RESEARCH REPORT

Norwegian Meteorological Institute

DNMI

Det norske meteorologiske institutt

No. 131

NLH AGRICULTURAL UNIVERSITY OF NORWAY

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Observational systems: Development, validation and implementation of new technical systems and methods for observation of meteorological parameters are areas which are continuously focused.

Gravitational settling of particles in dispersion model simulations using the Chernobyl Accident as a test case

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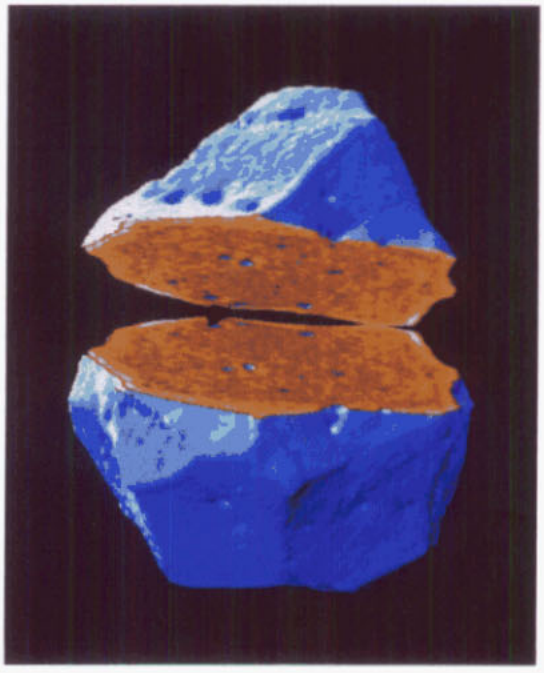

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Abstract

Atmospheric emission, transport and deposition of radioactive particles of different size and density is the main topic of the report. An important and innovative part of the report is a detailed description of the source term for particles released from the Chernobyl reactor, which is not typical for other studies dealing with simulation of atmospheric transport and deposition. The source term description led to 12 scenarios for the SNAP model runs, which included four different classes of particle size and three different classes of particle density. The SNAP model, developed originally at DNMI for operational applications, was modified mainly by implementing parameterization of gravitational settling velocity, the main process leading to dry deposition of larger particles. The results of the modified model simulations showed that the deposition of radionuclides was dependent mainly on particle size and that particles with the diameter of 10 μm were transported to Norway during the Chernobyl Accident. This finding is in agreement with the measurements performed at NLH. Particles with sizes larger than 50 μm are deposited relatively close (less than 100 km) to the source, contributing more to local than regional problems.

Keywords

Gravitational settling, hot particles, model simulations, Chernobyl Accident

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Preface

At the “NKS-SBA-1 2000” in Oslo 17-19 October 2000, a presentation given by Professor Brit Salbu of NLH (Agricultural University of Norway) initiated an exchange of views concerning the importance of including the process of gravitational settling of particles in dispersion model calculations. As experienced after the Chernobyl accident, radioactive debris from the accident in the form of particulate matter have been observed surprisingly far away from the site of release.

In most long range dispersion model calculations, releases of radionuclides in the form of particulate matter are treated as aerosol particles. The real-time dispersion model SNAP (Severe Nuclear Accident Program) in use at the Norwegian Meteorological Institute, is typical in that sense.

The particles are supposed to have similar properties to that of sulphate particulate matter; in the size range up to a few μm in diameter. Dry deposition (both as impaction and gravitational settling) is not supposed to be very efficient as a deposition process for particles of this size. This means that wet scavenging by precipitation is the more effective deposition process for this fraction of the releases. This is the background for excluding gravitational settling of particles as a deposition process in real time long range dispersion models in operational use.

As an example, we describe the situation from the Norwegian point of view: “Kriseutvalget” - is the central decision-making body in the national preparedness organization handling nuclear accidents. DNMI is a central adviser to “Kriseutvalget” in questions where meteorological information/data/prognoses are needed. One of DNMIs main operative ‘tools’ in this adviser role is SNAP; a real time dispersion model. The main use of SNAP is to provide transport simulations of radioactive debris from accidental releases far from Norway (~some 100 to 1000 *km* away). As such, SNAP is useful as a tool for supporting “Kriseutvalget” in its decision-making process.

NLH has worked with radioactive particles released from different sources since 1987, in particular in fallout from the Chernobyl accident. Based on several historical events, radioactive particles should be expected if refractory radionuclides are released during a severe nuclear accident. Furthermore, particle characteristics influencing mobilisation and the subsequent transfer of associated radionuclides are source and release scenario dependent. The relevance

of including radioactive particles (source term codes) in impact assessments has been acknowledged by EU (Nuclear Fission 4. Frame - RAFF and 5. Frame -Advance projects), and by IAEA initiating a new five year programme on radioactive particles. NLH is also an adviser to Kriseutvalget and consider that information on particle deposition would significantly reduce the uncertainties in ecosystem transfer estimates.

The information given by Professor B. Salbu raised some questions concerning assumptions and simplifications made in formulation of dispersion models in use at some meteorological centres, - including SNAP - used by DNMI. The inclusion of the process 'gravitational settling of particles' can be of high importance in some situations in the simulation of dispersion/transport events.

Based on this background, DNMI and NLH initiated a collaboration in which we intended to test more in detail the importance of including gravitational settling of particles in dispersion simulations.

The main objectives of the NKS-SBA-1GaP project are to:

- Test formulas/codes related to gravitational settling of particles in dispersion model calculations,
- To test the model with implemented codes on the Chernobyl accident case.

The project has been a joint collaboration between DNMI and NLH.

Table of Contents

1 INTRODUCTION

To assess the environmental impact of radionuclides released to the atmosphere from a known source, information on the source terms, the dispersion pattern and associated deposition pattern is needed. To improve predictions related to areas affected, the source term as input to the atmospheric dispersion model should be relevant. Furthermore, processes influencing radionuclides defined as the source term should be adequately treated in the models.

The source term includes information on the activity concentration of released radionuclides, activity or isotopic ratios and the physico-chemical form (speciation). Radionuclides released from different nuclear sources can be present in different physico-chemical forms, ranging from simple ions or molecules to colloids, particles and fuel fragments (Salbu, 2000a). A significant fraction of refractory radionuclides released by nuclear events, such as testing of nuclear weapons (Lokan *et al.* 1985; Simon *et al.*, 1994; Danesi *et al.*, 1998, Salbu *et al.*, 2000b) or use of Depleted Uranium (DU) in munition (Danesi *et al.1998*, Salbu *et al. 2001*), nuclear reactor accidents (Devell *et al.*, 1986, Salbu *et al.*, 1994), fires involving nuclear installations (Windscale), satellite and air craft accidents (Thule, Palomares, Cosmos) and the dumping of radioactive waste (Salbu *et al.,* 1997), is present as radioactive discrete particles of various size, composition and structure. Effluents released from nuclear reactors and reprocessing plants under normal operating conditions (e. g. Sellafield, UK; La Hague, France) also contain radionuclides as particles and colloids (Salbu *et al.*, 1993). Thus, the release of radioactive particles occurs more frequently than usually anticipated. Furthermore, release of radioactive particles should be expected if refractory radionuclides are identified in fallout from nuclear installations or devices.

The release of radioactive substances to the atmosphere from the accident at the Chernobyl nuclear power plant starting on the 26 April 1986, consisted to a large extent of particulate matter, in addition to gases including noble gases. These two forms of releases have been taken into account in modelling atmospheric transport and deposition of radioactive substances caused by the accident, but the main focus has been on the small particles (aerosol).

In most of the existing models developed to simulate long range transport (e. g. Maryon *et al.*, 1991; Valkama and Salonaja, 1993), the diameter of particles was limited to a few microns only, similar to SO_4 aerosol, assuming that larger particles were deposited close to the source. Such an assumption was reasonable for a large part of the emitted particles, but not for all of them. This and similar questions were considered by the EU RAFF project (RAFF, 1999).

Available measurements showed that large particles emitted from the Chernobyl Accident were found all over Europe (Pöllänen *et al.*, 1997). The possibility of atmospheric transport of large particles to distant locations was also confirmed by the results of the trajectory model developed in Finland (Pöllänen *et al.*, 1997). These particles, often referred to as "hot particles", are quite frequently found far away from the source and sometimes their activities may be so high that even a single particle can cause severe health effects. Following Pöllänen and Tuovinen (1994) and RAFF (1999), we consider a particle to be large if its aerodynamic diameter is 20 μm or larger, with the corresponding terminal settling velocity of the order of a few $cm\ s^{-1}$, and corresponding real particle diameter 7 μm or larger depending on its density.

After the Chernobyl Accident, Severe Nuclear Accident Program (SNAP) model was developed at DNMI in order to simulate emission, atmospheric transport and deposition of radioactive debris released to the air during potential nuclear accidents. As most models of this type, the operational version of the SNAP did not treat large particles with sufficient refinement to account for the observational facts mentioned above. Implementation of refined treatment of large particles into the model calculations has therefore been the main objective of the joint (DNMI + NLH) project SBA-1GaP: "Gravitational settling of particles in dispersion model calculations", performed during the year 2001 in the frame of the NKS Programme.

In order to estimate the range of large particle transport, several simulations of the Chernobyl accident have been made, using the modified version of SNAP. Description of the modifications of the model and results of the model simulations are included in the present report.

2 RELEASE OF PARTICLES DURING THE CHERNOBYL ACCIDENT

Noble gases, gases of volatile elements, aerosols, fuel particles and fuel fragments were released to the atmosphere during approximately 10 days of release from the Chernobyl Accident. General description of the release can be found in the OECD Report (OECD, 1996). Gaseous forms, such as krypton and xenon escaped completely from the fuel material while approximately 50-60% of gaseous iodine was released. A part of iodine, as well as cesium and tellurium were attached to aerosols, of typical size 0.5 - 1.0 μm. One of the features of the source term was release of fuel materials in form of mostly large but also smaller particles, which were often referred to as “hot particles”. Model simulations of emission, atmospheric transport and deposition of these particles is the main subject of the present study.

Early estimate for fuel material released to the atmosphere was 3 ± 1.5 % (IAEA, 1986). The revised estimate (Bedyaev *et al.*, 1991), 3.5 ± 0.5 %, corresponds to about 6 - 8 tonnes as fragmented UO_2 fuel material.

Following Buzulakow and Dobrynin (1993), estimation of daily release pattern is shown in Figure 1. High release rates at the beginning of the period were caused mainly by the mechanical fragmentation of the fuel during the explosion. The second period of high release rates, between day 7 and day 10, was associated with criticality and the high temperature reached in the core melt. The drop in release after 10 days was caused by fire extinction and rapid cooling of the fuel.

2.1 Sources of radioactive particles

Several sources have contributed to release of radioactive particles in the past. Following high temperature accident scenarios (e.g., the Chernobyl accident), a range of different uranium fuel particles and condensed particles were observed. The uranium particle matrix vary in composition, structure (e.g. crystalline and amorphous phases) and oxidation state. Following low temperature releases (e.g., early 1950s release from Windscale piles, UK), flake-like uranium fuel particles significantly different from those collected after the Chernobyl accident were identified (Salbu,1994). Radioactive particle releases have also occurred at the Windscale and Rocky Flats nuclear installations as a result of incidents involving fires. Accidents involving nuclear weapons near Thule (Greenland) and Palomores (Spain), an incident involving a rocket launch

on Johnson Atoll, and the failure of the Cosmos 954 satellite have all lead to the dispersion of significant quantities of particulate forms of plutonium into the environment. High specific activity particles have been identified in the fjords of Novaya Zemlya in close vicinity to dumped objects within radioactive waste dumping sites. Also, it is now well established that a major fraction of radionuclides released in effluents from reactors and reprocessing plants (Sellafield, UK; La Hague, France) are in the form of particles and colloidal material. To assess the long term impacts of radioactive particles at these sites within reasonable uncertainties, information on key particle characteristics, size distributions, transport and ultimate fate of particle associated radionuclides is essential.

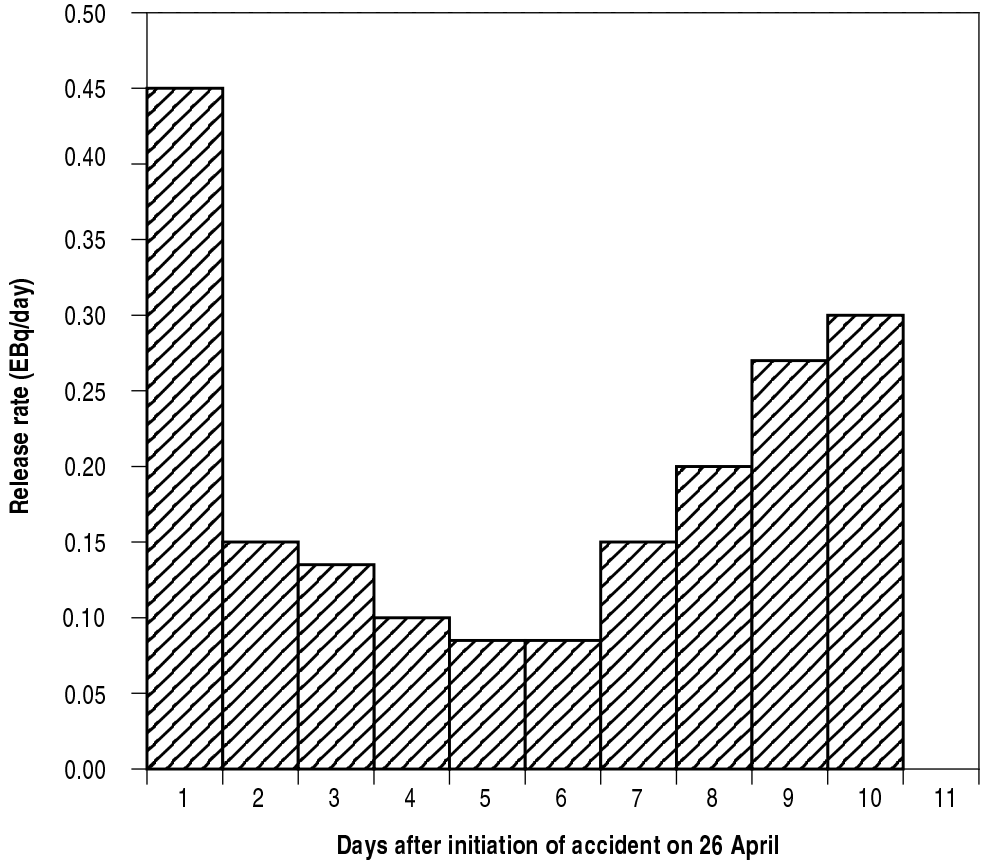


Figure 1. The daily release rate of radioactive substances into the atmosphere (not including noble gases). The range of uncertainty for all releases is ±50 %.

2.2 Formation of radioactive particles

Radioactive particles are defined as localised aggregates of radioactive atoms larger than 0.45 mm that give rise to an inhomogeneous distribution of radionuclides, significantly different from that of the matrix background (IAEA, 1999). Localised heterogeneities ranging within 1 *nm* - 0.45 μ*m* are referred to as radioactive colloids and pseudo colloids. Following a nuclear event, radioactive particles are formed due to:

- mechanical destruction of the fuel matrix,
- formation of clusters,
- condensation of volatiles on particle surfaces during release or,
- interactions with aerosols during atmospheric transport.

Release of fuel particles containing refractory elements reflecting burn-up (e.g. ^{95}Nb, ^{95}Zr, ^{144}Ce) and variable concentrations of volatiles (e.g. ^{137}Cs, ^{90}Sr) is attributed to mechanical destruction of fuel caused by explosions, fires or corrosion. Thus, radionuclide concentrations and activity ratios are source-specific, while different release conditions could influence micro-distributions, structures and oxidation states of matrix elements.

The source term speciation may include:

- Fuel particles or particles of fissile material formed by mechanical disruption, pulverisation and dispersion of the original bulk radioactive material, ranging in size from μm to several hundreds of μm (e. g., U or Pu-particles).
- Discrete radioactive particles or clusters formed within the fuel during normal operations or during the release (e. g. Ru-particles).
- Gasses of volatile radionuclides (e. g., noble gasses)
- Condensed particles or molecular aggregates and aerosols formed upon condensation of volatile radionuclides on available surfaces during the release or during transport.
- Colloids and/or pseudo colloids (e.g., 1-450 *nm*) in effluents either released directly or formed in the environment.
- Low molecular mass species (e. g., ions, complexes, cheletes) having a nominal molecular mass lower than 1 *nm* (<1 kDa).

After the release, deposited radioactive particles represent point sources of short- and long-term ecological significance. As these materials include high-specific activity particles, pseudo colloids or colloids, there is a risk from inhalation, dermal absorption, wound exposure and ingestion of the particles. For filter-feeders (e.g., molluscs), aggregates and colloids are retained from the aquatic environment. Being relatively inert, ecosystem transfer of particles associated radionuclides is delayed. Due to time-dependent weathering, however, associated radionuclides are mobilised and the ecosystem transfer increases. Thus, information on parti-

cle characteristics influencing weathering rates and subsequent radionuclide mobilisation is essential for impact assessments. Weathering rates will depend on the particle composition (e.g., UO_2 fuel), structural changes occurring during the event (e.g. transformation from UO_2 to U_3O_8), and on local chemical conditions and transformation processes occurring after deposition (e.g., pH, redox conditions, bioerosion). These processes will be particularly important in controlling the speciation, and transfer to biota of mobile radionuclides such as ^{90}Sr.

2.3 Release from the Chernobyl reactor

During the Chernobyl accident, about 6-8 tonnes of UO_2 fuel were released into the atmosphere (Victorova and Garger, 1990). Large fuel fragments varying in size and radionuclide composition were deposited within the 30 *km* zone (Loschilov *et al.*, 1992; Kashparov *et al.*, 1996), while small-sized fuel particles as well as ruthenium particles were identified in Scandinavia (Figure 2), more than 2000 *km* from the site (Devell *et al.*, 1986; Salbu *et al.*, 1988).

During the initial explosion in Unit 4 on April 26th 1986, mechanical destruction of the uranium dioxide fuel occurred at high temperature and pressure, and deposition of large fuel particles took place in a 100 *km* long and 10 *km* wide trail to the West of the reactor (Loschilow *et al.*, 1992; Kashparov *et al.*, 1996). Large fragments having radionuclide composition close to estimated fuel burn-up, deposited within 2-5 *km* from the site (Victorova and Garger, 1990; Loschilov *et al.*, 1992; Kashparov *et al.*, 1996). During the following days, April 27th-30th, volatile fission products and U fuel particles were released under high temperature and oxidising conditions due to the fire, and deposition of particles occurred along a trail to the Northwest, North and Northeast direction of the plant (Kashparov *et al.*, 1996). From April 30th to May 6th the temperature and subsequently the emission of volatiles decreased. It is assumed that a lowering in temperature (600-1200 *K*) enhanced the oxidation of exposed fuel. As the release scenario, including temperature, pressure and redox conditions, changed during the course of the event, released fuel particle characteristics are expected to change during the course of the event (Kashparov *et al.*, 1999). Thus, two source terms should be differentiated:

- release during the initial explosion in the Unit 4 Chernobyl reactor (high temperature, pressure, no oxygen),
- release during the subsequent fire in the reactor core (medium temperature, normal pressure, oxygen).

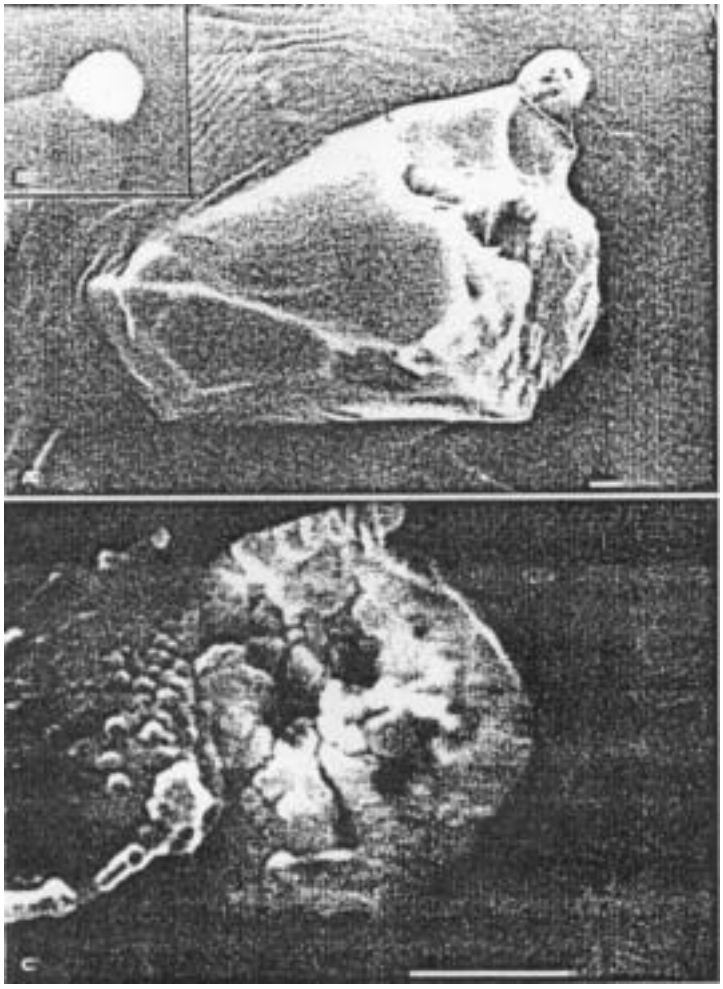
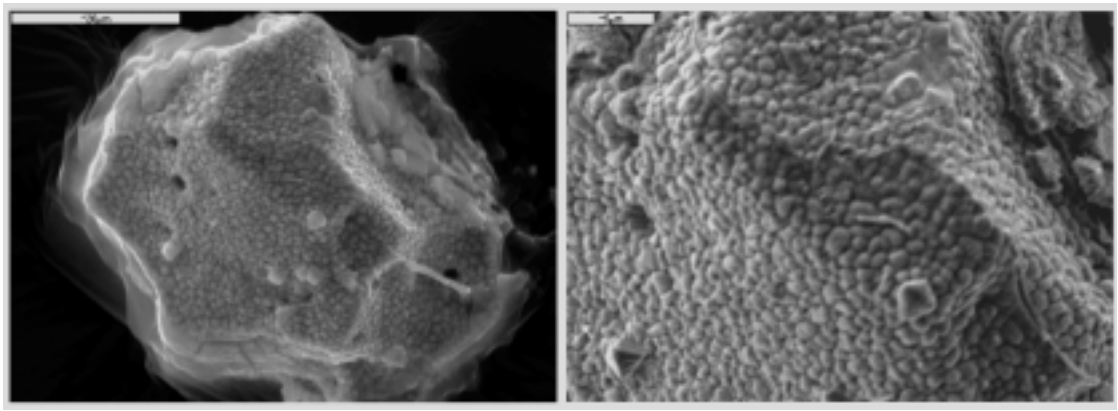


Figure 2. 106-Ru particle (with a spherical structure) attached to a larger Si particle. The particle was collected in Valdres, Norway, in 1997.

2.4 Characteristics of Chernobyl fallout

Radioactive particles collected during 1989-1998 to the West and to the North of the Chernobyl reactor and from within the damaged reactor, have been characterised with respect to radionuclide composition (gamma spectrometry), structure (Scanning Electron Microscopy (SEM) and synchrotron radiation X-ray micro X-ray techniques). SEM documented large aggregates, spherical grains or small crystallites varying in size from few to several hundred microns (Figure 3) could be identified (Salbu *et al.*, 1994; 1998). Backscattered electron imaging (BEI) showed fuel particle surfaces containing high atomic number elements, while X-ray mapping and element analysis with X-ray Micro-Analysis (XRMA) confirmed the presence of U and fission products (Figure 4). Based on more than 100 particles, a variety of surface structures and morphologies were observed, for instance, black coloured aggregates or hexahedral crystals, fragile white or brownish amorphous aggregates easily destroyed by handling and particle

surfaces with cavities and spherical holes, with traces of fission products (e.g., ^{106}Ru, Cs-isotopes, ^{144}Ce), activation products from construction materials (e. g., ^{95}Zr, ^{55}Fe), or metals from fire extinction materials (e. g., Pb). SR-based X-ray microscopic techniques were used to identify crystallographic structures (microscopic X-ray diffraction (μ-XRD)), obtaining elemental 2-D and 3-D distributions (microscopic X-ray absorption (μ-XAS), fluorescence spectroscopy (μ-XRF) and micro tomography) and to obtain information on the distribution of oxidation states for U (microscopic X-ray absorption near edge structure spectroscopy (μ-XANES)).



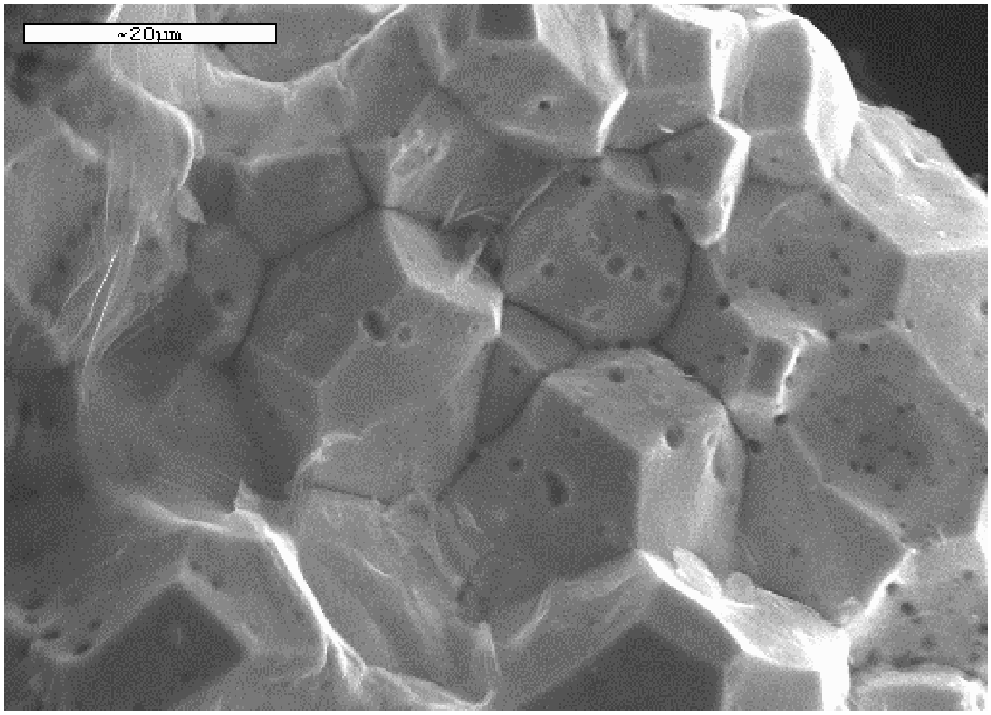


Figure 3. SEM documented large aggregates, spherical grains or small crystallites varying in size from few to several hundred microns (Salbu *et al.*, 1994).

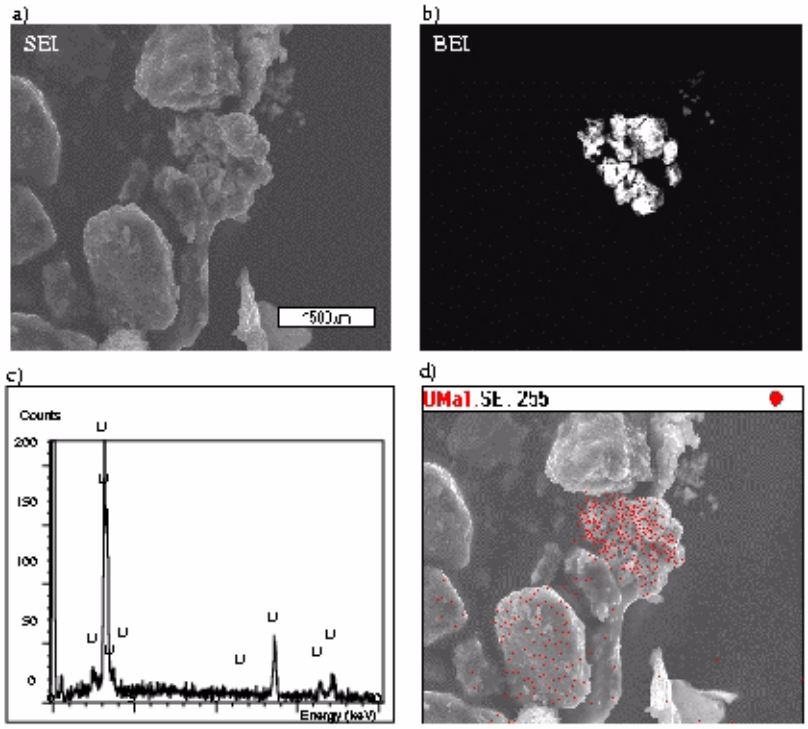


Figure 4. Oxidised fuel particles collected within the 30 *km* zone of Chernobyl, 1987 (Salbu, 2000): a) Secondary Electron imaging (SEI) mode, b) Backscattered Electron Imaging (BEI) mode, bright areas reflect high atomic number elements, c) Elemental analysis by XRMA d). X-ray mapping of uranium (Bar 500 *m*).

During the initial explosion in the Chernobyl reactor, apparently reduced U fuel particles with reminiscence of the UO_2 core were released and deposited to the west of the reactor. These particles are relatively inert, i. e. less susceptible to weathering (Figure 5). The weathering rate is relatively slow and subsequent mobilisation of associated radionuclides is delayed. During the fire, however, oxidised U_2O_5/U_3O_8 fuel particles with UO_2 cores were released and deposited to the North, and probably also to the East and South of the reactor. These particles are more susceptible to weathering (Figure 4). The weathering rate is relatively high and increased soil-to-plant transfer of ^{90}Sr have also been observed 10 years after the accident.

Being key factors in determining particle weathering rates, subsequent mobilisation and ecosystem transfer of associated radionuclides, particle characteristics are essential input parameters in impact assessment models for areas significantly contaminated with radioactive particles (nuclear test sites, accidental or waste sites). Unless particle characteristics and the impact of particles are taken into account, assessment model predictions suffer from large uncertainties.

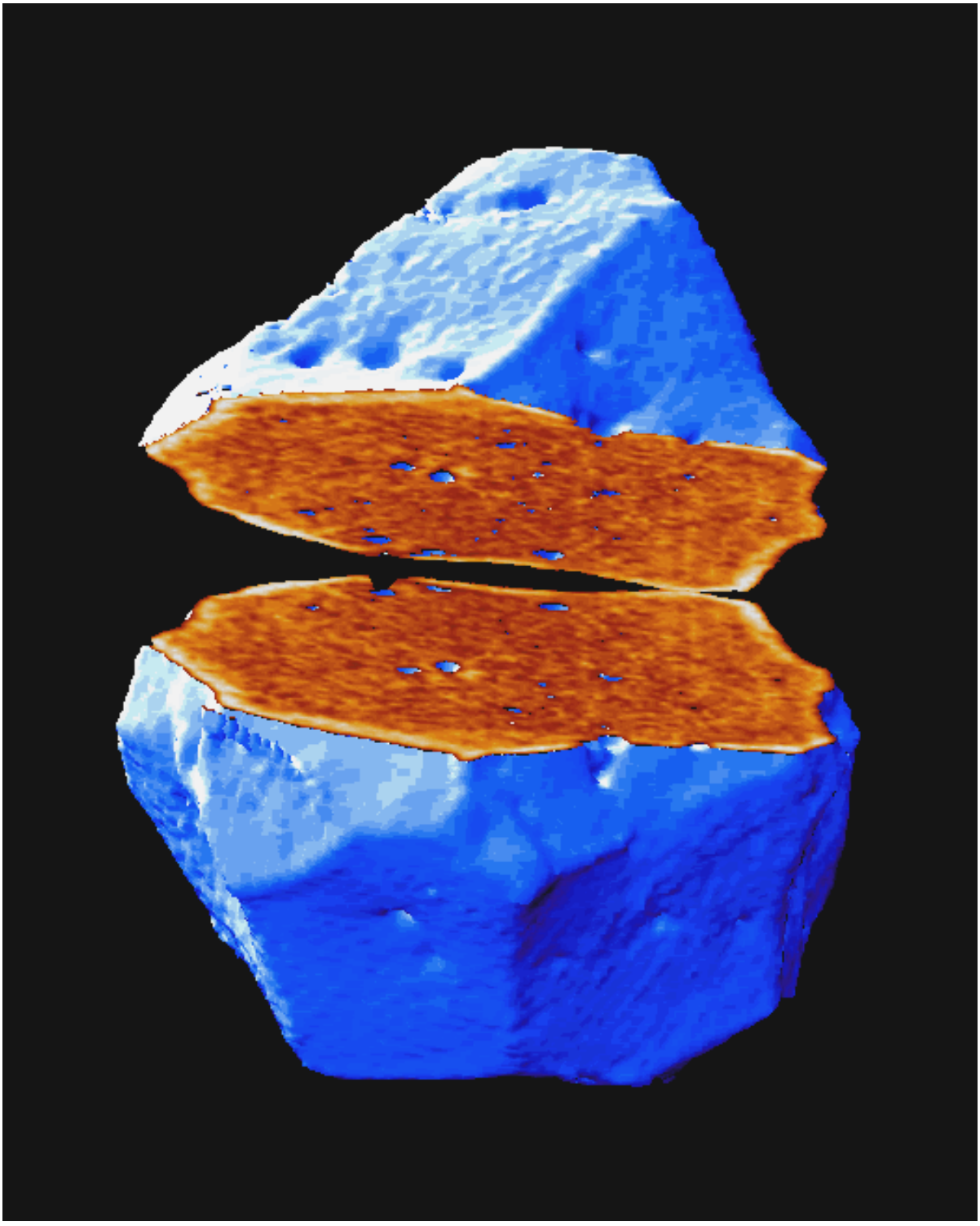


Figure 5. Synchrotron radiation based X-ray micro-tomography of apparently reduced Chernobyl fuel particle. The data block has been cut and opened to show the inner structure of the particle (Salbu *et al.*, 2001).

2.5 Source term descriptions for selected scenarios

To analyse the transport and deposition of particles released from Chernobyl reactor, 12 scenarios have been designed for the SNAP model runs. Based on the information presented in the previous paragraphs, the following assumptions were made for these scenarios:

1. The total mass of fragmented fuel material released to the atmosphere was about 6-8 tonnes. We assume that approximately 1/3 of this release (M_e~2 tonnes) was available for atmospheric transport out of the 30 *km* zone.

2. 30% of M_e was released the first day: 600 kg
   - 50% of this in the form of U metal particles: 300 kg
   - 50% of this in the form of UO_2 particles: 300 kg
   - effective height of release for this day was in the range 1000 m - 3000 m

3. 30% of M_e was released in days 2 - 6: 600 *kg*
   - 50% of this in the form of U metal particles: 300 *kg*
   - 50% of this in the form of UO_2 particles: 300 *kg*
   - effective height of release for these days was in the range 150 *m* - 1000 *m*

4. 40% of M_e was released in days 7 - 10: 800 *kg*
   - 100% of this in the form of U_3O_8 particles: 800 *kg*
   - effective height of release for these days was in the range 1000 *m* - 2000 *m*

5. The particle spectrum covers the range 0.1 μ*m* - 500 μ*m*, and is divided into four particle size classes:
   class 1: 10% of M_e: 200 *kg* in the range 100 μ*m* - 500 μ*m*, with geometric mean diameter 200 μ*m*
   class 2: 30% of M_e: 600 kg in the range 30 μ*m* - 100 μ*m*, with geometric mean diameter 50 μ*m*
   class 3: 45% of M_e: 900 kg in the range 1 μ*m* - 30 μ*m*, with geometric mean diameter 10 μ*m*
   class 4: 15% of M_e: 300 kg in the range 0.1 μ*m* - 1 μ*m*, with geometric mean diameter 0.2 μ*m*

6. Three density classes for the particles are taken into account:
   U metal: 19.0 $g\ cm^{-3}$ (inert U)
   UO_2: 10.96 $g\ cm^{-3}$
   U_3O_8: 8.3 $g\ cm^{-3}$ (oxidized U)

Using the above assumptions, emission rates were calculated for the 10 days the Chernobyl accidental releases lasted, separately for each size class and density class. These emission rates were then used in the model simulations and are summarized in Table 1. We have also assumed release height ranges for each of the 10 days in the following way:

Day of release	Height of release
1	1000 *m* - 2000 *m*
2 - 6	50 *m* - 1000 *m*
7 - 10	1000 *m* - 1500 *m*

Table 1: Source term data used in the model simulations of the Chernobyl Accident

Day of accident	**1. class:** 10% of M_e	**2. class:** 30% of M_e	**3. class:** 45% of M_e	**4. class:** 15% of M_e
	(100µm-500µm) d_p=200µm	(30µm-100µm) d_p=50µm	(1µm-30µm) d_p=10µm	(0.1µm-1µm) d_p=0.2µm
1 (1 day)	Total emission: 60 kg Emission rate: 60 kg/day=0.694 g/s divided in: **U** : 30 kg/day **UO_2** : 30 kg/day **U_3O_8**: 0 kg/day	Total emission: 180 kg Emission rate: 180 kg/day=2.083 g/s divided in: **U** : 90 kg/day **UO_2** : 90 kg/day **U_3O_8**: 0 kg/day	Total emission: 270 kg Emission rate: 270 kg/day=3.125 g/s divided in: **U** : 675 kg/day **UO_2** : 675 kg/day **U_3O_8**: 0 kg/day	Total emission: 90 kg Emission rate: 90 kg/day=1.042 g/s divided in: **U** : 45 kg/day **UO_2** : 45 kg/day **U_3O_8**: 0 kg/day
2 3 4 5 6 (5 days)	Total emission: 60 kg Emission rate: 12 kg/day=0.139 g/s divided in: **U** : 6 kg/day **UO_2** : 6 kg/day **U_3O_8**: 0 kg/day	Total emission: 180 kg Emission rate: 36 kg/day=0.417 g/s divided in: **U** : 18 kg/day **UO_2** : 18 kg/day **U_3O_8**: 0 kg/day	Total emission: 270 kg Emission rate: 54 kg/day=0.625 g/s divided in: **U** : 21.6 kg/day **UO_2** : 21.6 kg/day **U_3O_8**: 0 kg/day	Total emission: 60 kg Emission rate: 18 kg/day=0.208 g/s divided in: **U** : 9 kg/day **UO_2** : 9 kg/day **U_3O_8**: 0 kg/day
7 8 9 10 (4 days)	Total emission: 80 kg Emission rate: 20 kg/day=0.232 g/s divided in: **U** : 0 kg/day **UO_2** : 0 kg/day **U_3O_8**: 20 kg/day	Total emission: 240 kg Emission rate: 60 kg/day=0.694 g/s divided in: **U** : 0 kg/day **UO_2** : 0 kg/day **U_3O_8**: 60 kg/day	Total emission: 360 kg Emission rate: 90 kg/day=1.042 g/s divided in: **U** : 0 kg/day **UO_2** : 0 kg/day **U_3O_8**: 90 kg/day	Total emission: 120 kg Emission rate: 30 kg/day=0.347 g/s divided in: **U** : 0 kg/day **UO_2** : 0 kg/day **U_3O_8**: 30 kg/day

3 MODIFICATIONS OF THE SNAP MODEL

The development of SNAP dispersion model at DNMI started as a cooperation with the UK Meteorological Office in 1991 and was based on the same theoretical assumptions as the UK dispersion model NAME (Maryon *et al.*, 1991). The first version of SNAP (Saltbones, *et al.*, 1995), had the aim of providing decision makers and government officials with a tool for real-time simulation of nuclear accidents. The first version used meteorological data available from LAM-50 Numerical Weather Prediction (NWP) model. The present version is using meteorological input data from DNMI's operational NWP model HIRLAM. The SNAP model is fully operational at DNMI at present, and the meteorologist on duty can start the model (on request) at any time - 24 hours a day.

In order to take into account atmospheric transport and deposition of large particles, the existing operational version of SNAP had to be modified. The main modification was the implementation of the gravitational settling process in the model equations. For small particles (~1μm), included in the operational version, the wet deposition process was more effective in removing them from the air than the dry deposition process. Dry deposition, in the case of small particles was caused by turbulent and Brownian diffusion. In case of large particles, gravitational settling is the most effective process responsible for removing these particles from the air. This process is so fast, that dry deposition of large particles dominates over wet deposition.

Introduction of gravitational settling into the model involved several important modifications/ changes. These modifications are described in more details below.

3.1 Gravitational settling velocity

When the Stokes law is valid, gravitational settling velocity with spherical shape of particles is a function of particle size, particle density and air density. (Zannetti, 1990):

$$v_g = \frac{d_p^2 \cdot g \cdot (\rho_p - \rho_a) \cdot C(d_p)}{18\eta} \qquad (1)$$

where:

v_g - gravitational settling velocity ($m\ s^{-1}$)

d_p - particle diameter (m)

$g = 9.81$ - acceleration of gravity ($m\ s^{-2}$)
ρ_p - density of particle ($g\ m^{-3}$)
$\rho_a = \rho_a(p, T)$ - density of the air ($g\ m^{-3}$)
$C(d_p)$ - Cunningham correction factor
$\eta = \eta(T)$ - dynamic molecular viscosity of the air ($kg\ m^{-1}\ s^{-1}$)

Density of the air ρ_a is calculated from the equation of state:

$$\rho_a = \frac{p}{R \cdot T} \tag{2}$$

where:
p - atmospheric pressure (hPa)
T - absolute air temperature (K)
$R = 287.04$ - gas constant for dry air ($J\ kg^{-1}\ K^{-1}$)

Viscosity of the air is a function of temperature (RAFF, 1999):

$$\eta = 1.72 \times 10^{-5} \cdot \frac{393}{T + 120} \cdot \left(\frac{T}{273}\right)^{\frac{3}{2}} \tag{3}$$

and Cunningham correction factor for small particles is calculated as (Zannetti, 1990; Seinfeld, 1986):

$$C(d_p) = 1 + \frac{2\lambda}{d_p} \cdot \left(1.257 + 0.4e^{-0.55 \cdot \frac{d_p}{\lambda}}\right) \tag{4}$$

where $\lambda = 6.53 \times 10^{-8}\ m$ is the mean free path of air molecules.

Equation (1) is not valid in the turbulent regime for particles larger then 20-30 μm. In this case, correction to account for high Reynolds numbers is necessary leading to the following equations (Seinfeld, 1986):

$$\begin{aligned} v_g\left(1 + \frac{3}{16}Re + \frac{9}{160}Re^2 \ln(2Re)\right) &= \frac{d_p^2 \cdot g \cdot (\rho_p - \rho_a) \cdot C(d_p)}{18\eta} && 0.1 < Re \le 2 \\ v_g(1 + 0.15Re^{0.678}) &= \frac{d_p^2 \cdot g \cdot (\rho_p - \rho_a) \cdot C(d_p)}{18\eta} && 2 < Re \le 500 \end{aligned} \tag{5}$$

where $Re = \frac{v_g d_p \rho_a}{\eta}$ is the Reynolds number.

Equation (5) is non-linear and requires numerical solution, usually iterative (e.g Baklanov and Sørensen, 2001), which may significantly slow down the model performance, if Equation (5) is applied to each individual particle. Therefore, in this study we have applied a simplified solution of Equation (5). Following description of the scenarios for the model run, we have taken into account only three particle density classes and four classes of particle size. We have further approximated, air pressure and air temperature functions by 50 discrete classes for each of these variables. In this way we ended up with a matrix of 30 000 discrete values of the gravitational settling velocities. Between these discrete values gravitational settling velocity is linearly interpolated. This approach works well, both for small and large particles and does not slow down the model performance significantly, because all the values in the matrix are calculated before the model run.

If we compare gravitational settling velocities calculated from the Equation (1) and those calculated from the Equation (5), all velocities calculated using Equation (5) are smaller. For the first two particle size classes (0.2 μm and 10 μm), the difference is negligible (below 0.1%). For particle diameter 50 μm, the differences are also small (below 5%). However, for large particles in class 200 μm, the differences are large; Equation (5) gives values 70-80% lower than Equation (1).

Comparison of gravitational settling velocities for the three classes of particle densities as a function of particle size is shown in Figure 6. In this comparison, we have assumed air temperature $20^{o}C$ and atmospheric pressure 1013 *hPa*. The difference between gravitational settling velocity calculated from the analytical Stokes Equation (1) and numerical solution of Equation (5) is getting larger with the particle size.

3.2 Source term parameterization

In the operational version of the SNAP model, the “particles” had a meaning as carriers of the pollutant load. Now, in the modified version of SNAP, we will give the “particles” a more physical meaning: - size, density, amount of radioactivity etc. However, they still can be interpreted as small, homogeneous air parcels carrying a large number of real particles of the same type. In the operational model version, particles were assumed to be well mixed in so called “mixing layer”. It implied that, after each model time step, they were randomly distributed between the ground and the top of the mixing height. Therefore, the source height, as well as the vertical distribution of emissions were not important for the model results, as long as the sources were located within the mixing layer.

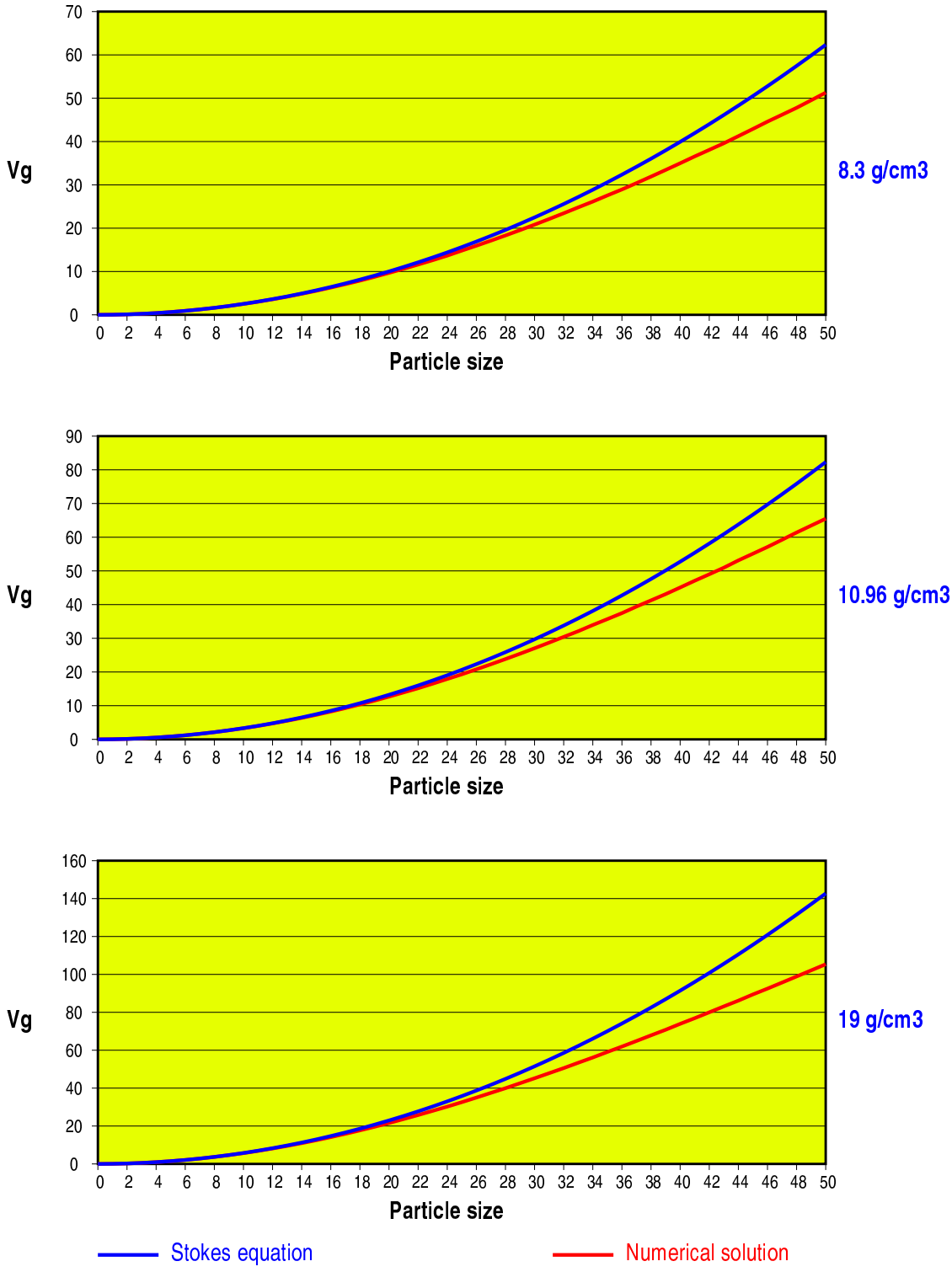


Figure 6. Comparison of gravitational settling velocities for three classes of particle densities as a function of particle size. Units for gravitational settling velocity: *cm* s^{-1}, for particle diameter: µ*m*.

Implementation of gravitational settling required changes in the parameterization of the mixing layer (described in the next section), and in the parameterization of emissions. In the modified model version, the vertical structure of the emissions play an important role. This vertical structure of the source is shown in Figure 7.

The location of the source is defined by geographical co-ordinates *(longitude, latitude)*, which are transformed to model Cartesian horizontal co-ordinates (x_l,y_l). Emissions are assumed to be uniformly distributed within the cylinder shown in Figure 7, with the radius r_e given in meters. The cylinder is limited from the top by the value of z_t and from the bottom by the value of z_b, both in meters. Emission rate for particles is given in $g\ s^{-1}$, and their properties can be defined, either by diameter and density, or by aerodynamic diameter with assumed unit density.

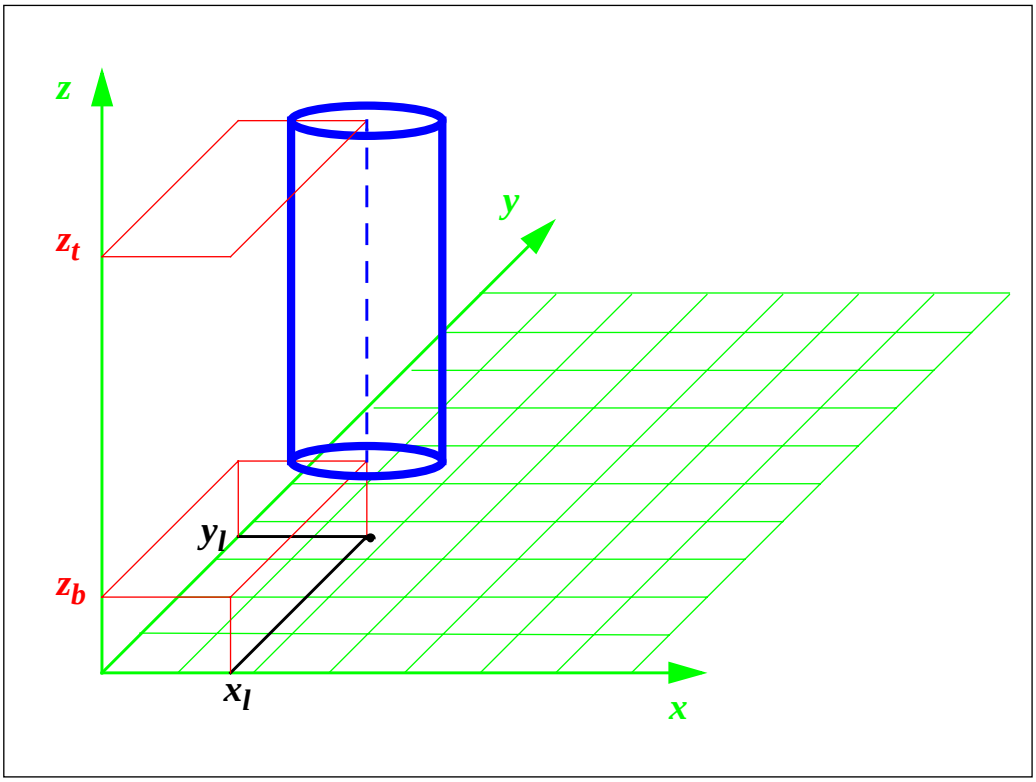


Figure 7. Illustration of the source shape parameterization in the modified version of the SNAP model.

Concerning time evolution, emission rate and all cylinder parameters can be a linear or a step function of time. In this study, emission rates and parameters of the cylinder were consistent with the source term description for Chernobyl Accident given in Chapter 2. They were kept constant for each of the three periods of release: day 1, days 2-6 and days 7-10. Obviously, the location of the source was kept constant during the release.

3.3 Vertical advection and diffusion

For simulating the Chernobyl Accident, a version of SNAP with the vertical σ coordinate has been used (Saltbones *et al.*, 1995). In this version, gravitational settling of particles was not taken into account, and in the mixing layer vertical diffusion and advection were parameterized as a completely random displacement of particles (with the length scale *l*) for each model time step.

In the modified version, both in the mixing layer and above, vertical advection and vertical diffusion are described by the same equation:

$$\sigma(t + \Delta t) = \sigma(t) + (\omega(t) + \omega_g(t)) \cdot \Delta t + r_\sigma \cdot l_\sigma \qquad (6)$$

where:

σ - model vertical coordinate

$\omega = \frac{D\sigma}{dt}$ - average vertical velocity in the model grid and over one model time step (s^{-1})

ω_g - average vertical velocity caused by gravitational settling (s^{-1})

$\Delta t = 900$ - model time step (s)

$r_\sigma \in [-0.5, 0.5]$ - random value

l_σ - length scale of the vertical turbulent transport during one model time step

Vertical model coordinate σ was defined as $\sigma \equiv \frac{p - p_T}{p_s - p_T}$

where:

p_T - pressure at the top of the model domain

p_S - surface pressure

Parameter l_σ has a different value in the mixing layer and above it:

$$l_\sigma = \begin{cases} 0.04 & \text{above the mixing layer} \\ 0.08 & \text{in the mixing layer} \end{cases}$$

Parameterization of the vertical diffusion in the modified SNAP version is simple, but it is consistent with the parameterizations of the other processes in the model, which make model performance very fast. This is the one of most important requirements for the operational applications of the model from the users side, mainly Norwegian Radiation Protection Agency.

3.4 Remarks about deep convection

One of the processes which may influence the range of atmospheric transport of large particles is the so called deep convection (Valkama and Pöllänen, 1996). This process is not directly taken into account in the operational version of the SNAP model, because we assumed that deep convection does not effect so much the long-range transport of gases and small particles/ aerosol. In case of large particles, deep convection can be important and we plan to introduce this process into the next version of the SNAP model.

4 RESULTS OF THE MODEL SIMULATIONS AND DISCUSSION

The modified version of the SNAP model, which included parameterization of gravitational settling velocity also for large Reynolds numbers, was run for 12 different classes of particles (three densities and four sizes). For each class of particles, there was a different source term, resulting in different atmospheric transport and deposition pattern. These runs were performed with the meteorological data from the NORLAM Weather Prediction Model (NWP).

For comparison, model runs with meteorological data from the HIRLAM NWP were also performed. The results were similar, but the range of the transport calculated with the HIRLAM data was slightly shorter, and especially some of the particles did not reach Norway at all in this case. This shows that model results are sensitive to meteorological input data.

Finally, model runs were performed with meteorological data from the NORLAM model, but with the emission sources located higher in the atmosphere than in the standard run. The results of this simulation are discussed in section 4.3

4.1 Dynamics of the transport

The results of both the operational and modified versions of the SNAP model are on-line displayed on the screen during the simulation in the form of the cloud of particles on the map of the model domain. This nice model feature gives a better insight into the dynamics of the atmospheric transport of radioactive debris than the static pictures are able to give.

An example of what can be seen on the screen during the model simulation is shown in Figure 8. In this figure, the locations of all classes of particles are shown (all heights) as seen from the above. It presents the first waves of particles released at day one, which reached Norway on the 1th of May, after approximately four days of transport. Red colour corresponds to particles affected by rain and black colour denotes dry particles. These particles travelled towards Finland and Sweden and approached Norway from the East.

Particles remaining in the air on the 7th May, approximately 11 days after the release started are shown in Figure 9. This shows the second wave, which reached Norway, coming from the South. It included different particles from the entire period of 10 days emission in Chernobyl.

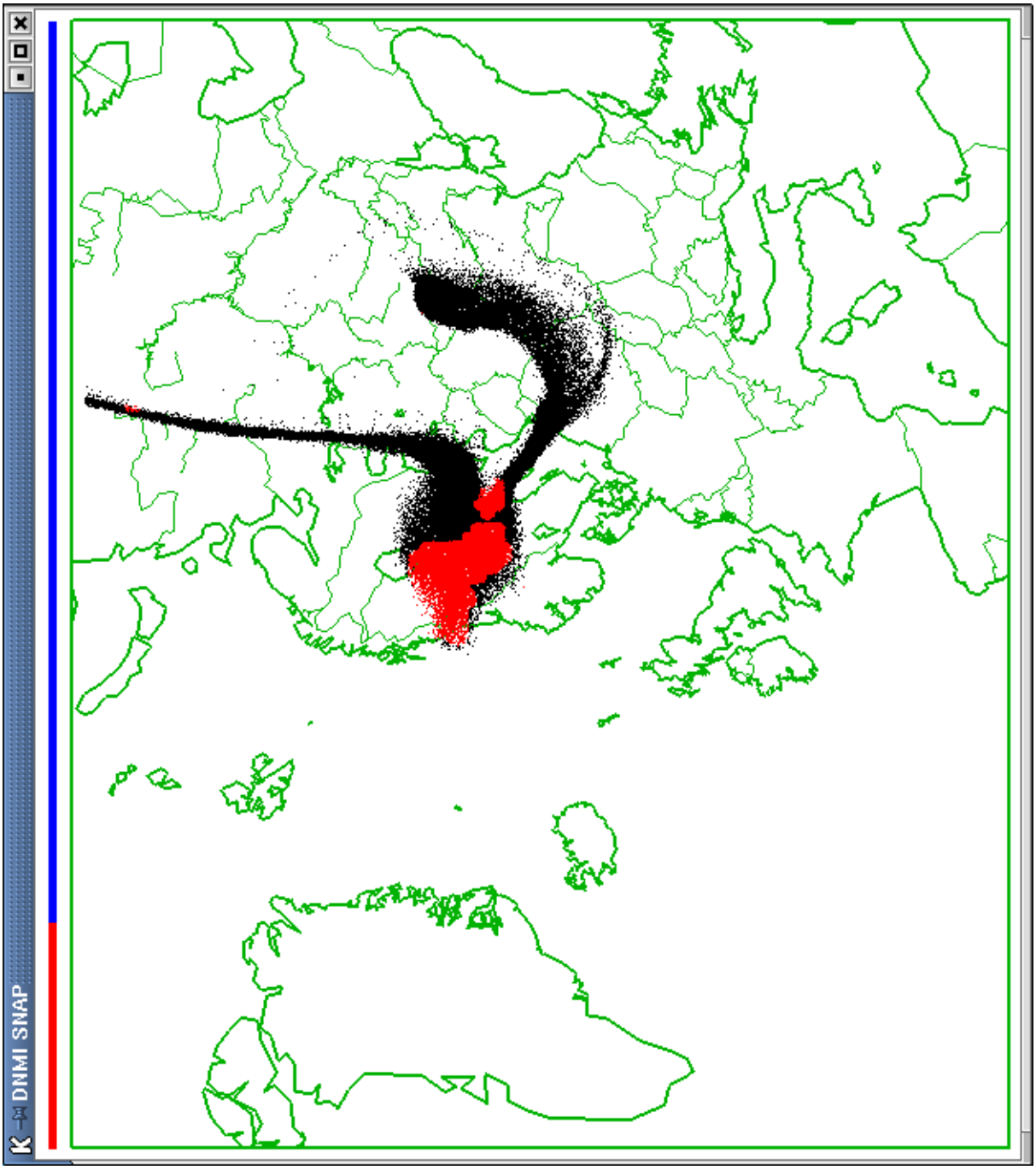


Figure 8. Results of the SNAP model simulation for the 1th of May 1986, 75 hours after the release start. All particles, including three density classes and four size classes, emitted from Chernobyl reactor and remaining in the air are shown in the figure. Black colour denotes dry particles and red colour denotes particles affected by rain.

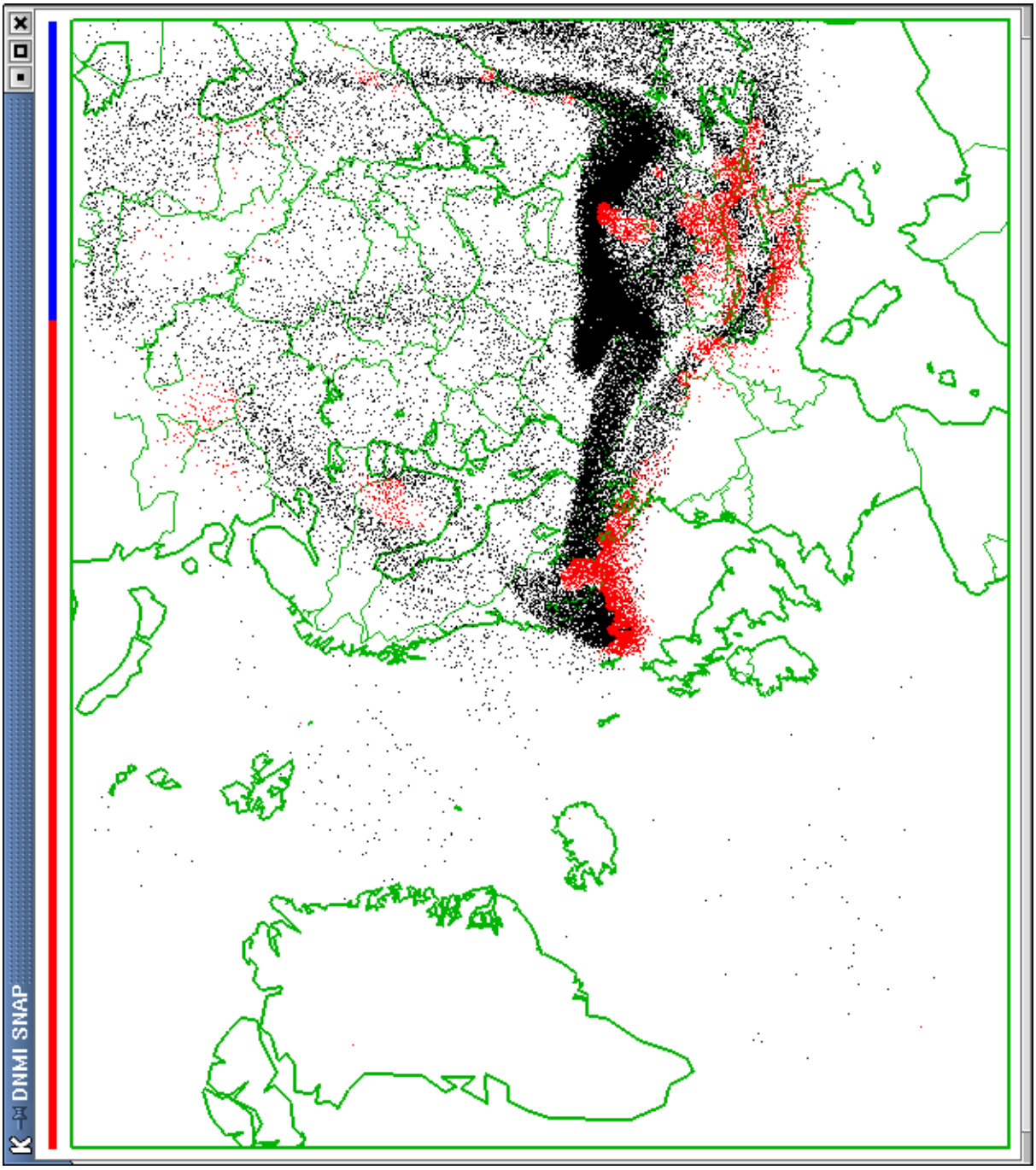


Figure 9. Results of the SNAP model simulation for the 7th of May 1986, 272 hours after the release start. All particles, including three density classes and four size classes, emitted from Chernobyl reactor and remaining in the air are shown in the figure. Black colour denotes dry particles and red colour denotes particles affected by rain.

In both cases shown in Figures 8 and 9, not only small particles (0.2 μm) but also larger particles (10 μm) are coming to Norway. This fact is in agreement with observations made by NLH. Spatial distribution of radioactive particles computed with the modified version of the SNAP model is different from the distribution of aerosols computed with the operational version.

4.2 Results for different particle classes

The modified SNAP model was run for each of 12 particle classes. The maps of accumulated total (wet + dry) deposition for each of two size classes, 0.2 μm and 10 μm, and for all three density classes are shown in Figures 10 to 15. As an example, accumulated total deposition for particles of the size 50 μm and density 10.96 $g\ cm^{-3}$ is shown in Figure 16.

There is a significant difference in deposition pattern for particle size 10 μm and below and for particle size 50 μm and above. Smaller particles are transported for long distances (more than 100 *km*), whereas larger particles are deposited relatively close to the source (less than 100 *km*). This is the reason why we are showing only one example of accumulated deposition map for the simulation of larger particles. It also shows that in order to simulate properly the transport of these larger particle classes, we need a better spatial resolution of the input meteorological data for the dispersion model.

There is a relatively small difference in the transport range for the three density classes of the smallest particles (0.2 μm), but this difference is quite important for the distant countries and Norway in particular. All three density types of the smallest particles reached Norway, but from different directions and in different periods. Two types of particles, U and UO_2 with densities 19 $g\ m^{-3}$ and 10.96 $g\ m^{-3}$, respectively, came to Norway mainly from the East at the beginning of the simulation period (emissions from the first day). Third type, U_3O_8 particles with density 8.3 $g\ m^{-3}$ reached Norway from the South, and at the end of the simulation (emissions from days 7-10). Transport and deposition of these particles is not much different in the operational and modified version of the model, because gravitational settling velocity in this case is small and it is not very important for the effectiveness of the dry deposition process. In addition, for such small particles, dry deposition is definitely less effective than wet deposition.

For particles of the size 10 μm, differences between density classes (and related emission periods) are even more pronounced than for smallest particles. In this case two types of particles, U_3O_8 and UO_2 with densities 8.3 $g\ m^{-3}$ and 10.96 $g\ m^{-3}$, respectively, also came to Norway, form different directions and in different periods. The third class, the heaviest U particles with density 19 $g\ m^{-3}$, did not reach Norway at all.

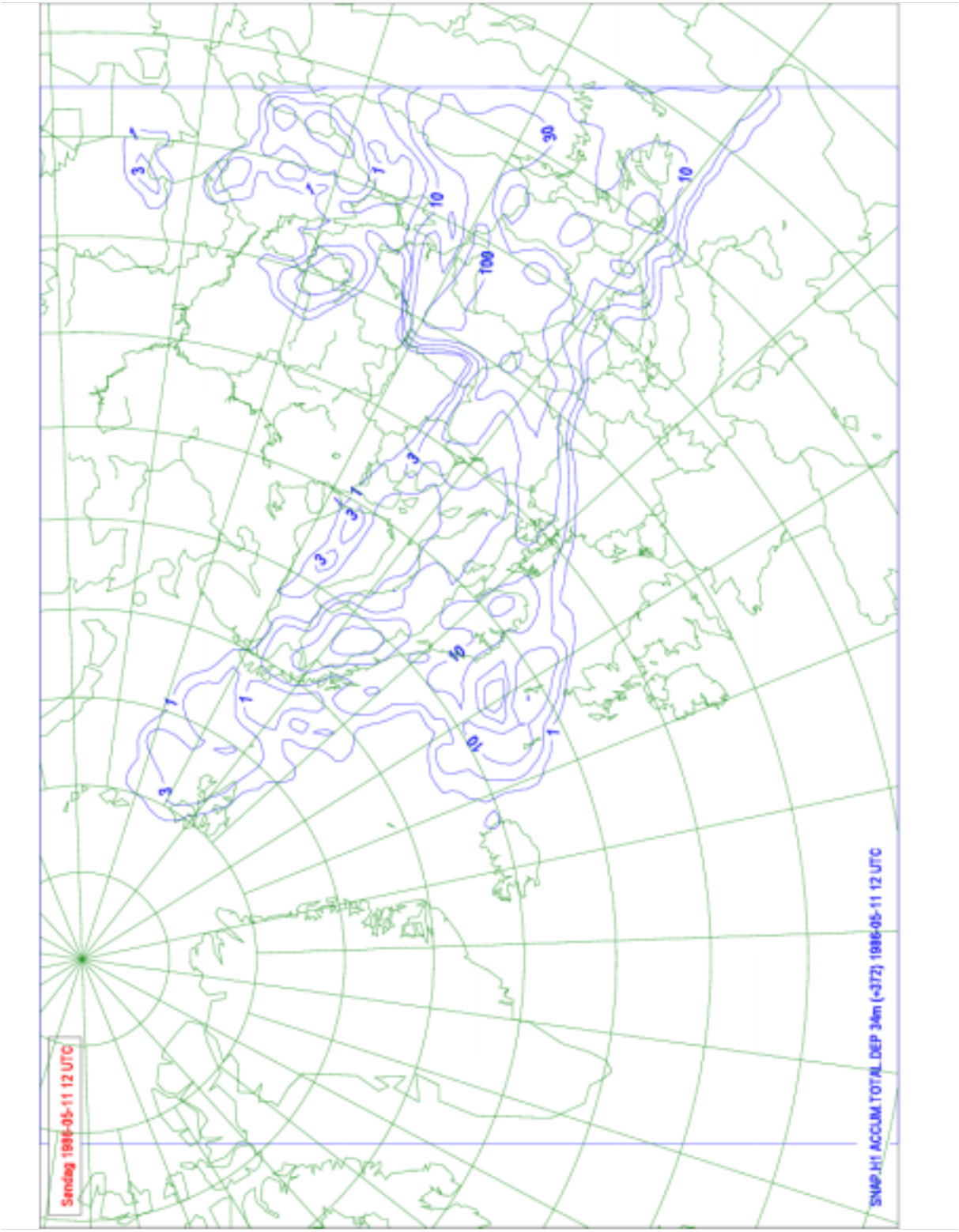


Figure 10. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 0.2 μ*m* and density 8.3 *g* cm^{-3} are shown for the entire period of the simulation. Units: *ng* m^{-2}.

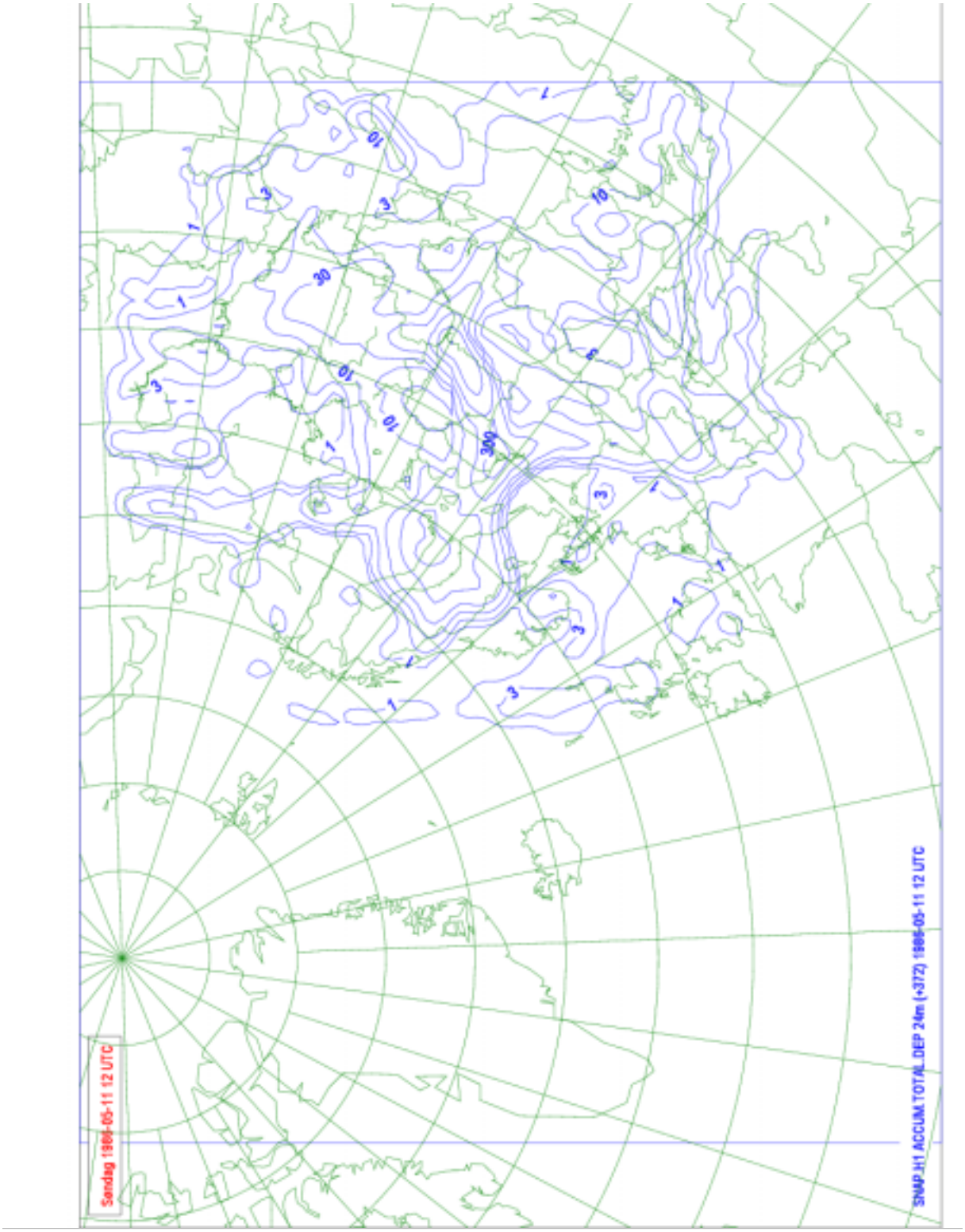


Figure 11. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 0.2 μ*m* and density 11 *g* cm^{-3} are shown for the entire period of the simulation. Units: *ng* m^{-2}.

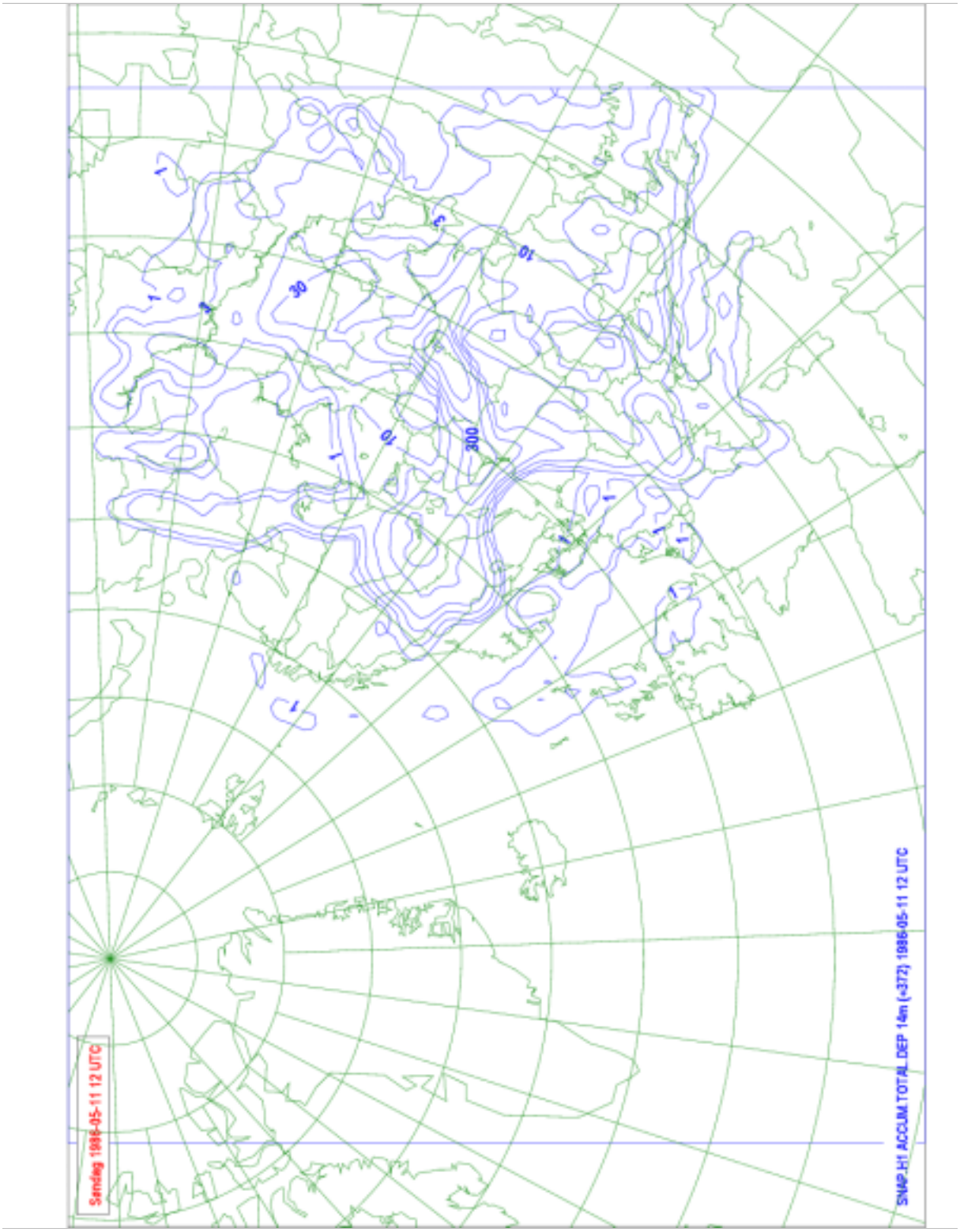


Figure 12. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 0.2 μ*m* and density 19 $g\ cm^{-3}$ are shown for the entire period of the simulation. Units: $ng\ m^{-2}$.

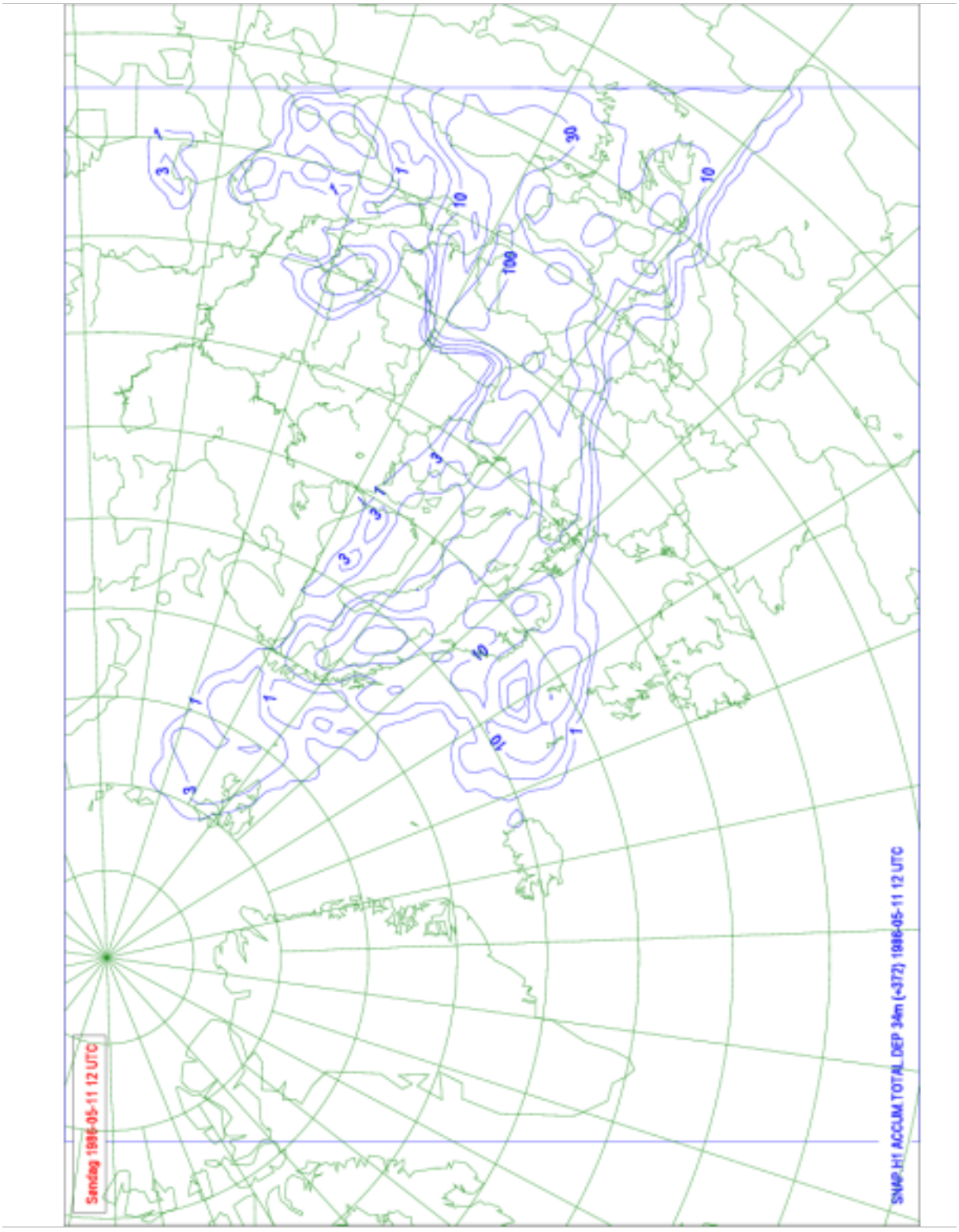


Figure 13. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 10 µ*m* and density 8.3 *g* cm^{-3} are shown for the entire period of the simulation. Units: *ng* m^{-2}.

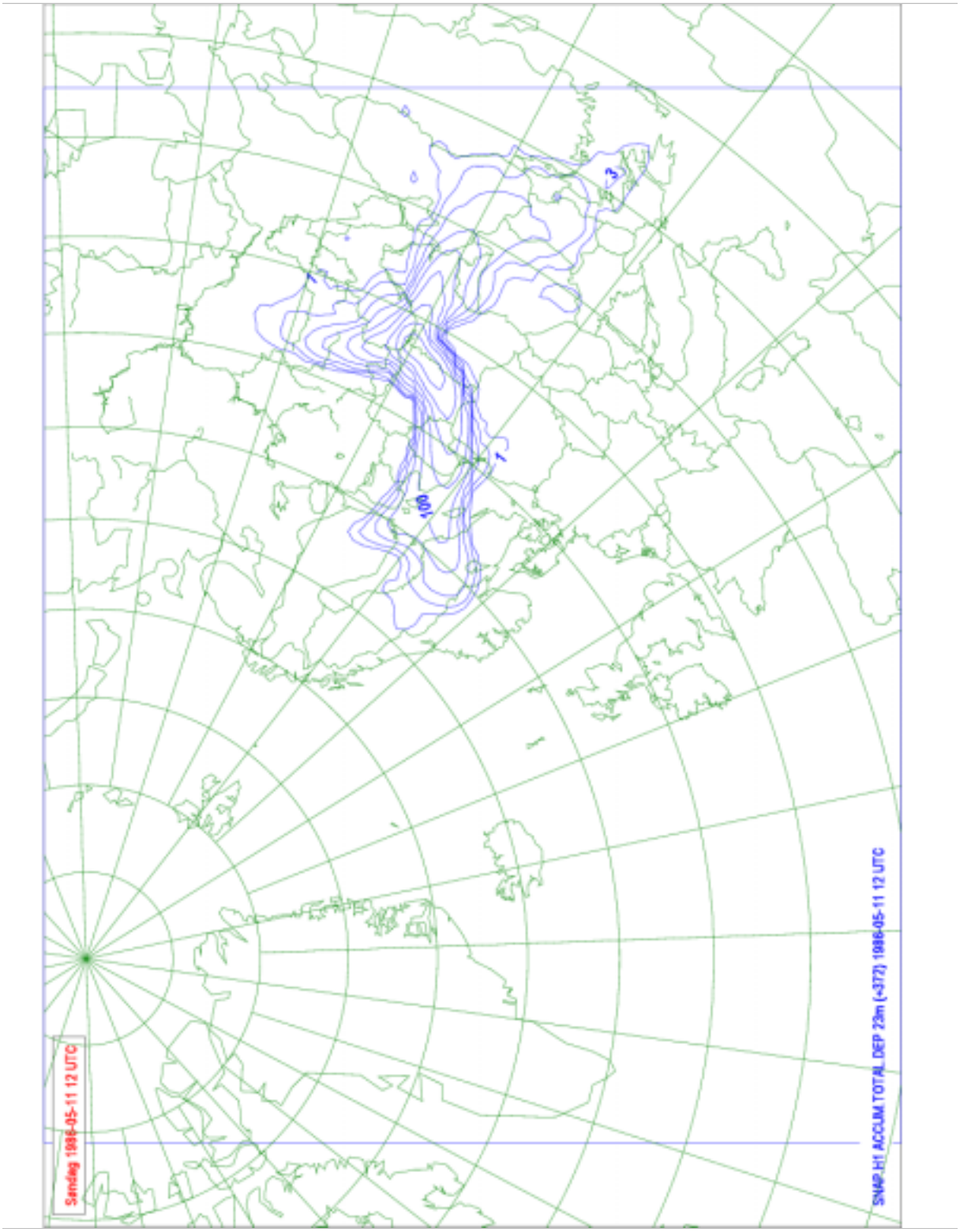


Figure 14. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 10 µ*m* and density 11 $g\ cm^{-3}$ are shown for the entire period of the simulation. Units: $ng\ m^{-2}$.

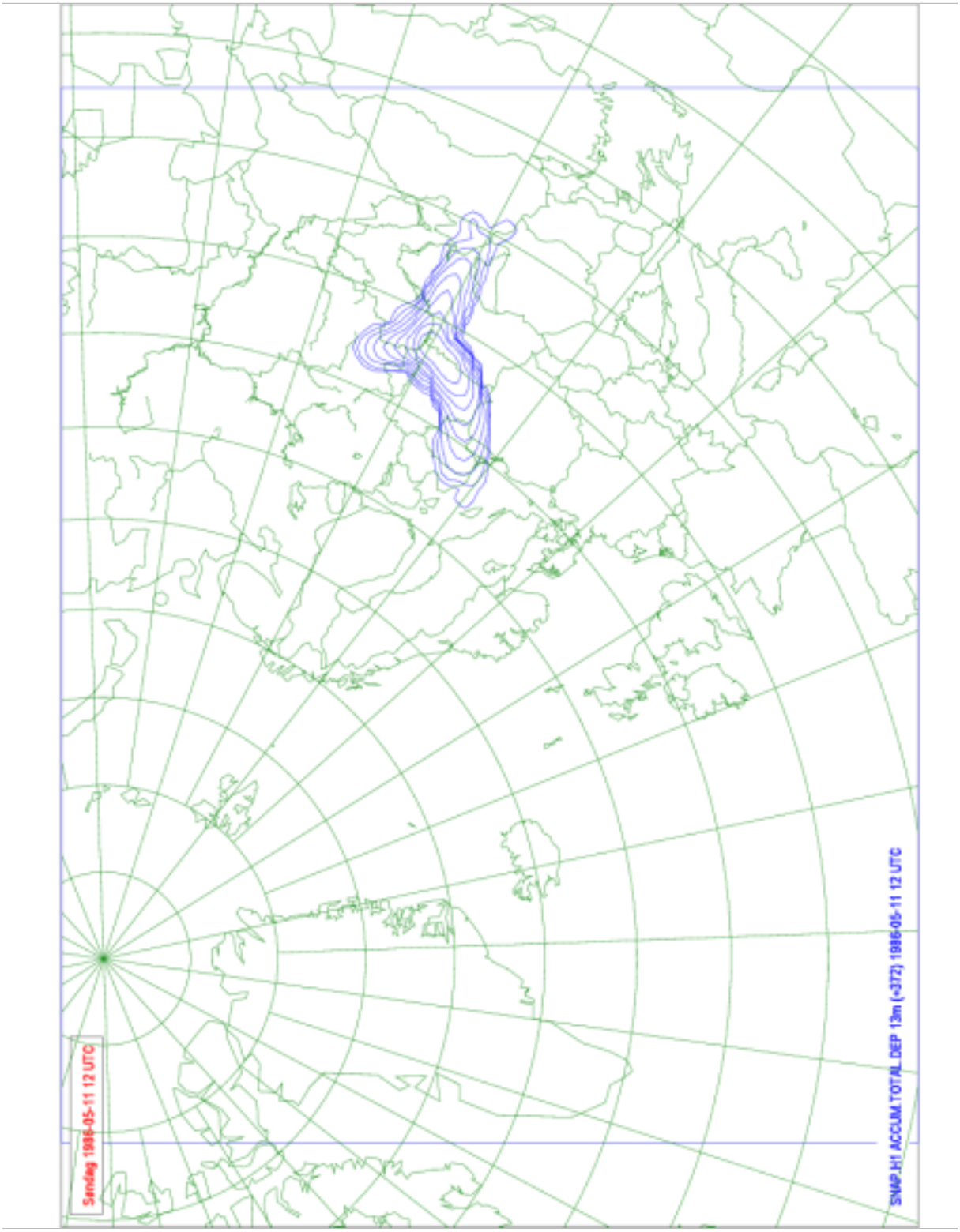


Figure 15. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 10 μ*m* and density 19 $g\ cm^{-3}$ are shown for the entire period of the simulation. Units: $ng\ m^{-2}$.

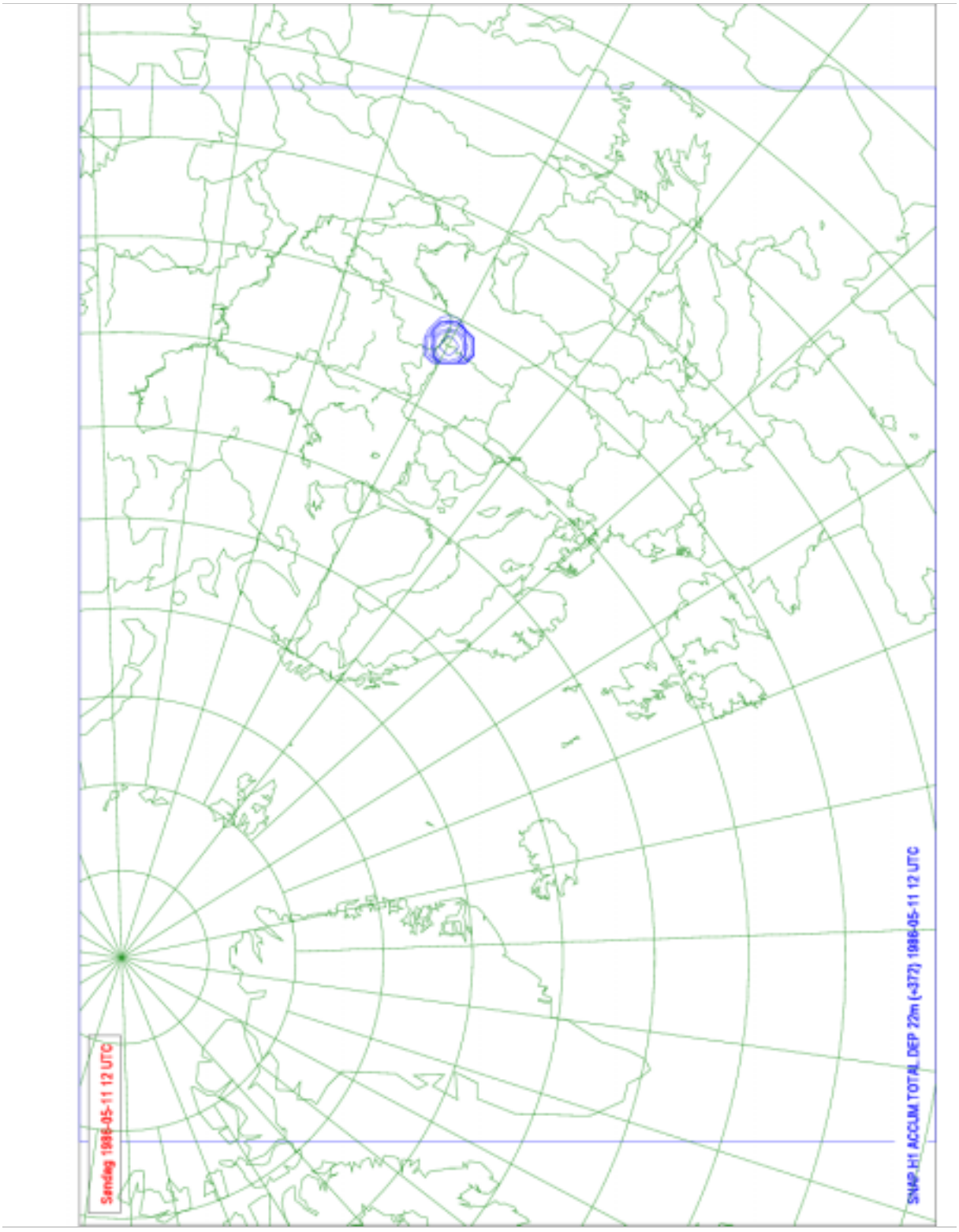


Figure 16. Results of the SNAP model simulation for the 11th of May 1986, 372 hours after the release start. Isolines of accumulated total (wet + dry) deposition for particle size 50 μ*m* and density 11 $g\ cm^{-3}$ are shown for the entire period of the simulation. Units: $ng\ m^{-2}$.

For all three density classes, the deposition patterns are different, because particles of this size (10 μm) have larger gravitational settling velocity than those for the less dense classes. In this size class gravitational settling velocity dominates dry deposition and makes the transport sensitive to wind directions on different levels. Particle with 10 μm diameter are not taken into account in the operational version of the SNAP model. The present results indicate that these particles can travel a long distance, longer than expected, and have to be included in the next version of the model.

The deposition pattern shown in Figure 16, for large particles of the size 50 μm and density 10.96 $g\ m^{-3}$, is very typical for all particles of the size 50 μm and 200 μm, indicating that transport and deposition of such particles are mainly a local scale problem because of the large gravitational settling velocities. However, in this case we have to be careful with definitive conclusions because the SNAP model was not designed to deal with such a small scale dispersion. Since there is a growing demand for local scale simulation, combined with the long range transport, we plan to modify our model also in this direction in the near future.

Comparison of Figures 10 - 15 with Figure 16 shows a dramatic difference in the deposition pattern between particles with 10 μm diameter and particles with 50 μm diameter. Several questions remain to be answered in connection with this difference. For example, where exactly (between 10 and 50 μm) is the border between long range and a local scale transport? Another question is: how important and efficient is the effect of deep convection, which certainly prolongs the transport of particles.

The answers to these questions, in our case, require further modification of the model and more systematic experiments with the setup of scenarios. We were not able to perform such experiments in the frame of the present project, because of the limited time and resources available. The main focus of the present study was on the implementation of gravitational settling velocity for particles. In the next phase, when the tool (modified model) is ready, we will be able to perform more experiments and hopefully answer the above questions.

4.3 Sensitivity to source height

As mentioned before, we have run the model for the same classes of particles and with the same source term specification, except for the height of release, which was between 2000 m and 3000 m for all 10 days with emissions from the Chernobyl reactor. The results of this model run were compared with the results of the standard model run, and are shown in Figure 17 - for small and light particles (0.2 μm and 8.3 $g\ m^{-3}$), and in Figure 18 for larger and heavier particles (10 μm and 10.96 $g\ m^{-3}$).

Small particles are not so much sensitive to the source height, especially far away from the source, because gravitational settling velocity is not an effective mechanism for dry deposition in this case. As we can see in Figure 17, the deposition pattern for two runs is quite similar, except for a small area in the Eastern part of the model domain.

The situation is different for the particles with 10 μm size (Figure 18). In this case, gravitational settling velocity dominates dry deposition, and depending on the wind strength and direction, results for low and high emission sources can be significantly different. This comparison confirms the need for gravitational settling velocity and larger particles in the simulations of nuclear accidents.

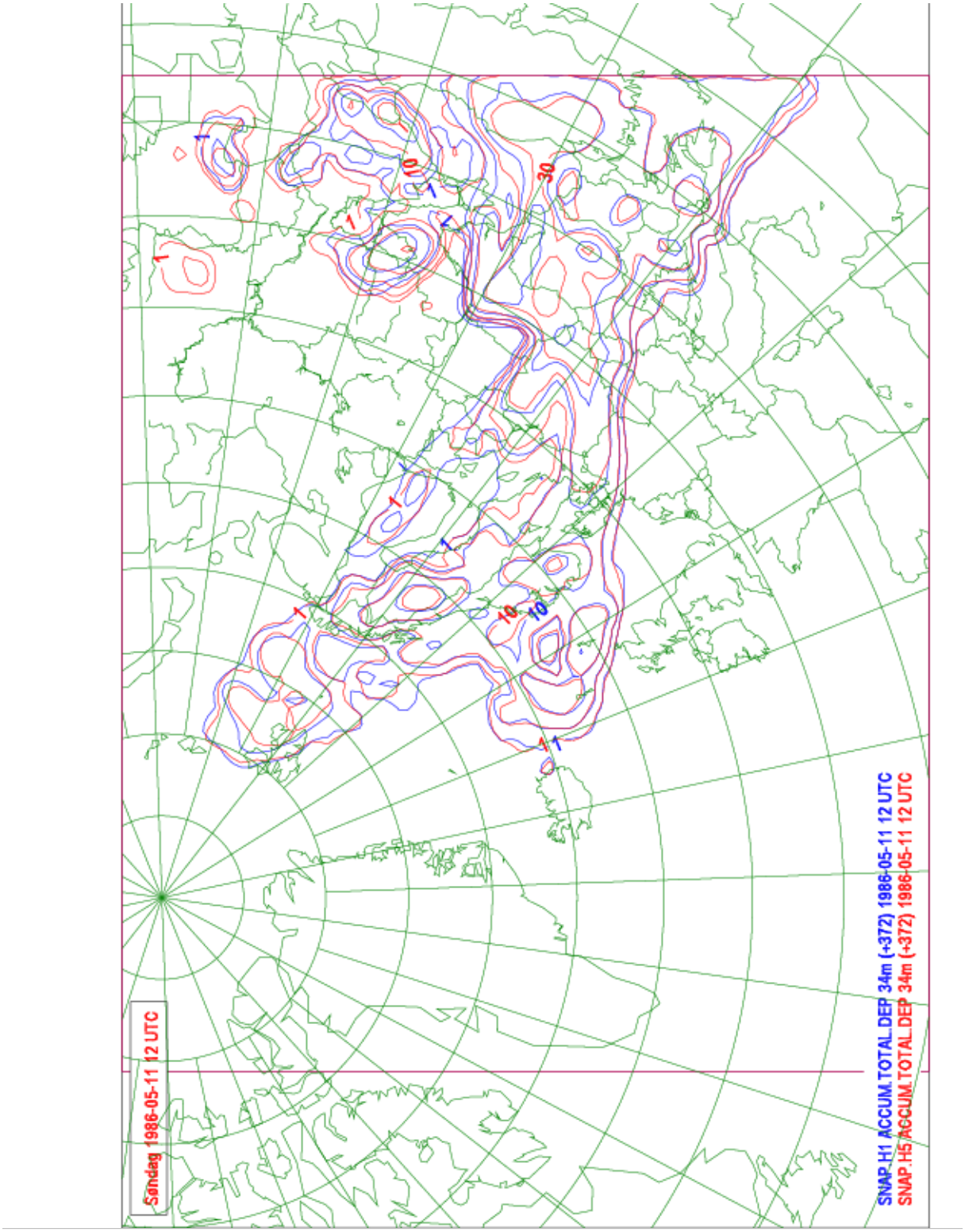


Figure 17. Comparison of the SNAP model simulations for the 11th of May 1986, 372 hours after the release start. Isolines of computed accumulated total (wet + dry) deposition for particle size 0.2 μ*m* and density 8.3 $g\ cm^{-3}$ are shown for the entire period of the simulation. Blue isolines: results of the standard model run. Red isolines: results of the model run with release height in the range 2000 *m* and 3000 *m* for the entire period of emissions - 10 days. Units: $ng\ m^{-2}$.

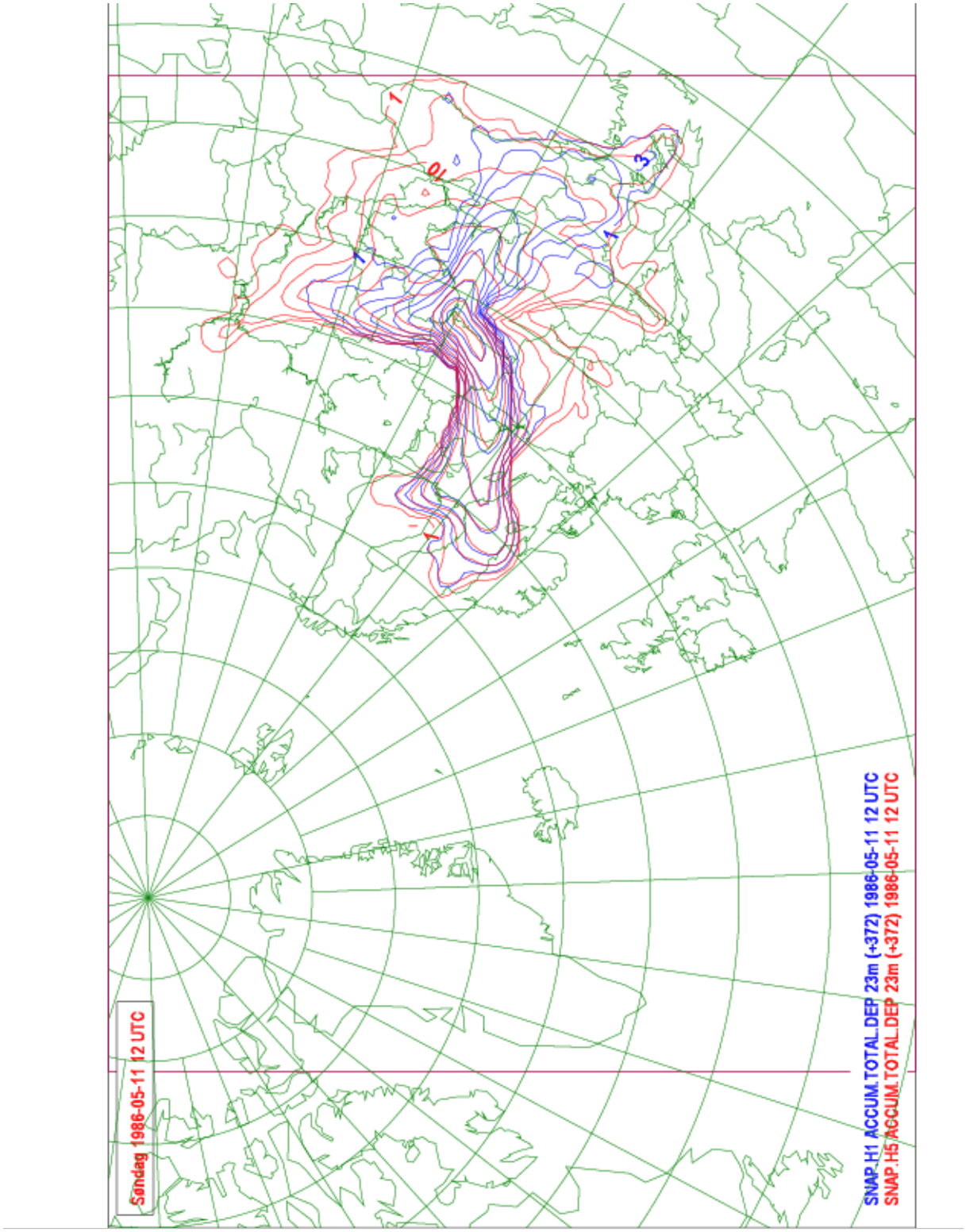


Figure 18. Comparison of the SNAP model simulations for the 11th of May 1986, 372 hours after the release start. Isolines of computed accumulated total (wet + dry) deposition for particle size 10 μm and density 11 $g\ cm^{-3}$ are shown for the entire period of the simulation. Blue isolines: results of the standard model run. Red isolines: results of the model run with release height in the range 2000 *m* and 3000 *m* for the entire period of emissions - 10 days. Units: $ng\ m^{-2}$.

5 CONCLUSIONS AND RECOMMENDATIONS

Our joint (DNMI + NLH) project produced several interesting results, probably more than expected. On one hand, meteorologist from DNMI modelling atmospheric transport have received innovative and unique description of the source term for Chernobyl accident based on detailed observations, which is quite different from the typical data used by most of the modellers. On the other hand, chemists from NLH who can see the real particles trough their microscopes, need information on transport and deposition pattern of particles to assess the environmental impact.

Our joint experiments and analysis of the results led to several interesting conclusions and recommendations for future research, which are listed below:

- In most of the transport models, only small particles/aerosol have been taken into account in simulation of nuclear accidents. This approach has to be changed, because both observations and model experiments presented in this study show that the large (10 μm) particles can travel long distances (more than 1000 *km*) before being deposited.

- Typical assumption in modelling atmospheric transport and deposition of radionuclides states that different elements are transported as individual particles. In light of the information given in this report, in most of the cases actual particles are complicated agglomerates of different radionuclides. This is a very important fact, which has to be taken into account by all modellers.

- The present version of the modified SNAP model is able to simulate atmospheric transport of 10 μm particles released during the Chernobyl accident to Norway (where such small particles actually have been observed). This means that the model is able to reproduce the measurements, because such particles were observed by scientists from NLH in Norway after the Chernobyl Accident.

- The present results show that deposition distribution of 10 μm particles computed with modified version of the SNAP model is different from the deposition distribution of aerosols computed with operational version.

- Some of the pictures presented in this report, show clearly that not all particles are spherical. If they are not, their gravitational settling velocity can be reduced and the range of their

atmospheric transport extended, in some cases significantly. Transport and deposition of such non-spherical particles should be a subject of the future studies.

- It still remains unclear where is the border (concerning particle size) between long range and local scale transport and deposition of particles. Answer to this question can only be given after more systematic model simulations with different particle size classes. The answer can be significantly influenced by the implementation of the deep convection process into the model, which we recommend here.

- It is essential to establish a possible relationship between particle source terms, deposition of particles and associated ecological impact.

- Conclusions, questions and suggestions presented above lead to one general recommendation, which is an extension of the joint project between DNMI and NLH in the future, with partners from other Nordic countries. In the present situation, source terms not only for nuclear power plants such as Chernobyl reactor or Kola reactors, but also source terms for nuclear explosions, together with model tests can be of great interest not only for scientists, but also for decision makers responsible for managing severe emergencies.

6 REFERENCES

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