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Parameterisation of the deposition processes and radioactive decay: A review and some preliminary results with the DERMA model

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Summary

The Danish Emergency Response Model of the Atmosphere (DERMA) (Sørensen, 1998, Sørensen and Rasmussen, 1998, Sørensen et al., 1998) is a 3D Lagrangian Gaussian puffs long-range dispersion model, developed by the Danish Meteorological Institute for nuclear emergency preparedness purposes. Previous versions of the model included simulations of transport and diffusion of passive tracers in the atmosphere. However, the removal processes into the atmosphere and interaction of radionuclides with the earth surface are very important for modelling of atmospheric transport of nuclear accidental releases.
Therefore, this report focuses on an analysis of different parameterisations of the removal processes by dry and wet deposition of radionuclides to the surface and suggests some parameterisations for deposition and radioactive decay for the DERMA model. For emergency response and for post-accidental analysis, different versions of the DERMA model are suggested using different approaches and complexities for the parameterisation of the deposition processes.

It was shown that the washout coefficient Λ strongly depends on the particle size (the so-called 'Greenfield gap'), although in most existing models of long-range pollution transport the washout coefficient does not depend on particle radius. Therefore, a revised formulation of the wet deposition parameterisation for particles of different size was suggested. For accidental releases from nuclear power plants, particles smaller than 0.5 μm radius can play an important role in long-range transport and dose forming, therefore the formulation suggested for the washout coefficient in the DERMA model, in contrast to the formulation in other models, includes the mechanism of the Brownian capture and covers a gap of the washout parameterisation for small particles.

For heavy particles ($r_p > 1$ μm), the gravitation settling essentially effects the process of deposition to the surface. Therefore, the effect of the gravitation settling was included into the dry deposition parameterisation through the gravitation settling velocity v_g. It was suggested to model the gravitation settling velocity v_g by a combination of the Stokes law (for particle diameters less than 3.5 μm) with the Cunningham correction for small particles ($r_p < 0.5$ μm) and an iterative procedure for the equation for the terminal settling velocity (for particles larger than 3.5 μm).

Earlier comparisons of simulations by the DERMA model versus the ETEX experiment with passive tracers gave very good results. In order to verify the deposition parameterisation for the DERMA model and to study effects of deposition, simulations for the INEX-2-Hun exercise and the Algeciras accidental Cs-137 release in Spain were made taking into account the deposition process. A comparison of the simulation results with measurements data from 9 European monitoring stations are presented and discussed in the report.

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I. Introduction

The Danish Emergency Response Model of the Atmosphere (DERMA) is a 3-D Lagrangian Gaussian puffs dispersion model, developed by the Danish Meteorological Institute (Sørensen 1998, Sørensen and Rasmussen 1998, Sørensen et al., 1998) for nuclear emergency preparedness purposes. Currently it includes a simulation of transport and diffusion of passive tracers in the atmosphere. However, the removal processes in the atmosphere and an interaction of radionuclides with the earth surface are very important for modelling of atmospheric transport of nuclear accidental releases. Usually, especially for long-term consequences, the radioactivity deposited contributes more to the total dose to humans than direct exposure from the air.
Earlier comparisons of simulations by the DERMA model versus the ETEX experiment with passive tracers gave very good results. 27 institutions from most European countries, USA, Canada and Japan contributed to the real-time model evaluation. Based on analyses from the first experiment, the DERMA model was emphasised as being very successful (Graziani et al., 1998). The RTMOD programme, as a continuation of the ETEX project, plans to include a comparison of existing long-transport models for different releases taking into account also the deposition process.

The objectives of the report are:

- to review the existing parametrisations of the removal processes in long-range radionuclide transport models;
- to develop a parametrisation of the radionuclide-removal processes by dry and wet deposition, suitable for the DERMA model;
- to verify the deposition parameterisation developed for the DERMA against the Algeciras accidental release, Spain, and hypothetical releases planned in the RTMOD programme and the INEX exercise.

II. Overview of deposition parameterisation in existing models

2.1. ADPIC model

The MATHEW/ADPIC model is a real time modelling system, developed by the Lawrence Livermore National Laboratory (Lange 1978, Forster 1992) for major emergency response and management. In the basic version of ADPIC (ARAC), dispersion is described by a particle-in-cell model (Foster, 1992). The 3D modelled wind field, in which the particles are advected, is mass consistent and is produced by the MATHEW model or by interpolating meteorological NWP data into the model grid. New generation of the model (ARAC-3) implemented a Monte-Carlo approach to diffusion processes using the random displacement method (RDM) (Ermak et al., 1995).

2.1.1. Dry deposition

Dry deposition is calculated separately for two mechanisms of deposition: gravitational settling and flux of materials from the surface layer to the ground. For particles the gravitational settling velocity is calculated by the Stocks law and for heavy particles (d_p >3.5 μm) by the McDonalds method, which is an extension of the Stokes law when the Reynolds number is greater than 1 (McDonalds, 1960). The dry deposition velocity approach is used for estimation of the flux of materials from the surface layer to the ground.

2.1.2. Wet deposition

The wet deposition is treated in a standard way with a constant-in-space washout coefficient. ADPIC in present form (Foster, 1992) doesn't treat in-cloud rainout, but has provisions for below-cloud washout (Crandall et al., 1973). Näslund & Holström (1993) improved the description of precipitation and wet deposition in ADPIC by introducing a space-varying coefficient. For the wet deposition parameterisation they expressed the washout coefficient as a polynomial function of particle radius and rainrates. However, for general purposes (a large spectrum of particles) they used the formula, received by Crandall et al. (1973) only for large particles of nuclear debrises. According to this formula, there is no wet deposition effect for particles with a radius less than 0.5 μm, but some experimental data (Burtsev et al., 1970; Hicks 1976; Radke et al., 1977) indicate the washout coefficient about $0.1\text{-}0.5*10^{-3}$ sec^{-1} for particle radius of 0.01-0.4 μm.

2.2. DEM/DREAM model

The Danish Eulerian model (DEM) is an air pollution model developed by the Danish Environment Research Institute (NERI) for studying different phenomena in regional scale connected with transport of air pollutants in the atmosphere. Main important physical processes (advection, diffusion, deposition, emissions and chemical reactions) are represented in the mathematical formulation of the model (Zlatev 1995, Zlatev et al. 1996).

The DREAM (the Danish Rimpuff and Eulerian Accidental release Model) is a high-resolution three-dimensional tracer model, which has been developed from the DEM model for studying short- and large-scale atmospheric transport, dispersion, and deposition (wet and dry) of radioactive air pollution caused by a single but strong source (Brant 1998). The model is based on a combination of a Lagrangian short-scale puff model and an Eulerian long-range transport model. The Lagrangian Rimpuff model of the Risø National Laboratory is used in an area near a source to describe the transport, dispersion, and deposition in the initial phase of a release and the Eulerian model is used

for long-range transport, dispersion, and deposition calculations in the model domain which covers the whole Europe.
The deposition scheme of the Eulerian model is based on almost the same theoretical assumptions as the NAME model (Maryon et al., 1996), however, cases of 'occult deposition' were not included into consideration (Maryon et al., 1992).

2.3. EMEP models

In the bounds of the EMEP programme, the Lagrangian (EMEP 1998) and Eulerian (EMEP 1998; Bartnicki et al. 1998) versions of the EMEP model were developed. The models were developed for gaseous non-radioactive pollutants, however the deposition parameterisations were also developed in details for particles (EMEP 1998; Bartnicki et al. 1998).

2.4. EURAD model

The European Air Pollution Dispersion Model (EURAD) is a three-dimensional Eulerian model developed by the Institut für Geophysik und Meteorologie, Universität zu Köln, Germany (Ebel et al., 1991).
EURAD has been designed to predict air pollution episodes, trends and to study emission reduction scenarios. Therefore, EURAD simulates transport, chemical transformations and depositions of atmospheric constituents in the troposphere over Europe. Its comprehensive description of atmospheric processes includes chemical processes in clouds and rain and also development of aerosols.
The EURAD model consists of several modules, which are developed by several institutions. The deposition module is developed for non-radioactive pollutants, first of all, and has a standard approach for the parameterisation. The temporarily and spatially varying dry deposition velocities of SO_4 are used for I-131 and a half of this value is employed for Cs-137 (Haas et al., 1990).

2.5. FOA RDM model

The Swedish Defence Research Establishment (FOA) developed a particle random displacement model (RDM) for long-range radionuclide transport and deposition (Linquist et al., 1998; Karlsson et al., 1996). The wet deposition is parameterised according to Näslund & Holström (1993) similar to the ADPIC model, and the dry deposition processes are parameterised through the gravitation settling for the Stocks and turbulent regimes - according to Näslund & Thaning (1991) and the surface resistance - according to Näslund & Karlsson (1995).

2.6. KNMI/RIVM model

The Netherlands nuclear accident model KNMI/RIVM (Van Rheineck et al., 1989) is a Lagrangian Gaussian puffs dispersion model, developed by the Netherlands Meteorological Institute (KNMI) and the National Institute of Public Health and the Environment (RIVM). The model is based on the similar theoretical assumptions as the DERMA model (Van Rheineck et al., 1989), therefore algorithms of parameterisation for deposition can be used for the DERMA model.
The dry deposition flux is calculated using a resistance model, however the deposition velocity is uniform over the model area. A correction of the concentration near the surface due to the dry deposition suggested in the model is reasonable to use in the DERMA model with the complete mixing of pollutants into ABL.
The wet deposition is proportional to the precipitation rate and the species-specific scavenging coefficients of washout and rainout.

2.7. MACCS model

The MELCOR Accident Consequence Code System (MACCS) has been developed by the Sandia National Laboratory, USA for analysis of risk levels and possible consequences for the population after radiation accidents (MACCS, 1992). It is a tool for probabilistic risk environment assessment. The input weather conditions for the probabilistic analysis are used from the statistical long-term meteorological data for many sectors. The early effects for the population of the nearest to a NPP zone are calculated by the MACCS codes 'Atmos' and 'Early'. The third part 'Chronic' is a tool for estimations of long-term effects for the population.

A simple Gaussian plume dispersion model is used for simulation of air concentration and deposition. Dry and wet depositions of nuclides are included to the model for several particle sizes using Chamberlain's source depletion approach for the dry deposition and the washout coefficient according to Brenk & Vogt for the wet deposition (MACCS, 1992).

2.8. MATCH model

The MATCH model of the Swedish Meteorological and Hydrological Institute (SMHI) (Langner et al., 1994; Person et al., 1994; Robertson et al., 1996) is a 3D Eulerian Meso-scale Atmospheric Transport and Chemistry model. The model is extended with a Lagrangian particle model for an emergency response system (Langner et al., 1997). The deposition scheme is more or less conventional/classical for such type of models (Person et al., 1994, Robertson et al., 1996).

Dry deposition is proportional to the concentration and a species-specific dry deposition velocity at 1 m height. The dry deposition flux calculation is transformed to the first model level using the standard similarity theory for the atmospheric surface layer. Dry deposition velocities are specified as a function of the surface characteristics (fraction forest, fields etc.).

The wet deposition of different species is proportional to the precipitation rate and a species-specific scavenging coefficient. Values of coefficients and velocities are close to those used in the EMEP calculations.

2.9. NAME model

The UK nuclear accident model NAME (Maryon et al., 1992, 1996a, 1996b) is a Lagrangian particle dispersion model. The model has a very good description of mechanisms of deposition for airborne radionuclides. Beside the dry and wet depositions, the authors of the model parameterised some cases of 'occult deposition' like orographic effects: capping cloud on hilltops and seeder-feeder effect, scavenging by snow, including the seeder-feeder effect and below-cloud scavenging by snow.

2.10. RODOS/RIMPUFF model

The Risø Mesoscale PUFF model (RIMPUFF) is a fast and operational puff diffusion model suitable for real-time simulation of puff and plume dispersion during time and space changing meteorology (Thykier-Nielsen & Mikkelsen, 1991). RIMPUFF includes fast subroutines for the calculation of gamma doses from airborne and deposited radioactive isotopes, released to the atmosphere from a nuclear power plant. For real-time applications, RIMPUFF can be driven by wind data from a combination of: - A permanent network of meteorological towers, - The flow models LINCOM, and - NWP data (DMI-HIRLAM).

The puff or plume diffusion process in RIMPUFF is controlled by the local turbulence levels, either are provided directly from on-site measurements, or provided via pre-processor calculations (Mikkelsen and Desiato, 1993). RIMPUFF is further equipped with plume rise formulas, inversion and ground level reflection capabilities, gamma dose algorithms and wet/dry depletion. The

RIMPUFF model is connected to the RODOS system for a local scale simulation (Thykier-Nielsen et al., 1997).
In the first version of RODOS, the deposition of radionuclides from the atmosphere to ground was considered in several atmospheric dispersion modules (ATSTEP, RIMPUFF and MATCH), and in the terrestrial food and dose module FDMT (Mikkelsen et al., 1997). Each of these modules has its own way of calculating deposition in order to estimate: plume depletion, exposure from radionuclides deposited on the ground, and contamination of crops as input to food chains.
This diversity of deposition calculation approaches does not guarantee a consistent approach throughout the whole RODOS system. Therefore, the last version (RODOS, 1999) describes changes in the deposition calculation which aim at a harmonisation of calculation methodologies within the atmospheric dispersion modules, terrestrial, aquatic and forest food chain modules.

2.11. SNAP model

The SNAP (Severe Nuclear Accident Program) model is a real-time Lagrangian particle dispersion model, developed by the Norwegian Meteorological Institute (DNMI) (Saltbones et al., 1995, Saltbones et al., 1996) for major emergency management. The first version of the model was based on the same theoretical assumptions as the NAME model. However, the deposition parameterisation in the last version of the model is original and different from the NAME. Both for wet and dry deposition, a probabilistic approach (Saltbones et al., 1995) is taken as a basis. These processes are applied for each particle at every time step. The approach is interesting and promising for particle models, but not suitable for the DERMA model.

2.12. TRADOS/SILAM models

The Finnish Meteorological Institute (FMI) and State technical research centre (VTT) developed two models for emergency preparedness items. The TRADOS model (Pöllänen et al., 1994; Pöllänen et al., 1997) has been a first standard long range model for nuclear accidents. It is a run-of-mill trajectory model, with mixed Gaussian and vertical K-profile solutions. SILAM is a new generation development project. The dispersion model framework designed for multi-purpose use, first of all for operational dispersion, fallout and radioactive dose calculation in case of an emergency in a nuclear power plant. The SILAM model contains a meso-scale/long-range random-walk particle model, high quality radioactive dose-calculation system and a three-dimensional trajectory model (Valkama, 1998). The deposition method used in the SILAM dispersion model is the standard scavenging coefficient approach. The respective effectiveness of rain and snow depletion is described by adopting different numerical values for the constants used in converting HIRLAM NWP-model precipitation rates to scavenging coefficients. The presence of water or snow is deduced from the ambient air temperatures on the HIRLAM model levels.

III. Deposition parameterisation approach for the DERMA model

The basic form of the equation for concentration of radioactive species in the atmosphere, taking into account removal processes in the atmosphere and interaction of the radionuclides with the earth surface, might be the following:

$$\partial c/\partial t = div\,(u \cdot c) - Ddep - Wdep - \lambda c + \lambda' c' + Res \qquad (1)$$

The DERMA model currently includes only the first term in the right part of the equation (1). This term describes the advection transport by velocity u and turbulent diffusion of passive tracers in the atmosphere. For simulation of the real processes of contamination by radionuclides (particles and gases) it is necessary to include the following five terms in the right part of equation (1):

- $Ddep$ - dry deposition on the surface,
- $Wdep$ - wet deposition processes,
- λc - radioactive decay to daughter,
- $-\lambda' c'$ - decay from parent,
- Res - resuspension processes and other sources.

Depending on the aims of the model: for the emergency response and for the post-accidental analysis versions of the DERMA model we will use different approaches and complexities for parameterisation of the deposition processes.

3.1. Dry deposition

Dry deposition is the removal of gaseous and particulate nuclides or other pollutants from the atmosphere to the earth surface by vegetation or other biological or mechanical means. It plays an important role for most of nuclides (excluding the noble gases) .

As a first simple parameterisation of dry deposition we can suggest a classic approach, based on a concept of the deposition velocity v_d. For the DERMA model, it is reasonable to include the mass loss due to the dry deposition in the calculation of source term Q_p - the amount of radionuclide associated with each puff p depending on the emission rate. The dry deposition takes place in the lower surface layers and is not valid in the free troposphere (z > h). Therefore, for the assumption - employed in the DERMA model - about a complete vertical mixing within ABL for each puff centre, we can obtain the following formula for the mass loss due to the dry deposition in Q_p:

$$Q_p|_{n+1} = Q_p|_n \cdot exp(-\Delta t \cdot v_d / h), \qquad (2)$$

where Δt - time step of the model, h - the ABL height.

Prahm and Berkowicz (1978) showed, that the source depletion method can give considerable errors of the surface air concentration in case of stable stratification of ABL. However, the approach of the complete vertical mixing within ABL for each puff centre will not lead to this problem in the DERMA model.

Calculation of the amount of radionuclides deposited on the surface due to dry deposition is reasonable to implement at the final part of the DERMA model, where the air concentration of species is calculated.

The dry deposition velocity depends on many parameters of particles, characteristics of the surface and surface layer. Methods of its parameterisation will be discussed below.
For the simplest case of dry deposition parameterisation for the emergency version of the DERMA model, we assumed that the dry deposition velocity is a constant for each nuclide and different surface type (see table 2). For example, for Cs-137 a reference value is v_d = 0.05 cm/s, for I-131 v_d = 0.5 cm/s (see below about I-131), for Sr-90 v_d = 2 cm/s, for the noble gases v_d = 0.

A more sophisticated approach for parameterisation of the dry deposition, is realised by defining the dry deposition velocity as the inverse of a sum of resistances r_a , r_b, r_c (see 3.1.2) in three sequential layers (Yamartino, 1989; Zanetti, 1990), in the following form for gaseous pollutants:

$$v_d = (r_a + r_b + r_c)^{-1}.$$

This formula for the dry deposition velocity of particles has an additional term, defined by the sedimentation/gravitation settling of particles:

$$v_d = (r_a + r_b + r_c)^{-1} + v_g\,, \tag{3}$$

where v_g is the gravitation settling velocity.
However, this resistance approach was developed for gaseous pollutants, first of all, therefore the gravitation settling velocity for particles is usually assumed to be constant due to problems with the empirical data.

3.1.1. Gravitational settling

For particles, especially for heavy particles (radius r_p > 1 μm), the gravitation settling strongly effect the process of deposition to the surface. As we suggested above, the effect of the gravitation settling is described in (3) through the gravitation settling velocity v_g.
For particle diameters less than approximately 3.5 μm (airflow around the failing particle is laminar) the gravitation settling velocity v_g is modelled by the Stokes law (Hinds, 1982):

$$v_g = [2C(\rho_p-\rho)g r_p^2]/9\nu\,, \tag{4}$$

where ρ_p and ρ are the particle- and air density, g is the gravitational acceleration, r_p is the particle radius, ν is the kinematic viscosity of air ($1.5\cdot10^{-5}$ m^2s^{-1}) and C is the Cunningham correction factor (Zanetti 1990) for small particles (r_p < 0.5 μm):

$$C = 1 + \frac{\lambda}{r}\left[a_1 + a_2 \exp\left(-\frac{2a_3 r}{\lambda}\right)\right], \tag{5}$$

where λ is the mean free path of air molecules (λ=$6.53\cdot10^{-5}$ m) and a_1=1.257, a_2=0.40, a_3=0.55 are constants.

However, for larger particles, the Stokes law does not work and in the turbulent regime we will use an iterative procedure to solve the equation for the terminal settling velocity according to Näslund & Thaning (1991):

$$\frac{dw_p}{dt} = \left(w - w_p\right) f(V) - \beta g, \tag{6}$$

$$f(V) = \frac{3\rho}{8r\rho_p} V C_d ,$$

$$V = \left(\left(u - u_p\right)^2 + \left(v - v_p\right)^2 + \left(w - w_p\right)^2\right)^{1/2},$$

$$C_d = \frac{24}{\mathrm{Re}}\left[1 + 0.173(\mathrm{Re})^{0.657}\right] + \frac{0.413}{1 + 16300(\mathrm{Re})^{-1.09}};$$

where V is the relative velocity of particles, u, v, w, u_p, v_p, w_p are the air and particle velocity components, C_d is the drag coefficient that is valid for the static case (Turton & Levenspiel 1986), $Re = 2Vr/\nu$. For most of situations we can suppose that $u = u_p$ and $v = v_p$ and consider only the vertical velocity w_p ($V \approx v_g$). The iteration procedure for calculation of the gravitation settling velocity v_g needs about 10 iterations for a necessary accuracy, and therefore is not time consuming for the modelling.

3.1.2. Material flux from the surface layer to the ground due to resistance

The resistance terms in (3) are: r_a – the aerodynamic resistance, which describes the turbulent transfer of the contaminant to the near-surface layer, r_b – the resistance to penetration across the atmospheric near-surface layer (with domination of the molecular transport) by convection, diffusion or inertial processes, and r_c – the resistance associated with pollutant-surface interaction.
The aerodynamic resistance r_a depends on meteorological parameters, such as wind speed, atmospheric stability and surface roughness, and can be derived (Wesely & Hicks 1977) by

$$r_a = \frac{1}{\kappa u_*}\left(\ln\frac{z_s}{z_0} - \psi_c\right), \tag{7}$$

where ψ_c is a stability function (Voldner et al. 1986),

$$\psi_c = -5\,{}^{z}\!/_{L} \qquad \text{for } z/L > 0,$$

$$\psi_c = \exp\left[0.598 + 0.390\ln\left(-{}^{z}\!/_{L}\right) - 0.09\left(\ln\left(-{}^{z}\!/_{L}\right)\right)^2\right] \qquad \text{for } z/L < 0, \tag{8}$$

L is the Monin-Obuhkov length, z_s is the height of the first reference level, z_0 is the roughness height, κ is fon Karman constant ($\kappa \approx 0.4$), u_* is the friction velocity.
The surface layer resistance r_b depends on parameters characterising diffusion across a laminar sublayer, e.g. on molecular rather than turbulent properties. Therefore, this parameter will be different for particles and gases.
For particles the surface layer resistance r_b can be expressed in a general form according to (Zannetti, 1990) as a function of the Schmidt number $Sc = \nu/D$:

$$r_b = \frac{d_1 (Sc)^{d_2}}{\kappa u_*}, \tag{9}$$

where D is the Brownian diffusivity of a pollutant: $D = {k_B TC}/{12\pi\mu r_p}$, r_p is the particle radius, C is the Cunningham correction factor, T is the temperature, k_B is the Boltzmann constant ($k_B = 1.38 \cdot 10^{-23}$), μ is the dynamic viscosity coefficient ($1.8 \cdot 10^{-5}$ kg/ms), the constants $d_1 = 5$ and $d_2 = 0.66$.
Calculation of r_b for gases (I-131 and others) can be done by the following formula:

$$r_b = 8/u_* \tag{10}$$

The transfer (canopy) resistance r_c depends on the physico-chemical interaction between the pollutant and the surface.
This term for particles is negligible, since once the particle encounters the surface it is considered to have deposited. Seinfield (1986) suggests for particles another term $r_a r_b v_g$ instead of the transfer resistance r_c.
For gaseous compounds in general, calculation of the transfer resistance r_c is a very complex problem. According to Erisman (1994) and Hongisto (1998), for estimation of r_c we can use the following formula:

$$r_c = \left[\frac{1}{r_s + r_m} + \frac{1}{r_{lu}} + \frac{1}{r_{ext}} + \frac{1}{r_{dc} + r_{cl}} + \frac{1}{r_{ac} + r_{gs}} \right]^{-1} \tag{11}$$

Whereby the transport of the pollutant to the canopy is approximated as following routes:
r_s and r_m – into the plants via stomata and through mesophyll,
r_{lu} and r_{ext}– into the plants through the cuticula cells (leaf surface resistance r_{lu} or trough external leaf surface in wet situations r_{ext}),
r_{dc} and r_{cl} – into the lower canopy against the resistance of buoyant convection in canopies r_{lu} and the surface resistance of leaves, twing and bark r_{cl},
r_{ac} and r_{gs} – into the ground against the resistance r_{ac}, which depends on the canopy height and density and surface resistance of the soil, leaf litter etc., r_{gs}.
However, most of the resistances depends very much upon the type of gas and can be estimated from some empirical or semiempirical parameterisations. Unfortunately, for radioactive gases it is very difficult to estimate correctly all those resistances, because of a lack of experimental data. Therefore at this stage, it is reasonable to limit the description of the canopy resistance r_c for radioactive gases (like I-131 and others) by some simpler parameterisation.
At the first step we use the simplest suggestion of Verner & De Leeuw (1992) for the canopy resistance r_c for the gaseous form of I-131: $r_c = 500$ s/m.
In addition, some nuclides can transform from a gaseous to particulate form. For example, it is very important for I-131, that can be released in different forms (molecular, organic and particles). The gaseous form of I-131 is usually dominated in releases (more than 90 %). However, the gaseous iodine has a significant tendency to become attached to other particles (usually to SO_4 particles). This is not an one-way process, there is a strong tendency for the effective deposition velocity to decrease with time (Maryon et al., 1992). This can be described in the model by a depletion equation:

$$\frac{dc}{dt} = \left(\frac{v_{dg}}{z_i} + \frac{1}{\tau} \right) C + \frac{C_g(0)}{\tau} \exp\left(- \frac{v_{dp}}{z_i} t \right), \qquad (12)$$

where v_{dg} and v_{dp} are the gaseous and particulate deposition velocities and τ is a timescale estimated to be 47 days from the gaseous/particulate ratio measured after the Chernobyl accident (Maryon et al. 1992).

However, for main modelling items the prediction time is much shorter (up to 5-7 days), therefore it is possible to simulate the dry deposition of iodines with the time-independent dry deposition velocities, but separately for the gaseous and particulate forms. In this case we have to know the I-131 releases for each form. For the gaseous form of I-131 the reference value for v_{dg} is close to values for SO_2 and forms about 0.5 cm/s and for the particle form v_{dp} is correspondent to the dry deposition velocity for SO_4 and equal to about 0.1 cm/s.

3.1.3. Land classification for parameterisation of dry deposition velocity

Since the dry deposition velocity and surface resistances depend on a type of the surface, it is reasonable to use some suitable classification of the land/sea surface. The existing operational version of the DMI-HIRLAM model uses a very simple classification of the earth surface for the calculation of the roughness and albedo parameters (HIRLAM, 1994). However, new research versions of HIRLAM (Bringfeld et al. 1995) improved the classification of the earth surface in many classes (more than 20). A new research version of DMI-HIRLAM (Sattler, 1999) develops the following Biosphere-Atmosphere Transfer Scheme Legend (Table 1) for the earth surface classification in the 20 classes. This classification, after including one additional class 'Urban areas', might be very suitable in our case for the calculation of the dry deposition velocities and surface resistances (see Table 1).

Table 1. Biosphere-Atmosphere Transfer Scheme Legend.

Value	Description
1	Crops, Mixed Farming
2	Short Grass
3	Evergreen Needleleaf Trees
4	Deciduous Needleleaf Tree
5	Deciduous Broadleaf Trees
6	Evergreen Broadleaf Trees
7	Tall Grass
8	Desert
9	Tundra
10	Irrigated Crops
11	Semidesert
12	Ice Caps and Glaciers
13	Bogs and Marshes
14	Inland Water
15	Ocean

16	Evergreen Shrubs
17	Deciduous Shrubs
18	Mixed Forest
19	Interrupted Forest
20	Water and Land Mixtures
21	Urban area

Unfortunately, this land classification is ready only for the 'D'-version of the DMI-HIRLAM model and currently does not cover all the European territory.
Therefore, as the first approximation we can use the existing classification of the land/sea surface from the operational version of the DMI-HIRLAM model. This simple classification includes the following parameters/fractions for each grid cell: the sea/ocean surface, land surface, ice-sheet, urbanisation, roughness and, probably in the nearest future, fraction of the forest (HIRLAM, 1994).
However, for the emergence response version of the model at present stage it is reasonable to use a simpler parameterisation for the dry deposition velocity for different types of surface (see table 2). Thus, v_d is assumed (in case when it is important) to be proportional to the leaf area index LAI for each surface type:

$$v_d = v_{dmax}\, LAI/LAI_{max} \tag{13}$$

where LAI is the leaf area index for each surface type at time of deposition, LAI_{max} is the leaf area index at time of fully developed foliage.

Table 2. Deposition velocities v_{dmax} for soil and fully developed plant canopies (according to Müller & Prön, 1993):

	Deposition velocity, cm/s		
Surface type	Aerosol bound radionuclides	Elemental iodine	Organic bound iodine
Soil	0.05	0.3	0.005
Grass	1.05	0.15	0.015
Trees	0.5	5	0.05
Other plants	0.2	2	0.02

3.2. Wet deposition

The wet deposition or pollutant scavenging by precipitation processes, as the Chernobyl accident showed, are very important for modelling of atmospheric transport of nuclear accidental releases and the deposited radioactivity pattern. Usually the wet deposition is treated in a standard way with a washout coefficient for below-cloud scavenging and a rainout coefficient for in-cloud scavenging (Yamartino, 1985; Seinfeld, 1986; Zanetti, 1990).
As the first approximation for the emergency response version of DERMA, for parameterisation of the wet deposition (absorption into droplets followed by droplet removal by precipitation) of aerosol particles or highly soluble gases, we can describe the local rate of material removal as a first-order process:

$$\frac{dC}{dt} = -\Lambda \cdot C(x_i, t), \tag{14}$$

where $\Lambda(r, x_i, t)$ is a washout coefficient, which depends in general on height above the surface and time.

The wet deposition flux to the surface, in contrast to the dry deposition flux, is the sum of wet removal from all volume elements aloft, assuming that the scavenged material comes down as precipitation. However, for the DERMA model, by the assumption of complete vertical mixing within ABL and if the height of rain clouds $H_r \leq h$, we can introduce a term of the wet deposition velocity:

$$v_w = \Lambda' \cdot H_r \tag{15}$$

So, we can yield the similar to (2) formula for calculation of the mass loss by the wet deposition in Q_p:

$$Q_p|_{n+1} = Q_p|_n \cdot exp(-\Delta t \cdot \Lambda' \cdot H_r / h). \tag{16}$$

If the height of rain clouds H_r is unknown, for simplification we can suppose that $H_r = h$.

3.2.1. Washout

Below-cloud scavenging (washout) coefficient Λ for aerosol particles of radius r_p can be expressed in a general form as

$$\Lambda = -\pi N_r \int a^2 w_r(a) E(r_p, a) f_a(a) da, \tag{17}$$

where N_r is the total number of raindrops residing in the unit volume, $E(r,a)$ is the aerosol capture efficiency term, a is the raindrop projected radius, w_r is the vertical velocity of the raindrops (negative downward), f_a is probability-density function of the raindrop size distribution.

The aerosol capture efficiency $E(r,a)$ is a function of the radius of particle, r and rain drops, a and depends upon several mechanisms (Hales 1986):

- impaction of aerosol particles on the rain drop,
- interception of particles by the rain drop,
- Brownian motion of particles to the rain drop,
- nucleation of a water drop by the particle,
- electrical attraction,
- thermal attraction,
- diffusioforesis.

The washout coefficient Λ varies spatially and temporally, however, in our case, for the emergency response version of DERMA, it is possible to use an average washout coefficient Λ' and the surface precipitation data from the DMI-HIRLAM model.

As it was shown above, in most of existing models of long-range pollution transport the washout coefficient does not depend on particle radius. However, as it was shown by many authors (Seinfeld 1986; Hales 1986; Burtsev et al., 1970; Hicks 1976; Radke et al., 1977), $E(r,a)$ and correspondingly Λ strongly depend on the particle size (so-called the 'Greenfield gap').

According to Näslund & Holmström (1993), we can express the washout coefficient Λ' as a polynomial function of particle radius r and rainrates q (mm/h):

$$
\begin{aligned}
&\Lambda'(r,q) = 0, && r < 0.5\ \mu m \\
&\Lambda'(r,q) = (b_0 + b_1 r + b_2 r^2 + b_3 r^3)\, f(q), && 0.5\ \mu m < r < 10\ \mu m \\
&\Lambda'(r,q) = f(q), && r > 10\ \mu m \\
&f(q) = a_1 q + a_2 q^2,
\end{aligned}
\tag{18}
$$

where $a_1 = 2.7 \cdot 10^{-4}$, $a_2 = -3.618 \cdot 10^{-6}$, $b_0 = -0.1483$, $b_1 = 322013.3$, $b_2 = -3.0062 \cdot 10^{10}$, $b_3 = 9.34458 \cdot 10^{14}$.

However, according to this formula, there is no wet deposition washout effect for particles with a radius less than 0.5 μm, but several authors (Seinfeld, 1986; Burtsev et al., 1970; Hicks, 1976; Radke et al., 1977) showed experimental data with the washout coefficient about 0.1-0.5 $\cdot 10^{-3}$ sec^{-1} for particle radius 0.01-0.5 μm. Therefore, as the first approximation, we suggest a revised formulation for particles of smaller size:

$$
\begin{aligned}
&\Lambda'(r,q) = a_0\, q^{0.79}, && r < 1.4\ \mu m \\
&\Lambda'(r,q) = (b_0 + b_1 r + b_2 r^2 + b_3 r^3)\, f(q), && 1.4\ \mu m < r < 10\ \mu m \\
&\Lambda'(r,q) = f(q), && r > 10\ \mu m
\end{aligned}
\tag{19}
$$

where, according to Maryon et al. (1996), $a_0 = 8.4 \cdot 10^{-5}$.

The reason, why Näslund & Holmström (1993) got that effect for small particles, was the following. They used results of investigations of Crandal et al. (1973) for free-air bursts, where very few debris particles were smaller than 0.1 μm radius. Therefore, Crandal et al. included into consideration the impaction of aerosol particles on the rain drop and interception of particles by the rain drop only and did not include the Brownian capture and other additional mechanisms.

For accidental releases from nuclear power plants, particles smaller than 0.1-0.5 μm radius can play an important role in long-range transport and dose forming. Therefore, for the DERMA model, especially for the post-accidental analysis version, it is reasonable to improve the washout parameterisation by including the mechanism of the Brownian capture.

The Brownian capture and diffusion mechanism give the following additional term to the aerosol capture efficiency:

$$
E = \frac{4}{Pe}\left(1 + 0.4\,\mathrm{Re}^{1/2}\, Sc^{1/3}\right), \tag{20}
$$

where Pe is the Peclet number ($a\, w_r/D$), Sc is the Schmidt number (ν/D)and Re is the Reynolds number ($a\, w_r/\nu$), ν is the kinematic viscosity of the air, D is the Brownian diffusivity of particles.

Based on a simplification of the equation (17) according to Slinn (1975) we can write

$$
\Lambda = qE(r_p,a)/2a_m \ , \tag{21}
$$

where the volume-mean raindrop radius $a_m = 0.35mm(p/1mm \cdot h^{-1})^{-1/4}$ for steady frontal rain. Accordingly, we can add the effect of the diffusion mechanism to (18) for Λ by the additional term (20)-(21), which plays role for particles with a radius smaller than 0.1 μm.

Crandal et al.(1973) and Slinn (1974) found the aerosol capture efficiency *E(r,a),* due to the impaction of aerosol particles on the rain drop and interception of particles by the rain drop, as an semi-analytical formulation:

$$E = \frac{4r_p}{a_m}\left(\frac{r_p}{a_m} + \frac{(1+2\,\mu_w r_p/\mu_a a_m)}{\left(1+\mathrm{Re}^{-1/2}\,\mu_w r_p/\mu_a a_m\right)}\right) + \left(\frac{St-St_*}{St-St_*+2/3}\right)^{\frac{3}{2}} \qquad (22)$$

where a_m is the volume-mean raindrop projected radius, *St* is the Stokes number ($-2r^2\rho_p w_r/9a\rho_a\nu$), *St** is the critical Stokes number (*1.2+ln(1+Re/2)/12)/(1+ln(1+Re/2)*), μ_w and μ_a are the dynamic viscosity of water and air, ρ_p and ρ_a are the density of particles and air.
Based on a simplification of the equations (20)-(22), we can receive the following formulation for the washout coefficient Λ:

$$\Lambda = \frac{q}{2a_m}\left[\frac{4}{Pe}\left(1+0.4\,\mathrm{Re}^{1/2}\,Sc^{1/3}\right) + \frac{4r_p}{a_m}\left(\frac{r_p}{a_m} + \frac{(1+2\,\mu_w r_p/\mu_a a_m)}{\left(1+\mathrm{Re}^{-1/2}\,\mu_w r_p/\mu_a a_m\right)}\right) + \left(\frac{St-St_*}{St-St_*+2/3}\right)^{\frac{3}{2}}\right] \qquad (23)$$

This parameterisation will be developed and verified versus empirical data.

3.2.2. Rainout

There are, beside the wet deposition effect by the washout below a cloud base (see 3.2.1), the following additional effects of the wet deposition when air pollutants are into the clouds:

- rainout between the cloud base and top (scavenging into the cloud),
- wet deposition caused by surface deposition of fog and cloud droplets.

The first process of the rainout between the cloud base and top depends on the *convective* or *dynamic* type of precipitation.
The rainout coefficient for the *convective* precipitation is more effective/intensive than the washout coefficient and can be estimated according to Maryon et al. (1996) by the following formula:

$$\Lambda'(r,q) = a_0\, q^{0.79}, \qquad (24)$$

where $a_0 = 3.36\cdot10^{-4}$. Crandall et al. (1973) showed simulations for rainout in which the scavenging coefficient is not a strong function of particle size.
The rainout coefficient for the *dynamic* precipitation is equal to the washout coefficient and the rainout effect in this case can be also estimated by the formula (19).
However, for correct simulation of the effects of the rainout between the cloud base and top, it is necessary to know from a NWP model the cloud base and top heights for each horizontal grid cell. At the moment we have not such output data from the DMI-HIRLAM model. Therefore, we use another possibility to calculate the heights of cloud base and top from 3D fields of humidity, assuming that the cloud is present in areas where the relative humidity is above 80 % (Podyukevich, 1989). It is necessary to mention that this method can give a very rough estimation, therefore it is planned to include the additional output information from the DMI-HIRLAM model for simulation of scavenging into clouds.

3.2.3. Scavenging by snow

According to publications (Hongisto 1998, Maryon et al. 1996) in most of the existing models processes of the scavenging by snow are described by the same formulae for a case of rain precipitation, like the formulae (16), (19) and (24) in our case, but with other values of the scavenging coefficient Λ. Diapason of values of Λ for snow differs from 2 up to 10 times lower than the washout coefficient Λ for a case of rain with the equivalent precipitation rates q.

For scavenging by snow, according to Maryon et al. (1996), we use the following simple formulation without any dependence of the coefficient Λ' on the particle radius:

$$\Lambda'(q) = a_0 q^b, \tag{25}$$

where $a_0 = 8.0 \cdot 10^{-5}$, $b = 0.305$ for scavenging by snow below the cloud base and between the cloud base and top for the *dynamic* precipitation, and $a_0 = 3.36 \cdot 10^{-4}$, $b = 0.79$ for scavenging by snow between the cloud base and top for the *convective* precipitation.

As a criterion for distinguishing between rain or snow precipitation, air temperature is used.

Unfortunately, the processes of scavenging by snow are not well studied. For example, there are some effects of water content in snowflakes on the scavenging by snow, especially for temperatures close to 0°C (Karlsson and Nyholm, 1998), but there are no relevant experimental data for taking this effect into account.

3.2.4. Orographic and other effects

Beside the wet deposition effects, described above, it is reasonable to include additional effects and to parameterise some cases of 'occult deposition' like orographic effects: capping cloud on hilltops and seeder-feeder effect, scavenging by snow, including the seeder-feeder effect and below-cloud scavenging by snow. It is very suitable for the DERMA model to use the parameterisations of such effects from the NAME model (Marion et al., 1995). However, it needs special data from satellite remote sensing monitoring system, now available for the UK territory only, but not for other European regions.

3.3. Total deposition to the ground

Because the mass loss due to the dry and wet deposition is calculated in the DERMA model using the source depletion concept for each individual puff (Q_p - the amount of radionuclide associated with each puff p depending on the emission rate), the total deposition of nuclides to the ground will be calculated by the integration of the total mass loss due to the deposition over each horizontal grid cell for the time period.

Calculation of the amount of radionuclide deposited on the surface due to the dry and wet deposition is implemented in the DERMA model at the final stage, where the air concentration of species is calculated. The choice of the time step for calculation of the total amount of a nuclide deposited is very important and demands further investigation.

IV. Nuclides of main concern and radioactive decay

4.1. Nuclides of main concern

For an analysis of the environment and human health consequences after accidents with nuclear reactors we can focus only on simulation of the atmospheric transport, deposition and effects of main dose-contributing nuclides. An analysis of published risk assessments for different nuclear power plants shows (ATMES 1992, Slaper et al. 1994, Thaning & Baklanov 1997) the following main dose- contributing nuclides:

Table 3. Characteristics of the main dose- contributing nuclides.

Nuclides	Half live, hrs	Median radius, μm	v_d, cm/s **	Form	Comments
Cs-137	2.628 e05	0.5	0.1	=> Ba-137	ρ = 1.88 g/cm^3
Cs-134	1.805 e04	0.5	0.12		
I-131	1.93 e02	gas/0.7 *	0.6 *	molec/org/part.	ρ = 4.93 g/cm^3 (particl.)
Te-132	7.82 e01	0.6	0.3	=> I-132	
I-133	2.08 e01	gas/0.7 (?)	0.7		
Ba-140	3.048 e02	~ 1.8	0.9	=> La-140	
Sr-90	2.549 e05	3(?) => 20	2 (?)		
Ru-103	9.459 e02	0.7(?) => 10	0.5	RuO_3, RuO_4 > 'hot part'	Chernobyl case
Pu-238	7.683 e05	3 (?)	2 (?)		for the FBR type
Pu-239	2.111 e08	3 (?)	2 (?)		long-term effects
Sr-89	1.212 e03	3 (?)	2 (?)		nearest zone

* during first week the gaseous form (elementary and organic) dominates for I-131, v_d is corrected/recalculated for 70% gaseous I-131, the dry deposition velocity for I-131 particles ≈ v_d for SO_4;
** the typical deposition velocities are given for a grass surface for most of the nuclides.

The physical and chemical form of a nuclide effects its behaviour in the atmosphere and may itself be subjected to transformation during atmospheric transport. The radioactive decay is the main mechanism of transformation. However, the radionuclides might be emitted in various forms, including (Warner & Harrison 1993):

1. an inert gas, lost from the atmosphere only by decay, but possibly producing daughter nuclides with different properties;
2. a more reactive species such as elemental I-131 which may interact physically or chemically with other species, for example by absorption on the surface of the ambient aerosol (SO_4 and other);
3. as small nuclei-mode particles (~0.01 μm diameter) which interact with the ambient aerosol;
4. as in the case of the actinides and more refractory nuclides from Chernobyl, attached to coarse fuel particles which sediment out rapidly.

4.2. Radioactive decay simulation

The radioactive decay is the main mechanism of transformation of many basic dose- contributing nuclides and should be taken into consideration for simulation of the possible radioactive contamination. The decay effects in the following ways:

- simple decay;

- daughter nuclides (B, or B1, B2)
- secondary decay of daughter nuclides (C, D,…).

It is possible to split decay simulation in the model into two basic steps:
1. during the airborne transport for short-living nuclides (like I-131) into the DERMA model;
2. after the airborne transport simulation for deposited to the surface long living-nuclides (like Cs-137) in a separate subprogram/model.

Simulation of physical and chemical form of nuclides includes for I-131: 3 forms of Iodine (gaseous and particles):
- Elemental Iodine,
- Organic Iodine (CH_3I),
- Attached to aerosol particles.

During the first week the gaseous form dominates for I-131, v_d is corrected/recalculated for 70% gaseous I-131, the dry deposition velocity for I-131 particles is almost equal to v_d for particles of SO_4 .

The radioactive decay is taken into account through mother (A) and possible daughter nuclides (B, C, …) by the following formulae:

$$dA/dt = -\lambda_a A \;;$$

$$dB/dt = -\lambda_b B + \lambda_a A \;; \qquad (26)$$

$$dC/dt = -\lambda_c C + \lambda_b B + \lambda_a A \;;$$

where λ_c, λ_b, λ_a are the decay constants for corresponding nuclides.

The second step of the decay simulation after the airborne transport for deposited to the surface long-living nuclides is not included into the DERMA simulations at the current stage.

4.3. Resuspension processes

Atmospheric resuspension of radionuclides can be a secondary source of contamination after an accidental release has stopped or a primary source for dusting tailing dumps on mining or ore-dressing works (Garger et al., 1999, Gavrilov et al., 1998). However, resuspension processes play an important role for the local scale contamination, first of all, therefore at this stage we can omit these processes in the regional scale model.

V. Examples of deposition simulation by the DERMA model

5.1. Algeciras accident, Spain

Earlier comparisons of simulations by the DERMA model versus the ETEX experiment involving passive tracers gave very good results (Graziani et al., 1998), however they did not include into account the deposition processes. As the first step to verify the deposition parametrisations implemented in the DERMA model, and in order to study effects of deposition, model simulations have been performed for the Algeciras accidental release of radioactivity. For the simulations we used the emergency response version of the DERMA with the simple parametrisation of deposition and decay, according to (2), (4), (16), (19), (24)-(26).

On 30 May, 1998, a Cs-137 source was accidentally melted in one of the furnaces at the Acerinox stainless steel production plant in Los Barrios, near Algeciras, province of Cádiz, Spain. The source was contained in scrap used as raw material for steel production (CSN, 1998; Vogt et al., 1998). The release parameters were not well estimated. According to the Spanish Nuclear Security Agency (CSN), the most likely release rate was 8-80 Ci (1 Ci = $3.7 \cdot 10^{10}$ Bq) between 01-03 UTC on 30 May.

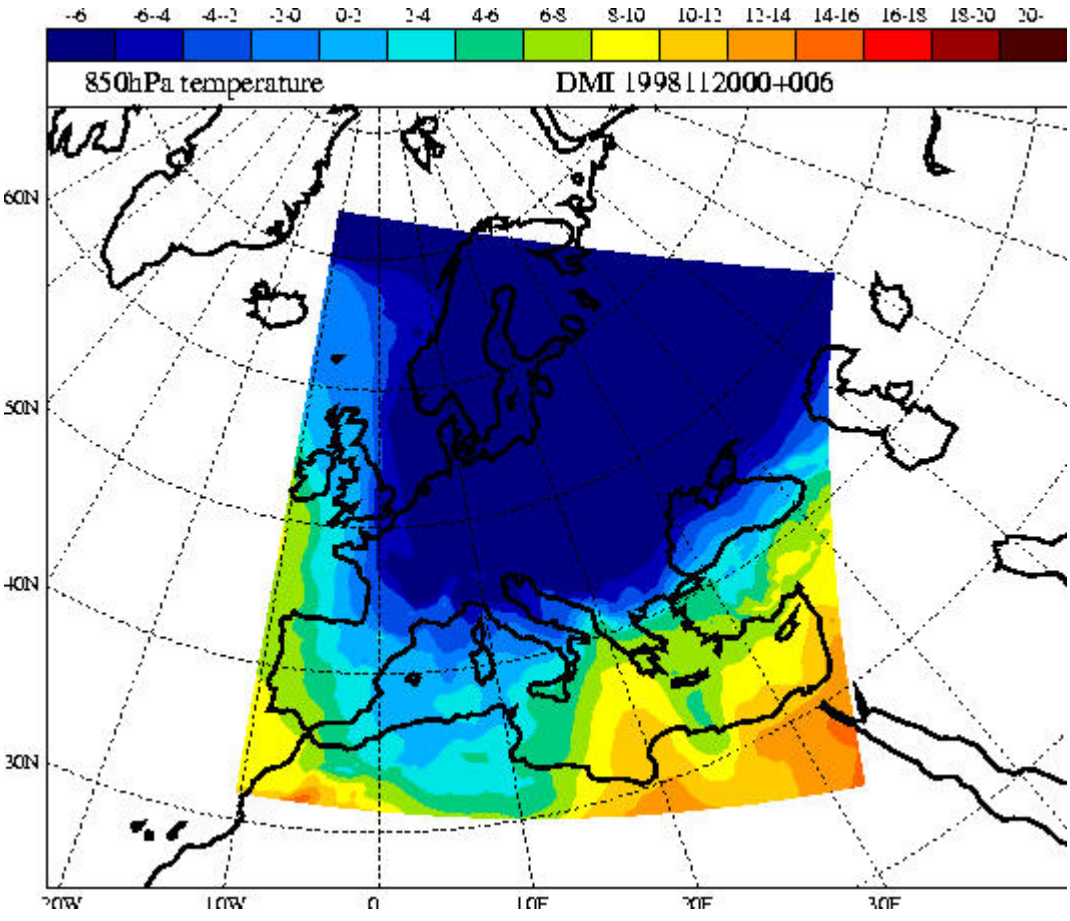

Figure 1. The area for the RTMOD simulations for the Algeciras accidental release.

The study area encompasses the European region between 8° W to 20°E and 35°N to 55° N. The meteorological analysed data set for 30 May - 05 June, 1998 from the 0.45 degree (approximately 46 km) horizontal resolution version of the DMI-HIRLAM model was used for the simulation. Figure 1 shows the area (a sub-region of the DMI-HIRLAM area), which was used for simulations of the Algeciras accidental release.
The meteorological data consist of analysed data from 00, 06, 12 and 18 UTC, and +3 and +6 hour forecast data filling out the gaps between analyses thus producing a 3-hourly time resolution (Sørensen, 1999). The following multi-level fields:

- *u* wind component,
- *v* wind component,
- temperature,
- specific humidity,

and single-level fields:

- surface geopotential,
- surface pressure,
- sea surface temperature,
- surface temperature,
- 2-meter temperature,
- 2-meter specific humidity,
- 10-meter u wind component,
- 10-meter v wind component,
- fraction of ice,
- fraction of land,
- albedo,
- dynamic (sea) surface roughness,
- climatological surface roughness,
- surface sensible heat flux,
- surface latent heat flux,
- surface momentum flux,
- accumulated stratiform precipitation,
- accumulated convective precipitation

from the DMI-HIRLAM model were used.

According to the first requirements to the participants of the Algeciras RTMOD inter-comparison, a Cs-137 release of ~50 Ci starts on 30 May 01:30 UTC, 0.5 hours duration at 5° 26' 00" W and 36° 10' 00" N. The source material is depositing (V_d =0.1 cm/sec).
Next requirements to the RTMOD participants of the Algeciras inter-comparison were the following: the release has taken place at 100 m above ground. The source strength was decided to be 50 Ci/hr of Cs-137, the duration of one hour. The suggested deposition velocity was 0.1 cm/s and the suggested wash out coefficient was 1.E-04 s^{-1} corresponding to 1 mm hr^{-1} precipitation intensity. However, participants were left free to use any value different from those suggested.
Figures 2 and 3 show the DERMA simulation results for the first release scenario of the surface air concentrations of ^{137}Cs (Bq/m^3) for the first 5 days: during 30 May-05 June 1998, each 12 hours (12 & 00 UTC).

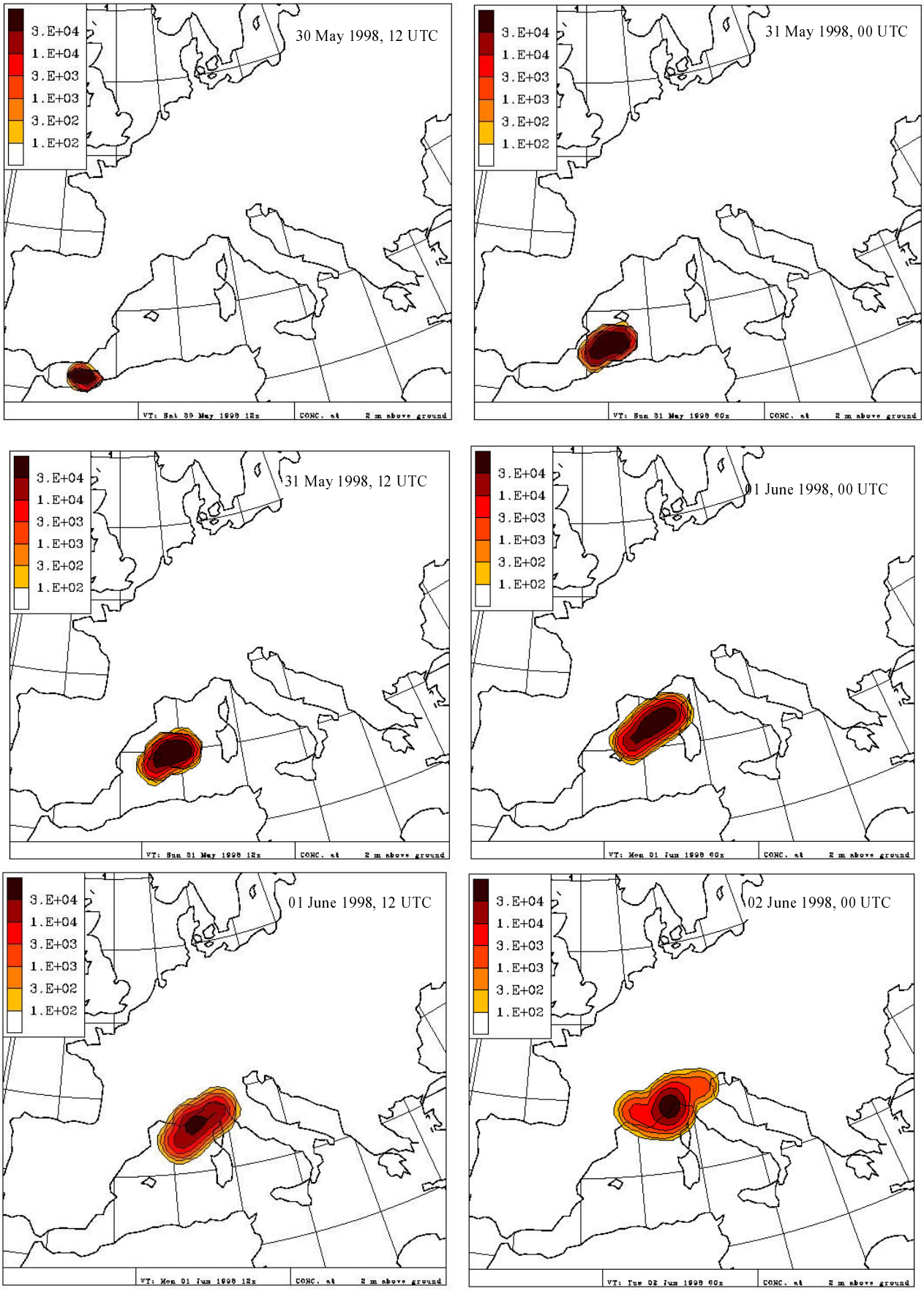

Figure 2. Air concentration of ^{137}Cs (μBq/m^3) during 30 May-02 June 1998, 12 & 00 UTC.

The first map of the figure 2 shows the concentration pattern 10 hours after the end of the release at 12 UTC on 30 May. The released plume extends over the Mediterranean sea and only slightly touches the territory of Africa (Algeria) and the south-west cost of the Iberian peninsula close to Palomares. Next maps of the figure 2 show transport and dispersion of the plume each 12 hours.

Next hours the plume extends over the western part of the Mediterranean sea and around 18 UTC on 31 May it reaches the Sardinia island and three hours later - the coast of south-east France and about 23-24 UTC - the north-west extremity of Italy. After that, a northern periphery of the plume covers a land territory of south-east France and north Italy (two last maps in the figure 2). Since 12 UTC on 01 June the main part of the plume extends over France, Italy, Switzerland and Austria (Figure 3). Then, during the morning hours on 03 June the Cs concentration of the plume strongly decreased, mostly due to the scavenging by precipitation.

Figures 4-6 show the DERMA simulation results of the cumulative deposition pattern of ^{137}Cs (Bq/m^2) by the dry (Figure 4) and wet depositions (Figure 5) and by the total deposition (Figure 6) 6 days after the release, 05 June 1998, 00 UTC for the first release scenario.
During the first two and a half days, the dry deposition totally dominated in the deposition pattern, therefore the total deposition to the Mediterranean sea is conditioned by the dry deposition (Fig. 4-6). Over the European territory, the wet deposition plays an important role due to the precipitation over France, Germany, Italy, Switzerland, Austria and other central European countries during 02-04 June. Over this territory the wet deposition plays as important role as the dry deposition or even dominates over the dry deposition, especially for the northern areas, covered by the plume.

Neither the radioactivity levels detected in Europe nor those detected close to the source were such as to have any effect on the health of individuals or the environment. The radioactive plume was detected at a number of European radiological stations (Figure 7).
Simulation results have been compared with measurement data from nine European monitoring stations, that measured a significant level of Cs-137 concentration (Fig. 7 shows the names of those stations). Table 3 presents the average concentration of Cs-137 measured at the monitoring stations in comparison with that simulated by DERMA for the second scenario.

Several examples of a simulation and measurement comparison are shown as graphs in Figures 8-15. The figures present hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average or several-days-average concentrations measured at different European monitoring stations (thin curve) for different periods between 25 May and 8 June.

Analysis of Table 3 and the figures indicates a fair agreement in general with the simulated concentration. However, the model significantly overestimates concentrations for Ispra, Milano, Nice, Cadarache, Marcoule: the modelled concentrations are 1.5-3 times higher than measured levels. Average concentrations of Cs-137 measured at Toulon/La Seyne, Montpellier, Montfaucon are 1.5-4 times lower in comparison with the calculations by DERMA. The maximum of daily-average concentration, measured at Nice is 11 times lower than the modelled concentrations for 1-2 June and 4.5 times lower for 2-3 June.

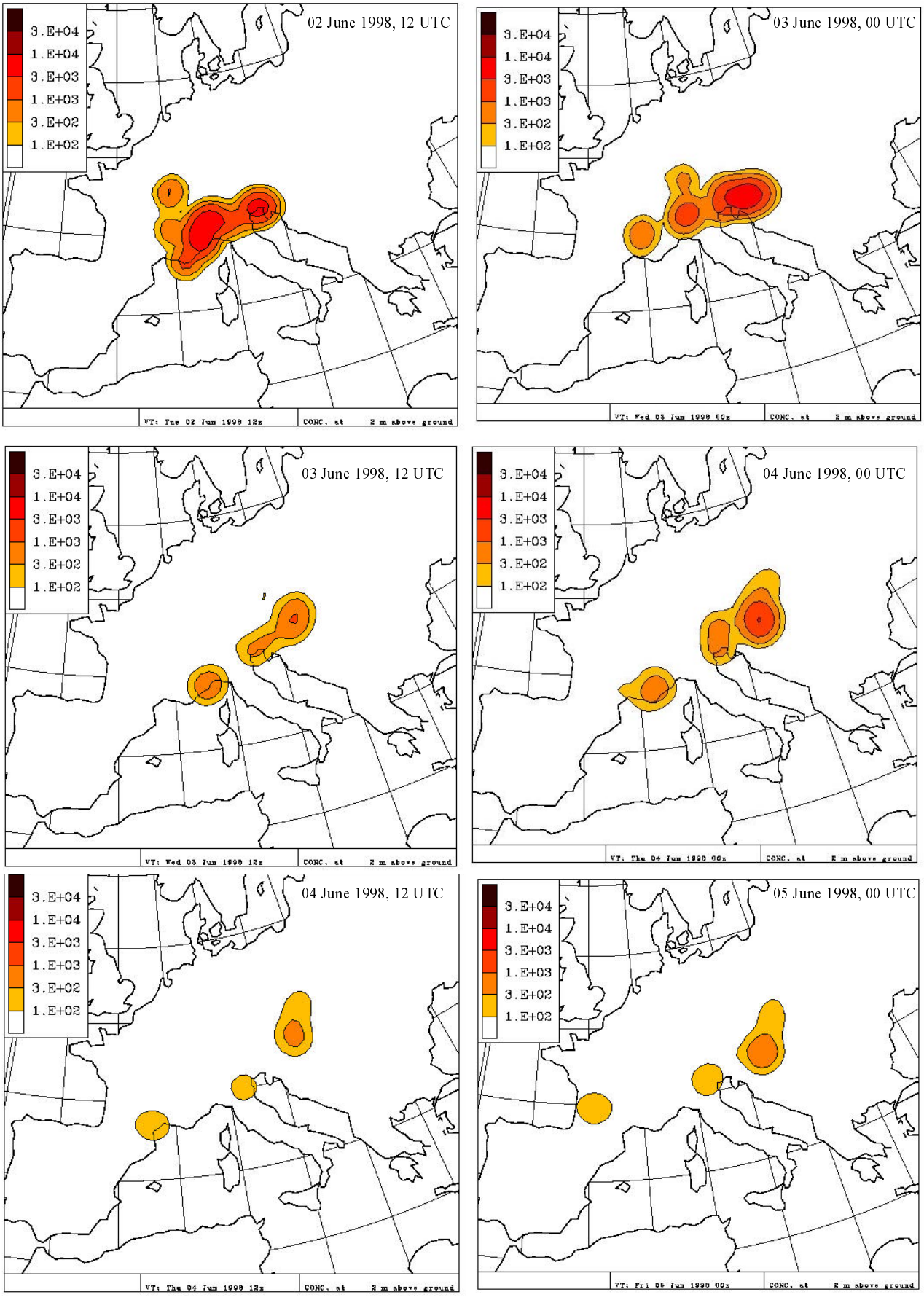

Figure 3. Air concentration of ^{137}Cs (μBq/m^3) during 02-05 June 1998, 12 & 00 UTC.

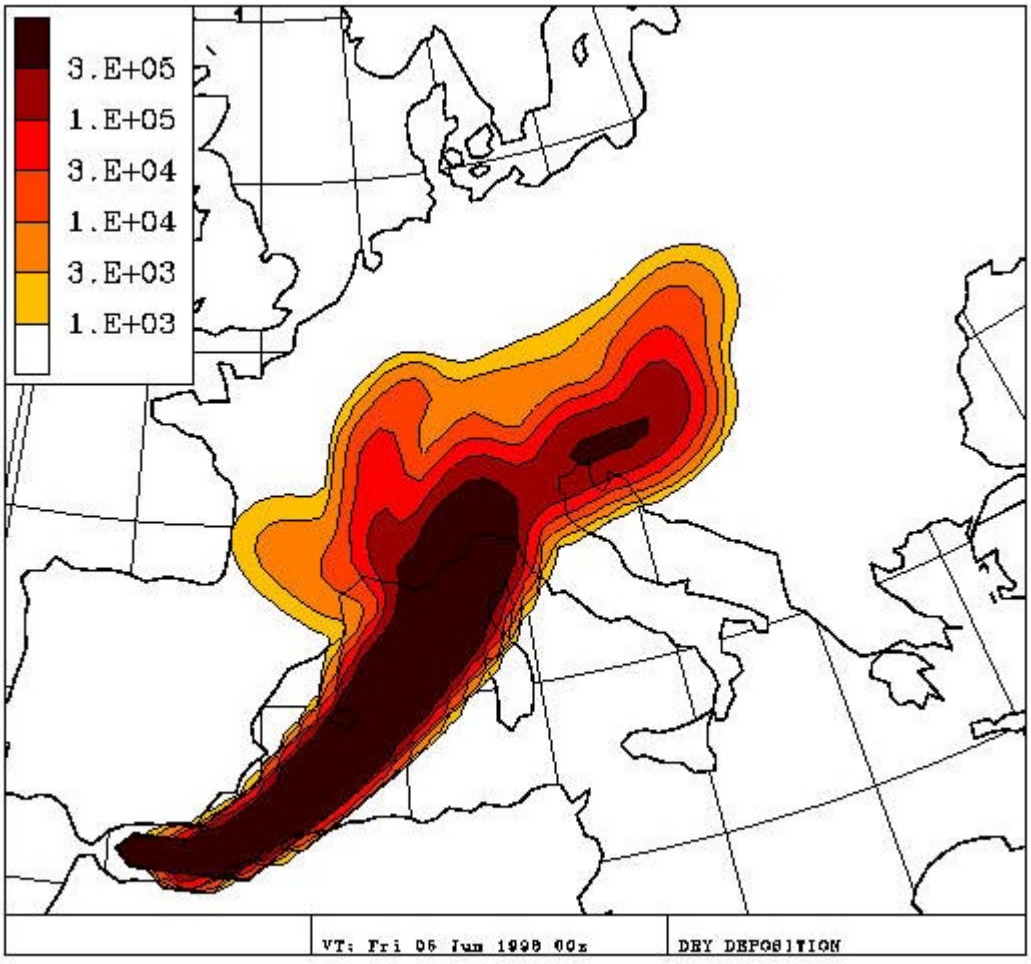

Figure 4. Cumulative deposition pattern of ^{137}Cs ($\mu Bq/m^2$) by the dry deposition 6 days after the release, 05 June 1998, 00 UTC.

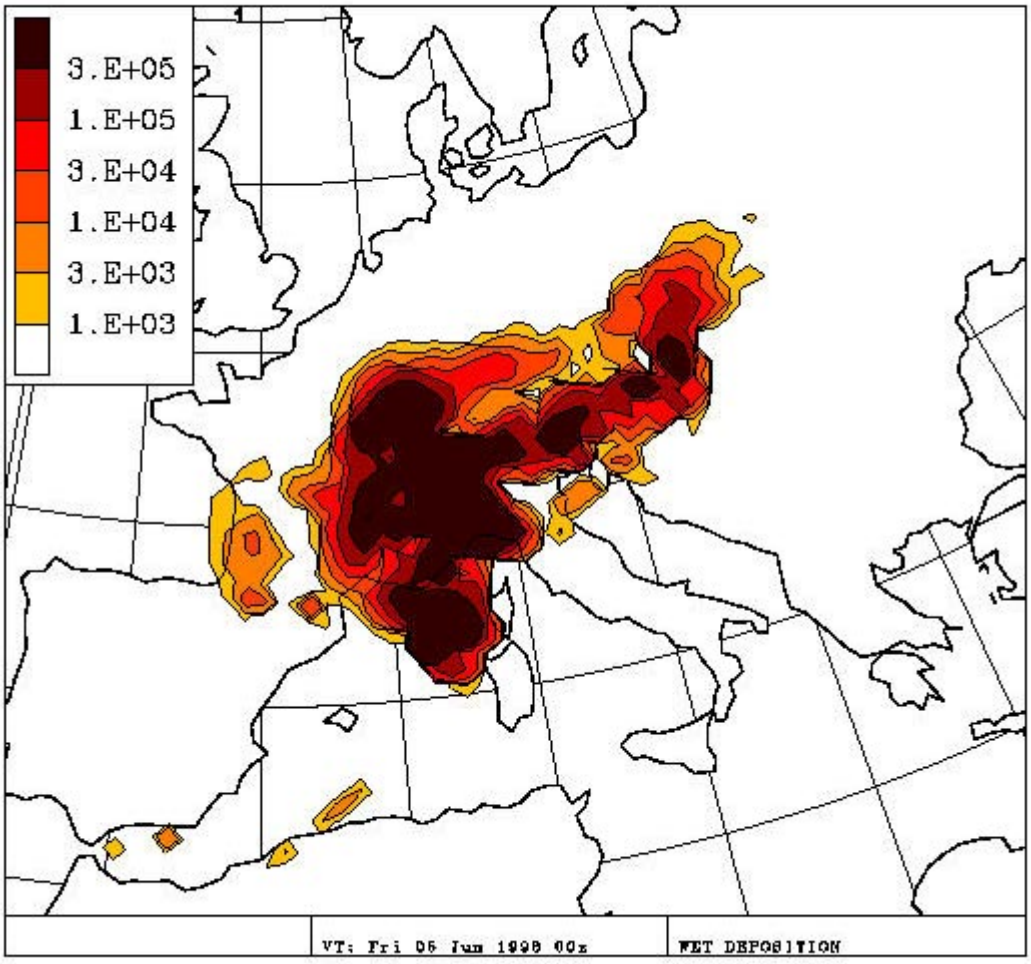

Figure 5. Cumulative deposition pattern of ^{137}Cs ($\mu Bq/m^2$) by the wet deposition 6 days after the release, 05 June 1998, 00 UTC.

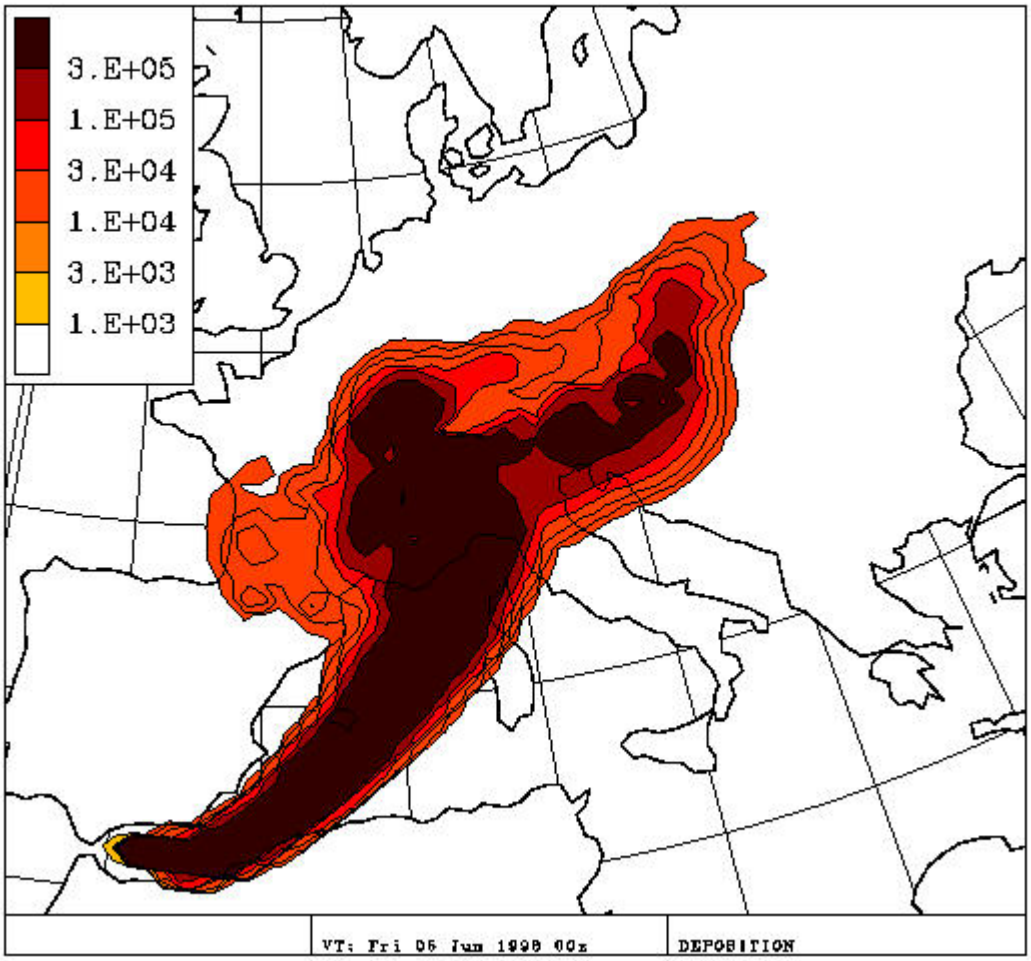

Figure 6. Cumulative total deposition pattern of ^{137}Cs ($\mu Bq/m^2$) by the dry and wet deposition 6 days after the release, 05 June 1998, 00 UTC.

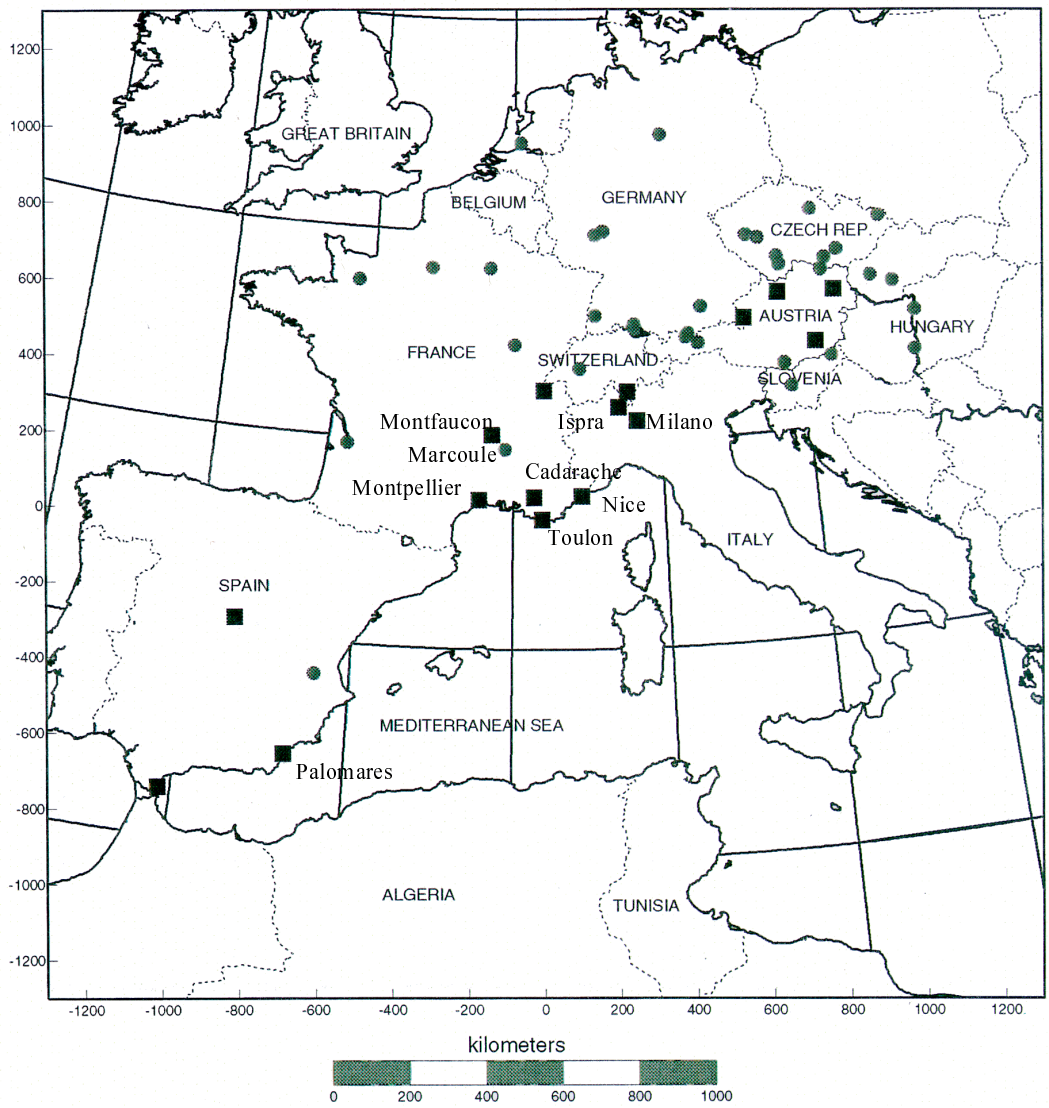

Figure 7. Locations of the European radiological monitoring stations, measured Cs137 air concentration after the Algeciras accidental release. The names indicated only for the stations, that were used for our comparison.

Table 3. Average concentration of Cs-137 measured at nine European monitoring stations in comparison with a calculation by DERMA.

#	Site name	Averaging period	Measured (μBq/m^3)	Modelled (μBq/m^3)
1	Ispra	1-2 June	390	959
		2-3 June	900	2886
		3-4 June	460	21.5
		4-5 June	140	3.9
		5-6 June	150	-
		2-5 June	740	970
		29 May-5 June	220	645
		2-4 June	670	1454
2	Milano	1-3 June	1700	2995
3	Nice	1-2 June	2100	24318
		2-3 June	330	1530
4	Toulon/La Seyne	25 May-2 June	2430	1229
		2-6 June	200	179
		5-10 June	5.5	-
		10-15 June	2.9	-
5	Cadarache	1-2 June	1600	4244
		2-3 June	510	505
6	Montpellier	1-2 June	600	179
		2-3 June	460	110
7	Montfaucon	2-3 June	870	345
8	Marcoule	1-2 June	560	814
9	Palomares	25 May-2 June	890	max 0.03
		2-8 June	4	0

The main maxima of concentration, predicted by the DERMA model, well coincide temporally with the measurement peaks for the monitoring stations Ispra and others. Comparisons of the estimated ABL height in DERMA with measurement data for the Gibraltar aerological station at the release time show very good agreement (810m to 820m).

Let's consider in more details the results for the station Ispra, where daily measurements were made. A considerable overestimation of the model concentration takes place only for the main peaks on 1-3 June (in 2.5-3 times). It seems, the release lasted for a longer time and continued after 02 UTC, because the model concentrations for 3-4 and 4-5 June were considerably lower than those measured, and the mean values for 2-5 June differs on 33 %. Other reason for this difference might be a simplification of the description of the vertical distribution of pollutants as evenly-mixed along ABL.

For the station Nice, the situation was different: the centre of the model plume passed exactly over Nice 1-2.06, so we got a very high concentration that day. It seems, in the reality the centre of the plume with maximal concentration passed aside the station. Besides, this strong difference might be explained by the poorly-defined time or duration of the release.

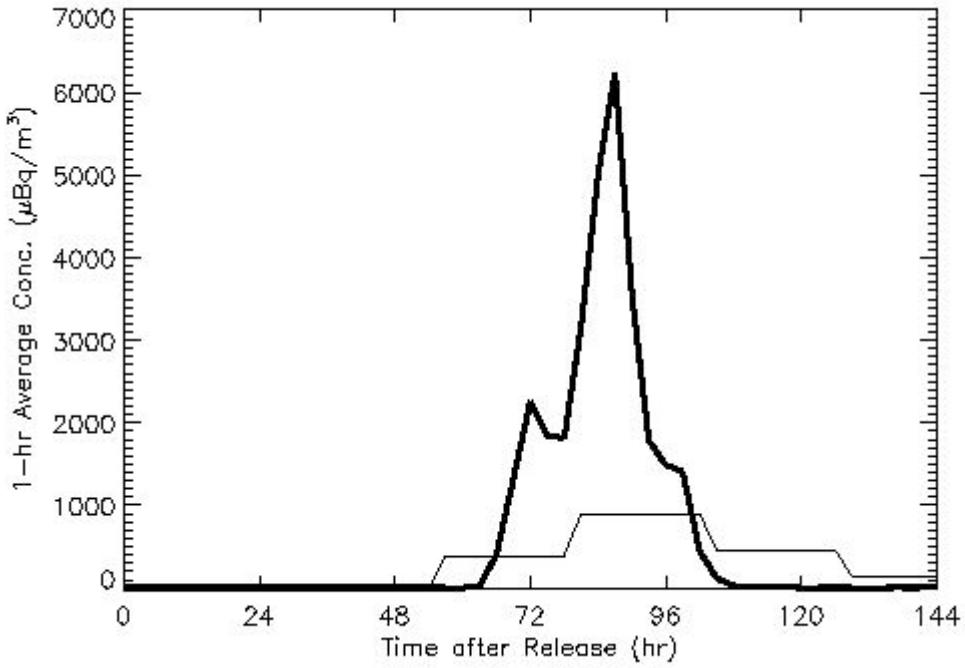

Figure 8. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average concentration measured at JRC, Ispra, Italy(thin curve).

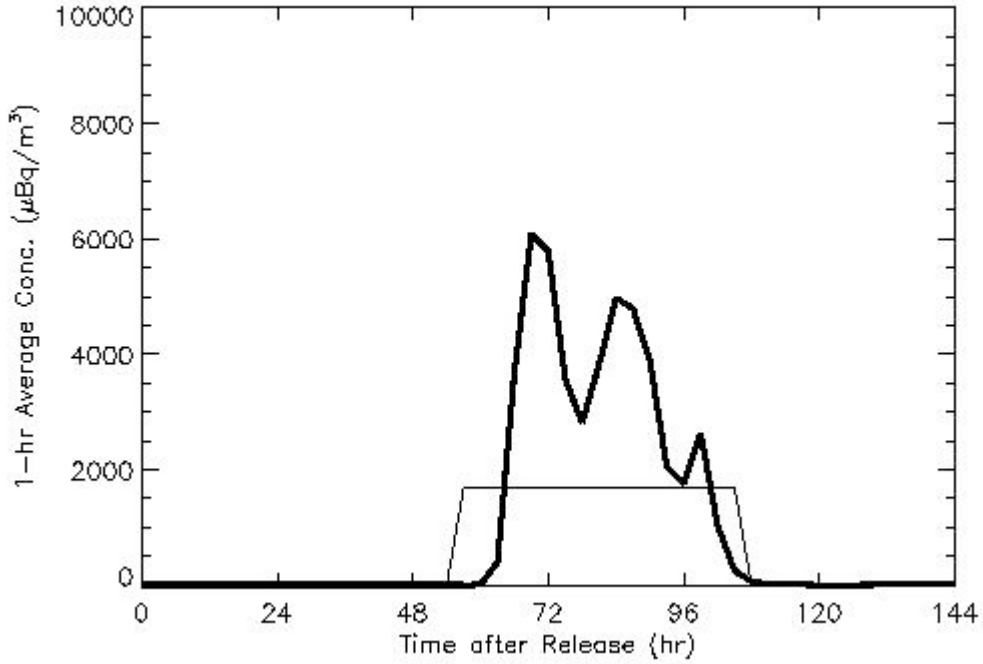

Figure 9. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and two-day-average concentration for 1-3 June measured at Milano, Italy (thin curve).

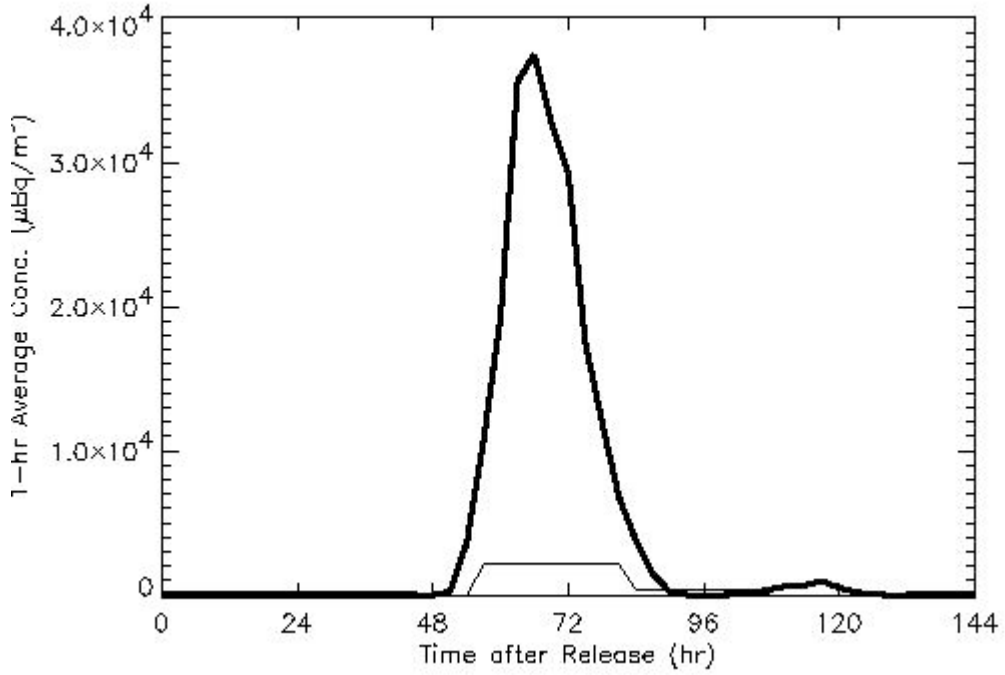

Figure 10. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average concentration for 1-3 June measured at Nice, France (thin curve).

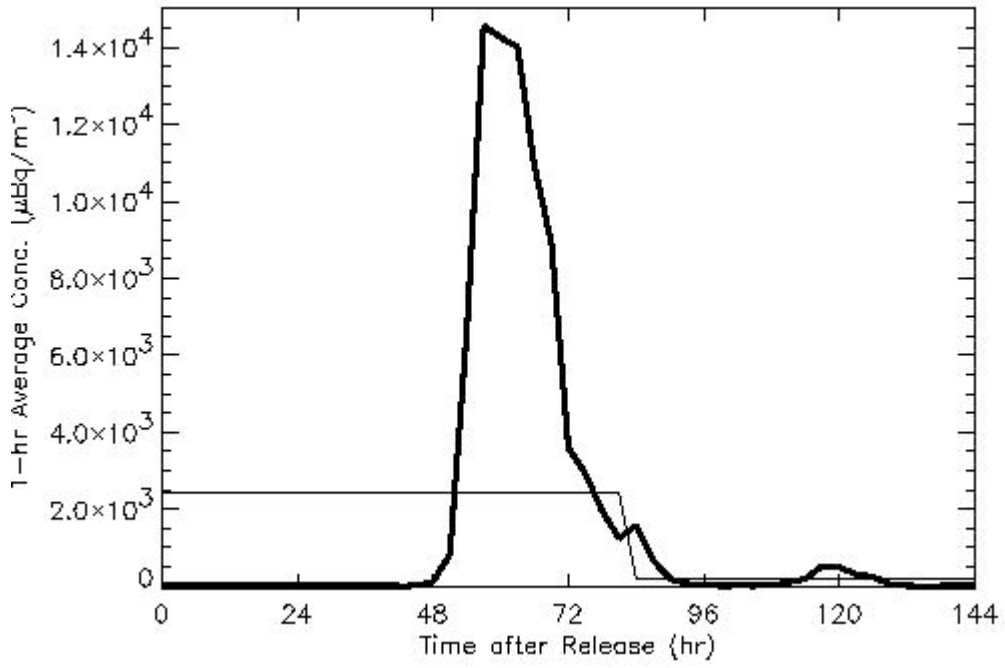

Figure 11. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and several-day-average concentrations for 25 May - 2 June and 2-6 June measured at Toulon/La Seyne, France (thin curve).

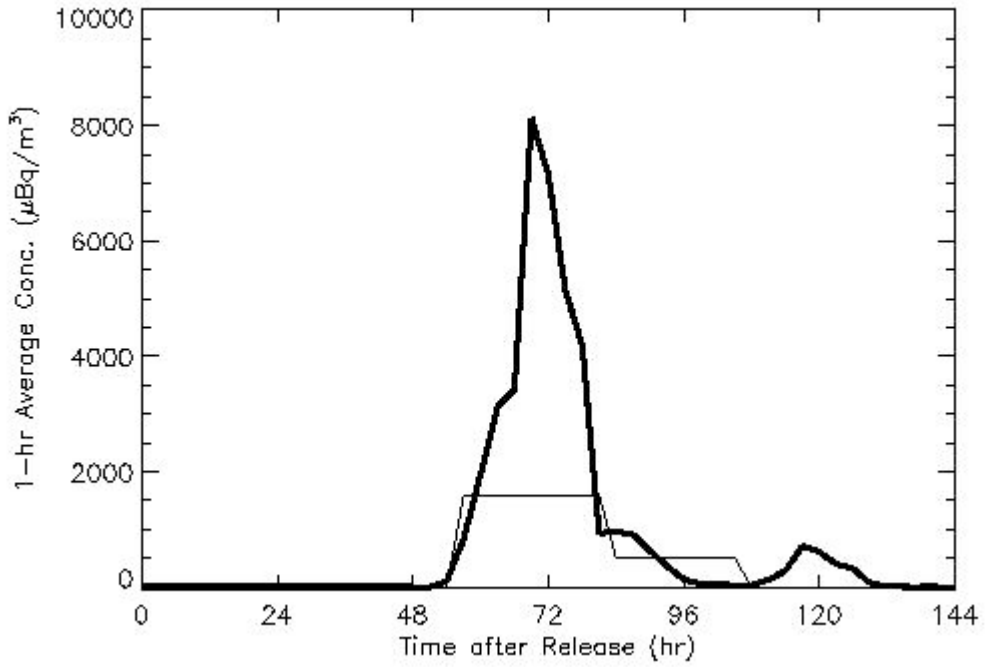

Figure 12. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average concentration for 1-3 June measured at Cadarache, France (thin curve).

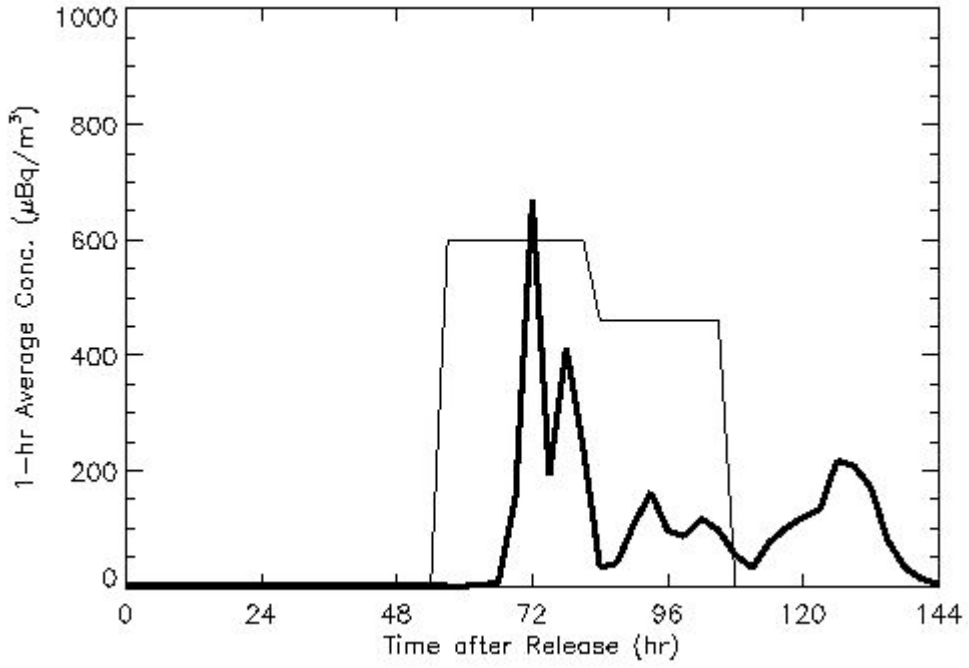

Figure 13. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average concentration for 1-3 June measured at Montpellier, France (thin curve).

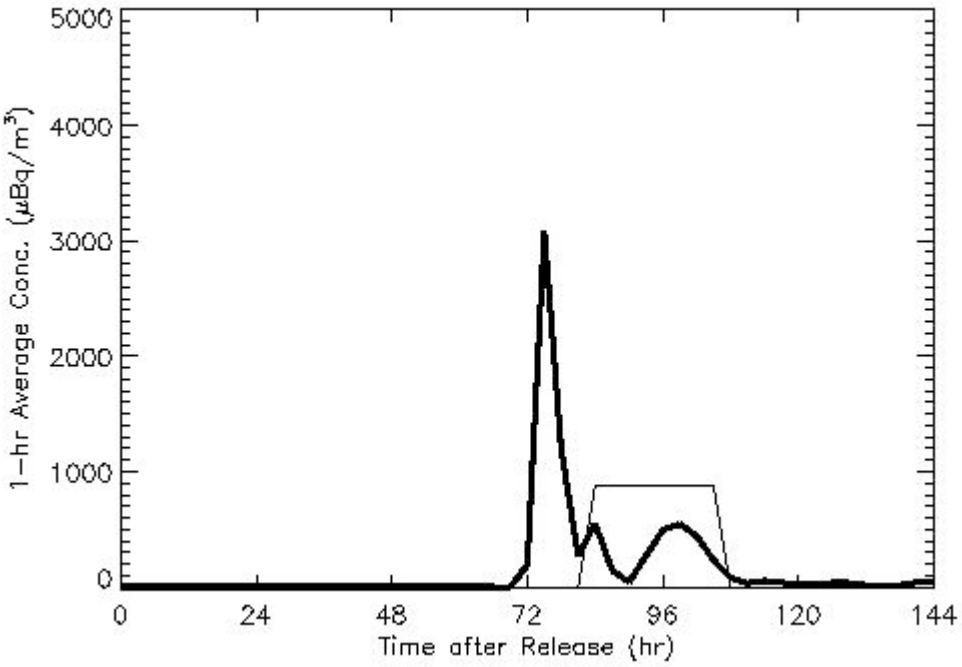

Figure 14. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average concentration for 2-3 June measured at Montfaucon, France (thin curve).

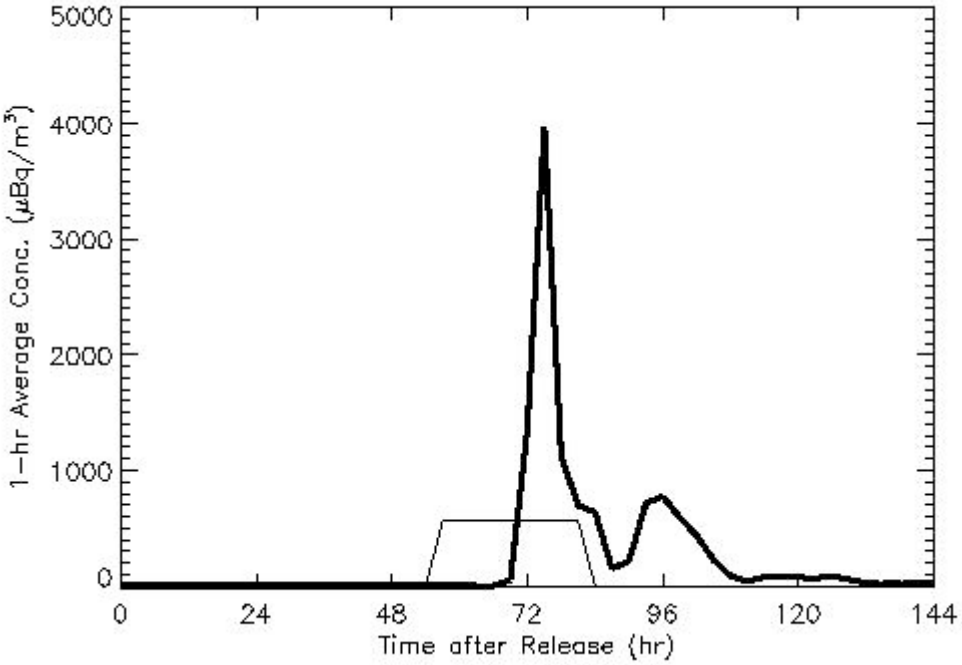

Figure 15. Hourly-average concentration of Cs-137 calculated by DERMA (thick curve) and daily-average concentration for 1-2 June measured at Marcoule, France (thin curve).

High concentration of Cs, registrated at Palomares for the period of 25 May - 02 June was not reproduced by the model (Table 3), because the monitoring station is situated on the periphery of the plume and the modelled plume only slightly touches the south-west cost of the Iberian peninsula and Palomares by 21 UTC on 31 May with a very low concentration (max 0.03 $\mu Bq/m^3$), probably, due to limited resolution of the meteorological fields.

Due to the poor estimation of the release parameters (according to CSN the release rate was 8-80 Ci between 01 and 03 UTC on 30 May), a main possible reason of the model overestimation is an overestimation of the release by a factor of 1.5-2 (the proposed release was about 25 Ci) or a different time or duration of the release. To answer the last question, it is necessary to study a sensitivity of the plume concentration to the time and duration of the release. Moreover, additional DERMA simulations with better/higher resolution of the meteorological DMI-HIRLAM data (the 0.15 degree horizontal resolution grid) show a high sensitivity of the concentration and deposition fields to the wind field resolution.

5.2. INEX 2 exercise: the Perth NPP, Hungary

On 3 November, 1998, an international nuclear emergency exercise, INEX-2-Hun, took place with a hypothetical release from the nuclear power plant in Paks, Hungary. DMI took part in this exercise providing the Danish Emergency Management Agency (DEMA) with predicted concentration and deposition fields calculated by the DERMA model using DMI-HIRLAM data.

The source term information was used from different sources: the first scenario from DEMA on the first step of the forecast and from the RODOS programme - the second scenario. The information we got from DEMA on the source term was different from that estimated by RODOS.

According to the first scenario, the hypothetical release started at 7.15 UTC on 03.11.1998 and lasted for 6 hours, the released amount was 150 PBq of I-131.

The second corrected scenario of the release is the following: starts at 12.00 UTC, duration is 6 hours, released amounts are based on the following IAEA information: 7 PBq of I-131, 0.4 PBq of Cs-137, 0.6 PBq of Cs-134, 10 PBq of the noble gases.

For those two case studies the DERMA long-range dispersion model was run using DMI-HIRLAM data with15 km horizontal resolution.

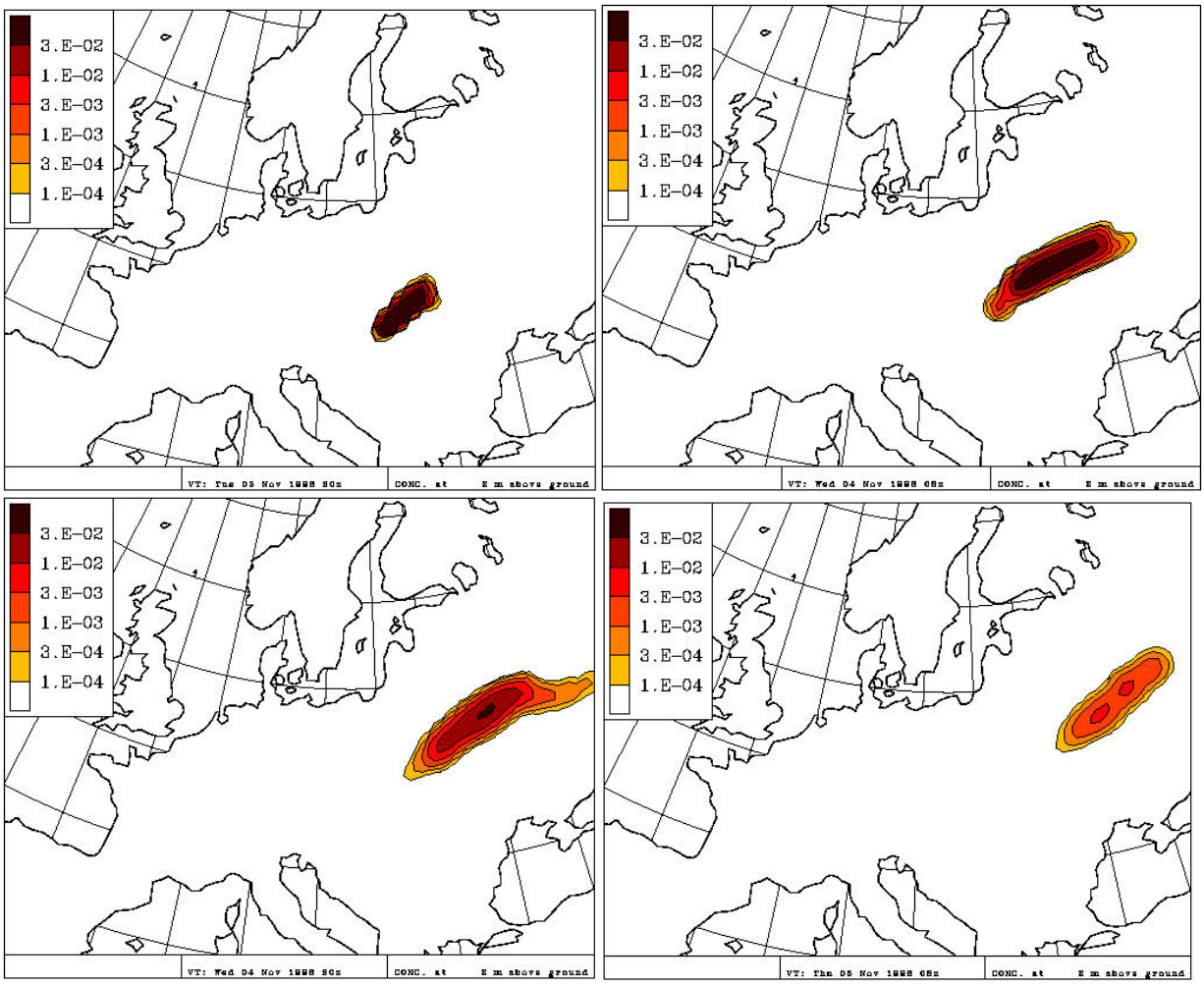

Figure 16. DERMA results. Air concentration of ^{137}Cs (kBq/m^3) 8, 20, 32 and 44 hours after the start of the release on 03-05 November 1998, 20 & 08 UTC. The second scenario.

Figure 16 shows the DERMA simulation results for the second release scenario of the surface air concentrations of ^{137}Cs for the first 2 days: 8, 20, 32 and 44 hours after the start of the release on 03-05 November 1998 (20 & 08 UTC).
The first map of Figure 16 shows the air concentration pattern of ^{137}Cs 8 hours after the start of the release at 20 UTC on 03 November. The released plume extends over the eastern part of the Hungarian territory, the territory of Ukraine and slightly touches the south-west part of Belarus. Next maps of the figure 16 show transport and dispersion of the plume each 12 hours. Next hours the plume extends over the territory of Ukraine, Russia and partly over Belarus.
Figure 17 shows the DERMA simulation results of the external dose (left) and the total deposition (right) of ^{137}Cs by the dry and wet depositions 44 hours after the start of the release on 05 November 1998, 08 UTC for the second release scenario.

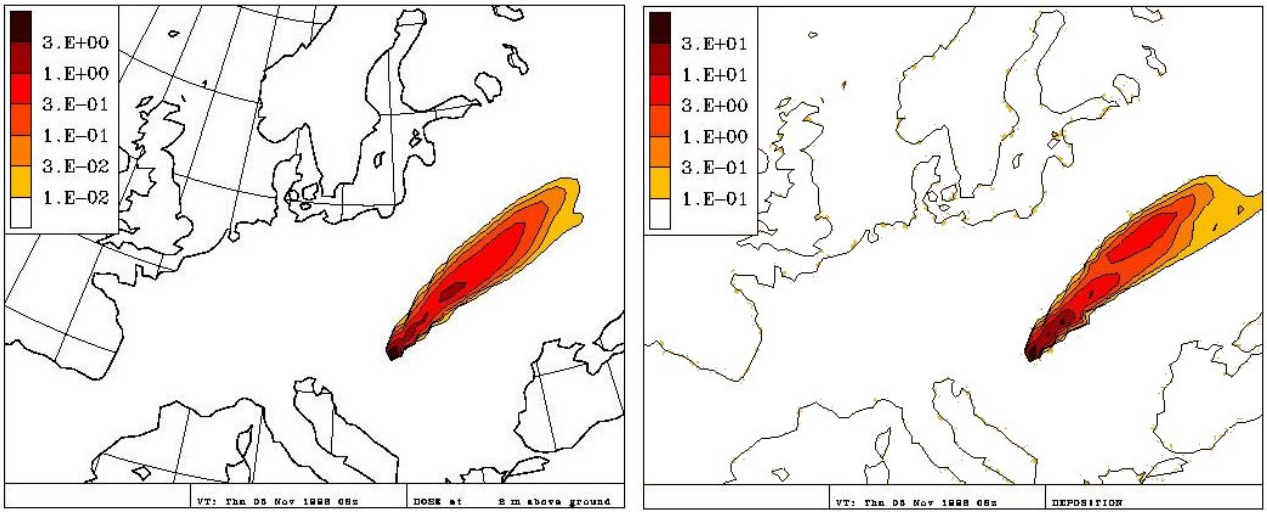

Figure 17. DERMA results. External dose (left) and total deposition of ^{137}Cs (kBq/m^2) (right) 44 hours after the start of the release on 05 November 1998, 08 UTC. The second scenario.

For a comparison we show relevant simulation results of the Finish Meteorological Institute (FMI) by the SILAM model (FMI, 1999). Figure 18 shows the air concentration +6 & +12 hours after the start of the release for the second scenario (FMI, 1999). Figure 19 shows the external dose rate (cloud + fallout) at + 24 & +48 h after the start of the release (FMI, 1999).
As it seen, our DERMA results do not seem to differ much from what can be seen from FMI by the SILAM model and from the Swedish Meteorological and Hydrological Institute simulations by the MATCH model (SMHI, 1999).

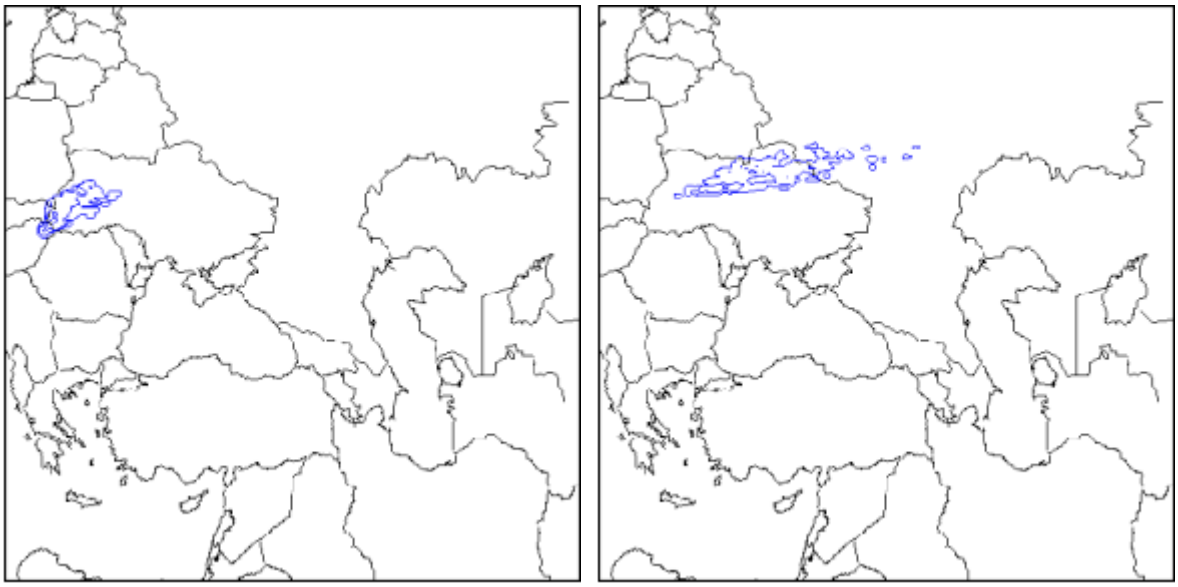

Figure 18. SILAM results. Air concentration +6 & +12 hours after the start of the release. The second scenario (FMI, 1999).

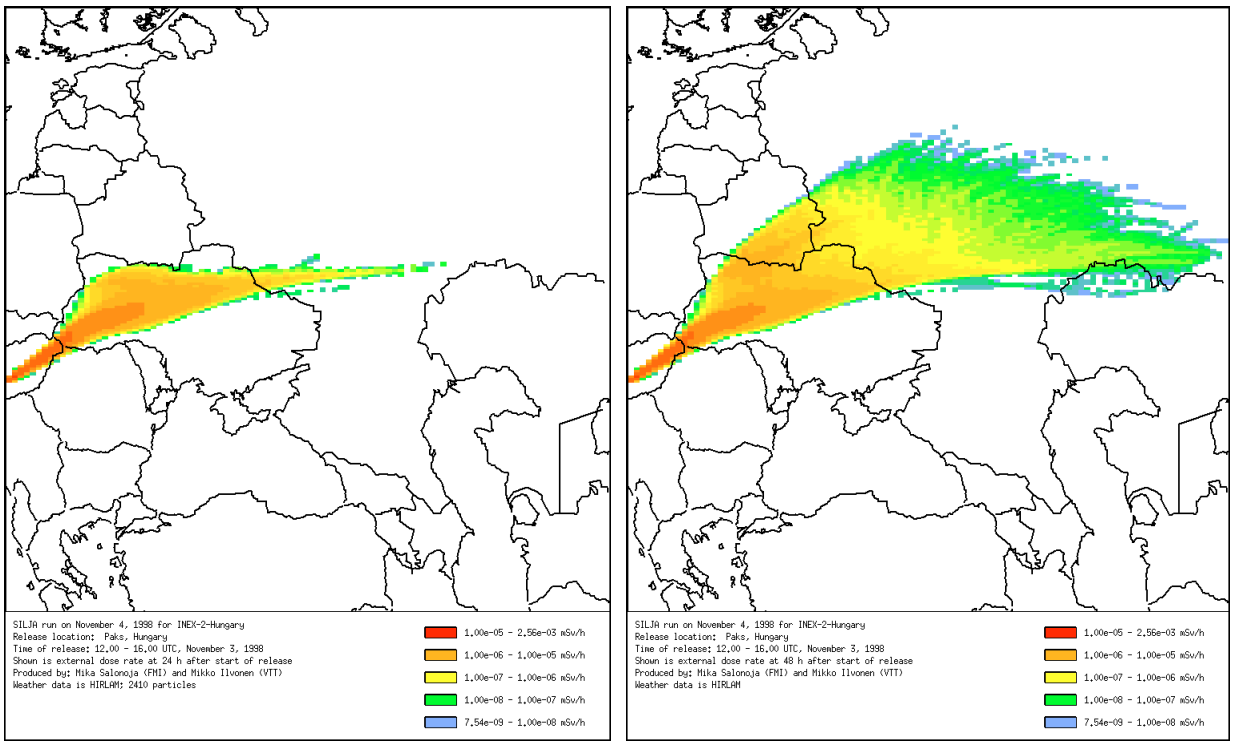

Figure 19. SILAM results. External dose rate (cloud + fallout) at + 24 & +48 h after the start of the release. The second scenario (FMI, 1999).

VI. CONCLUSIONS

The Danish Emergency Response Model of the Atmosphere (DERMA) was improved by including processes of the deposition and radioactive decay. Different parameterisations of the removal processes by the dry and wet deposition of radionuclides to the surface were analysed. For emergency response and for post-accidental analysis, different versions of the DERMA model are suggested using different approaches and complexities for the parameterisation of the deposition processes.

It was shown that the washout coefficient Λ strongly depends on the particle size (the so-called 'Greenfield gap'), although in most existing models of long-range pollution transport the washout coefficient does not depend on particle radius. Therefore, a revised formulation of the wet deposition parameterisation for particles of different size was suggested.
For accidental releases from nuclear power plants, particles smaller than 0.5 μm radius can play an important role in long-range transport and dose forming, therefore the formulation suggested for the washout coefficient in the DERMA model, in contrast to the formulation in the ADPIC model, includes the mechanism of the Brownian capture and covers a gap of the washout parameterisation for small particles.

For heavy particles (r_p > 1 μm), the gravitation settling essentially effects the process of deposition to the surface. Therefore, the effect of the gravitation settling was included into the dry deposition parameterisation through the gravitation settling velocity v_g. It was suggested to model the gravitation settling velocity v_g by a combination of the Stokes law (for particle diameters less than 3.5 μm) with the Cunningham correction for small particles (r_p < 0.5 μm) and an iterative procedure (Näslund & Thaning, 1991) for the equation for the terminal settling velocity (for larger than 3.5 μm particles).

As the first step to verify the deposition parametrisation for the DERMA model and to study effects of deposition, simulations for the INEX-2-Hun exercise and the Algeciras accidental Cs-137 release in Spain, taking into account the deposition process, and a comparison of the simulation results with measurements data from 9 European monitoring stations were presented. The comparison shows in general a good agreement of the simulated concentration with the monitoring data.

The main maxima of concentration, predicted by the DERMA model, coincide rather good temporally with the measurement peaks for the monitoring station Ispra, as well for other stations.
Due to the poor estimation of the release parameters, a possible reason for an overestimation by the model for some monitoring stations is an overestimation of the release by a factor of 1.5-2 (the proposed release was about 25 Ci) or incorrect time and duration of the release. To answer this question, it is necessary to additionally study a sensitivity of the plume concentration to the time and duration of the release.
Other reason for this difference might be a simplification of the description of the vertical distribution of pollutants as evenly-mixed along ABL. Comparisons of the estimated ABL height in the DERMA with measurement data for the Gibraltar aerological station at the release time show a good agreement.

Moreover, DERMA simulations with higher resolutions of the meteorological DMI-HIRLAM data show a high sensitivity of the concentration and deposition fields to the wind field resolution.

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The comparisons of the simulations with the measurement data for the Algeciras accident were possible thanks to the RTMOD project (co-ordinator Giovanni Graziani) and LLNL. I also thank Ilkka Valkama from the Finish Meteorological Institute for giving his results of INEX2-Hun simulation for the comparison.

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