

The role of deposition in atmospheric transport of radionuclides



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Introduction

Current atmospheric transport modelling (ATM) at the CTBTO/IDC does not consider dry or wet deposition of radionuclides. While this is appropriate for noble gases, it is well known that particulate or particle-borne nuclides (and also gaseous iodine (I_2 , which is however not measured in the CTBT network) are subject to dry as well as wet deposition. It is mainly wet deposition which limits the atmospheric half-life of accumulation-mode aerosols in the midlatitude troposphere to a couple of days.

As a part of the project flexRISK (<http://flexrisk.boku.ac.at/>) which studied the risk in Europe due to severe nuclear power plant (NPP) accidents, ATM calculations were carried out for all NPP sites in Europe and 2,788 cases in the years 1995 and 2000–2009. This gives a climatologically well representative sample. As both a noble gas and an aerosol species were tracked for 15 days, it is a good base to investigate the deviation between calculations with and without deposition.

Transport calculations

- Model used: Lagrangian particle dispersion model FLEXPART v8 (<http://flexpart.eu>)
- Domain: Europe and surroundings, see Figure 1
- Meteorological input data: ERA interim, horizontal resolution 0.75°, temporal resolution 3 h, all model levels.
- Duration of calculations: 15 days forward
- Number of calculations: 88 in the year 1995, further 2700 in the years 2000–2009 (spread evenly over seasons and time of the day), for all NPPs in Europe
- Species considered: noble gas (zero deposition), aerosol (deposition details see below)
- Release: Spread evenly between 0 and 50 m above ground level, duration 4 h. Unit release for both species, thus output concentration gives **source-receptor sensitivity (SRS)**
- Concentration output: coarse grid with 1° resolution (used here), fine grid over inner part of domain with 10 km resolution. Temporal resolution 3 h (values averaged over 3 h).

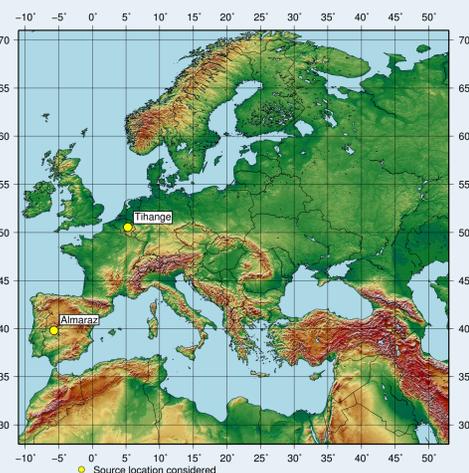


Figure 1 : FLEXPART output domain. Release (NPP) sites Tihange (Belgium, maritime climate) and Almaraz (Spain, Mediterranean climate) were used for the evaluations.

Evaluation methods

Main parameter: ratio r between the concentration in air of the aerosol species c_p to that of the noble gas species c_{NG} , which is a function of time:

$$r(t) = \frac{c_p(t)}{c_{NG}(t)} \leq 1 \quad (1)$$

The removal of aerosols from the atmosphere can be approximately described by an exponential law. As the initial concentration of both species is identical, and as their transport is identical except for the deposition effect, we can write

$$r(t) \approx \exp(-\alpha t), T = \alpha^{-1} \quad (2)$$

where α is an atmospheric removal constant and thus T the atmospheric residence time (reduction to $1/e \approx 0.37$).

Percentiles and mean values of r as well as residence times were calculated.

Deposition in FLEXPART

Dry deposition

The dry deposition velocity for particles is calculated with a resistance approach as

$$v_d(z) = [r_a(z) + r_b + r_a(z)r_b v_g]^{-1} + v_g \quad (3)$$

For details, see Stohl et al. (2005).

Wet deposition

Wet deposition in FLEXPART is calculated by applying a scavenging rate Λ to the mass of the species associated with each particle, at each time step

$$m(t + \Delta t) = m(t) \exp(-\Lambda \Delta t) \text{ with } \Lambda = AI^B \quad (4)$$

with I being the precipitation intensity in mm/h, and A, B being empirical constants. For full details, see again Stohl et al. (2005).

Prior to version 8 (here: **scheme A**):

- Eq. 4 applied to the total column of air with (values chosen in flexRISK) $A = 1 \cdot 10^{-4}, B = 0.8$ (gives Λ_{old}).

Versions ≥ 8 : new scheme which differentiates between *sub-cloud scavenging (low efficiency)* and *in-cloud scavenging (high efficiency)* (Eckhardt et al., 2008, here with fix by Seibert and Arnold (2013)) (here: **scheme B**):

- Eq. 4 for below-cloud scavenging with reduced constants, use $A_{bc} = 2 \cdot 10^{-6}, B_{bc} = 0.62$ to obtain Λ_{bc} (index bc for below-cloud)
- Areas where clouds could not be properly diagnosed: apply schema A with Λ_{old}
- In-cloud scavenging (index ic = in-cloud, H cloud height in m):

$$\Lambda_{ic} = \frac{A_{ic} I^{B_{ic}}}{H} \text{ where } A_{ic} = 1.25, B_{ic} = 0.64 \quad (5)$$

Thus, for a cloud height of 10 km, the result is similar to Eq. 4 within the cloud, and below-cloud scavenging is reduced by a factor 100. However, if the diagnosed cloud height is much less, unreasonable high scavenging rates may result, as for numerical reasons, consistency between cloud height and precipitation rate is not guaranteed.

Thus, modified **schemes C–H** were introduced and tested.

Results – various tests

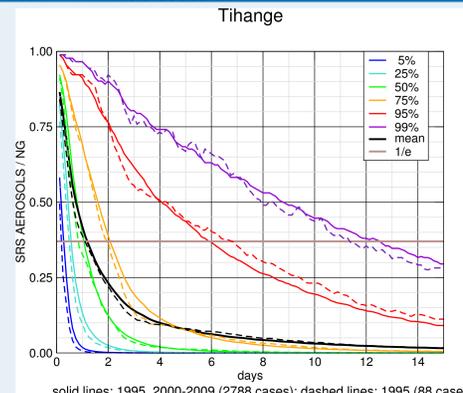


Figure 2 : Percentiles and means of the ratio r at Tihange, **scheme A**.

Scheme A, Fig. 2: The aerosols are scavenged very rapidly, $1/e$ reached after 1.2 d on average (year 1995 is sufficiently representative). Therefore, various modified schemes were tested (Fig. 3):

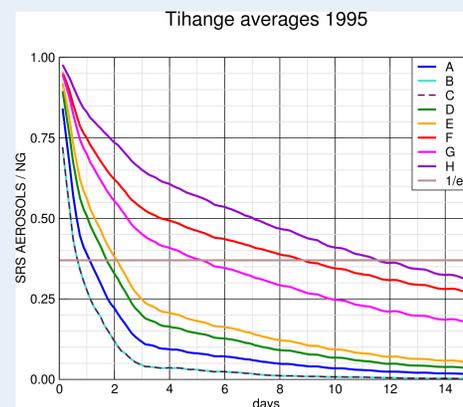


Figure 3 : Mean value of the ratio r at Tihange, **schemes A–H**.

- A** Old scheme, just 1.2 d lifetime!
- B** New scheme – even shorter lifetime!
- C** New scheme, but in areas without diagnosed cloud, $A = 1 \cdot 10^{-5}, B = 0.8$. Only insignificant changes. \Rightarrow failure to diagnose clouds is very rare.
- D** Limit $\Lambda_{ic} \leq \Lambda_{old}$. Lifetime increases, but still below 2 d.
- E** $A_{ic} = 0.125$ (reduction by factor 10!). Surprisingly, only moderate increase of T to about 2 d.
- F** Limit $\Lambda_{ic} \leq 0.1 \Lambda_{old}$. Big increase of T to 9 d. \Rightarrow Too thin clouds are a major problem. New lifetimes probably a bit too long.
- G** Keep principle of scheme H, but double Λ_{old} and A_{bc} . $\Rightarrow T=5$ d, very reasonable! However, r falls off very quickly at the beginning (when dry deposition plays a more important role because plume is still near ground).
- H** Like G, but no dry deposition. This leads to a slower decay of r until about day 4. $T \approx 9$ d. \Rightarrow dry deposition is important, maybe a bit too strong. If that's true, wet scavenging parameters could be moved up closer to original values while maintaining realistic residence times.

Results – best scheme

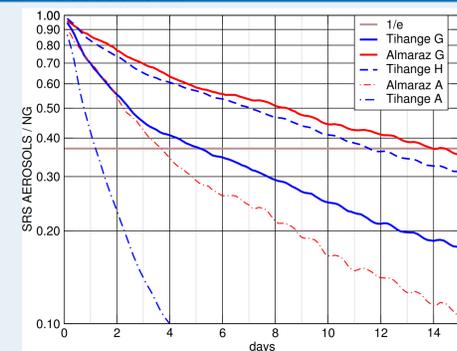


Figure 4 : Mean values of the ratio r in logarithmic scale at Tihange (**schemes A, G and H**) and Almaraz (**schemes A, G**). The slope of the curves is proportional to the removal rate $\alpha(t)$. Scheme A is for comparison.

Table 1 : Atmospheric residence times T (defined as $1/e$ -life, not half-life!) and removal rates in %/day are shown at specific transport times. The “total” column has two values: the first one is the time when the line in Fig.4 crosses $1/e$, the second is a resultant value after from 0–15 d (or until plume left domain), as if one would connect the $d=0$ and $d=15$ values by a straight line in this diagram.

	Transport time								
	1 d		5 d		12 d		total		
	T (d)	%	T (d)	%	T (d)	%	T (d)	%	
Almaraz G	6.4	15.5	12.2	8.2	29.2	3.4	14.0	14.5	6.9
Tihange G	2.8	35.7	11.3	8.8	12.3	8.1	5.2	8.7	11.5
Tihange H	5.4	18.7	15.8	6.3	15.8	6.3	11.6	12.9	7.8

Figure 4 and Table 1 allow a more quantitative view on the temporal change of α , and compare Tihange with Almaraz.

- At Tihange, α is stabilised after ca. 4 d at a value corresponding to $T \approx 12$ d, after initially 3 d.
- Even without dry deposition (scheme H), removal rates are higher at the very beginning ($T=5.4$ d) than later ($T \approx 16$ d). Maybe because then the particles spread into regions with less precipitation, or weather situations where they stay within the domain are more anticyclonic (less wind and less rain).
- Scheme G for Almaraz and scheme H for Tihange give very similar results. This is probably coincidence. Average wet scavenging should be lower at the dry site of Almaraz.
- The residence time at Almaraz (scheme G) is about 14 days.
- Due to the faster removal at the beginning, the actual residence time is better reproduced by the $1/e$ crossing time (first value under total in Table 1). It is about **5 d for Tihange and 12 d for Almaraz**.

Table 2 : Percentiles and mean of $1/\alpha$ for **scheme G** (most realistic). This is the ratio of aerosol SRS (deposition ignored) to SRS (deposition included).

	Transport time		
	1 d	5 d	10 d
5th percentile	3.72	14.96	29.51
25th percentile	1.79	4.89	9.33
median	1.33	2.85	4.95
75th percentile	1.14	1.91	2.82
90th percentile	1.04	1.32	1.62
average	1.43	2.67	4.04

Table 2 shows that:

- already after 1 day, neglecting deposition leads to an average overprediction of the aerosol SRS by 40%;
- after 5 d, values are overpredicted by a factor of 2.67, which may vary (5th–95th percentile) between 1.32 and 15!

Conclusions

- In general, the atmospheric lifetime of accumulation-mode aerosol ($0.1-1 \mu\text{m}$) is estimated to be on the order of 3–12 d (Warneck, 1988; Kristiansen et al., 2012).
- Standard FLEXPART wet deposition for aerosols is substantially too strong, with lifetimes of 1–4 d both in the old (pre-version 8) and new (V8+quick fix) scheme.
- Reasonable aerosol lifetimes were achieved by
 1. reducing all the A parameters by about a factor 5, except for below-cloud scavenging (this very small value was doubled)
 2. introducing a strict upper bound for the in-cloud scavenging ratio to prevent low cloud heights from amplifying this ratio in unreasonable ways
- With new scheme G, lifetimes are realistic with $\approx 5-10$ d.
- Longer times appear at the drier Spanish site compared to the maritime site in Belgium.
- A significant decrease of removal rates during the transport time was found. It is related to dry deposition, possibly also other factors.
- The reason for the overestimation of wet scavenging is different in the **old (A)** and **new (B)** scheme: in **A**, the parameter A was too conservative while in **B**, clouds diagnosed as too shallow amplify Λ .
- More investigations on the variability of both dry and wet deposition and their causes in FLEXPART are desirable.
- Nevertheless, it is clear that totally neglecting dry and wet deposition leads to massive overprediction of SRS values for particulates after a few days, and in some cases even after a single day.

References

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