The efficiency of below-cloud and in-cloud scavenging processes is known to depend on the aerosol size, the aerosol properties, the cloud phase or the precipitation type and the precipitation rate. Good cloud condensation nuclei are submicron in size (~ 0.08 – 1.0 μm) and hygroscopic (water absorbing). Below-cloud scavenging is efficient for very small particles (< ~ 0.01 μm), due to their Brownian motion, and for coarse particles (of the order of a few microns), which are readily collected by impaction due to their inertia. Below-cloud scavenging is much less efficient for aerosols in the intermediate size range, which tend to be swept along streamlines and around falling precipitation elements, thereby escaping capture.

Traditionally, atmospheric dispersion models use bulk wet scavenging parametrizations, where the change in mass (M) is modelled by a simple loss equation using the concept of a scavenging coefficient (Λ),

$$\frac{dM}{dt} = -\Lambda M. \qquad (1)$$

Here the scavenging coefficient represents the mean scavenging rate over all particle sizes and takes the form

$$\Lambda = ar^{b}, \qquad (2)$$

where r is the precipitation rate in mm hr^{-1} and a and b are parameters which can vary for different types of precipitation (rain / snow) or cloud phase (liquid / ice), for different scavenging processes (in-cloud / below-cloud) and for different species. Feng (2007) showed that bulk parametrizations can significantly overestimate the scavenging of aerosol mass in heavy or long-duration medium rains since they do not take into account reductions in the scavenging coefficient with time due to changes in the particle size distribution of the atmospheric aerosol.

A SIMPLE PARTICLE SIZE DEPENDENT WET DEPOSITION SCHEME

Explicit calculation of wet scavenging processes requires detailed aerosol and cloud microphysics schemes, representing the formation and growth of cloud droplets and cloud ice crystals and the life cycle of aerosols including their emission or formation, growth, aggregation and chemical transformations. There are, however, limitations in the level of sophistication possible in offline dispersion models, which generally use basic cloud and precipitation information and are often required to be efficient for emergency response purposes. Variations in the scavenging coefficient with particle size can, however, be included in offline Lagrangian dispersion models (Feng, 2007; Feng, 2009; Grythe et al., 2017). Grythe et al. (2017) describe a recent development to include a particle size dependency to the impaction removal scheme in FLEXPART. Feng (2007) designed a 3-mode below-cloud parametrization for MLDP0 with bin-defined wet scavenging coefficients for particles within the nucleation, accummulation and coarse bins. The particle-size dependent wet deposition scheme introduced into NAME and described here is based on the work of Feng (2007; 2009) and uses information on in-cloud aerosols from Stier et al. (2005) and Croft et al. (2010).

Below-cloud scavenging

Below-cloud scavenging by precipitation is efficient for small particles with diameters < 0.01 μm and large particles with diameters greater than a few micrometers. For particles with diameters between 0.01μm and a few micrometres, the scavenging coefficient can be smaller by up to a few orders of magnitude (see Figure 2 in Feng (2007) and Figure 5 in Feng (2009)). For frozen precipitation, below-cloud scavenging depends on the shape of the snow particles or ice crystals (e.g., column or plane) and scavenging coefficients obtained from experiments are highly variable and uncertain.

In the NAME particle size dependent below-cloud scavenging scheme a modification of Feng's 3-mode approach is adopted. Figure 1 shows the variation with particle size of the below-cloud scavenging parameters (a and b) for rain (shown in red) and snow (shown in blue), together with, for comparison, NAME's default bulk scavenging parameters (which do not vary with particle size). A few key aerosol diameter values $(d_p)_i$ are chosen from Figure 2 in Feng (2007) and Figure 5 in Feng (2009) with appropriate scavenging coefficients $\Lambda_i = a_i r^{b_i}$. For aerosol diameters between the chosen key values, linear interpolation in terms of ln a or ln b versus ln d_p is used (see Figure 1).

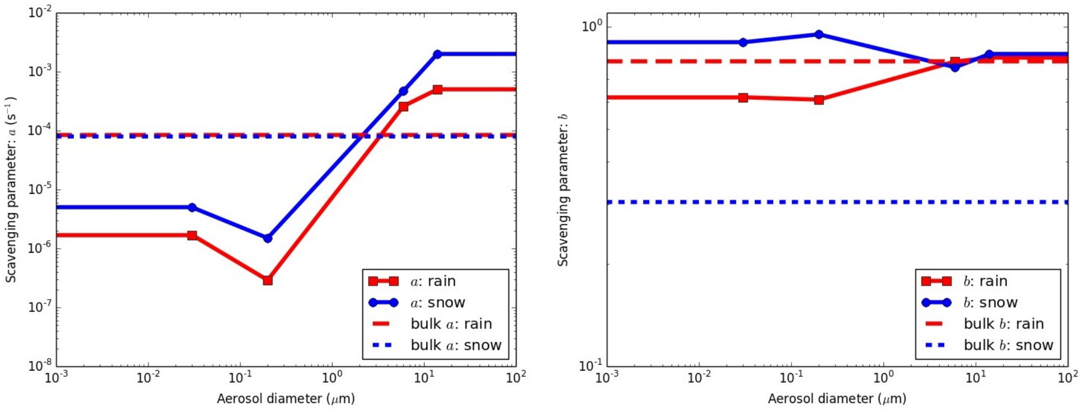

Figure 1. Variation of below-cloud scavenging parameters, a (left) and b (right), with particle diameter. For comparison, the bulk scavenging parameters used by default in NAME are shown.

In-cloud scavenging

In cloud, the NAME particle size dependent wet deposition scheme uses bin-defined scavenging coefficients (Λ_i) for different particle size bins (nucleation: $d_p \leq 0.01$ μm, Aitken: $0.01 < d_p \leq 0.1$ μm, accummulation: $0.1 < d_p \leq 1.0$ μm and coarse: $d_p > 1.0$ μm). The bin-defined scavenging coefficient takes the form

$$\Lambda_i = a_i r^b = AR_i r^b, \tag{3}$$

where $A = 5.2 \times 10^{-4}\ s^{-1}$ and $b = 0.79$ are constants which do not vary with aerosol size and are based on below-cloud scavenging parameters for particles equivalent in size to typical cloud droplets (Feng, personal communication). R_i is the in-cloud scavenging ratio for aerosol mode i and is defined as the fraction of the in-cloud aerosol in mode i that is embedded within the cloud liquid water / cloud ice. R_i takes the values for stratiform clouds given by Stier et al. (2005). The in-cloud scavenging ratio depends on the phase of the cloud and whether the aerosol is hygroscopic (soluble) or hydrophobic (insoluble). Figure 2 shows the variation of the NAME particle size dependent in-cloud scavenging parameter *a* with particle size in liquid, mixed and ice phase clouds, for soluble and insoluble aerosols. Constant values are adopted within each particle size bin, with discontinuities in the scavenging parameter at the bin boundaries. For comparison, the bulk in-cloud scavenging parameters (which do not vary with particle size or solubility) are also shown.

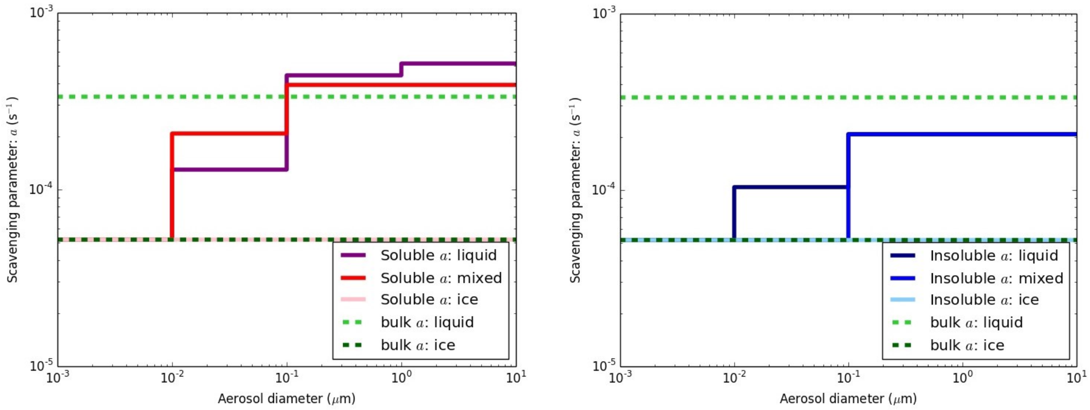

Figure 2. Variation of in-cloud scavenging parameter *a* with particle diameter in liquid, mixed and ice phase clouds for soluble (left) and insoluble (right). For comparison, the in-cloud bulk scavenging parameters used by default in NAME are shown.

TESTING

A range of typical NAME simulations modelling different particle species have been used to assess the effect of the particle size dependent wet deposition scheme and to compare to simulations conducted using the default bulk scavenging parametrization.

Ammonium sulphate

A simple continuous point release from ground level of a soluble atmospheric aerosol (ammonium sulphate, $(NH_4)_2SO_4$) with a typical atmospheric aerosol diameter of 0.4 µm was modelled and the wet deposition fields obtained using both schemes compared. Particles of this size are inefficiently scavenged by falling precipitation elements and the particle size dependent wet deposition scheme assumes smaller below-cloud scavenging coefficients than the bulk parametrization. In-cloud, the scavenging coefficients are similar in both wet deposition schemes for soluble aerosols of this size, with slightly higher scavenging coefficients adopted by the particle size dependent wet deposition scheme in liquid phase clouds than in the bulk parametrization. Figure 3 shows the time integrated wet deposition fields obtained using both wet scavenging schemes. The predicted fields are very similar with just some subtle differences. Near to the release point deposition amounts are slightly lower from the particle size dependent scheme, due to the smaller below-cloud scavenging coefficients. Since less aerosol is deposited near to source by the particle size dependent scheme, atmospheric concentrations are higher downwind and this can result in an increase in wet deposition amounts further afield.

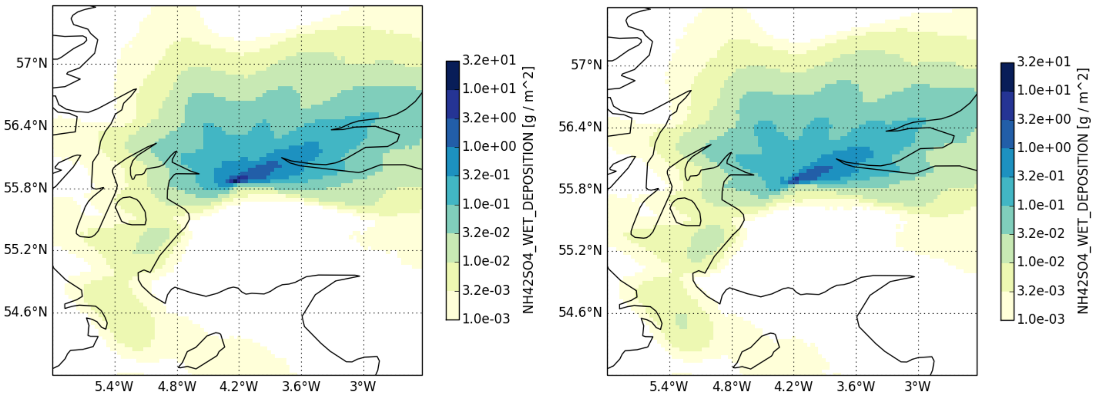

Figure 3. A comparison of NAME predicted total wet deposition of $(NH_4)_2SO_4$ obtained using the bulk wet scavenging scheme (left) and the new particle size dependent wet deposition scheme (right).

Volcanic ash

The eruption of Eyjafjallajökull in 2010 was used as an example to assess the impact of the particle size dependent wet depositon scheme on volcanic ash simulations. Volcanic ash is assumed to be hygroscopic. The particle size distribution commonly used in NAME for volcanic ash represents ash particles up to 100 µm in diameter and is intended to represent the fine ash fraction which survives near source fallout. The majority of the mass (70%) is placed in the coarse particle size range 10 - 30 µm. Figures 1 and 2 show that the particle size dependent wet deposition scheme for soluble aerosols adopts larger scavenging parameters than the bulk scavenging parametrization in this size range. Consequently the particle size dependent wet deposition scheme deposits more volcanic ash, resulting, when there is significant wet deposition, in lower air concentrations than when the bulk scavenging parametrization is used (see Figure 4). Comparing against observations of peak ash concentrations (as done by Webster et al., 2012), this slight reduction in predicted ash concentrations is apparent with an increase in the number of underpredictions and a decrease in the number of overpredictions. The particle size dependent wet deposition scheme removes the tendency of the method described in Webster et al. (2012) to overpredict airborne ash concentrations for the eruption of Eyjafjallajökull in 2010, giving a similar percentage of model over- and under-predictions.

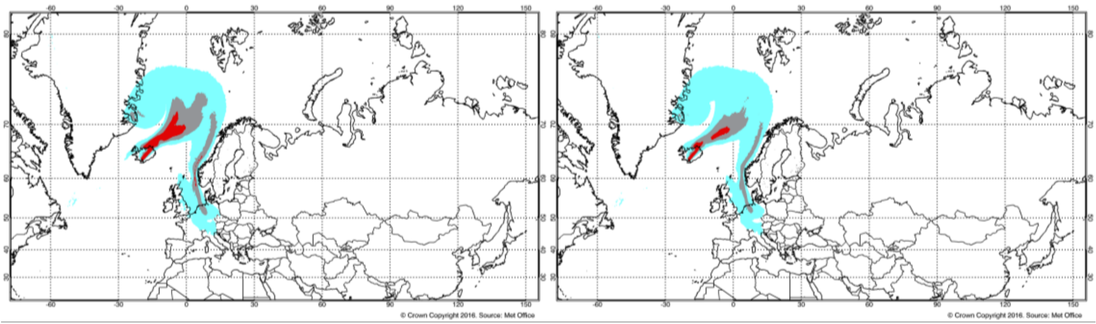

Figure 4. A comparison of NAME predicted 6-hourly averaged ash concentrations from the eruption of Eyjafjallajökull in 2010 using the bulk wet scavenging scheme (left) and the new particle size dependent wet deposition scheme (right). Ash concentrations between 12:00 - 18:00 UTC on 18/05/2012 and from FL000 - FL200 are shown in three concentration ranges: 200 - 2000 µg m^{-3} (cyan), 2000 - 4000 µg m^{-3} (grey) and > 4000 µg m^{-3} (red).

Dust

The NAME dust scheme calculates a source term based on the underlying land surface and meteorological information (for example, near surface wind speed). A particle size distribution is used, with particle diameters ranging from 0.0632 µm to 63.2 µm. Mass is mainly released in the coarse size range on particles with diameters between 6.32 µm and 20 µm. Dust is hydrophobic and hence the NAME particle size dependent wet deposition scheme for insoluble aerosols is selected. In the particle size dependent wet deposition scheme, below-cloud scavenging coefficients for coarse particles are larger than those used in the bulk scavenging scheme (see Figure 1). For insoluble aerosols in this coarse size range, the in-cloud scavenging coefficients in the particle size dependent wet deposition scheme are slightly smaller for liquid phase clouds than those in the bulk scavenging scheme (see Figure 2b). Figure 5 compares the NAME predicted total wet deposition of dust over a 24-hour period using both wet deposition schemes. The particle size dependent wet deposition scheme deposits slightly more dust, due to larger below-cloud scavenging parameters.

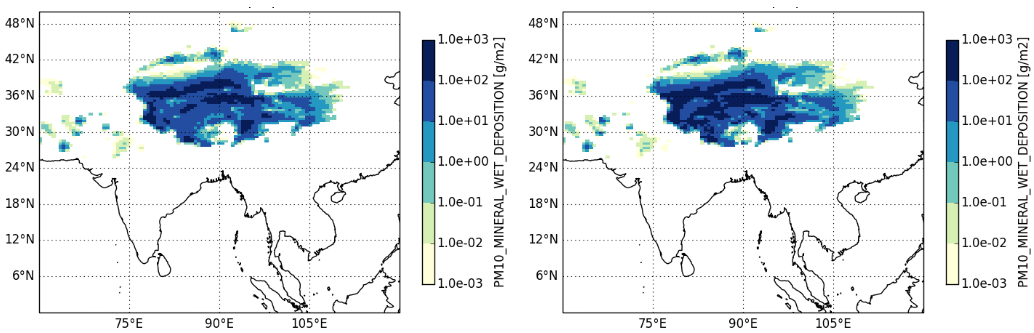

Figure 5. A comparison of NAME predicted 24-hour integrated wet deposition of dust obtained using the bulk wet scavenging parametrization (left) and the particle size dependent wet deposition scheme (right).

CONCLUDING REMARKS

A particle size dependent wet deposition scheme has been added into NAME. The scheme takes into account the known dependency of the scavenging coefficient on particle size and solubility. The effect on predicted wet deposition and air concentration fields for a range of aerosols has been assessed. Simulation runtime is an important consideration for models which are used in operational settings and the particle size dependent wet deposition scheme has no noticeable impact on the run-time of NAME.

Particle size information may not always be well known and errors in the particle size distribution will translate into errors in the scavenging rate. Furthermore input cloud and precipitation data used by offline dispersion models is often crude and the accuracy of predicted wet deposition fields will largely be driven by the accuracy of this input data, regardless of the wet deposition scheme used.

REFERENCES

Croft, B., U. Lohmann, R.V. Martin, P. Stier, S. Wurzler, J. Feichter, C. Hoose, U. Heikkilä, A. van Donkelaar and S. Ferrachat, 2010: Influences of in-cloud aerosol scavenging parameterizations on aerosol concentrations and wet deposition in ECHAM5-HAM. *Atmos. Chem. Phys.*, **10**, 1511-1543.

Feng, J., 2007: A 3-mode parameterization of below-cloud scavenging of aerosols for use in atmospheric dispersion models, *Atmos. Environ.*, **41**, 6808-6822.

Feng, J., 2009: A size resolved model for below-cloud scavenging of aerosols by snowfall, *J. Geophys. Res.*, **114**, D08203, doi:10.1029/2008JD011012.

Grythe, H., N.I. Kristiansen, C.D. Groot Zwaaftink, S. Eckhardt, J. Ström, P. Tunved, R. Krejci and A. Stohl, 2017: A new aerosol wet removal scheme for the Lagrangian particle model FLEXPART v10, *Geosci. Model Dev.*, **10**, 1447-1466.

Stier, P., J. Feichter, S. Kinne, S. Kloster, E. Vignati, J. Wilson, L. Ganzeveld, I. Tegen, M. Werner, Y. Balkanski, M. Schulz, O. Boucher, A. Minikin and A. Petzold, 2005: The aerosol-climate model ECHAM5-HAM. *Atmos. Chem. Phys.*, **5**, 1125-1156.

Webster, H.N., D.J. Thomson, B.T. Johnson, I.P.C. Heard, K. Turnbull, F. Marenco, N.I. Kristiansen, J. Dorsey, A. Minikin, B. Weinzierl, U. Schumann, R.S.J. Sparks, S.C. Loughlin, M.C. Hort, S.J. Leadbetter, B.J. Devenish, A.J. Manning, C.S. Witham, J.M. Haywood and B.W. Golding, 2012: Operational prediction of ash concentrations in the distal volcanic cloud from the 2010 Eyjafjallajökull eruption. *J. Geophys. Res.,* **117**, D00U08, doi:10.1029/2011JD016790.