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Nuclear fuel particles in the environment – characteristics, atmospheric transport and skin doses

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SUMMARY

In the present thesis, nuclear fuel particles are studied from the perspective of their characteristics, atmospheric transport and possible skin doses. These particles, often referred to as 'hot' particles, can be released into the environment, as has happened in past years, through human activities, incidents and accidents, such as the Chernobyl nuclear power plant accident in 1986. Nuclear fuel particles with a diameter of tens of micrometers, referred to here as large particles, may be hundreds of kilobecquerels in activity and even an individual particle may present a quantifiable health hazard.

The detection of individual nuclear fuel particles in the environment, their isolation for subsequent analysis and their characterisation are complicated and require well-designed sampling and tailored analytical methods. In the present study, the need to develop particle analysis methods is highlighted. It is shown that complementary analytical techniques are necessary for proper characterisation of the particles. Methods routinely used for homogeneous samples may produce erroneous results if they are carelessly applied to radioactive particles.

Large nuclear fuel particles are transported differently in the atmosphere compared with small particles or gaseous species. Thus, the trajectories of gaseous species are not necessarily appropriate for calculating the areas that may receive large particle fallout. A simplified model and a more advanced model based on the data on real weather conditions were applied in the case of the Chernobyl accident to calculate the transport of the particles of different sizes. The models were appropriate in characterising general transport

properties but were not able to properly predict the transport of the particles with an aerodynamic diameter of tens of micrometers, detected at distances of hundreds of kilometres from the source, using only the current knowledge of the source term. Either the effective release height has been higher than reported previously or convective updraft may have influenced the transport. Models applicable to large particle dispersion in a turbulent atmosphere should be further developed.

The health threat from large nuclear fuel particles differs from that of uniform contamination. In contact with human tissue such as skin, a highly active beta-emitting particle may cause a large but localised dose to the tissue, whereas at distances of more than about one centimetre from the source the dose is negligible. Large particles are poorly inhalable because of their size. They may be deposited in the upper airways but are not easily transported deep into the lungs. Instead, deposition onto the surface of skin is of more relevance with respect to acute deterministic health effects. In the present work, skin doses are calculated for particles of different sizes and different types by assuming the particles are deposited on the body surface. The deposition probability as a function of the number concentration of the particles in air is not estimated.

The doses are calculated at the nominal depth of the basal cell layer and averaged over a square centimetre of the skin. Calculated doses are compared with the annual skin dose limit for the public (50 mGy at a depth of 0.07 mm and averaged over 1 cm^2). After the Chernobyl accident the most active nuclear fuel particles detected in Europe, hundreds of kilometres from the source, would have been able to produce a skin dose exceeding this limit within one hour when deposited onto skin. However, the appearance of deterministic effects necessitates skin contact lasting more than one day.

The health hazards of nuclear fuel particles must be taken into account in estimating the consequences of a severe nuclear accident and planning countermeasures to protect the rescue workers and the general public.

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Avainsanat Ydinpolttoainehiukkanen, kuuma hiukkanen, radionuklidi, kulkeutuminen ilmakehässä, aktiivisuus, ihoannos.

YHTEENVETO

Väitöskirjassa tutkitaan ydinpolttoainehiukkasia - niiden ominaisuuksia, kulkeutumista ilmakehässä ja mahdollisia ihoannoksia. Näitä ns. kuumia hiukkasia voi vapautua ympäristöön ihmisten toiminnan sekä erilaisten tapahtumien ja onnettomuuksien seurauksena kuten esimerkiksi Tshernobylin onnettomuudessa vuonna 1986. Suuret, halkaisijaltaan kymmenien mikrometrien ydinpolttoainehiukkaset, voivat olla aktiivisuudeltaan satoja kilobecquerellejä ja jopa yksittäiset hiukkaset voivat aiheuttaa välittömiä säteilyhaittoja.

Yksittäisten ydinpolttoainehiukkasten havainnointi ympäristöstä, hiukkasten eristäminen analyysejä varten sekä hiukkasten ominaisuuksien luonnehdinta eivät ole suoraviivaisia, vaan edellyttävät hyvin suunniteltua näytteenottoa ja räätälöityjä analyysimenetelmiä. Tutkimuksessa tuodaan esille menetelmäkehityksen tarve sekä osoitetaan, että toisiaan täydentävien analyysimenetelmien käyttö on välttämätöntä radioaktiivisten hiukkasten ominaisuuksien kartoittamiseksi. Homogeenisille näytteille tarkoitetut menetelmät rutiininomaisesti sovellettuna voivat tuottaa virheellisiä tuloksia ja voivat siten vaikuttaa vääristävästi hiukkasten aiheuttaman terveyshaitan arviointiin.

Tutkimuksessa osoitetaan, että suuret ydinpolttoainehiukkaset kulkeutuvat ilmakehässä toisin kuin pienet hiukkaset ja kaasumaiset aineet. Kaasumaisten aineiden trajektorit eivät siten välttämättä ole riittäviä arvioitaessa onnettomuudessa vapautuvien radioaktiivisten aineiden kulkeutumista ja aluetta, jonne kuumat hiukkaset voivat kantautua. Sekä yksinkertaistettua että kehittynyttä, todellisiin säätietoihin perustuvaa

laskentamallia sovellettiin erikokoisten hiukkasten kulkeutumiseen Tshernobylin onnettomuudesta. Mallit kykenivät varsin hyvin ennustamaan suurten hiukkasten kulkeutumista, mutta aerodynaamiselta halkaisijaltaan jopa kymmenien mikrometrien hiukkasten löytymistä useiden satojen kilometrien etäisyydeltä päästölähteestä ei pystytty kunnollisesti selittämään olemassaolevan tiedon perusteella. Joko vapautumiskorkeus on ollut aiemmin raportoitua suurempi tai konvektiiviset pystysuuntaiset virtaukset ovat vaikuttaneet kulkeutumiseen. Leviämismalleissa todettiin olevan kehittämisen tarvetta erityisesti turbulenttisen dispersion ja suurten hiukkasten kulkeutumisen osalta.

Ydinpolttoainehiukkasten aiheuttama uhka poikkeaa tasaisen kontaminaation tapauksesta. Kudoksen kanssa kosketuksessa oleva beetasäteilyä lähettävä hiukkanen voi aiheuttaa suuren mutta erittäin paikallisen säteilyannoksen, kun taas kauempana, noin yli senttimetrin päässä, annos on vähäinen. Kokonsa takia suuret ydinpolttoainehiukkaset eivät hengitysilman mukana kulkeudu syvälle keuhkoihin, vaan jäävät esimerkiksi ylähengitystiehyeisiin. Akuutteja säteilyseurauksia tarkasteltaessa hiukkasten depositio ihoon ja tätä kautta saatava säteilyannos onkin varteenotettava altistusreitti. Työssä laskettiin mahdollisia ihoannoksia erityyppisille ja erikokoisille ydinpolttoainehiukkasille olettaen hiukkasten olevan kiinnittyneenä ihon pintaan. Deposition todennäköisyyttä hiukkasten lukumääräpitoisuuden funktiona ei arvioitu.

Ihoannokset laskettiin tyvisolukerroksen nimellissyvyydelle ja keskiarvoistettuna neliösenttimetrin pinta-alalle. Laskennallisten annosten suuruutta verrattiin väestön ihoannosrajaan (50 mGy vuodessa ihon neliösenttimetriä kohden syvyydelle 0,07 mm). Tshernobylin onnettomuuden jälkeen aktiivisimmat Euroopasta, satojen kilometrien etäisyydeltä päästöpaikasta löydetyt ydinpolttoainehiukkaset olisivat voineet jopa alle tunnissa aiheuttaa tämän rajan ylityksen deponoiduttuaan ihon pintaan. Deterministisen terveyshaittojen ilmaantumiseen olisi kuitenkin vaadittu kuitenkin yli vuorokauden kestävä ihokontakti.

Ydinpolttoainehiukkasten aiheuttama terveysuhka täytyy ottaa huomioon arvioitaessa mahdollisen vakavan ydinonnettomuuden säteilyseurauksia ja suunniteltaessa suojelutoimenpiteitä pelastustyöntekijöille ja väestölle.

CONTENTS

ORIGINAL PUBLICATIONS

The present thesis is based on the following original publications, which are referred to in the text as roman numerals I – VIII. In addition, the results of the licentiate thesis (Pöllänen 1997) in particular and other publications of the disputant mentioned in the reference list are also taken into account.

I Toivonen H, Pöllänen R, Leppänen A, Klemola S, Lahtinen J, Servomaa K, Savolainen A L, Valkama I. A nuclear incident at a power plant in Sosnovyy Bor, Russia. Health Physics 1992; 63: 571 - 573.

II Pöllänen R, Toivonen H. Skin doses from large uranium fuel particles - application to the Chernobyl accident. Radiation Protection Dosimetry 1994; 54: 127 - 132.

III Pöllänen R, Toivonen H. Transport of large uranium fuel particles released from a nuclear power plant in a severe accident. Journal of Radiological Protection 1994; 14: 55 - 65.

IV Pöllänen R, Toivonen H. Skin dose calculations for uranium fuel particles below 500 μm in diameter. Health Physics 1995; 68: 401 - 405.

V Pöllänen R, Valkama I, Toivonen H. Transport of radioactive particles from the Chernobyl accident. Atmospheric Environment 1997; 31: 3575-3590.

VI Pöllänen R. Highly radioactive ruthenium particles released from the Chernobyl accident: particle characteristics and radiological hazard. Radiation Protection Dosimetry 1997; 71: 23 - 32.

VII Pöllänen R, Ikäheimonen T K, Klemola S, Juhanoja J. Identification and analysis of a radioactive particle in a marine sediment sample. Journal of Environmental Radioactivity 1999; 45: 149 - 160.

VIII Pöllänen R, Klemola S, Ikäheimonen T K, Rissanen K, Juhanoja J, Paavolainen S, Likonen J. Analysis of radioactive particles from the Kola Bay area. Analyst 2001; 126: 724 - 730.

This work was conducted in the Laboratory for Airborne Radioactivity at STUK - Radiation and Nuclear Safety Authority in 1992 – 2001. The disputant is the sole author of publication VI and, except for publication I, the leading writer of other publications. In publication I, the disputant performed nuclide ratio calculations and contributed the preparation of the paper.

The disputant designed publications II, III and IV together with the co-author, and was responsible for the calculations and the overall preparation of the papers.

Intense co-operation with Finnish Meteorological Institute (FMI) was a prerequisite for achieving the results of the atmospheric transport calculations in publication V. The disputant designed the research, performed dose calculations, made the review of particles from the Chernobyl accident, and interpreted the results. FMI performed atmospheric transport calculations.

Close collaboration with other co-authors of publications VII and VIII was necessary in characterising the properties of radioactive particles. The disputant designed the research and prepared the papers as well as performed the identification and isolation of the particles. The disputant took the need and use of other analytical methods under advisement and interpreted the results. A notable part of experimental particle characterisation was performed by co-authors.

1 INTRODUCTION

Radioactive particles considered in the present thesis are tiny pieces of artificial radioactive material, mainly nuclear fuel particles, dispersed into the environment because of different human activities. Radioactive particles are considered here as aggregates of radioactive atoms that give rise to inhomogeneous distribution of radionuclides significantly different from that of the natural background particles of the sample. Nuclear fuel particles are those originating from nuclear fuel. Although particle size may vary considerably mainly large fuel fragments, diameter from a few micrometers up to tens of micrometers and often referred to as 'hot particles', are of special concern here. Radioactive particles smaller than about 1 μm in diameter and generated by various nucleation/condensation processes are not treated.

There exists no widely accepted definition for the concept of a 'hot particle'. It is often used in the meaning that the particle is highly active; sometimes it is used for particles having high specific activity. Khitrov *et al*. (1994) have suggested the following definition: a hot particle is a particle with any radionuclide or composition with size up to 50 - 80 μm and activity over 4 Bq. The National Council on Radiation Protection and Measurements (NCRP 1999) states that "hot particles are considered to be > 10 μm but < 3000 μm in any dimension. Hot particles smaller than 10 μm may be treated as general contamination…". Radioactive particles originating from atmospheric nuclear tests are historically referred to as hot particles. This concept was later attributed to fuel fragments originating from the Chernobyl accident.

Hot particles have been frequently identified after past nuclear incidents as shown e.g. by Salbu (2000): "A significant fraction of radionuclides released by nuclear events, for instance nuclear weapon tests, nuclear reactor accidents, and fires involving nuclear installations is associated with particles. Furthermore, effluents from nuclear reactors and reprocessing plants under normal operating conditions contain radionuclides in particulate and colloidal form. Radioactive particles are also observed in the vicinity of radioactive waste dumped at sea." Hot particles are also regularly detected in normally operating nuclear power plants. The NCRP (1999) concluded that "hot particles associated with nuclear facilities have been mostly an 'inplant' problem

associated with nuclear reactors, but the possibility that hot particles could inadvertently escape to the outside environment cannot be entirely dismissed".

Although the presence and release of hot particles into the environment from past nuclear incidents are widely documented, they are often considered as a unique and "rare form of radioactivity" (Kutkov *et al.* 1995) and treated something like a scientific curiosity (Sandalls *et al.* 1993) which may lead to an underestimation of their significance. The particulate nature of radioactive substances is not necessarily taken properly into account in environmental monitoring and consequence analyses of nuclear accidents. In traditional consequence analyses 'radioactivity' (not necessarily particles) is assumed to be released, transported, dispersed, and deposited on a target area. Existing monitoring systems are rather designed to detect 'radioactivity' than individual particles; and in laboratory analyses the radioactive species are often considered as 'becquerels' more or less evenly distributed in a sample. As regards the abovementioned issues, the results of the present thesis give a new perspective and ideas for subsequent studies.

The role of hot particles in estimating the significance of radioactive releases is far from clear. Vajda (2001) concluded that "there is still a lack of knowledge to fully understand the processes of hot particle formation, transport, migration, bioavailability and health hazards". The aim of the present thesis is to study nuclear fuel particles from the perspective of health hazards. The studies focuses on

- the characterisation of nuclear fuel particles,
- their transport and dispersion in the atmosphere and
- the estimation of possible skin doses.

The hazard cannot be considered hypothetical. At the time of the Chernobyl accident, six persons were near the plant at different distances in the direction to which the radioactive plume was moving (Barabanova and Osanov 1990). They were exposed to fallout particles which covered their skin and the ground. The person who received the highest γ-dose had very severe and widespread β-radiation burns (the β-dose was more than 20 times higher than the γ-dose). At the time of the accident, he was 1 km from the plant and remained there for an hour. He was covered with black dust and died 17 d after the exposure. Except for one person with the smallest dose, other victims in this area also died. No beta burns were reported in distant areas.

Highly active particles (activity of ^{137}Cs up to 10^8 Bq), originating from the disposal site of manipulated nuclear fuel of the material test reactor, were observed in the Dounreay, Scotland (COMARE 1999). COMARE performed risk estimation for ingested radioactive particles and arrived at the conclusion that "the particles, if encountered, present a hazard to health, and the hottest particles could induce serious acute radiation effects. Whilst the probability of encountering a particle is small, it is not negligible."

Since nuclear fuel particles may pose a health hazard, there is a need to develop methods for atmospheric transport and dispersion that take into account realistic weather conditions and the particulate nature of the release plume. As shown in publication V, nuclear fuel particles may present a potential health threat far from the source. Particles may be transported hundreds of kilometres in the atmosphere and, depending on wind conditions during the transport, also to other areas than gaseous species.

The health hazard caused by nuclear fuel particles is different compared with uniform contamination. Even if their number concentration in air is low, it is possible some individuals will receive a highly active particle deposited for example on the skin. Consequently, only a small fraction of the tissue may be exposed at levels that may cause severe health damage. Since the activity of a particle may be more than 1 MBq, the dose in its immediate vicinity may be very large. Risk estimations have been performed for inhalable hot particles from Chernobyl (e.g. Hofmann *et al.* 1988) but some of the particles detected after past nuclear incidents were much more than 10 μm in aerodynamic diameter and, consequently, are poorly inhaled (NCRP 1999). Instead, when deposited on the skin they may produce a notable skin dose within a short time (Publication VI). This fact is often disregarded in assessing the significance of large radioactive particles (see e.g. Garland and Nicholson, 1991).

The health threat that large nuclear fuel particles may pose is determined by particle properties such as composition, activity and size and their concentration in the environment. In practice, quantitative risk estimation may be useless owing to the very large uncertainties related to particle release, characteristics, appearance, and bioavailability. In the present thesis, rather an identification of short-term hazards than a detailed risk analysis is performed using the Chernobyl accident and nuclear fuel fragments originating from it as an example. A proper understanding of the nature of the hazard caused by highly active particles may facilitate designing the countermeasures that are needed in future nuclear incidents.

2 NUCLEAR FUEL PARTICLES IN THE ENVIRONMENT

Radioactive particles were distributed worldwide as a result of atmospheric nuclear tests mainly in 1950s and 1960s (see e.g. Salbu 2000, Vajda 2001). Local contamination due to nuclear bomb debris occurred as a result of aircraft crashes in Thule, Greenland, in 1968 (Moring *et al.* 2001) and Palomares, Spain, in 1966 (Romero 2001). The uncontrolled re-entry of nuclear-powered satellites into the atmosphere, such as Cosmos 954 in Canada in 1978 (Gummer *et al.* 1980), led to the dispersion of highly active particles into the environment.

The nuclear fuel particles considered in the present study may be generated in severe nuclear accidents and released into the environment, as occurred in the Chernobyl accident. The release of nuclear fuel particles cannot be considered unique to severe nuclear accidents only. After the Sosnovyy Bor incident in Russia 1992 nuclear fuel particles were detected in air samples collected along the coast of the Gulf of Finland (publication I, Paatero and Hatakka 1997). In addition, particulate radioactive materials have been detected in the environment after the releases from reprocessing plants, such as in Tomsk in 1993 (Hyder *et al.* 1996). Particles have been observed in aquatic environments (Salbu 2000) and in the areas in which the storage of nuclear fuel and radioactive waste exists (publication VIII).

Nuclear fuel particles have been also detected in nuclear power plants during their normal operation. NCRP (1999) reviewed in-plant radioactive particles found in U.S.A. Comprehensive surveys in nuclear power plants revealed the presence of fuel type particles as well as those composed of activation products. Although the review concentrated on the particles found in nuclear power plants NCRP concluded that "hot particles can occur in research reactor facilities and other nuclear facilities". Highly active particles are treated as an occupational safety problem rather than as a hazard to the public. However, "minimising the production and release of hot particles are clearly the preferred control methods, but the possibility of such events occurring cannot be ignored" (NCRP 1999). Mandjukov *et al.* (1994) concluded that "the presence of hot particles in the NPPs seems to be the rule rather than the exception."

However, "radioactive particles, tens of μm or more in diameter, are unlikely to be emitted directly from nuclear facilities with exhaust gas cleansing systems, but may arise in the case of an accident or where resuspension from contaminated surfaces is significant" (Garland and Nicholson, 1991).

Although highly active particles have been frequently detected after past nuclear incidents, their appearance and distribution even in the vicinity of the release site is generally not known. Published deposition maps of radionuclides such as ^{137}Cs, based for example on an environmental survey using mobile radiation detection systems, do not necessarily reflect the distribution of hot particles. Their presence must be verified using a well-defined sampling and sample analysis programme (Fig 1).

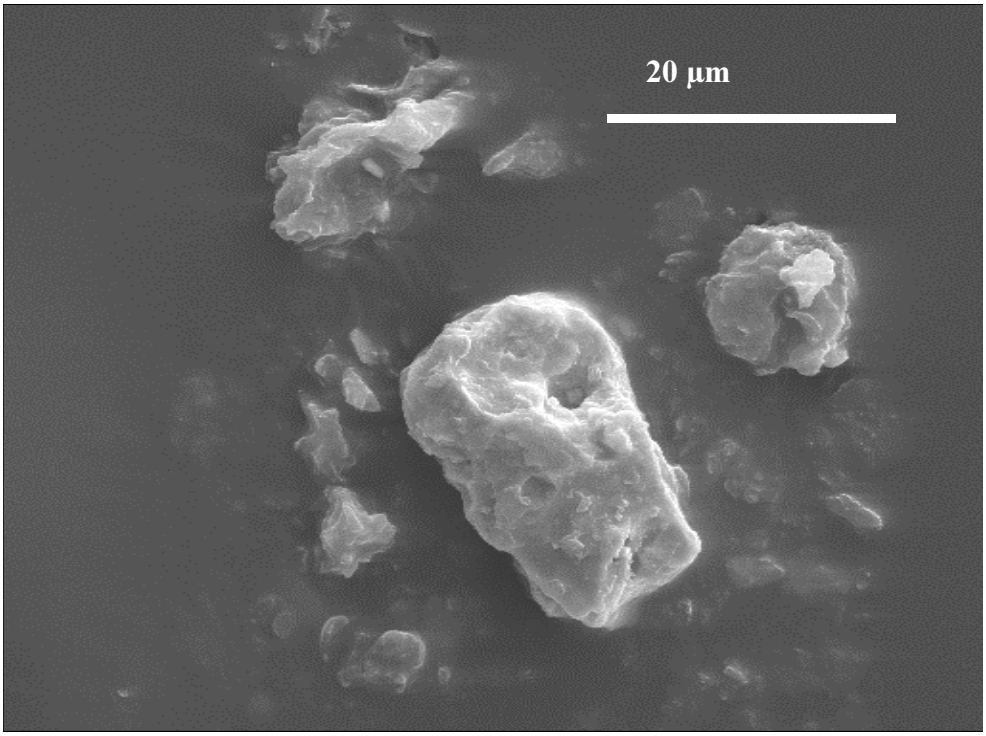

Figure 1. *Nuclear fuel particle (large object in the middle) detected in a marine sediment sample (publication VII).*

Despite the fact that more than a hundred scientific papers dealing with radioactive particles in the environment have been published (see for example the reference list by Pöllänen (1997)), the particle data from past nuclear incidents is far from complete owing to sparse sampling and the difficulties in analysing particle properties. Usually only a limited number of individual particles have been identified and, subsequently, characterised by different analytical means. Nevertheless, the number of identified radioactive particles especially from the Chernobyl accident is large enough to facilitate their classification into different groups. This makes it possible to understand the processes of particle formation and release.

2.1 Formation and release of particles from nuclear fuel

The characteristics of nuclear fuel particles reflect the properties of the source material and provide detailed information of their origin, formation, transport, reactivity, transformation reactions, and environmental impact (Jambers *et al.* 1995). The radionuclide composition of nuclear fuel particles characterises the nuclear fuel and different physical processes occurring during irradiation; elemental composition refers to the mechanisms during particle formation. The particle may contain nuclides that are already present in the source material except when nuclides from other sources are attached to the particle during transport or generated through radioactive decay.

In a severe nuclear accident, radioactive materials emitted from nuclear fuel and released into the environment may be in the form of gases, vapours and particles of different sizes. Soon after the release, the gaseous materials may be converted into solid species because of radioactive decay (for example ^{87}Kr $\rightarrow$ ^{87}Rb). High temperature may lead to the emission of volatile materials such as I, Cs, or their compounds and lead to the subsequent generation of small (diameter less than 1 μm) radioactive particles via nucleation, condensation, and coagulation. The nuclide composition of these 'condensation-type' particles deviates from that of nuclear fuel. After the Chernobyl accident, nuclides such as ^{103}Ru, ^{131}I, ^{132}Te, ^{134}Cs and ^{37}Cs were generally detected in these particles.

Energetic events may lead to the fragmentation of nuclear fuel, as occurred in the Chernobyl accident. Particles generated through mechanical fuel disintegration are usually large (diameter larger than 1 μm) and their nuclide

composition resembles that of the source material. This does not necessarily mean that each particle has the same composition because the source material is usually somewhat inhomogeneous. In the Chernobyl accident, 'U-type' fuel fragments with composition similar to nuclear fuel on the average were released into the environment. In addition, 'Ru-type' particles composed mainly of elements such as Ru, Rh, Mo, Tc and Pd were released from the damaged plant. These particles were often identified as the metallic precipitates present in irradiated nuclear fuel (publication VI). However, their formation/release mechanisms are still unclear. Mechanical emission is an obvious mechanism to explain the characteristics of some of the Ru-type particles found in the environment (publication VI).

Complex physical-chemical phenomena may affect the composition of fuel fragments if the temperature during the particle formation/emission is high. The elements or compounds, which have sufficiently high vapour pressure, may be easily vaporised from the particles that are then depleted with respect to these materials. U-type fuel fragments from the Chernobyl accident were generally depleted with respect to elements such as I and Cs.

2.2 Nuclear fuel particles from the Chernobyl accident

Nuclear fuel particles from the Chernobyl nuclear power plant were identified in several European countries. Pöllänen (1997) summarised those findings in which single particles or their detailed properties were analysed (also publication V). The particles were found in air filters or were collected from different surfaces. The method and the time of collection and the location of sampling have a crucial effect on the detectable characteristics of the particles. At the time of the Chernobyl accident, only a few laboratories were aware of the occurrence of nuclear fuel particles and thus their findings were clustered and sparse. Particles were systematically collected in only a few locations, not over wide areas, and the characteristics of only a few hundred radioactive particles have been reported in literature. The reported range of values of the number of deposited particles and especially the number concentrations in air is wide (Tables I and II) because of difficulties in locating the particles, different threshold values in detecting the particles, different analysis methods, and different timing of the estimates.

***Table I**. Number of deposited hot particles per square meter (Pöllänen 1997). Some authors have not reported the detection threshold (in terms of activity). In this case the reported detection limit is denoted as a question mark.*

Reference	Location	Reported detection threshold	m^{-2}
Rytömaa *et al*., 1986	Open areas in Finland	1 Bq	up to 10000 (mean<1000)
Robertson 1986,	Stockholm	?	5 - 8
Perkins *et al*., 1989	Öland	?	< 1
Kerekes *et al*., 1991	Stockholm, Gotland, Gävle	90...220 Bq	~ 0.2
Devell 1988	Studsvik	100 - 200 (^{141}Ce) Bq	1
Osuch *et al*., 1989	North-eastern Poland	50 Bq	0.007 - 1.9
Broda 1987	Mikolajki	100 Bq	0.1
Petryaev *et al*., 1990	40 - 250 km from the plant	?	10^5 - 10^3
Wahl *et al*., 1989	Konstanz	1 - 10 Bq	170
		10 - 50 Bq	38
		> 50 Bq	3
Kritidis *et al*., 1988	Athens	1-10 Bq (7.5.1986)	130
		10-100 Bq (7.5.1986)	13
		100-1000Bq (7.5.1986)	1.3
Lindner *et al*., 1992	4 km from the plant	d_p < 5 μm	> 10000
		d_p > 5 μm	5000
Viktorova and Garger 1990	5 km from the plant	?	50000
	90 km from the plant	?	15000
Khitrov *et al*., 1994	20 km from the plant	2 × background	50 - 60
	40 km from the plant	2 × background	30 - 40
(Sandalls *et al*., 1993)	60 km from the plant	2 × background	10
	Kiev	2 × background	1 - 2

A simple order-of-magnitude-estimate of the number of deposited U-type particles per unit area can be achieved as follows: by assuming that the amount of nuclear fuel released into the environment in the form of particles is 7000 kg (Sandalls *et al*. 1993) and by assuming that the equivalent volume diameter of the particles is 10 μm and their density 10500 kg m^{-3}, the number of released particles is then approximately 10^{15}. The sum activity of relatively

long-lived isotopes is 1300 Bq per particle (see Table IV, decay time 10 d). If particles are deposited within a zone of 30 km in radius the number of deposited particles per square meter is on the average 450 000 m^{-2} (which refers to the activity of about 500 MBq m^{-2}). This approximate estimate is by a factor of ten higher than the numbers reported near the Chernobyl nuclear power plant (Table I).

Table II. *Number concentration of hot particles in ambient air (Pöllänen 1997).*

Reference	Location	Time	Detection threshold (Bq)	m^{-3}
Toivonen *et al*., 1988	Helsinki	28.4.1986	10 (May 1987)	0.08
Mattsson *et al*., 1986	Helsinki	28 - 30.4.1986	50	0.0011 - 0.020
	Nurmijärvi	27 - 30.4.1986	50	0.0022 - 0.022
Sinkko *et al*., 1987	Nurmijärvi	27 - 28.4.1986	?	0.019 - 0.08
Lujanas *et al*., 1994	Vilnius	28 - 29.4.1986	?	10000
		29.4 - 1.5.1986	?	4 - 866
Kolb 1986	Brunswick	1.5 - 9.5.1986	1 - 15	0.0002
Wahl *et al*., 1989	Konstanz	May 1986	1 - 10	0.068
			10 - 50	0.015
			> 50	0.0011
Balashazy *et al*., 1988	Budapest	29.4 - 8.5.1986	10 (2.7.1986)	5×10^{-5}

Large-scale fallout from the Chernobyl accident was spatially heterogeneous, i.e. the territorial distribution of nuclides such as ^{95}Zr, ^{141}Ce, ^{144}Ce, ^{134}Cs and ^{137}Cs varied widely (Arvela *et al.* 1990; Jantunen *et al.* 1991, Mietelski and Was 1995). Smaller inhomogeneities with elevated amounts of radioactive materials were also identified (Luokkanen *et al.* 1988); these 'hot spots' were a few kilometres in diameter. In Lithuania, a small number of hot spots (several tens of square metres in area) were found on the ground (Lujanas *et al.* 1994). Near the Chernobyl power plant, hot spot areas with a marked occurrence of radioactive particles were detected (IAEA, 1991). In Poland, spots as small as 30 cm in diameter were identified (Broda 1987).

The local distribution of nuclear fuel particles was extremely inhomogeneous. The relative occurrence of U-type and Ru-type particles differs. This observation suggests that they were not released in the same way or their transport was different. It is possible that the particles deposited in Poland originated from a different part of the reactor compared to those which were transported to Scandinavia. This was deduced by different depletion of Cs and different isotope ratios of $^{103}Ru/^{106}Ru$ and ^{144}Ce (Broda 1987). Large objects may be transported hundreds of kilometres from the plant. For example, Broda *et al.* (1989) reported even 600 µm particles in Poland, suggesting that the bulk of these particles comprised fragments of the reactor graphite moderator. Radioactive particles were also found in human tissue.

The content of nuclear fuel particles, their size and isotopic composition depend on the distance from the Chernobyl nuclear power plant. The nuclear fuel particle contribution in the 30-km zone around the plant was at least 65 % of the total activity (Tcherkezian *et al.* 1994). The territorial distribution of nuclear fuel particles and condensable particles differ. The proportion of condensation particles is estimated as 60 % and 98 % at distances up to 25 and 60 km from the NPP (Pavlotskaya et al. 1994). Within 10 km from the plant, less than 3 % were attributed to condensed particles and more than 95 % to fuel particles (Salbu et al. 1994).

3 CHARACTERISATION OF RADIOACTIVE PARTICLES

The prerequisites for achieving proper results in particle analysis are the awareness of the possible existence of hot particles in the environment, well-designed sampling procedures and the capability of identifying and isolating particles from the sample. In addition, the availability of proper equipment for particle analysis is necessary, as is a well-trained and experienced staff that is able to apply the methods normally used in bulk analyses to the analysis of individual particles.

The analysis of individual particles complements conventional bulk analysis. However, the measured properties of individual particles are not necessarily sufficient in estimating the relevant characteristics of the particles, their transport in the environment, and the possible threats to health. Sometimes calculative methods must be applied. This is especially the case when the appropriate methods of analysis are not available or they are used too late: nuclear fuel particles have been identified and analysed typically weeks or even years after the accidents, preventing the detection of short-lived nuclides. In addition, the particles were usually analysed with gamma-ray spectrometers only. Although nuclides with low gamma yield or pure beta emitters as well as those with low activity were not generally detected, they may essentially contribute to possible health hazards (publication VI).

Particle analysis can be divided into the following phases:

- Identification of the possible presence of individual radioactive particles in a sample; estimation of their number in the sample and, possibly, the activity of individual particles.
- Isolation and separation of the radioactive particles from the sample that may contain billions of inactive uninteresting background particles.
- Analysis of particle characteristics using different methods.
- Interpretation of the analysis results and comparison with calculated particle properties.

3.1 Sampling, identification, and isolation

Standard radiation monitoring techniques may provide information about areas with elevated amount of contamination (hot spots) but they are not necessarily appropriate for the identification of individual hot particles. Sampling techniques are needed. Direct identification of the presence of hot particles in the environment requires devices that are able to scan the inhomogeneities of the contamination (e.g. Khitrov *et al.* 1994).

The procedures for particle sampling (method, time and location) and analysis (method and time of analysis) determine those properties of the particles that are possible to detect. Especially the sampling distance from the plant and the time and methods of analyses make a notable contribution in obtaining a representative sample of radioactive particles. For example, coarse particles cannot be detected far from the plant owing to their short residence time in air and short-lived nuclides cannot be detected if sampling and analyses are performed too late.

A range of sampling and sample manipulation techniques exists for different types of sample materials (Vajda 2001). For example, airborne radioactive particles are usually collected using air filtration or deposition samplers. Without particle isolation/separation, average quantities, such as activity concentrations in air, are then obtained. However, the size distribution of the particles can be measured using impactors. Single particles from other types of samples can be identified as deposited onto different surfaces or incorporated in the matrix of a bulk sample (publications VII and VIII).

The presence of non-volatile nuclides, such as ^{154}Eu (publication VII), detected by traditional bulk sample analysis may give a hint of the presence of hot particles. Another possibility for making a preliminary identification of the presence of hot particles in the samples is to perform repeated mixing of the samples and subsequent counting (Bunzl 1998). Sample splitting into smaller subsamples and their analysis is another option. In addition, autoradiography (Pöllänen *et al.* 1996) or imaging plate techniques (Moring *et al*. 2001; Zeissler *et al.* 1998) are appropriate methods that also provide data on the particle distribution in a sample (Fig. 2).

A prerequisite for analysing the properties of individual particles is isolation and further separation of the particles in question from the sample. The advantages of isolation are apparent: by analysing individual particles, it is

possible to achieve results that are otherwise not possible owing to the interference of non-relevant bulk particles in the sample. Adhesive tapes are often used in the isolation (publications VII and VIII). The identification and analysis of individual hot particles may sometimes be possible without manual particle isolation using scanning electron microscopy or secondary ion mass spectrometry. A sufficiently large number of hot particles must then be present in the sample and the physical size of the sample should be small enough.

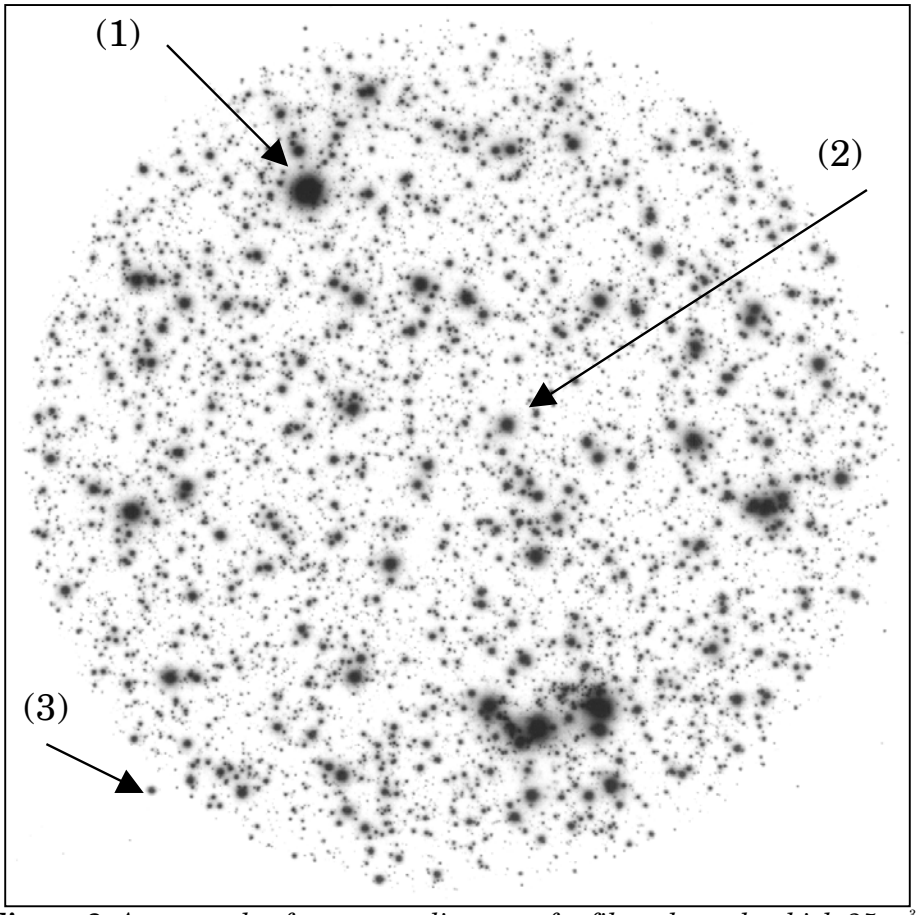

Figure 2. *An example of an autoradiogram of a filter through which 25 m^3 air has passed (Pöllänen et al. 1996). The sample containing activation-type radioactive particles was collected in a nuclear power plant. The film exposure time was 10 d. The total activity of the filter was about 0.5 kBq. Three particles were separated from the filter and analysed using a gamma-ray spectrometer and nuclides ^{54}Mn, ^{58}Co, ^{59}Fe, ^{60}Co and ^{95}Zr were identified. The sum activity of these nuclides in the particles 1, 2 and 3 were 25 Bq, 12 Bq and 0.7 Bq, respectively.*

3.2 Analysis methods

A range of assay methods is needed for the complete characterisation of hot particles. The information that can be obtained in analysing the particles depends on the method of analysis. The information collected from the inactive nuclides of the particle may be as valuable as that obtained from radioactive nuclides. In forensic analyses, the particle characteristics may function as a fingerprint of their origin. For example, the comprehensive analyses of the hot particle detected in the marine sediment sample from Gotland deep (publication VII) were of fundamental importance in estimating the particle's origin.

The scientific and technical challenges of hot particle manipulation and analysis are associated with the small size of the particles and their identification from (inactive) non-interesting background particles. Different techniques have specific limitations of their own, which may lead to difficulties in determining the optimum method for analysing the particle in question. However, if they are appropriately used, the results of their analysis may complement each other. Special emphasis must be placed on the order of different analysis methods. The order depends on the type of information needed in each case (Table III).

Vajda (2001) classified hot particle analysis techniques into nuclear analytical techniques and microanalytical techniques. The main advantages of using nuclear analytical techniques compared to microanalytical ones are their sensitivity and relatively simple use. Radiation emitted by radioactive materials in the sample is 'passively' registered whereas microanalytical techniques need stimulation, typically a particle beam ejected into the specified object to be analysed, before detecting the response signal. This means that in microanalytical techniques the hot particle in question must be usually unequivocally identified before the analysis. In nuclear analysis techniques, the particle does not necessarily have to be identified provided the background particles in the sample do not contain significant amounts of the same radionuclides as the particle itself.

Nuclear analytical techniques commonly used in particle analysis, and also utilised in publications VII and VIII, are autoradiography or similar types of imaging systems, and α, β and γ-ray spectrometry. The outcome of these methods is the nuclide composition of the particles and in the case of autoradiographic/imaging techniques, the particle distribution in the sample.

Other methods, such as different types of dose rate meters, are used especially during sample manipulation.

Table III. *Methods of analyses used in the present thesis (publications VII and VIII).*

Method of analysis	Purpose	Property to be measured
Autoradiography	Identification, localisation, and isolation of radioactive particles.	Number of radioactive particles, total beta activity of a particle.
Gamma-ray spectrometry	Presence and amount of gamma-emitting radionuclides in a particle/sample.	Activity of gamma-emitting radionuclides.
Beta spectrometry	Presence and amount of beta-emitting radionuclides in a particle/sample.	Activity of beta-emitting radionuclides (entire or dissolved particle).
Alpha spectrometry	Presence and amount of alpha-emitting radionuclides in a particle/sample.	Activity of alpha-emitting radionuclides (entire or dissolved particle).
Scanning electron microscopy with X-ray analysis	Visualisation and elemental composition of radioactive particles.	Particle size, concentration of elements.
Secondary ion mass spectrometry	Visualisation and nuclide composition of a radioactive particle.	Concentration of nuclides.
Infrared spectrometry	Material characterisation.	Chemical composition.

Microanalytical techniques are frequently used in material sciences but their applications to radioactive particles are rare. The small size of the particles, the absence of suitable standards and difficulties in determining the interaction volume (the volume from which measurable information can be achieved) make it difficult to obtain quantitative results. Depending on the type of beam used, the information may come from the surface of the particle or from the deeper parts. The key characteristics of the hot particles that can be detected using microanalytical techniques are their element (or sometimes

nuclide) composition and structure. In addition, the chemical composition, crystal structure, oxidation state etc. may be obtained.

Since most microanalytical techniques require special and usually very expensive equipment, co-operation between different research centres is often necessary. Their use in particle analysis is also very expensive because of the amount of manual work with respect to the number of analysed particles is often very small (publications VII and VIII). A notable drawback of most microbeam techniques is the need to operate under vacuum, which may cause the loss and transformation of volatile and unstable compounds.

Different types of electron microscopes are widely used in particle analysis. They use an electron beam to excite the atoms in the target sample. Characteristic X-rays as well as secondary electrons, backscattered electrons, transmitted electrons, or diffracted electrons are detected. Scanning electron microscopy with an energy dispersive X-ray spectrometer is used in the present analyses (publications VII and VIII). Equipment using other types of beams, such as X-rays, protons, sychroton radiation etc. was not available.

Mass spectrometers are used to determine the elemental and nuclide composition of bulk materials, but applications to particle analysis are scarce. In mass spectrometry, the samples (or the target in question) are decomposed and ionised and then ejected into the unit which separates the ions with different mass-to-charge ratios. Mass spectrometers can be used in hot particle analysis provided the particle in question can be separated from the bulk sample (publication VIII). Other techniques, such as infrared spectroscopy (publication VIII), are very seldom used in particle analysis.

3.3 Calculation of particle characteristics

The characteristics of nuclear fuel particles can be predicted in certain situations. In order to estimate particle properties, the primary assumption is that nuclear fuel particles reflect the characteristics of nuclear fuel. Another assumption is that the proportions of certain elements remain unchanged during particle formation and release. This, of course, is not valid for gaseous (or volatile) species. The third assumption is that other materials are not significantly agglomerated/condensed into an existing nuclear fuel particle or materials are not evaporated, i.e. particle composition does not change remarkably during transport and deposition. However, when particles are

deposited on the ground their weathering leads to mobilisation and the possible intake of the radionuclides incorporated in the particles.

The characteristics of nuclear fuel particles as well as their abundance in various media must be known in order to estimate their potential health effects and risks. The dose calculations cannot be fully performed using the information gathered merely from particles detected in the environment since particle sampling and detection may not be necessarily representative and the analytical methods may not be necessarily adequate for estimating all the relevant properties of the particles (publication VI). Sometimes prompt activity analyses of the particles are not possible because of the tedious procedures needed for particle sampling, isolation, and analysis. Calculative methods are needed.

An approach based on fuel inventory calculations is used to supplement characteristics that cannot be obtained from measurements. A typical example of this procedure is that the activity of a relatively short-lived nuclide is calculated from the activity of a more long-lived nuclide of the same element, provided the decay time and burnup of the fuel from which the particle has been originated are known.

The amount of (radioactive) materials in a reactor core, the 'inventory', is estimated using the ORIGEN2 (Croff 1983) computer code designed to calculate the different characteristics of nuclear fuel. It calculates numerically the concentrations of different elements and nuclides in the nuclear fuel. The main phenomena considered in ORIGEN2 are nuclear fission, neutron-induced transmutation, and radioactive decay. A set of ORIGEN2 calculations for different reactor types was performed as a function of fuel burnup and decay time. The results of the calculations are stored in a database that is used to estimate the characteristics of irradiated fuel and particles.

Since the output of ORIGEN2 is very large and the code itself does not include the possibilities to treat all this information in a modern way, a tool for data management, known as OTUS, was developed (Pöllänen *et al.* 1995). Inventory data were calculated for PWR (VVER-440), BWR (Swedish type) and RBMK (Chernobyl) reactors using simplified operation histories (Pöllänen 1997), i.e. reactor power was assumed to be constant during the irradiation, and fuel outages were taken into account except for the RBMK reactor. The database contains the concentrations of the elements and nuclides present in the reactor fuel. The activity of a specified nuclide per unit volume of the reactor fuel in

question is obtained multiplying fuel density by activity concentration (Fig. 3). Since the specific power used in the calculations was assumed to be constant during irradiation, the calculated concentrations are rather indicative than true estimates of the amount of materials present in the reactor fuel.

The Chernobyl accident showed that the isotopic composition of U-type particles generally reflects the core inventory (Osuch *et al.* 1989). The proportions of certain elements in a nuclear fuel fragment, e.g. Zr and Ce, are similar to those in nuclear fuel. The proportions of their nuclides are determined only by the burnup and cooling time (decay time) of the specified reactor fuel provided that isotopes of the same element behave similarly during particle formation and release. In addition, the activities per unit volume may differ by a factor of about two in different NPP reactor types (Pöllänen 1997).

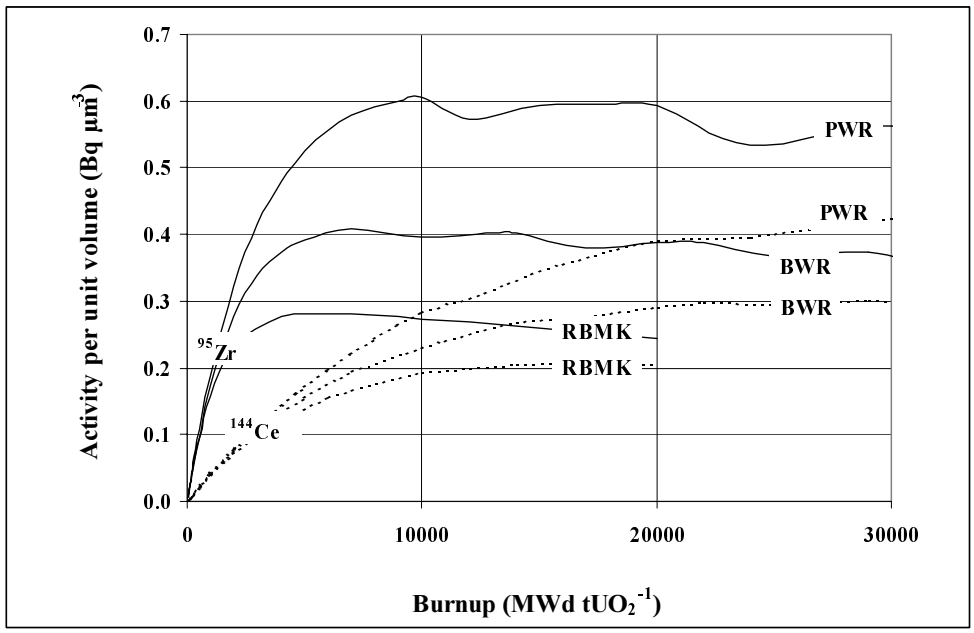

Figure 3. *An example of the activity per unit volume (Bq μm^{-3}) of ^{95}Zr and ^{144}Ce in PWR (VVER440), BWR and RBMK (Chernobyl) fuel as a function of fuel burnup. Activities are shown up to the average burnup of exhausted fuel (Pöllänen 1997).*

The particle size estimation from nuclide-specific activities is based on the assumption that the nuclide ratios of non-volatile elements are not changed during the fuel fractionation processes (e.g. Piasecki *et al.* 1990, Jaracz *et al.* 1990). Pöllänen (1997) calculated the equivalent volume diameter of Chernobyl particles from the known activity per unit volume of a nuclide (or from the sum activity of several nuclides). Particle sizes were estimated separately for U-type and Ru-type particles because their activities per unit volume were different.

Activity ratios are of special importance. In general, the activity ratios can be used in two ways:

1) If two different isotopes of the same element, e.g. ^{141}Ce and ^{144}Ce, are detected in a particle then it is possible to estimate the burnup of the fuel from which the particle has originated. This was applied by Pöllänen (1997) in making hot particle size estimation detected in the environment for which the sizes were not reported in the original publications. The method was also used in publication VII.
2) If the burnup and decay time of the fuel is known then it is possible to estimate the activity of the nuclides (e.g. ^{90}Sr) that cannot be detected using a gamma-ray spectrometer. This was applied in publication VI where the activities of short-lived nuclides, not reported in the original papers, were estimated.

For homogeneous particles, the activity is directly proportional to particle mass which, in turn, is proportional to the cube of particle linear dimension. Particle size (volume or equivalent volume diameter) is often determined using electron microscopy, which in addition to activity analysis allows determining the activities of various nuclides per unit volume of the particle. On the other hand, particle size can be estimated using gamma-ray analysis only, if activity per unit volume is known.

4 ATMOSPHERIC TRANSPORT

In atmospheric transport and dispersion, large nuclear fuel particles pose different problems compared with small radioactive particles and gaseous species (publications III and V). In the atmosphere, they behave differently and they are not distributed within a release plume in the same way. Soon after their release, the large particles (aerodynamic diameter $d_a \gtrsim 20$ μm) leave the main aerosol stream mainly by sedimentation. In weather types where wind speed and direction change significantly with height, large particles and gaseous species or small particles may be transported separately. It is even possible that in some areas the fall-out contains mainly large particles, neither gaseous species nor small particles.

In addition to the sedimentation, atmospheric turbulence contributes notably on the deposition of large particles. Particles with a sedimentation velocity greater than 1 m s^{-1} ($d_a \gtrsim 200$ μm) fall so fast that turbulent dispersion is no longer important (Van der Hoven 1968). For particles of a few μm in aerodynamic diameter, turbulent dispersion is certainly an important deposition mechanism but a question arises over the net effect of the sedimentation and the turbulent dispersion for particles of sizes between these limits. The threshold value of the sedimentation velocity, at which settling becomes important, depends on the magnitude of the velocity of turbulent motions in the air (Garland and Nicholson 1991).

Despite the uncertainties associated with the role of sedimentation and turbulent dispersion, it is important to discover the areas that may receive the fall-out of highly active particles in a severe nuclear accident. However, gravitational settling of particles as a deposition process in real time long-range dispersion models in operational use is often excluded (Bartnicki *et al.* 2001). For the purposes of emergency preparedness, it is crucial to establish the maximum transport range of particles that may produce an acute health hazard, although they are of special concern near the release site. The transport range is defined as the distance from the point of release to the point of deposition (Pöllänen *et al.* 1995, publication III). Range estimates are based on the time difference between the release and deposition. During this time, the particles, originally lifted up to the effective release height, are transported over a distance determined by the wind velocity. Only dry deposition is

considered (also in publications III and V). A user-friendly computer code using simplified atmospheric conditions was developed to estimate the transport range of large particles (Pöllänen *et al.* 1995). The code, known as TROP, is intended mainly for use in emergency preparedness but it can also be used for research purposes.

TROP does not take into account varying wind conditions during transport and, thus, it cannot be fairly used in operational situations. The transport of large particles must be connected to the prevailing weather (Fig. 4). A long-range transport, dispersion and dose model, TRADOS[1], was developed for real-time calculations in realistic atmospheric conditions (Valkama and Salonoja 1993). The transport and dispersion of the particles is described by 3-dimensional trajectories and the resultant vertical velocity is the sum of the sedimentation velocity and the velocity of ascending or descending airflow.

The code uses numerical weather prediction model data to calculate trajectories. Weather parameters for different altitudes are stored routinely four times a day in a database that contains weather parameters, such as wind components, air temperature, relative humidity etc. The size and density of the particles as well as their effective release height are required as trajectory model input. However, in operational use, these quantities may be very uncertain and the calculated fallout areas are then only suggestive.

Particle trajectories were calculated for the Chernobyl accident in publication V and in Valkama *et al.* (1995). The effective release height of the particles and the atmospheric phenomena related to their transport were investigated by comparing particle findings in the environment with the locations given by particle trajectories. Since the size of the nuclear fuel particles detected in the environment is usually not documented, it was estimated using calculative means.

Unlike air parcel trajectories, the trajectories of large particles terminate when they hit the ground. However, the point of deposition cannot be determined accurately because of atmospheric turbulence. TRADOS is unable to take turbulence into account and the conclusion that the maximum effective release height must either have been considerably higher than previously reported or particles may have been lifted up to higher altitudes in deep convective cells (publication V) must be further validated. Valkama and Pöllänen (1996)

[1] SILAM computer code has replaced TRADOS, which is no longer in operational use.

concluded that convective lifting might have affected the dispersion and deposition of radioactive particles from the Chernobyl accident (Fig. 5) but the effects of atmospheric turbulence on the transport of large particles is still an open question. A novel computer code known as SILAM is under development (Valkama and Ilvonen, 2000) and it may validate the conclusions presented above.

The Norwegian Meteorological Institute has taken into account the ideas presented in publication V and simulated the transport of the large particles in the Chernobyl accident (Bartnicki *et al*. 2001). They verified that large particles could travel long distances before being deposited (publication V).

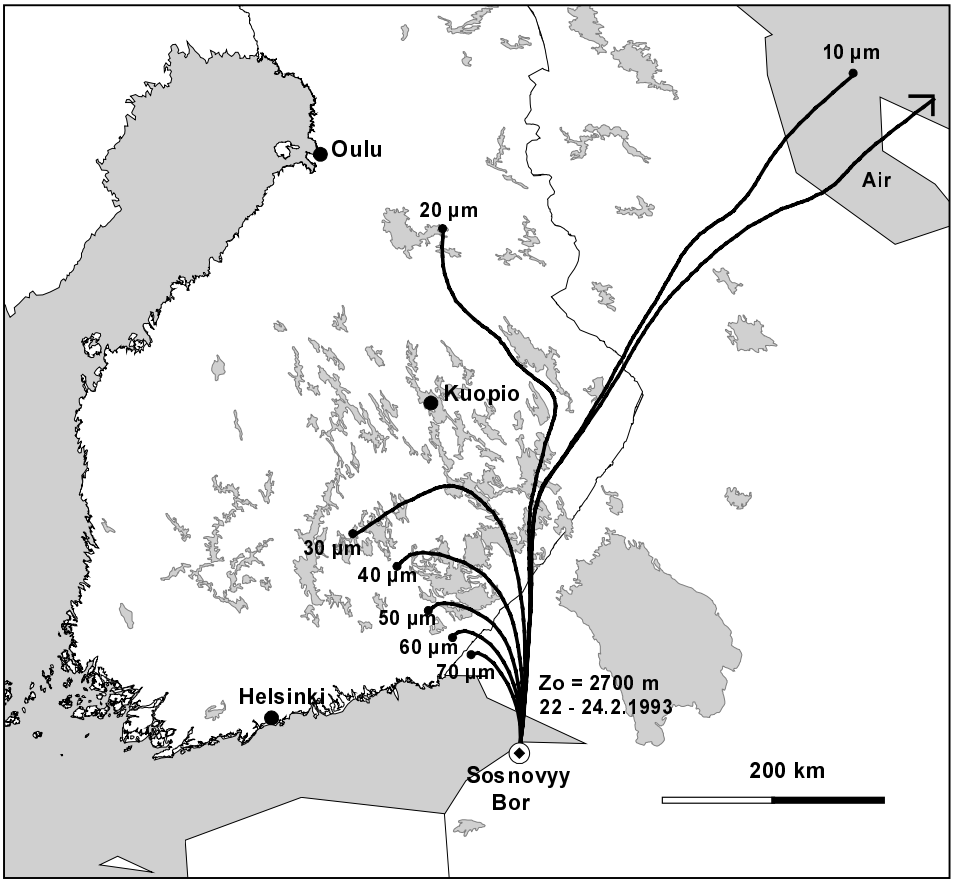

Figure 4. *Trajectories of an air parcel and a group of particles of different aerodynamic diameters released hypothetically from Sosnovyy Bor (Pöllänen et al. 1993). The release height is 2700 m. The average wind velocity along the particle trajectories is between 3.5 to 6 m* s^{-1}*. The transport situation refers to the days Feb 22-24, 1993.*

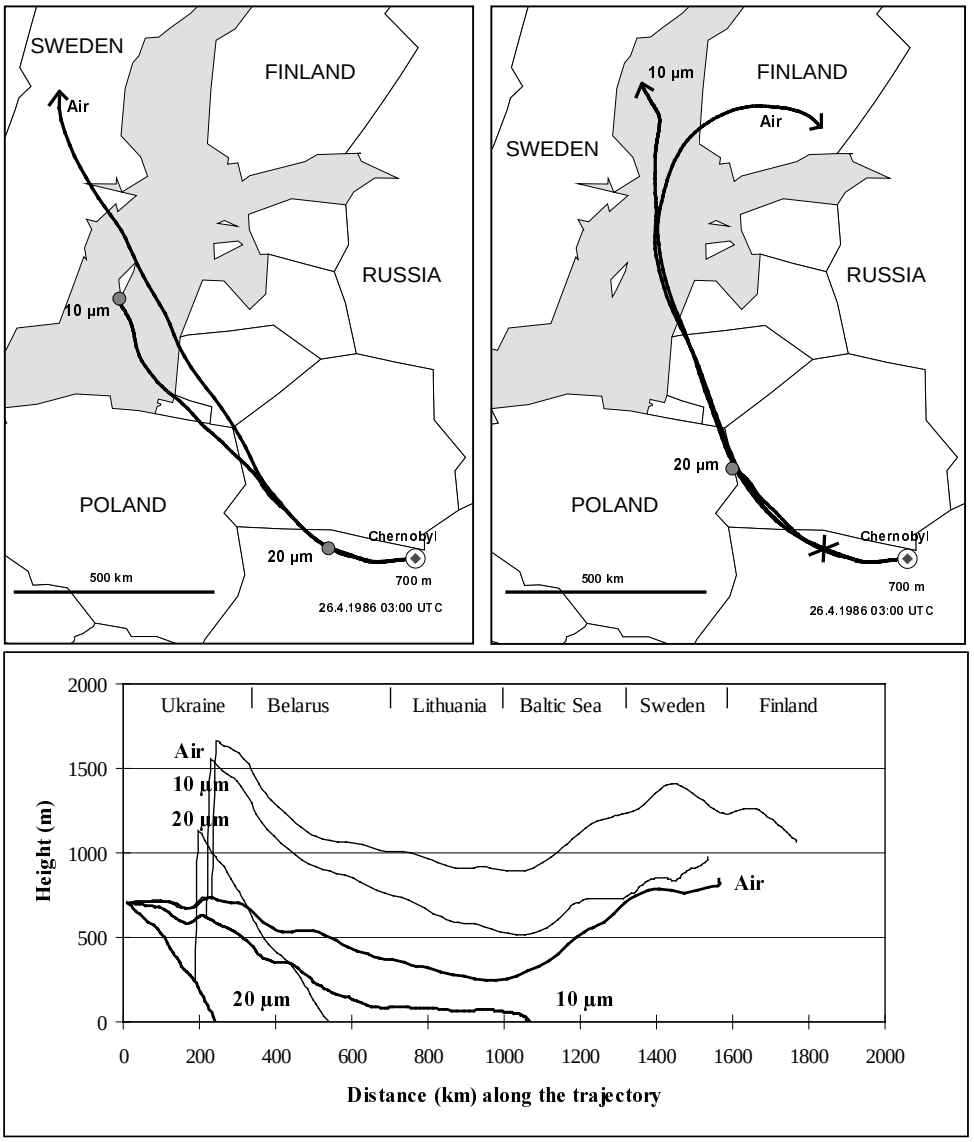

Figure 5. *Trajectories of an air parcel and particles 10 µm and 20 µm in aerodynamic diameter originating from Chernobyl NPP on April 26, 1986 at 03:00 UTC (Valkama and Pöllänen 1996). The effective release height is 700 m. On the left above, convective uplift is not assumed, whereas air parcel and particles on the right above experience an uplift of 1000 m. The location of the updraft is shown as a cross near Chernobyl. The heights of the trajectories are presented below. The thin lines refer to air parcel and particles that experience updraft whereas the thick lines represent those that experience no vertical uplift.*

5 CALCULATION OF SKIN DOSES

As regards the health hazards associated with large nuclear fuel particles, the skin doses are of special importance. The particles are poorly respirable because of their size and as a source of external radiation, they can often be treated in the same way as homogeneous contamination. As deposited into the environment, the mobility and bioavailability of the radioactive material incorporated in the particles are different compared to contamination that is more homogeneous. The uptake of radionuclides via ingestion of contaminated food or water and their subsequent absorption in the body are also different. Although above-mentioned matters must be taken into account in the consequence analyses of nuclear accidents, and are worth further research, they are not considered here. Instead, the most prominent feature of large nuclear fuel particles with respect to health threats, the possibility of producing acute deterministic radiation effects, is studied.

5.1 Ionising radiation and skin

Beta particles of various energies and low energy gamma rays are of greatest concern and importance in radiation protection of the skin. Damage that may be caused by more penetrating X and gamma rays will generally be limited by dose limits to other organs. Radiation doses from alpha particles may be high in the superficial layers of the skin without a notable dose in the deeper layers. This is due to the very low penetration of alpha particles. Exposure to very high doses over a very small area poses a particular problem.

The characteristics of the skin (Fig. 6) and its response to ionising radiation considerably affect the health hazard caused by highly active particles. The mechanisms of radiation effects on the skin are studied in numerous experimental investigations. However, the primary aim of most of these studies is related to radiotherapy, i.e. relatively large areas of the skin are irradiated with X or gamma rays. Specific radiation protection problems, such as skin irradiation with highly radioactive particles, cannot be necessarily explained by extrapolation from results of these experiments. The ionising radiation emitted from radioactive particles may induce deterministic effects and cancer of the exposed skin. A review of these effects as well as dosimetric

quantities and skin dose limits (chapter 5.2), based on ICRP (1990 and 1991), are presented in the following.

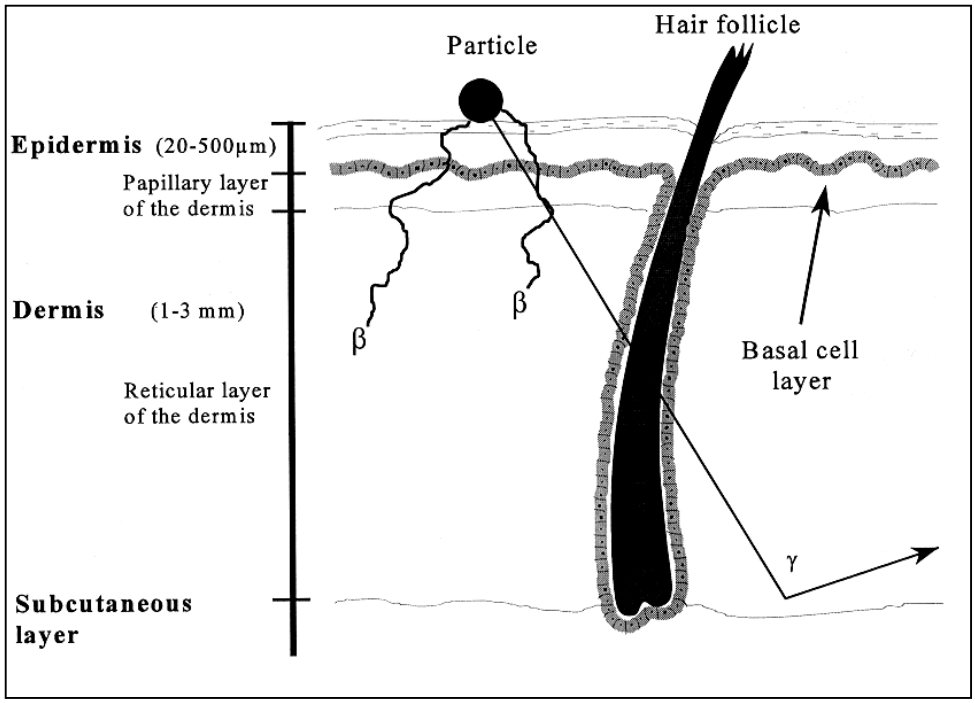

Figure 6. *Schematic cross-section of skin on which a radioactive particle is deposited (Pöllänen 1997). The routes of beta particles and photons originating from the particle are shown schematically. As a comparison for the thicknesses of the skin layers marked above, the distances in water at which 90% of the beta energy of ^{144}Ce, ^{90}Sr and ^{106}Rh is absorbed are 0.28 mm, 0.79 mm and 7.9 mm, respectively.*

Deterministic radiation-induced changes in the skin show several distinct phases of damage. Their severity depends on irradiation conditions, such as the type, energy, and intensity of the radiation and exposure time, and type and area of the exposed skin. The main phases of the damage are:

- Reddening of the skin (an early transient erythema) seen within a few hours of irradiation, which usually subsides after 1 - 2 d.
- Loss of the basal cells leads to the main erythematous reactions: keratinisation of the skin after moderate doses (dry desquamation), loss of the epidermis after high doses (moist desquamation) or hair loss may

result after 3 - 6 weeks. A skin wound (secondary ulceration) may develop if moist desquamation is severe.

- Late phase erythema is associated with blood vessel damage (dermal ischemia) and possible necrosis of skin between 8 - 20 weeks or more after irradiation.
- Late skin damage such as the thinning of dermal tissues after 6 months (atrophy), dilatation of superficial dermal capillaries (telangiectasia) and necrosis.

Nuclear fuel particles are of special radiological concern because of their small size ($\lesssim$1 mm) and high activity. When deposited on the skin they may produce very high and localised doses (Fig. 7). The nuclide composition of the particles may vary greatly, i.e., the particles may contain alpha emitters, low- and high-energy beta emitters, as well as gamma-ray emitters. The energy of alphas and very low-energy betas is absorbed in the epidermis above basal cell layer whereas gammas and high-energy betas may penetrate deep ($\gtrsim$ 1 cm) in the skin.

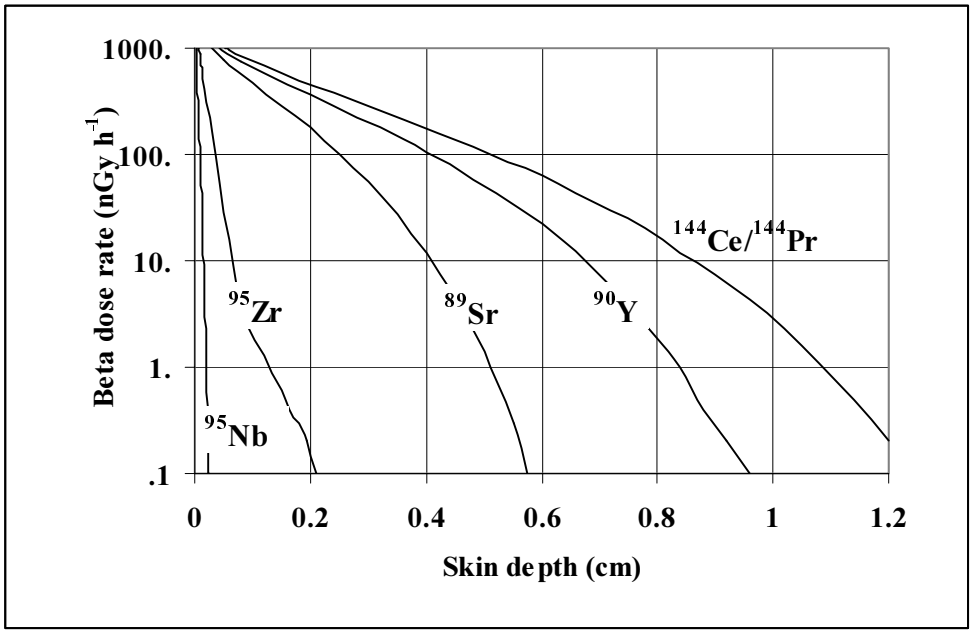

Figure 7. *Dose rate in water from a 1 Bq point isotropic beta source on the air-water boundary, averaged over 1 cm^2 circular area at different skin depths (Pöllänen 1997).*

Acute ulceration (skin wound) is the primary lesion resulting from irradiation with radioactive particles. The skin surface dose and the energy of the emissions from the particle determine the depth and size of the ulcer. The full lesion usually develops within 2 weeks of irradiation. A small pale and circular area with a slight blue tinge, surrounded by a halo of erythema, can be detected prior to the development of the ulcer. The estimated threshold dose for acute ulceration is ~75 Gy, measured over an area of 1.1 mm^2 at skin depth of 16 μm. This corresponds to a dose of ~1 Gy averaged over an area of 1 cm^2 at depth of 100 - 150 μm. A dose below 220 Gy (at depth of 16 μm over 1.1 mm^2) may lead to an ulcer that is likely to last for less than 1 week. A larger area erythema will also occur. Acute epithelial necrosis may be produced because of irradiation caused by low-energy beta particles. The dose from a point source to produce a 50 percent incidence of acute epidermal necrosis or acute ulceration varies from approximately 5 Gy to approximately 10 Gy, averaged over 1 cm^2 at a depth of 70 μm (NCRP 1999). This is to be compared e.g. with the effect from homogeneous X-ray irradiation, in which case a skin entrance dose of 5 Gy would only cause transient erythema.

The principal stochastic risk associated with irradiation of the skin is basal cell and squamuos cell skin cancers. The risk for radiation-induced skin cancer does not increase by very non-uniform radiation, i.e., radioactive particles do not cause more skin cancers per unit average dose compared with that caused by uniformly distributed doses in the skin.

5.2 Dosimetric quantities and skin dose limits

The fundamental dosimetric quantity in radiological protection is the absorbed dose, which is the energy absorbed per unit mass (unit Gy). The absorbed dose may be defined at a specified point but usually it is used to mean the average dose over a tissue or organ. The use of the average dose as an indicator of the probability of subsequent stochastic effects is a reasonable approximation over a limited range of dose. The average absorbed dose is not directly relevant to deterministic effects since the dose-response relationship is not linear.

The probability of stochastic effects depends on the absorbed dose as well as on the type and energy of radiation causing the dose. The absorbed dose averaged over a tissue or organ and weighted for the quality of the radiation, the equivalent dose (unit Sv), is of interest in radiological protection. The relationship between the equivalent dose and the probability of stochastic effects also depends on the organ or tissue irradiated. It is useful to indicate

the combination of different doses to several different tissues in a way that is likely to correlate well with the total of the stochastic effects. The effective dose is the sum of weighted equivalent doses in all tissues and organs of the body.

The equivalent dose and effective dose provide a basis for estimating the probability of stochastic effects only for absorbed doses well below the thresholds for deterministic effects. The ICRP recommendations for effective dose limits are sufficient to ensure the avoidance of deterministic effects in all body tissues and organs except the lens of the eye and the skin (ICRP 1991). The skin is not adequately protected by a limit of effective dose because it may be subject to localised exposure. Separate limits are needed.

In the case of stochastic effects, the equivalent dose can be averaged over the whole area of the skin. The stochastic effects are expected to arise in the basal cell layer. Some deterministic effects also arise in the basal cell layer, others in the deeper layer of the dermis. The ICRP (1990) recommends that the annual limit for occupational exposure is 500 mSv for the skin (150 mSv for the lens of the eye), averaged over any 1 cm^2, regardless of the area exposed. The nominal depth is 7 mg cm^{-2}. This limit is intended to protect the skin against deterministic effects. The limit for the public is reduced by an arbitrary reduction factor of 10 because the total period of exposure may be nearly twice as long as for occupational exposure, and because the exposed individuals may show a wider range of sensitivity than a more limited population of workers. The recommended annual limit for the public is 50 mSv for the skin (15 mSv for the lens of the eye), averaged over any 1 cm^2 area of the skin and regardless of the area exposed.

The NCRP (1989) recommendation on an occupational radiation exposure limit for a hot particle on the skin is intended to prevent acute deep ulceration of the skin, and is based on the time integral of the beta particles emitted from a radioactive particle. The NCRP recommends that exposure to the skin must be limited to 10^{10} beta particles emitted from the surface of a radioactive particle. This limit is obtained primarily from beta particle emissions from irradiated fuel particles, and refers to about 5 Gy averaged over 1 cm^2 at depth 70 μm in tissue (Baum and Kaurin 1991). Recently, the NCRP (1999) recommended that "the dose to skin at a depth of 70 μm from hot particles on skin (including ear), hair or clothing be limited to no more than 0.5 Gy averaged over the most highly exposed 10 cm^2 of skin". The NCRP skin dose limits are higher than those of the ICRP by a factor of 10 (Charles *et al*. 2000). Thus, there is disparity between the skin dose limits applied in USA and Europe.

5.3 Method of skin dose calculation

Determination of a skin dose caused by a radioactive particle deposited on the skin represents a complex problem. Although the geometrical arrangement appears to be simple, several problems arise in the dose calculation. The dose gradient is very sharp due to the short range of beta particles in tissue (Fig. 7). The energy spectrum of beta particles is continuous in contrast to gammas. Beta particles of varying energy interact with the surrounding materials, i.e. air, skin, and the particle matrix itself, in such a manner that analytical dose calculations may not be feasible.

The dose caused by a radioactive particle deposited on the skin depends primarily on the manner in which photons and beta particles interact with skin. Alpha particles are neglected in the dose calculations owing to their short range (up to ~50 μm in the skin). The dose to the basal cell layer of the epidermis is determined mainly by beta radiation; photons usually play a negligible role. This difference is smaller for large nuclear fuel particles because of the significant self-absorption of betas.

Several methods have been developed to calculate skin doses (see Pöllänen 1997). Loevinger (1956) proposed an analytical empirical function that can be used for estimating the point-source beta dose distribution in tissue. Later, the parameters of the empirical equation were revised and other point source dose distribution functions have been developed. The dose distribution in tissue around a point source of a beta emitting radionuclide, often referred to as 'point kernel', is also determined by numerically solving the transport equation or using a simplified form of the equation. The more accurate dose distribution data of betas and photons are generated using Monte Carlo calculations. Precalculated dose distributions are utilized in some methods.

Although there are many different methods of calculating skin doses produced by beta particles and photons, most of them are appropriate only for some predetermined purposes. For example, Monte Carlo methods, which are appropriate for detailed dose analyses in specified situations, are not necessarily suitable in routine dose calculations. Analytic representations of point source dose distributions may be useful only for certain nuclides; their validity for other nuclides is questionable. In addition, they can be used only for point sources, not necessarily for three-dimensional particles. Extensive tabulations, such as those presented by Cross *et al.* (1992) and Rohloff and

Heinzelmann (1996), can be used for dose estimation but their applicability for routine dose estimation is limited.

Skin doses are often calculated by assuming the skin is uniformly contaminated by beta active nuclides, not by individual particles. Moreover, the calculations are often based on the assumption that the particles are surrounded by an infinite homogeneous medium (water). Because of diminished backscattering in the air-tissue interface, the skin doses are then overestimated up to about 35 % (Cross *et al.* 1992). The dimensions of the particles are also frequently neglected. Especially for low energy beta emitters, the doses are overestimated if the effect of self-shielding is omitted. Gamma rays are often neglected, which may lead to underestimation of doses in certain cases.

A new model (PSS, Point Source and Self-absorption) has been developed to calculate doses caused by radioactive particles deposited on the skin (publication IV, Pöllänen 1997). The model, calculates the beta and photon dose rate to the skin at definite depths and averaged over a circular area of 1 cm^2. The source particle is assumed spherical and homogeneous. Neither protective clothes nor air gaps are assumed. The model uses point-source dose conversion factors that are analytically corrected for the self-shielding of beta particles. In the following, the doses are calculated for the basal cell layer of the skin (nominal depth 70 μm, circular target area) although any depth and any target area is possible provided appropriate point-source conversion factors are available.

The PSS model was compared to VARSKIN Mod 2 (Durham 1992) in publication IV. Skin doses, calculated with the PSS and VARSKIN Mod 2, were compared for spherical uranium dioxide particles of different sizes and unit activity by assuming the particles are deposited on the skin. The differences between the calculated doses are generally below two; the results were nearly equal deep (in terms of range of beta particles) in the skin. Both models show that the self-absorption of beta particles must be taken into account in skin dose calculations.

5.4 Skin doses from nuclear fuel particles

Radiation doses caused by uranium fuel fragments with composition of long-lived nuclides similar to nuclear fuel and deposited on the skin were estimated in publication II. Skin doses were calculated for particles of different sizes originating from the Chernobyl reactor by assuming the particles are composed of long-lived nuclides of some low-volatile elements. However, radioactive decay and dependency on the amount of nuclides in the particles as a function of fuel burnup and reactor type were neglected in the calculations. In addition, short-lived nuclides and some important low-volatile elements were not considered at all. Pöllänen (1997) considered these deficiencies for U-type particles and in publication VI for Ru-type particles generated in the fuel fragmentation of the Chernobyl accident. A summary of the results is presented in the following.

The nuclides of non-volatile elements incorporated in the calculations were selected based on their radiological importance (Table IV). The nuclide composition of low-volatile elements in reactor fuel as a function of fuel burnup and decay time was computed using the ORIGEN2 and OTUS codes. Activities were calculated separately for U-type and Ru-type particles (Figure 8, Table V).

The elements considered are alkaline earths Sr, Ba; refractory oxides Zr, Nb; noble metals Ru, Rh, Tc, Mo, Pd; rare earths + others Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Np, Pu, U (classification from WASH-1400, 1975). Their release fraction in fuel meltdown and vaporisation processes is below 20 % (WASH-1400, 1975). Since the gaseous species and volatile elements in the nuclear fuel particles observed in the environment were strongly depleted or totally missing, they were omitted in the calculations. This assumption may underestimate the doses. The criteria for the selection of the nuclides presented in Table IV were as follows: a) half-life is more than a few hours (some daughter nuclides may have shorter half-life), b) the amount in the nuclear fuel is sufficiently large, and c) beta energies are high enough to produce a notable dose on the skin.

Table IV. *Nuclides considered in the dose calculations. The maximum energy of the beta particles, E_{max}, refers to the most probable decay branch. CF_β is the beta dose rate from a point source of 1 Bq at the air-water boundary averaged over 1 cm^2 at the basal cell layer of the skin (Cross et al. 1992). The values denoted by * are calculated using SADDE Mod2 and VARSKIN Mod2 (Durham 1992). CF_γ refers to the gamma dose rate from a point source of 1 Bq averaged over 1 cm^2 at the basal cell layer of the skin (Rohloff and Heinzelmann 1996).*

Nuclide	$t_{1/2}$	E_{max} (MeV)	Probability per decay	CF_β (μGy h^{-1} Bq^{-1})	CF_γ (μGy h^{-1} Bq^{-1})
^{89}Sr	50.6 d	1.49	0.999	1.67	1.17×10^{-6}
^{90}Sr	28.6 y	0.546	1.00	1.38	-
^{90}Y	64.1 h	2.28	0.999	1.76	1.80×10^{-6}
^{91}Sr	9.5 h	1.10	0.339	1.57 *	1.01×10^{-2}
^{91}Y	58.5 d	1.54	0.997	1.67	3.60×10^{-5}
^{92}Y	3.54 h	3.63	0.857	1.76 *	3.33×10^{-3}
^{93}Y	10.1 h	2.89	0.902	1.73 *	1.32×10^{-3}
^{95}Zr	64.0 h	0.366	0.554	1.06	1.30×10^{-2}
^{95}Nb	35.1 d	0.160	0.999	0.23	1.30×10^{-2}
^{97}Zr	16.9 h	1.91	0.860	1.70 *	2.77×10^{-3}
^{97}Nb	72.1 min	1.28	0.983	1.58 *	1.31×10^{-2}
^{99}Mo	66.0 h	1.21	0.827	1.54	3.49×10^{-3}
^{103}Ru	39.4 d	0.226	0.900	0.568	1.25×10^{-2}
^{105}Ru	4.44 h	1.19	0.499	1.63 *	1.67×10^{-2}
^{105}Rh	35.4 h	0.567	0.750	1.17	2.80×10^{-3}
^{106}Ru	368 d	0.0394	1.00	-	-
^{106}Rh	30 s	3.54	0.787	1.85	4.53×10^{-3}
^{109}Pd	13.5 h	1.03	0.999	1.55 *	4.11×10^{-3}
^{140}Ba	12.8 d	0.991	0.370	1.46	1.00×10^{-2}
^{140}La	40.2 h	1.35	0.445	1.64	2.61×10^{-2}
^{141}La	3.94 h	2.43	0.970	1.73 *	3.58×10^{-4}
^{141}Ce	32.5 d	0.435	0.705	1.54	3.12×10^{-3}
^{142}Pr	19.1 h	2.16	0.963	1.69 *	4.32×10^{-4}
^{143}Ce	33.0 h	1.10	0.480	1.48 *	9.78×10^{-3}
^{143}Pr	13.6 d	0.935	1.00	1.52	1.57×10^{-10}
^{144}Ce	284.3 d	0.318	0.772	0.815	1.77×10^{-3}
^{144}Pr	17.3 min	3.00	0.977	1.82	3.15×10^{-4}
^{145}Pr	5.98 h	1.81	0.970	1.62 *	2.20×10^{-4}
^{147}Nd	11.0 d	0.805	0.811	1.31 *	8.08×10^{-3}
^{147}Pm	2.62 y	0.225	0.999	0.535	3.54×10^{-7}
^{148}Pm	5.37 d	2.46	0.555	1.60 *	7.48×10^{-3}
^{149}Pm	53.1 h	1.07	0.962	1.54 *	3.53×10^{-4}
^{151}Pm	28.4 h	0.843	0.427	1.65 *	9.60×10^{-3}
^{153}Sm	46.7 h	0.702	0.441	1.43	7.77×10^{-3}
^{156}Eu	15.2d	0.487	0.320	1.36	1.29×10^{-2}
^{237}U	6.75 d	0.238	0.531	0.632 *	1.62×10^{-2}
^{238}Np	2.12 d	1.25	0.450	0.770 *	1.23×10^{-2}
^{239}Np	2.36 d	0.436	0.520	1.09 *	1.51×10^{-2}

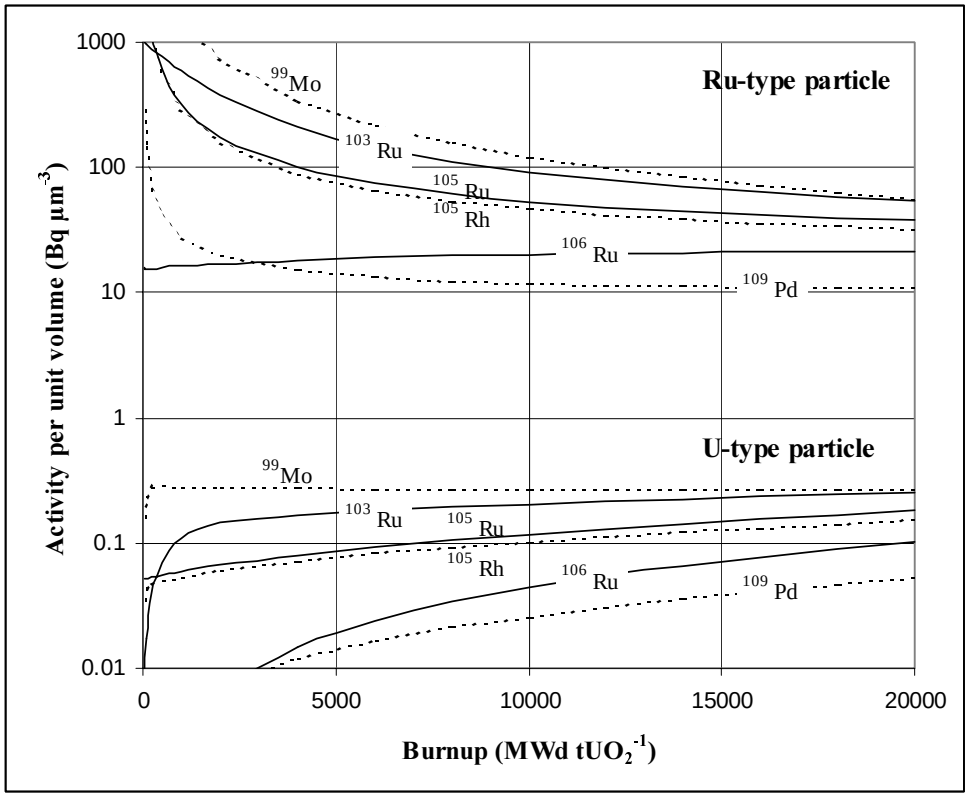

***Figure 8**. Calculated activity of ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{105}Rh, ^{106}Ru and ^{109}Pd per unit volume of an Ru-type particle and a U-type particle as a function of fuel burnup (Chernobyl reactor assumed). The activity of ^{106}Rh is not presented because of its short half-life. In practice, its activity is equal to the activity of ^{106}Ru.*

In general, the activity of most of the nuclides presented in Table IV per unit volume in a U-type particle is of the order of 0.05 - 0.3 Bq μm^{-3} (a complete set of activities is presented in Pöllänen, 1997; see also Fig. 3) whereas in the case of an Ru-type particle the activities of the nuclides of noble metals are higher by a factor of 100 - 1000. This is due to the fact that Ru-type particles are composed almost entirely of fission products whereas the U-type particles are composed mainly of bulk uranium dioxide. This is also the reason for the different behaviour of activities per unit volume as a function of fuel burnup (Fig. 8).

Table V. *Total activity of nuclear fuel particles as a function of equivalent volume diameter, d_p, for the decay times of 1 d and 10 d. The particles are assumed to originate from RBMK fuel irradiated to average burnup. The density of the particles is assumed to be 10500 kg m^{-3}. For U-type particles, the total activity, A_{tot}, is the sum of the activities of the nuclides presented in Table III, whereas only nuclides of noble metals Ru, Rh, Tc, Mo and Pd are taken into account for Ru-type particles (publication VI).*

	Decay time 1 d		Decay time 10 d	
d_p (μm)	U-type A_{tot} (Bq)	Ru-type A_{tot} (Bq)	U-type A_{tot} (Bq)	Ru-type A_{tot} (Bq)
1	3.0	130	1.3	57
2	24	1000	10	460
3	80	3500	35	1520
4	190	8000	83	3600
5	370	16000	160	7100
6	640	28000	280	12000
7	1000	43000	440	20000
8	1500	65000	660	29000
9	2200	94000	940	42000
10	3000	130000	1300	57000
15	9900	430000	4400	200000
20	24000	1000000	10000	460000

In addition to particle type, size is the most relevant quantity with respect to dose (Table VI) since for homogeneous particles the activity is directly proportional to particle mass, which is consequently proportional to the cube of the diameter (Table V). The composition and burnup and decay time of the fuel (= time of deposition) from which the particles have originated have a notable influence on the dose rates (Figs. 9 and 10).

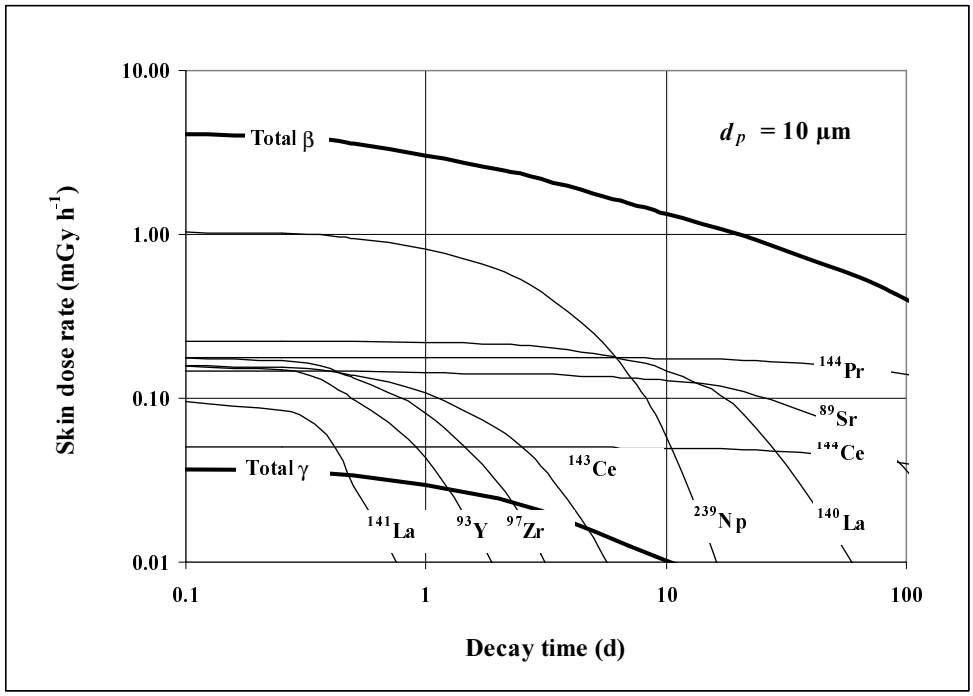

Figure 9. *Beta and gamma (total y) dose rates to the basal cell layer as a function of time, caused by a d_p = 10 µm U-type nuclear fuel particle deposited on the skin. The particle is assumed to originate from RBMK fuel irradiated to average burnup. All nuclides in Table IV are considered in the curves 'Total β' and 'Total γ', whereas other curves are for the beta dose rate of individual nuclides (not all nuclides are shown).*

The burnup of the fuel has a notable influence on the dose rates, although the effect for U-type is far less dramatic than for Ru-type particles (Fig. 10). Contrary to Ru-type particles, the more irradiated the fuel the more active the U-type particles.

Table VI. *Beta dose rate to the basal cell layer of the skin, caused by U-type and Ru-type particles deposited on the skin, as a function of particle diameter, d_p, for two decay times. The particles are assumed to originate from RBMK fuel irradiated to average burnup. The dose rates are averaged over 1 cm^2 at a skin depth of 70 μm. For U-type particles, all the nuclides presented in Table IV are taken into account, whereas only the nuclides of noble metals Ru, Rh, Tc, Mo and Pd are taken into account for Ru-type particles.*

	Beta dose rate at decay time 1 d		Beta dose rate at decay time 10 d	
d_p (μm)	U-type (mGy h^{-1})	Ru-type (mGy h^{-1})	U-type (mGy h^{-1})	Ru-type (mGy h^{-1})
1	0.0036	0.15	0.0016	0.049
2	0.028	1.2	0.012	0.38
3	0.093	3.8	0.040	1.2
4	0.22	8.8	0.094	2.9
5	0.41	17	0.18	5.4
6	0.70	29	0.31	9.1
7	1.1	45	0.48	14
8	1.6	66	0.70	21
9	2.2	93	0.99	29
10	3.0	130	1.3	39
15	9.4	390	4.2	120
20	21	890	9.4	270

Short-lived nuclides, such as ^{239}Np, are of primary importance within a few days from the end of the chain reaction (Fig. 9); their contribution is negligible later. The proportion of gamma rays emitted from nuclear fuel particles deposited on the skin is negligible; the dose caused by beta particles is 100 times larger than the dose caused by gamma rays. Reactors with high specific power may generate particles that have much higher activity per unit volume than particles produced by reactors with low specific power. A U-type particle originating for example from a PWR reactor may produce twice as large a skin dose than one of the same size originating from a RBMK reactor (Fig. 11).

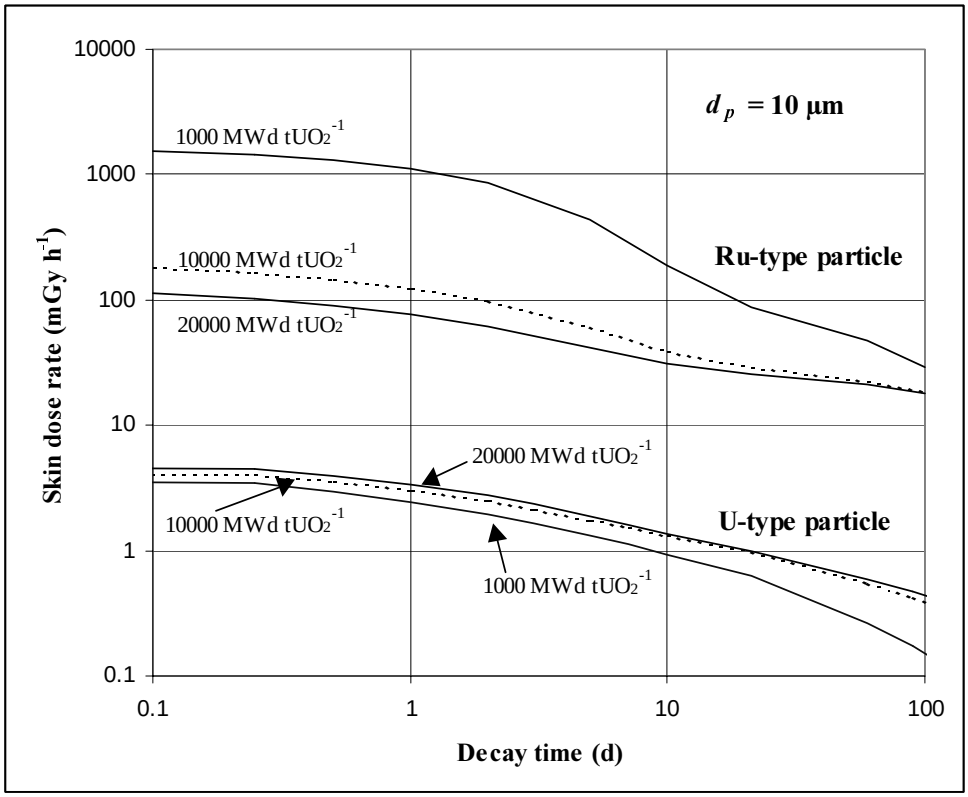

Figure 10. *Beta dose rate to the basal cell layer (averaged over 1 cm^2 at a nominal depth of 70 µm) as a function of decay time caused by U-type and Ru-type particles of 10 µm in equivalent volume diameter deposited on the skin. A Chernobyl type RBMK reactor is assumed in the calculations. The centremost curves refer to fuel of average burnup (10000 MWd tUO_2^{-1}) from which the particles are assumed to have originated. The upper and lower curves are for the particles emitted from low-burnup fuel (1000 MWd tUO_2^{-1}) and high-burnup fuel (20000 MWd tUO_2^{-1}).*

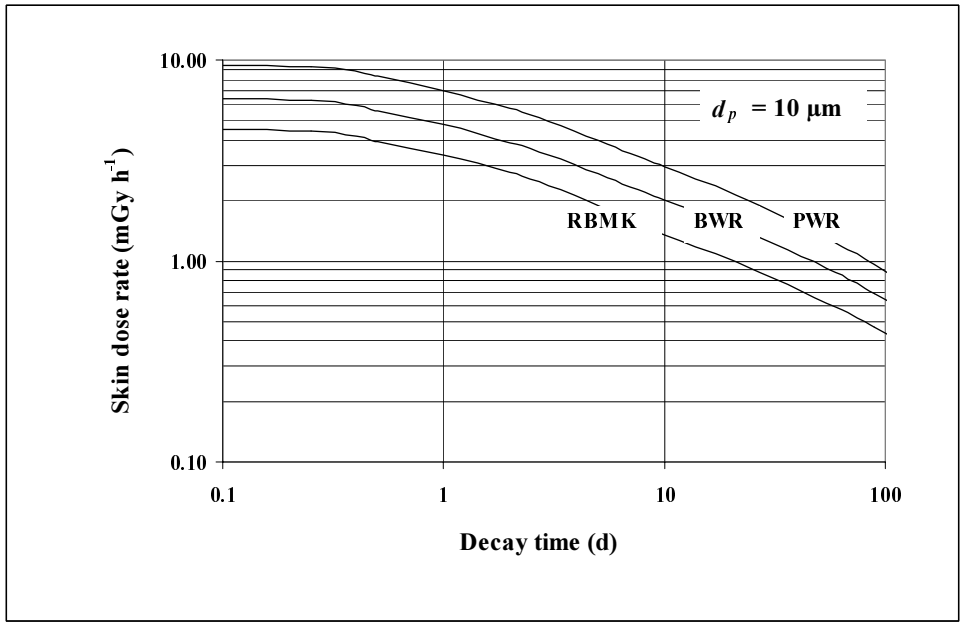

Figure 11. *Beta dose rates to the basal cell layer as a function of time, caused by a d_p = 10 µm U-type nuclear fuel particle deposited on the skin. The particles are assumed to originate from RBMK, BWR and PWR fuel.*

5.5 Skin doses from selected nuclear fuel particles released in the Chernobyl accident

The results of the calculations presented above are for hypothetical particles, i.e. particle characteristics, from which the skin doses are calculated, are derived from the properties of the nuclear fuel. Here skin doses are estimated for some U-type nuclear fuel particles detected in various European countries after the Chernobyl accident by assuming the particles are deposited on the skin (results are from Pöllänen 1997). Similar calculations are presented in publication VI for Ru-type particles detected in Poland.

For calculating the particle properties, the primary assumption is that nuclides of the same element behave similarly in a fuel fragmentation process. This makes it possible to estimate the amount of short-lived nuclides in the particle from the measured amount of long-lived nuclides. The activities of the nuclides that belong to other elements are included by assuming negligible fractionation between different non-volatile elements.

The calculations are performed as follows: the activity ratios of observed nuclides of the same element, such as $^{141}Ce/^{144}Ce$, are used first in estimating the burnup of the fuel from which the particle originated (see appendix H in Pöllänen 1997). The equivalent volume diameter of the particles is then estimated from the sum of the burnup-dependent activities of ^{95}Zr, ^{141}Ce and ^{144}Ce per unit volume. This size is then used to estimate the proportions of nuclides presented in Table IV, which are for some reason not detected. Usually, the reason is that the activity analyses were performed too late to detect short-lived nuclides.

The activities are computed by assuming a decay time of 1 d. If several activity ratios, e.g. $^{141}Ce/^{144}Ce$ and $^{103}Ru/^{106}Ru$, give different burnups, their average value is used. The fractionation of detected elements is taken into account in the computation. For example, the nuclides of ^{103}Ru and ^{106}Ru are enriched by a factor of about 5 in the particle detected by Rytömaa *et al.* (1986) (appendix H, Table H1 in Pöllänen 1997). This enrichment factor is then taken into account in estimating the proportions of the short-lived nuclides of Ru and Rh. If volatile nuclides, such as ^{131}I or ^{137}Cs, not mentioned in Table IV are observed in the particles, they are taken into account in the dose calculation. The other important nuclides of these elements are taken into account by assuming the same fractionation as for the observed elements.

The reported total activities of the particles are typically by a factor of 2 - 4 smaller than those obtained by calculations because of the presence of short-lived nuclides, mainly ^{239}Np (Table VII). The differences in the dose rates are approximately the same. The U-type nuclear fuel particles detected in north-western Europe after the Chernobyl accident and potentially deposited on the skin do not produce such a beta dose that the ICRP annual dose limit of 50 mGy at skin depth of 70 μm would be exceeded within 1 h. This is not the case for Ru particles (publication VI). However, nuclear fuel particles near the Chernobyl nuclear power plant were active enough to produce severe skin damage in a short time, provided the particles would have been deposited on the skin.

***Table VII**. The total activity of some U-type nuclear fuel particles detected in Europe after the Chernobyl accident and the beta dose rate that they may cause on the skin by assuming a decay time of 1 d (Pöllänen 1997). One particle is selected from each author and the particle code is that used by the original authors. A_{det} is the total activity of nuclides detected in the particles and $\dot{D}_{det}$ is the respective beta dose rate to the basal cell layer of the skin averaged over 1 cm^2. If daughter nuclides, such as ^{95}Nb, ^{106}Rh, ^{140}La and ^{144}Pr, are not reported by original authors they are included in the computation by assuming equilibrium with their parents. A_{all} is the calculated total activity of nuclides presented in Table IV including detected volatile nuclides and $\dot{D}_{all}$ is the respective basal cell beta dose rate averaged over 1 cm^2.*

Reference and location	Code	A_{det} (Bq)	A_{all} (Bq)	$\dot{D}_{det}$ (mGy h^{-1})	$\dot{D}_{all}$ (mGy h^{-1})
Rytömaa *et al.* 1986 Finland, Uusikaupunki	F2	430	1200	0.39	1.2
Saari *et al.* 1989 Finland, Uusikaupunki	U37(2)	620	2500	0.46	2.6
Devell 1987 Sweden, Studsvik	HP-9	5400	7400	4.5	7.3
Robertson 1986 Sweden, Älvkarleby	HP-3	3000	4500	2.4	4.6
Van der Wijk *et al*. 1987 Ukraine, Kiev	GHP1	1900	7400	1.4	7.2
Broda 1987 Poland, Mikolajki	M4	1900	7200	1.4	7.0
Balashazy *et al.* 1988 Hungary, Budapest	No 1	680	1700	0.68	1.8
Khitrov *et al*. 1994 Ukraine, Vil'cha	-	110000	450000	43	250
Salbu *et al.* 1994 Ukraine, near Chernobyl	No 1	2700000	29000000	1700	8800

6 SUMMARY OF THE RESULTS AND DISCUSSION

In the present thesis, nuclear fuel particles are investigated from the point of view of health hazards, the main focus being on their characterisation, their transport and dispersion in the atmosphere and the estimation of possible skin doses. The basic statement is that the release of nuclear fuel particles into the environment cannot be considered unique to severe accidents only (publication I, Pöllänen 1997). They were frequently identified in the environment after past nuclear accidents, especially in the Chernobyl accident, and in incidents from a number of other sources (Pöllänen 1997). Their possible release in future nuclear incidents cannot be totally dismissed.

The main results can be summarised as follows:

- Nuclear fuel particles should not be considered as 'becquerels' distributed homogeneously either in the environment or in a sample. Routinely used analysis procedures designed for bulk samples are not necessarily appropriate for nuclear fuel particles. Routine bulk analysis methods may lead to meaningless or even erroneous results. (Publications VII and VIII)
- The identification, isolation, and analysis of individual particles enables results to be obtained that are otherwise inaccessible owing to the interference of non-relevant bulk particles in the sample. Several complementary analysis techniques are needed to characterise particle properties thoroughly. The order in which different analysis techniques are used should be carefully thought through especially in the case when destructive analysis methods will be applied. (Publications VII and VIII)
- The characteristics of radioactive particles reflect the properties of the source material, which allows the performance of forensic analyses. Although complicated physical-chemical phenomena during the release may affect particle characteristics, it is possible to calculate the properties of individual particles in certain cases. These calculations are needed for the complete evaluation of the threats to health caused by nuclear fuel particles. (Publications VII and VIII)
- In a severe nuclear accident, large (aerodynamic diameter more than 20 μm) and highly active particles (activity even hundreds of kBq's) may be

transported hundreds of kilometres in the air before deposition. Effective release heights are then several hundreds of meters. (Publications II, III and V)

- The transport of particulate materials differs to that of gaseous species. Air parcel trajectories are not necessarily sufficient to identify the areas that may receive radioactive materials. Thus, in operational use particle trajectory or dispersion model calculations are needed. (Publications II, III and V)
- Realistic atmospheric conditions and the effects of turbulent dispersion must be taken into account in calculating the transport of radioactive particles should a nuclear incident occur. Simplified transport range calculations are adequate only in limited cases. (Publications II, III and V)
- In the Chernobyl accident, the effective release height may have been considerably higher than reported previously (up to 2 km) or particles may have been lifted up to higher altitudes in deep convective cells. (Publication V, Valkama and Pöllänen 1996)
- The composition of the particles may have an essential influence on skin doses. The presence of short-lived nuclides in particles emitted from low burnup fuel in particular contributes notably to skin beta dose rates. The contribution of gamma rays is often negligible. The self-absorption of beta particles in the nuclear fuel particle itself must be taken into account in dose calculations. (Publications IV and VI, Pöllänen 1997)
- The specific activity of the Ru-type particles found in the environment after the Chernobyl accident may be by a factor of about 100 higher than those composed mainly (in terms of mass) of bulk U. Ru-type particles are almost entirely composed of fission products. In a severe nuclear accident, reactors operating with high specific power may generate fuel particles that consequently may have high specific activity. (Publication VI, Pöllänen 1997)
- Contrary to particles composed mainly of bulk U, the specific activity of the Ru-type particles emitted from low burnup fuel may be considerable higher than that emitted from high burnup fuel. Thus, health threats are not necessarily the greatest for particles originating from high burnup fuel. (Publication VI, Pöllänen 1997)
- Even individual nuclear fuel particles, released uncontrolled into the environment in a severe nuclear accident, may represent an acute health hazard. When deposited on the body they may produce a high but much localised dose to the skin. Compared to the ICRP annual occupational dose limit for the public (50 mSv averaged over 1 cm^2 at a depth of 70 μm and intended to protect skin against deterministic effects) they may produce a

basal cell beta dose that exceeds this limit in a short time. This dose may be exceeded in 1 h provided that an Ru-type particle larger than 8 μm in diameter is deposited onto the skin (RBMK fuel assumed) whereas for U-type particles the dose refers to the diameter of about 30 μm. (Publication VI, Pöllänen 1997)

In order to assess the significance of the radioactive material that may be released into the environment with respect to radiation hazards, it is crucial to take into account the possibility that the material may be in the form of highly active particles. An awareness of this possibility is a prerequisite for taking appropriate countermeasures in a nuclear incident. As regards preparedness, the existence of a radiological hazard due to the presence of highly active particulate materials in a release plume must be realised and taken into account in contingency plans.

The possible release of nuclear fuel particles in the environment represents a technical, analytical, and even philosophical challenge for radiation protection. Their identification and detection in the environment necessitates properly designed environmental radiation monitoring and sampling systems that take the particulate nature of the releases into account. Traditional laboratory practices are designed for bulk sample activity analysis rather than for the analysis of the individual particles that have to be taken into account in estimating the relevance of the analysis results. Finally, the interpretation of the hazard, i.e. the possibility of receiving nuclear fuel particles deposited onto skin, which subsequently may produce high local doses, is far from clear with respect to radiological protection.

The Radiation and Nuclear Safety Authority (STUK 2001) has published a guide for the protective actions of members of the public and generic intervention levels to be applied in a state of a radiation emergency. This guide acts as a design basis for other authorities for weighting between different intervention operations. The principle is to prevent acute severe deterministic effects and to keep late stochastic effects as low as reasonably achievable. The practical question evokes whether the threat of nuclear fuel particles should be separately evaluated in the guide.

In a state of an acute radiation emergency, the recommended intervention actions such as sheltering and evacuation are based on the measurement of the external dose rate. Basic protective actions against hot particles are presumably appropriate in almost all practical situations. However, the

problem that highly active particles may be present in the air although the external dose rate is below the recommended operative action level (for example, the recommended external dose rate limit for sheltering is 100 μSv h^{-1}) is not only theoretical. The management of this situation requires special knowledge and equipment that are not necessarily available to the staff operating in field conditions. The possibility that highly active particles may serve as an additional health threat must be evaluated case by case based on expert judgement by the authorities familiar with radiation protection issues.

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Errata:

Publication II

Second sentence in the text of Fig. 3 should be: Figures (a), (b) and (c) represent effective release heights 100, 500 and 2000 m, respectively.

Reference [12]: The correct volume of Health Physics is 57.

Publication III

The text of Table 1: Uranium dioxide fuel mass is 219 000 kg.

Table 2: Half -life of ^{144}Ce is 284.3 d.

Discussion: The activity of single ruthenium particles found in Poland was even more than 100 kBq (Schubert and Behrend 1987).

Reference [19]: The year of publication is 1988.

Publication VII

Table 2: Half -life of ^{137}Cs is 30 y.

The second sentence of the text of Fig. 3 should be: The 'α particles' show the presence of alpha-active materials.