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# **Radiation exposure of human populations in villages in Russia and Belarus affected by fallout from the Chernobyl reactor**

**Measurements using optically stimulated luminescence in NaCl, TL-dosemeters and portable survey instruments**

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## Abstract

A quarter of a century has elapsed since the catastrophe at the nuclear power plant in Chernobyl. The radioactive fallout affected all the European countries and most severely the three countries of Belarus, Russia and Ukraine. In the aftermath of this devastating event, the scientific knowledge on the various radiological impacts of such a large scale accident and means to remediate these impacts has increased and will be essential for the future. The inhabitants that live in the most contaminated areas are, however, still exposed to elevated levels from the residuals of the Chernobyl fallout. This means that continued assessment of the long-term impact still is required to meet the concern of the affected people.

In this thesis, the current day exposure to inhabitants living in some of the villages in the Bryansk region (Russia) have been assessed using similar methods of individual monitoring as during earlier phases of the project (1991-2000). This included personal TLDs and individual whole body burden estimates of  $^{137}\text{Cs}$  by NaI(Tl) measurements. A method applicable for prospective as well as retrospective absorbed dose estimates has also been evaluated by using the optically stimulated luminescence in ordinary household salt. The dosimetric potential of salt was investigated in the laboratory with the aim to use salt as a retrospective dosimeter. The salt dosimeters were also tested for prospective *in situ* measurements together with TLDs and model estimates based on point measurements by means of a high pressure ionisation chamber and ordinary radiation protection instruments. These measurements were carried out at various (stationary) positions in a contaminated village in Belarus, not far from the villages in the Bryansk region.

Today the radiological importance is completely dominated by  $^{137}\text{Cs}$ . The contamination level of  $^{137}\text{Cs}$  varies significantly between different villages, within the villages and even within the gardens of individual residences. The external exposure is thus dependent on where an individual resides. The internal exposure is associated with the intake of forest food (mushroom, berries and game), rather than the contamination level within a specific village. As an average, the inhabitants in the Russian villages received a total annual effective dose due to Chernobyl caesium of  $0.5 \text{ mSv y}^{-1}$  and  $0.4 \text{ mSv y}^{-1}$  in 2006 and 2008, respectively. This corresponds to a reduction of more than 60% over 10 years. In 2006 and 2008 the internal component was 1/3 of the total effective dose and the remaining external component was around  $0.35 \text{ mSv}$ . As the rate of decrease of the external exposure is relatively stable the internal effective dose, which has a different temporal behaviour, is predicted to become of increasing importance in the future.

The comparative measurements in the Belarusian village showed a good agreement, and predicted an external effective dose of about  $1 \text{ mSv y}^{-1}$ . The overall results show that the effective dose to the inhabitants in the investigated area is now close to a level that is comparable to the natural background radiation. The study also shows that the potential of ordinary household salt as a tool for dosimetry is promising, also for prospective measurements *in situ* and at low absorbed doses and dose rates.

## Summary in Swedish

Människan har alltid exponerats för joniserande strålning från radioaktiva ämnen i naturen och i sin egen kropp, samt från kosmisk strålning. Summan av exponeringen från dessa strålkällor blir ungefär 1 mSv per år. Till detta kommer exponering av lungor och luftvägar från radon och radondöttrar, särskilt i inomhusluft. Som jämförelse kan nämnas att en tandröntgen ger en stråldos på ca. 0,01 mSv, en ”vanlig” röntgenundersökning 0,1-1 mSv och en CT-undersökning 1-10 mSv. Att man beräknar och försöker uppskatta stråldoser beror på att dessa ökar risken för att senare i livet drabbas av en cancersjukdom. Genom studier av sena hälsoeffekter hos individer som exponerats för höga stråldoser (överlevande från kärnvapenbombningarna i Hiroshima och Nagasaki) har man kommit fram till ett samband som visar att risken ökar linjärt med stråldosen.

Efter ett kärnkraftshaveri eller annan händelse som innebär en mera omfattande spridning av radioaktiva ämnen kommer man behöva vidta åtgärder för att reducera exponeringen och därmed hålla nere risken för framtida effekter på befolkningens hälsa. Det kommer också att finnas ett stort behov av att minska oron i samhället då den upplevda risken med joniserande strålning ofta är mycket större än den verkliga risken. Eftersom personer bland allmänheten inte normalt har egna dosmätare måste stråldoserna uppskattas i efterhand. Detta kan göras med hjälp av punktmätningar i omgivningen, mätningar av kroppsinnehållet av olika radioaktiva ämnen eller genom datormodeller. En annan metod som kallas för retrospektiv dosimetri utnyttjar en speciell minneseffekt som finns i vissa material och som gör att materialet minns om, och hur mycket, det exponerats för joniserande strålning. Genom att samla in föremål med speciella (fysikaliska) egenskaper (salt, mediciner i tablettform, mobiltelefonchips, armbandsur mm) som människor bär på sig eller som finns där de vistas kan man sedan genom att tömma föremålen på minnet, skapa en bild över strålningssituationen på dessa platser, i efterhand.

Katastrofen vid kärnkraftverket i Tjernobyl (Ukraina) 1986 gav upphov till en spridning av radioaktiva ämnen över ett enormt område (Vitryssland, Ryssland, Ukraina, Skandinavien och andra delar av Europa) och kom att påverka många miljoner människor. I de fyra arbeten som ingår i denna avhandling har strålningssituationen för invånarna i några av de mest radioaktivt kontaminerade byarna i Ryssland och Vitryssland utanför 3-milszonen studerats på olika sätt.

Radioaktiva ämnen i kroppen samt strålning från markbeläggningen har mätts på samma sätt som under en tidigare del (1990-2000) av projektet. Dessutom har nya dosimetrar bl.a. sådana som utnyttjar vanligt bordssalt använts. Resultaten från byarna visar att stråldosen varierar beroende på om man befinner sig utomhus (högst) eller inomhus (lägst) och vad man äter. De årliga dosbidragen från Tjernobyl ligger i dag på 0,5-1 mSv, motsvarande en minskning med en faktor (över) 60 sedan 1986. Upprepade mätningar under tre år i samma område har också bekräftat saltets potential som en retrospektiv dosimeter, även vid låga stråldoser. Detta skulle i framtida händelser innebära ett snabbt, enkelt och billigt komplement till annan dosimetri för att uppskatta stråldoser till personer bland allmänheten och därmed bidra till stöd i såväl information som fortsatta insatser för att mildra konsekvenserna av den joniserande strålningen och oron.

## Summary in Russian

Человечество всегда было подвержено воздействию ионизирующего излучения от радионуклидов, имеющих в природе и в организме самого человека, а также от космического излучения, постоянно действующего на Землю. Суммарное воздействие от этих источников составляет около 1 мЗв в год. Кроме того, легкие и дыхательные пути подвергаются воздействию радона и дочерних продуктов радона, присутствующих, в частности, в воздухе внутри помещений. Для сравнения, рентген зуба дает дозу примерно 0,01 мЗв, обычный рентген от 0,1 до 1 мЗв, а сканирование с использованием компьютерного томографа от 1 до 10 мЗв. Основанием для расчета и попытки оценки доз облучения является то, что они повышают риск отдаленного развития раковых заболеваний. На основании исследований отдаленных последствий для здоровья людей, подвергшихся воздействию высоких доз облучения (жертв ядерной бомбардировки Хиросимы и Нагасаки, многие из которых подверглись воздействию облучения свыше 200 мЗв) можно сделать вывод, что повышение риска находится в линейной зависимости от дозы облучения.

После аварии на ядерной установке или другого события, связанного с крупномасштабным рассеиванием радиоактивных материалов, требуется принять меры по уменьшению воздействия ионизирующего излучения на людей и, следовательно, не допустить повышения риска последующего воздействия на здоровье общества. Кроме того, в значительной мере потребуются снятие напряжения в обществе, так как зачастую предполагаемый риск ионизирующего излучения оказывается значительно серьезнее фактического риска. Так как отдельные представители общества обычно не располагают индивидуальными дозиметрами, то оценку доз облучения в таких случаях следует производить ретроспективно. Это можно делать посредством точечных измерений в окружающей среде, оценки содержания различных радионуклидов в организме человека или посредством компьютерного моделирования. Еще один метод, так называемая ретроспективная дозиметрия, использует специальный эффект запоминания, присущий определенным материалам, который позволяет материалам запоминать факты и интенсивность воздействия ионизирующего облучения. Собрав предметы, обладающие специфическими (физическими) свойствами (соль, медицинские препараты в виде таблеток, чипы мобильных телефонов, часы и т.п.), используемые людьми или находящиеся в их окружении, можно выделить память предметов в виде светового сигнала и воссоздать картину радиационной обстановки на месте в ретроспективе.

Авария на атомной станции в Чернобыле (Украина) в 1986 г. вызвала рассеяние радионуклидов на громадной территории (Беларусь, Россия, Украина, Скандинавия и другие европейские регионы) и оказала воздействие на многие миллионы людей. В рамках работы над этой диссертацией с применением различных методов исследования была изучена радиоактивная обстановка в местах проживания в наиболее загрязненных селениях в Беларуси и России за пределами 30-километровой зоны.

Для измерения радионуклидов в организме человека и излучения от поверхности почвы использовались традиционные дозиметры и другие детекторы таким же образом, как и в предыдущей части проекта (1990-2000). Кроме того, использовались новые дозиметры, включая модели, в которых применялась обычная столовая соль. Полученные в населенных пунктах результаты показывают, что доза облучения меняется в зависимости от того, производились ли замеры на открытом воздухе (выше)

или же внутри помещений (ниже), а также от того, что употреблялось в пищу. Годовая доза облучения от Чернобыля сегодня составляет 0,5 – 1 мЗв, что соответствует уменьшению в 60 раз по сравнению с 1986 г. Повторяющиеся измерения в течение трех лет в одной и той же местности также подтвердили потенциальные возможности обычной соли в качестве ретроспективного дозиметра, даже при малых дозах. В подобных ситуациях в будущем это может стать быстрым, простым и недорогим дополнением к другим дозиметрическим мероприятиям для оценки доз облучения обычных людей, способствуя информационной и непрерывающейся деятельности по смягчению воздействия ионизирующего облучения и напряженности в обществе.



## Original papers and preliminary reports

This thesis is based on the following papers, which will be referred to in the text by their Roman numerals.

- I. *Household salt as a retrospective dosemeter using optically stimulated luminescence*  
Bernhardsson C, Christiansson M, Mattsson S, Rääf C L  
Radiat Environ Biophys 2009;48(1):21-28
- II. *Comparative measurements of the external radiation exposure in a  $^{137}\text{Cs}$  contaminated village in Belarus based on optically stimulated luminescence in NaCl and thermoluminescence in LiF*  
Bernhardsson C, Matskevich S, Mattsson S, Rääf C L  
Submitted to Health Phys
- III. *In situ study of the small scale variability of the external radiation field in Russian and Belarusian villages after the Chernobyl accident*  
Bernhardsson C, Rääf C L, Mattsson S  
Manuscript
- IV. *Measurements of long-term external and internal radiation exposure of inhabitants of some villages of the Bryansk region of Russia after the Chernobyl accident*  
Bernhardsson C, Zvonova I, Rääf C L, Mattsson S  
Submitted to the Sci Total Environ

Preliminary reports were given at the following international meetings:

NKS-B status seminar, Oslo (Norway), 28-29 August, 2007. Bernhardsson C, Christiansson M, Zvonova I, Jesko T, Jakovlev V, Rääf C L, Mattsson S. *Internal and external doses to inhabitants of selected villages in the Bryansk region in Russia 20 years after the Chernobyl accident.*

Medical Physics in Baltic States 5, Kaunas (Lithuania), 5-6 October, 2007. Bernhardsson C, Christiansson M, Zvonova I, Jesko T, Jakovlev V, Rääf C L, Mattsson S. *The radiation environment in Bryansk villages: 20 years after Chernobyl.*

International conference on radioecology and environmental radioactivity, Bergen (Norway), 15 –20 June, 2008. Christiansson M, Bernhardsson C, Mattsson S, Rääf C L. *Optimization of read-out sequences for optically stimulated luminescence (OSL) of household salt for retrospective dosimetry.*

IRPA12, 12th International congress of the International Radiation Protection Association, Buenos Aires (Argentina), 19-24 October, 2008. Bernhardsson C, Christiansson M, Zvonova I, Jesko T, Jakovlev V, Rääf C L, Mattsson S. *Long-term radiation exposure of inhabitants in the Bryansk region in Southwestern Russia.*

IRPA12, 12th International congress of the International Radiation Protection Association, Buenos Aires (Argentina), 19-24 October, 2008. Christiansson M, Bernhardsson C, Rääf C L, Mattsson S. *Test of household salt read by optically stimulated luminescence (OSL) as a personal dosemeter.*

Medical physics in the Baltic states 7, 9-10 October, 2009, Kaunas (Lithuania). Bernhardsson C, Christiansson M, Rääf C L, Mattsson S. *OSL in household salt (NaCl) for environmental, occupational and medical exposure.*

Third European IRPA congress, 14-18 June 2010, Helsinki (Finland). Bernhardsson C, Matskevich S, Mattsson S, Rääf C L. *Indoor- and outdoor exposure in a Cs-137 contaminated village in Belarus measured using luminescence dosimetry (OSL in NaCl and TL in LiF).*

## Abbreviations

CNPP	Chernobyl nuclear power plant
$c_{specific}$	Specific luminescence
$C_v$	Coefficient of variation
CW-OSL	Continuous wave optically stimulated luminescence
$E_{ext}$	External effective dose
$E_{int}$	Internal effective dose
LM-OSL	Linearly modulated optically stimulated luminescence
MDD	Minimum detectable dose
OSL	Optically stimulated luminescence
OSLD	Optically stimulated luminescence dosimeter
SAR	Single aliquot regenerated-dose protocol
SD	Standard deviation
SEM	Standard error of the mean value
TL	Thermoluminescence
TLD	Thermoluminescent dosimeter
WBC	Whole body concentration



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## <sup>137</sup>Cs deposition over Europe after the Chernobyl disaster

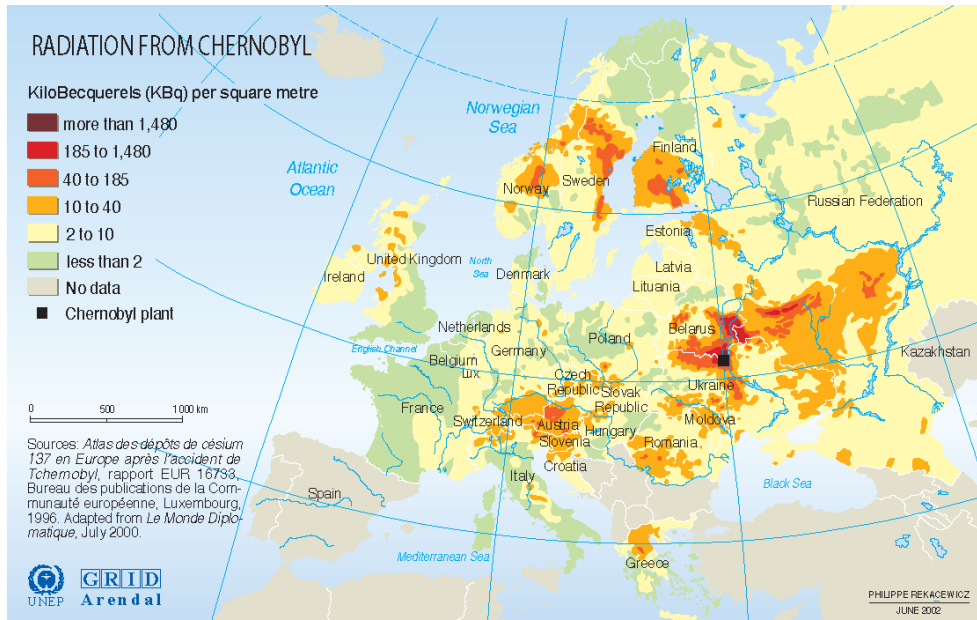


Figure 1. The total deposition ( $\text{kBq m}^{-2}$ ) of <sup>137</sup>Cs distributed over Europe as a consequence of the Chernobyl disaster [UNEP/GRID-Arendal 2007].

# 1. Introduction and aims

Humans have always been subjected to ionising radiation – natural and now also artificial. At some locations the natural level is elevated due to the composition of the ground and the soil, and due to high altitude. Artificial sources, like fallout from the previous testing of nuclear weapons, debris from the destroyed Chernobyl reactor etc. give rise to an additional exposure that varies a lot. Although the initial source may be confined to a limited area or surface, the radioactive substances can be dispersed over large areas. For example, a nuclear detonation in the atmosphere dispersed radioactive fallout all over the hemisphere where the detonations took place. Similarly, after a surface detonation of a nuclear device, or a nuclear reactor accident as in Chernobyl 1986, radioactive material is transported large distances from the source and hence, affects large areas and many people. The current unstable situation at the Japanese nuclear power plant Fukushima I (*Fukushima Daiichi*) with continuous leakages of radioactive material is another example.

There is today an increasing awareness of the potential threat of nuclear energy accidents. Another threat is the possible antagonistic use of radioactive materials and even the use of fissile material. To be able to minimize the damage of such events in terms of casualties, disturbances in the society, environmental and socio-economic effects, there is a need for preparedness programs including different means of assessing the exposure of the affected inhabitants.

When radioactive material is dispersed in the environment, simplified and generalised methods are needed to rapidly and efficiently mitigate the consequences of the event. In a severe scenario, persons with acute radiation syndrome have to be taken care of directly for medical treatment and continued follow-up. Inhabitants close to the source might have to be evacuated during a limited period (or for ever) to reduce health effects and others may be able to continue to live in the affected area.

In the initial phase after an event as described above, it is important to provide the affected persons with adequate information. However, when no direct radiation dose measurements are available, which is normally the situation for the general public, other methods will be needed to assess and confirm values of the cumulated absorbed dose of the affected people. For this purpose various techniques may be employed, which enable accumulated dose estimates retrospectively. Several such techniques are already in use and associated methods developed, such as *i*) detection of radiation induced biological effects on individual humans, *ii*) luminescent signals stored in different crystalline materials that are close to man and, *iii*) model calculations. Common to the retrospective dosimetry techniques is that they have stored some kind of response to ionising radiation that is proportional to the cumulated absorbed dose in the material.

After the initial phase of a severe incident, the mitigation process for reducing the health consequences must continue. Countermeasures for reducing the exposure of external as well as internal irradiation may be needed and the efforts of such must be balanced between the



social and economic costs of the adverted population dose. In the aftermath of a radiological and nuclear event it is also important to regularly monitor the radiation exposure of the inhabitants still living in the area and to continuously inform about the exposure situation. The exposure situation is influenced by physical factors such as half-life of the released radionuclides, weathering such as rain, snow and wind and human processes such as decontamination and methods used to reduce the intake of deposited radionuclides by the food. The radiation environment and the human exposure will therefore gradually change over time.

## 1.1 Objectives

The overall aim of this work was to investigate the radiation exposure of the population in villages of Russia and Belarus and to develop and test a method for retrospective determination of absorbed doses to affected individuals. It included the understanding of the major factors influencing the exposure in the long-term period after a nuclear power plant disaster such as the one in Chernobyl 1986. The specific objectives were:

- To develop a method for retrospective dosimetry by using optically stimulated luminescence in household salt by investigating the dosimetric properties of salt in the laboratory.
- To study the applicability of salt as a dosimeter *in situ*, as compared to conventional methods used for assessing radiation exposure.
- To study and evaluate the variability of the contamination pattern in the villages, 20-24 years after the deposition.
- To determine the radiation exposure of populations in villages in the Bryansk (Russia) and the Gomel (Belarus) areas and to study its variability in terms of external and internal radiation exposure by means of conventional and retrospective methods.

## 2. Background

### 2.1 Exposure from radiation in the environment

There are several sources of ionising radiation in the environment, both of natural and artificial origin. The natural sources can be divided into three main categories; 1) cosmic; 2) internal; 3) external. Cosmic radiation originates from our and other galaxies and mainly consists of protons (90%), the remaining 10% consisting of electrons, alpha-particles and heavier nuclides. When the cosmic radiation reaches the atmosphere a number of interactions create other elementary particles, mainly muons and neutrons as well as gamma radiation. Depending on latitude and height above the sea level the intensity of the cosmic radiation varies. The external sources, other than from cosmic radiation, are from naturally occurring radionuclides in the soil such as primordial  $^{40}\text{K}$  and decay products of uranium ( $^{238}\text{U}$  and  $^{235}\text{U}$ ) and  $^{232}\text{Th}$ . Common to the terrestrial radionuclides is that they all have long physical half-lives ( $10^8$ - $10^{10}$  y). The internal exposure is mainly from naturally occurring  $^{40}\text{K}$  ( $t_{1/2} = 1.25 \times 10^9$  y) within the human body and  $^{222}\text{Rn}$  and its daughters may also be considered as an internal source of ionising radiation when inhaled.

In Sweden the magnitude of the annual effective dose varies significantly between individuals, mainly due to the previously mentioned radon component. A general estimate assumes that a representative person of the Swedish population receives an average effective dose of 2.4 mSv per year [Andersson et al. 2007]. Included in this estimate is exposure from cosmic radiation ( $0.3 \text{ mSv y}^{-1}$ ) and potassium in the body ( $0.2 \text{ mSv y}^{-1}$ ), which are about the same for all persons. Higher individual variations are expected of the average dose contributions from natural radionuclides in ground and buildings ( $0.6 \text{ mSv y}^{-1}$ ), radon in the indoor air ( $0.2 \text{ mSv y}^{-1}$ ), artificial radionuclides in food ( $0.2 \text{ mSv y}^{-1}$ ) and medical exams ( $0.9 \text{ mSv y}^{-1}$ ). In certain cases much larger doses can be expected, above  $10 \text{ mSv y}^{-1}$ , for e.g. residents in houses with extreme levels of radon. For comparison, the average annual effective dose is  $2.4 \text{ mSv y}^{-1}$  globally (ranging from  $1$ - $13 \text{ mSv y}^{-1}$ ) and is composed of external contributions (terrestrial  $0.48 \text{ mSv y}^{-1}$ , cosmic  $0.39 \text{ mSv y}^{-1}$ ) and internal contributions (ingestion  $0.29 \text{ mSv y}^{-1}$ , inhalation  $1.26 \text{ mSv y}^{-1}$ ) [UNSCEAR, 2008].

Today there are still some areas in Europe with elevated levels of radioactive fallout from the Chernobyl release in 1986 that contribute to the external and internal effective dose to the population. The variation and the magnitude of this source in some of the most affected villages in Russia and Belarus are estimated and described in the present thesis.

### 2.2 The Chernobyl accident/disaster

On 26 of April 1986, Unit 4 at the Chernobyl Nuclear Power Plant (CNPP) in the Ukrainian SSR (now Ukraine) exploded during an unauthorised low-power engineering test. It was the worst nuclear power plant accidents/disasters ever, and is one of two events being classified

as a level 7 event on the International Nuclear Event Scale (INES<sup>1</sup>). The other one being the disaster at the nuclear power plant in Fukushima, Japan, following the earthquake and tsunami on 11 March 2011. At the CNPP a steam explosion ruptured the vessel of reactor 4, destroyed the reactor core and severely damaged the reactor building. Subsequently, the graphite moderator in the reactor caught fire and continued to burn for several days. In combination with the heat from the burning reactor, radioactive material (in the form of gases, condensed particles and fuel particles) was dispersed into the atmosphere. As the release to air took place for about 10 days [Buzulukov and Dobrynin 1993], with varying weather conditions, the fallout was dispersed unevenly over the former Soviet Union and in the rest of Europe (Fig. 1).

As a result of the accident, approximately 12.5 EBq of radioactive material (half of the amount being noble gases) and with a half-life more than one day, was released into the environment [IAEA 2001]. The composition of the fallout varied considerably during the active release due to variations in temperature and other parameters [Buzulukov and Dobrynin 1993]. The total inventory of noble gases was released as well as large fractions of the inventory of <sup>131</sup>I (50%-60%), <sup>134,137</sup>Cs (20-40%) with an average <sup>134</sup>Cs/<sup>137</sup>Cs activity ratio of 0.55, <sup>132</sup>Te (25-60%), <sup>89,90</sup>Sr and <sup>140</sup>Ba (4-6%) [OECD 2002]. In the initial period after the accident the dose rate in air was dominated by <sup>132</sup>Te, <sup>132</sup>I and <sup>131</sup>I [Golikov et al. 1993]. A schematic illustration of the temporal variation of the relative dose rate in air as measured over a contaminated area in Russia is shown in Fig. 2.

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<sup>1</sup> <http://www.iaea.org/Publications/Factsheets/English/ines.pdf>

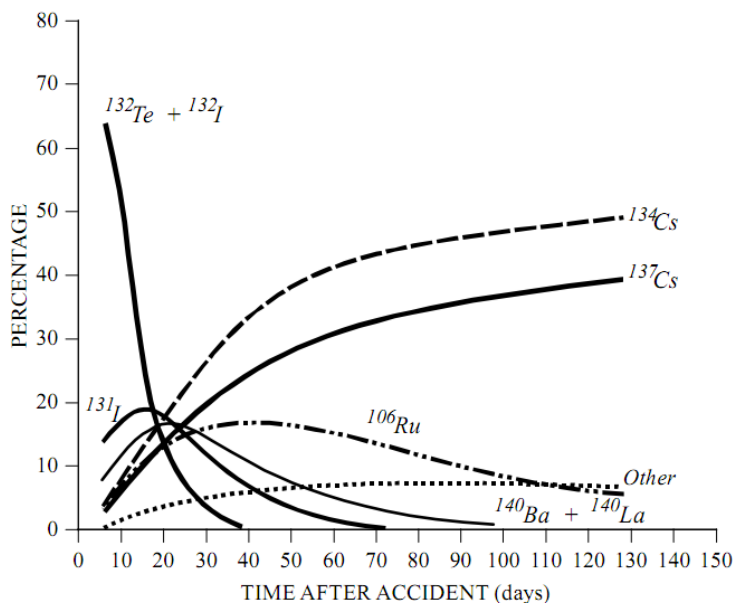


Figure 2. Relative contribution to the dose rate in air of various radionuclides from a contaminated area in Russia [Golikov et al. 1993].

In a few months after the accident the dose rate in air ways mainly attributable to  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  [Golikov et al. 1993]. Early activity measurements accomplished with specially equipped aircraft measurements defined the contaminated areas which were used to establish maps of the  $^{137}\text{Cs}$  contamination of the three republics of the former Soviet Union and Europe [Izrael 2007; De Cort et al. 1998]. Such maps were used to set the borders of the contaminated areas on the basis of the defined exposure limits. In the 1990s four contaminated zones were defined, in multiples of  $37 \text{ kBq m}^{-2}$  ( $1 \text{ Ci km}^{-2}$ ) of  $^{137}\text{Cs}$  contamination [EMERCOM 1996]. These zones have continued to play an important role in the decision-making processes regarding remedial and mitigating actions.

Although the contamination levels have been greatly reduced since 1986 there are today, 25 years after the event, still persons belonging to a so-called critical group that annually receives an effective dose above  $1 \text{ mSv}$  in the countries of Belarus, Russia and Ukraine. The results discussed in the papers of the present thesis will mainly concern the current-day situations, and hence dose contributions that predominantly originates from the long-lived fission product of  $^{137}\text{Cs}$ .

### 2.2.1 $^{137}\text{Cs}$

Caesium-137 is a fission product with a half-life of 30.2 years. The main (95%) decay is to  $^{137}\text{Ba}^{\text{m}}$  by beta emission and at the following deexcitation of  $^{137}\text{Ba}^{\text{m}}$  to the ground state, a  $661.7 \text{ keV}$  gamma photon is emitted. Caesium is chemically analogous to potassium which is an essential substance for humans, animals and plants. When ingested, caesium can substitute

potassium and will behave as the analogous nuclide when inside the human body. The caesium thus becomes distributed similarly as potassium, mainly in the intracellular fluid in muscles and organs, uniformly throughout the body. For a given intake of caesium it will be retained inside the body with a biological halftime of 2 days for 10% of the activity and 110 days for 90% of the activity, the latter half-life being much slower for women and for children the retention is mass dependent [ICRP 1989, 1997].

$^{137}\text{Cs}$  is of special importance in the human exposure as it is found in large amounts over a large area of Europe and will be present in the environment for many years to come. There are two main sources for the current inventory: atmospheric nuclear weapons tests carried out mainly in the 1950s and 1960s, and atmospheric releases from the Chernobyl accident. The relative contribution from these two sources varies geographically and depends also on the size of the area due to the inhomogeneous Chernobyl deposition. As a comparison, in the southern part of Sweden the ratio is about 1, and in the community of Gävle (central Sweden) which was the most affected area in Sweden by the Chernobyl fallout, the ratio is about 40 whereas it is between 400 and 1400 in the Gomel-Bryansk spot.

### 2.2.2 The Bryansk-Gomel spot

Two of the most heavily affected areas after the Chernobyl accident were the Bryansk region in south western Russia and the Gomel region in eastern Belarus, often referred to as the Bryansk-Gomel spot or the Bryansk-Belarus spot (Fig. 1). This spot is located about 200 km north-northeast from the CNPP and it was formed on April 28-29, 1986, when rain out of the passing plume of Chernobyl occurred. The  $^{137}\text{Cs}$  surface activity in the most heavily contaminated villages in this spot exceeded  $1.48 \text{ MBq m}^{-2}$ , equivalent to  $40 \text{ } \mu\text{Ci m}^{-2}$  (classified as a zone of resettlement [EMERCOM 1996]). The area is characterised by rural settlements where the population to a large extent uses locally produced foodstuff. The soil types of the area are predominantly turf-podzol sandy and sandy-loam, characterised by low natural fertility, high acidity and low mineral content [Balonov 1993]. These soils have an extraordinarily high transfer of caesium from soil to plants [OECD 2002].

The cumulated exposure from the Chernobyl fallout to the population in the Bryansk-Belarus spot can be divided into two main components: a) sources in the air and b) sources on and in the ground.

- a) *Sources in the air:* As long as the plume is moving the irradiation geometry is isotropic in the  $2\pi$  direction above ground (when the sources are distributed uniformly in horizontal directions). Material is removed from the plume by precipitation or as dry deposition. Resuspension of previously deposited radionuclides may to some extent also contribute to the dose in air.
- b) *Sources in the ground:* The depth distribution of radionuclides deposited on the ground from 'sources in the air' varies depending on the deposition pattern. Initially, dry deposited radionuclides are often situated on the surface whereas wet deposited radionuclides often lead to a deeper penetration into soil already at the deposition event. After some time, the depth distribution may be described by an exponential function.

In the initial (critical) period after the accident sources in the air were the most significant contribution to the dose rate. After a few weeks the ground deposition of caesium dominated the dose rate.

### *2.2.3 Long-term monitoring program*

Large scale radiation monitoring programs have been conducted since 1986 by different organizations of the former Soviet Union. After the disintegration of the Soviet Union it was up to the individual countries of Belarus, Russia and Ukraine, respectively, to continue to monitor the affected population. As a support to these measurements and to provide the population with independent dose estimates from researchers outside the former Soviet Union, a joint Soviet-Nordic monitoring program was initiated in 1990 by the late professor P. Ramzaev. Some years after launching the project it became a joint Russian-Swedish program. The external and internal annual effective doses were measured and estimated on a yearly basis from 1990 to 2000 (except in 1999) [Erkin et al. 1994; Wallström et al. 1995; Thornberg et al. 2001, 2005]. The measurements were always carried out during late summer in rural villages in the Bryansk area. All villages were similar in size and soil contamination level, although it was not possible to visit the same villages every year. Parallel programs in this area were also conducted (e.g. Fogh et al. 1999; Fesenko et al. 2000; Golikov et al. 2002; Ramzaev et al. 2006).

In 2006 the Swedish-Russian program was re-initiated. The ambition was to revisit the same villages as during previous years, using similar methods and equipment for the assessments. The effective doses more than 20 years after the accident were investigated in 2006 and 2008 and compared to the previous period from 1986 to 2000 (**Paper IV**). The highly inhomogeneous radiation environment in the area is highlighted in **Paper III**. Since 2008 similar studies have been carried out in a village in the Gomel area in Belarus (**Paper II**).

## **2.3 Effective dose and methods to estimate its magnitude**

There are two types of quantities used for radiation protection: protection quantities [ICRP 2007] and operational quantities [ICRU 1993]. The risk related protection quantities are not directly measurable, but can be estimated by conversions from operational quantities, which can be determined using various types of radiation detectors. In this section the approach used to link those two types of quantities is described.

### *2.3.1 Operational and physical quantities*

The International Commission on Radiological Protection (ICRP) has specified protection quantities for the purpose of reducing the occurrence of health effects due to exposure of ionising radiation. As tissues and organs have varying sensitivity to radiation and as the biological effect is different for different radiation qualities, the effective dose,  $E$ , is defined (Eq. 1) as the weighted sum of the average absorbed dose,  $D_{T,R}$ , in the specified organ or tissue,  $T$ , due to radiation of quality,  $R$ :

$$E = \sum_T w_T \sum_R w_R D_{T,R} \quad \text{Eq. 1}$$

where  $w$  are the weighting factors, determined mainly from epidemiological data. As the effective dose is not directly measurable a set of operational quantities have been defined by the International Commission on Radiation Units and Measurements (ICRU) for area and individual monitoring which are designed to give an (over-) estimate of the protection quantities. The operational quantities can be related by the physical quantities fluence,  $\Phi$ ; air kerma,  $K_{air}$ ; and absorbed dose in air or tissue,  $D$ .

For practical radiation protection measurements ICRU [ICRU 1993] have defined three measurable quantities; ambient dose equivalent  $H^*$  (e.g. the calibration of ionisation chambers, GM-tubes), directional dose equivalent  $H'$  (e.g. GM-tubes) and personal dose equivalent  $H_p$  (e.g. personal dosimeter). As the human body absorbs and scatters the incident radiation the definition of the measurable quantities are defined based on the ICRU-sphere. In this terminology the ambient dose equivalent is defined as the dose equivalent at a depth of 10 mm in the ICRU-sphere and the directional dose equivalent at a depth of 0.07 mm in the ICRU-sphere. In the definitions there is a condition on the quantities that  $H^*$  is independent of the incident angle of the radiation whereas  $H'$  varies with the angle of incidence. The personal dose equivalent  $H_p$  is defined in soft tissue at the depth  $d$  under a certain point of the body, normally 0.07 mm (relevant for soft penetrating radiation) and 10 mm (deeper penetrating radiation), respectively. The personal dose equivalent depends on the angle of the incident radiation. The definitions leads to that  $H^*(10) \geq H_p(10)$ .

The conversion that links the measurable and risk related quantities have been extensively investigated (ICRP 1996; ICRU 1998). The dose distribution in the human body after exposure to ionising radiation depends on several factors such as the incident radiation (type of radiation, energy and angular distribution) and the exposed body (size, orientation). Thus, the complex absorbed dose distributions and dose-related quantities are often approximately resolved by computational models using mathematically described anthropomorphic phantoms and Monte Carlo transport codes [e.g. Jacob et al. 1986; Zankl et al. 1988; Saito et al. 1998]. Such studies show that the body size (i.e. baby, child, adult) affects the conversion factors significantly [ICRU 1998]. It is assumed that the build-up effect of incident gamma photons with energies of about 100 keV, is responsible for an enhanced absorbed dose near the body surface. In combination with a reduced shielding of organs in a smaller body this gives rise to large difference in  $E$ ; 80-90% higher at 50 keV and less than 40% higher above 100 keV, comparing infant to adult [Saito et al. 1998]. At energies above 0.2 MeV, the photon cross section does not change significantly and the conversion coefficients for a given body size does not vary much up to about 1 MeV. The conversion coefficients are given for a limited number of whole-body irradiation geometries of the incident photons. For an infinite plane source of deposited radionuclides on the ground, the photons at 1 m above the ground come from an angle of incidence just below the horizontal plane, which is close to a rotational invariant geometry (ROT) (Jacob et al. 1986). Due to body self-shielding, conversion coefficients for other irradiation geometries will be lower, except for anterior-posterior and posterior-anterior geometries. Hence, as the activity migrates down in the soil the irradiation

geometry changes from ROT to a situation when the gamma rays are coming from the lower hemisphere of space which results in a higher self-shielding of the body and the conversion coefficient will thus decrease. Recently, Golikov et al. (2007) published results from measurements on anthropomorphic phantoms in the Bryansk region (carried out in summer-autumn 1991-1993). This study confirmed studies by Eckerman et al. (1993), that the conversion coefficients for a plane source at the air-soil interface (fresh deposition in the open) and a semi-infinite volume source homogeneously contaminated (multiple ploughed layer of soil) differ not more than 10% in the energy range from 0.1 to 2 MeV.

### 2.3.2 Estimating the radiation exposure

Occupational exposure is usually monitored by some sort of standardised dosimetry system with active or passive dosimeters. The exposure is registered and stored for comparison with the dose limits recommended by the ICRP [ICRP 2007]. The individual exposure of the general public is however, not monitored. After an event that disperses radioactive material in the environment, such as a major nuclear power plant accident, the radiation doses to the members of the public might be highly elevated and there will be a need to monitor and to follow-up a number of individuals. For the purpose of supplying the public with information and for the assessment of health effects when conventional dosimetric data is unavailable or inadequate, alternative methods are used to assess or verify the dose, retrospectively. Below is a short summary of approaches to assess absorbed doses to the general public.

Prospective measurements: To determine the exposure from *external irradiation* to individuals and population groups, individual dosimeters (e.g. TLDs, film dosimeters, electronic dosimeters) may be used. For such assessment, the dosimeters are distributed to a representative group of individuals and are then carried by the persons throughout the measuring period, normally during a few weeks to months. This requires that the participant carries the dosimeter throughout the measuring period and that the dosimeters are treated properly. Hence, this technique is rather time consuming and requires many dosimeters to be distributed to give an accurate estimate of the average doses. By the use of different conversion coefficients the dosimeter reading can thereafter be converted to a corresponding effective dose.

There are many methods to determine the body burden of radionuclides in humans and from that estimate the *internal exposure*. *In vivo* methods with direct gamma counting is a convenient and fast method with either a well-shielded whole-body counter [Mattsson et al. 1989; Lebedev and Yakovlev 1993] or with semi-mobile equipment with various degree of shielding (or without shielding) [Palmer 1966; Zvonova et al. 2000]. Another method is by measuring the radionuclide contents in urine for assessing the body burden and effective dose [Räaf et al. 1999].

Retrospective measurements: When no dosimeters were worn during the main exposure phase the acquired absorbed dose may still be estimated, retrospectively. There are several different biological and physical methods described in the literature for this purpose (see e.g. the review by Ainsbury et al. (2010)). The common requirement of these is that the indicator or the material should have a reproducible (biological, chemical or physical) signal response



to ionising radiation, preferably over a wide range of absorbed doses. This signal should also have a low (or known) fading and it is desirable if the sampling is non-invasive and the read-out is rapid. Among the methods of biological dosimetry there are three main techniques; chromosome aberration analyses, micronuclei analyses and somatic-mutation analyses. The time from sample receipt to dose estimate is often in the order of hours to days and the detection limit varies from 0.1 Gy to 1 Gy. Among the physical techniques, much attention have been focused on electron paramagnetic resonance (EPR) of tooth enamel which has shown several promising properties, such as a signal stability of  $\sim 10^6$  years and a detection limit of about 100 mGy [Fattibene and Callens 2010]. One of the drawbacks of using tooth enamel is the invasive sampling of tooth material and hence other materials (e.g. sugar, plastics, hair and nails) have also been suggested for use with EPR, albeit with a considerably higher detection limit ( $> 2$  Gy), compared with tooth enamel. The EPR technique also requires relatively expensive equipment and qualified personal to operate it. Stimulable luminescence of certain materials is another technique for retrospective dose estimations. Luminescence dosimetry, mainly thermoluminescence (TL), is widely used for prospective dosimetry for occupational exposure by TLDs. Optically stimulated luminescence (OSL) is another method to retrospectively estimate absorbed dose from e.g. building materials, personal belonging and everyday used items. An advantage of this technique is that the useful dose range is relatively wide (from about 0.03 Gy to several Gy) and the read-out is simple and inexpensive. A drawback is, however, that these materials must be shielded from light exposure in order not to lose the signal. As an accuracy of 50% for a retrospective dose estimate is considered sufficient for an individual assessment [ICRU 2002], different techniques should be combined as a complement to each other in different exposure scenarios, in order to give the most accurate estimate of the true exposure. Considerable uncertainty is associated with the relation between the measured signal in the media and the corresponding dose in organs and tissues, which still is an area for development in retrospective dosimetry.

In terms of internal exposure, the effective dose may be estimated from repeated measurements of the whole body content or of the excretion of radionuclides in urine and faeces combined with retention models such as the Integrated Modules for Bioassay Analysis (IMBA) software. Alternatively the internal effective dose may be assessed by estimating the daily intake of radionuclides. This intake can be estimated by evaluating the known radionuclide contents in the consumed food through e.g. food enquiries, or by model calculations based on information on the total ground deposition and known transfer factors from soil to plants, however, these estimates tend to overestimate the actual body content [UNSCEAR 1988; Balonov and Travnikova 1993].

Calculations/modelling: Models are aimed to give an estimate of the reality and certain assumptions and/or simplifications are needed to make the calculations feasible. Two different approaches are generally used: analytical dose reconstruction and numerical dose reconstruction. They are both similar in the sense that they use input data from *in situ* measurements in the area of interest. The difference is how the input data is processed together with other parameters used to describe the exposure situation.

### 3. Material and methods

In the four papers constituting the base for this thesis, dosimeters of lithium fluoride (LiF) and sodium chloride (NaCl) have been used. These luminescent detectors have been used for the estimation of the external exposure in combination with a variety of ordinary radiation protection instruments for comparative assessment. The luminescent detectors were used to study the time integrated radiation levels in the surveyed areas, whereas the radiation protection instruments have provided an instantaneous dose rate reading on-site. For the estimation of the internal exposure the whole body contents of caesium have been measured in residents of the surveyed area.

The radiation levels in the environment and the exposure of individuals have been measured to estimate annual external and internal effective doses to members of the public in the Bryansk (Russia) and Gomel (Belarus) regions. The approach adopted and the instruments used are briefly described below.

#### 3.1 Villages included in the study

In **Papers II-IV** *in situ* measurements have been carried out in a number of villages in Russia and Belarus (Table 1). They were all located in the Bryansk-Gomel spot, a few tenths of km apart, and had an initial contamination level of  $^{137}\text{Cs}$  ranging from 0.8 MBq m<sup>-2</sup> to 2.7 MBq m<sup>-2</sup>. These levels can be compared to the highest levels found in Sweden at the same time, in the Gävle area, where the average level was 0.085 MBq m<sup>-2</sup> [Edvardsson et al. 1991; SGU 2005] over an area of some hundreds of square kilometres.

*Table 1. Some characteristics of the rural villages in Russia and Belarus (Svetilovichi), visited during the surveys.*

	Demenka	Starye Bobovichie	Stary Vishkov	Veprin	Yalovka	Svetilovichi
Mean $^{137}\text{Cs}$ deposition in 1986 [MBq m <sup>-2</sup> ]	1.2	1.1	1.3	0.9	2.7	0.8
Countermeasures	N.D. <sup>a</sup>	P.D. <sup>b</sup>	P.D. <sup>b</sup>	N.D. <sup>a</sup>	F.D. <sup>c</sup>	F.D. <sup>c</sup>
Population in 2008	300	1050	523	N/A	620	~1000

*a) N.D. non-decontaminated (no systematic measures have been taken to reduce the radiation level).*

*b) P.D. partly decontaminated (roads (mainly unpaved) were covered with gravel).*

*c) F.D. fully decontaminated (topsoil around public buildings have been removed and the roads covered with asphalt as well as cleaning of some residential buildings and gardens).*

The villages were located in the Bryansk-Gomel spot between the two towns Novozybkov and Klintsy in Russia and the city of Gomel in Belarus. After the Chernobyl catastrophe, a wide range of countermeasures were initiated to protect the population from radiological impacts on their health [IAEA 2006]. Direct evacuation of the inhabitants of the most heavily

affected areas in the former Soviet Union was one of the major measures, which included the relocation of over 500,000 people during May and June 1986 [Balonov 1993]. The other measures can be separated in methods of reducing the external as well as internal exposure. The external exposure was reduced, apart from resettlement, by e.g. cleaning of residential areas (with special attention to public areas/buildings), cover roads with gravel and asphalt. The internal exposure was reduced by e.g. providing non-contaminated food, information on consumption and preparation of certain foodstuffs, measures for reducing transfer of caesium and strontium radionuclides from soil to plants and providing clean fodder to livestock.

Large scale decontaminations were performed between 1986 and 1989 in the most contaminated cities, towns and villages of the former Soviet Union. About one thousand settlements were treated during these years. After 1989 the large scale decontaminations were stopped, but continued on a smaller scale. The three countries Belarus, Russia and Ukraine decided individually in the mid-1990s, to continue such measures [Jacob et al. 2001]. In Belarus it was decided that optimised radiation protection measures are to be carried out in settlements with an expected average annual effective dose of 1-5 mSv. In Russia and Ukraine the remedial action level of an additional annual dose that exceeds 1 mSv was adopted.

## 3.2 Methods for external effective dose estimations from personal dosimeters

### 3.2.1 *Luminescence dosimetry*

Luminescence is the electromagnetic radiation emitted as a consequence of an atomic or molecular excitation. Materials with luminescence properties have the ability to store energy and later release part of that energy as light. An illustration of the simple band model showing the principles for the thermally and optically stimuable processes that generate the luminescence is shown in Fig. 3.

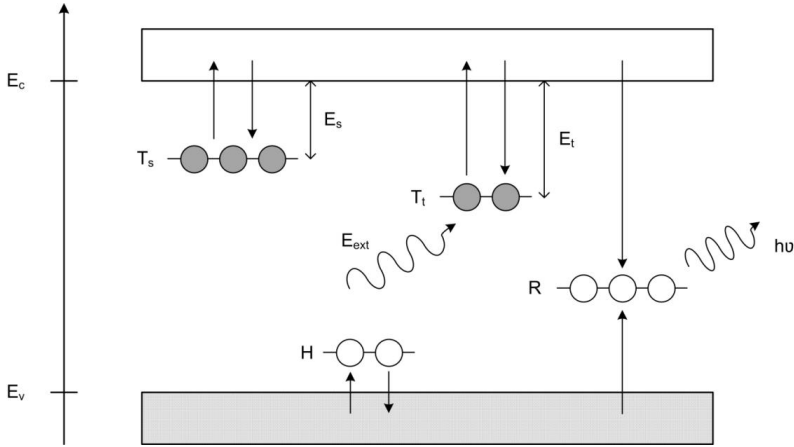


Figure 3. Simple band model illustrating the TL and OSL processes. The valence band at energy level  $E_v$  is separated from the conduction band at energy level  $E_c$  by an ideally forbidden gap, containing intermediate states at energy levels  $E_s$  (shallow trap,  $T_s$ ),  $E_t$  (deeper trap,  $T_t$ ) and recombination centres  $H$  and  $R$ .

When ionising radiation is absorbed by an insulator or semiconductor, free charge carriers are produced which can become trapped in the crystal defects of the material. In Fig. 3, the valence band at energy level  $E_v$  is separated from the conduction band at energy level  $E_c$  by a forbidden gap, where electrons can become trapped at the intermediate states (defects)  $T$  and  $H$ . The shallow level at  $E_s$  is not a stable trap and the probability of the electrons to escape is large even at room temperature (detrapping by lattice vibrations). The deeper trap at  $T_t$  is stable and electrons may be stored for shorter or longer times. In order for these electrons to escape the trap, stimulation by external energy is needed which may release the electrons to the conduction band. Thermal stimulation is referred to as thermoluminescence (TL) and light stimulation is referred to as optically stimulated luminescence (OSL). Part of the released electrons recombines with holes at  $R$  and luminescence is emitted.

The property making such materials interesting for dosimetry is that part of the energy deposited in the material, when exposed to ionising radiation, is stored. The amount of energy stored is proportional to the absorbed dose, up to a certain level, and can be released as luminescence (light) if the material is stimulated by additionally supplied energy (heat or light). Some materials exhibit both TL and OSL properties but are generally more suitable and optimised for one of the stimulation modes. An advantage of OSL over TL is that only<sup>2</sup> trapping levels most sensitive to light are sampled in OSL which correspond to the charge population most efficiently zeroed [Thomsen 2004]. Another advantage is that no heating to high temperatures is required and hence, materials with OSL active defects that do not tolerate heating can still be used as a luminescent dosimeter by OSL.

<sup>2</sup> This is valid for unheated materials, which are zeroed by light exposure rather than heat.

For the purpose of prospective dosimetry several commercial configurations of both TL (e.g. LiF- and CaF<sub>2</sub>-based) and OSL (e.g. Al<sub>2</sub>O<sub>3</sub>-based) dosimeters are available. For the use of TL and OSL as a tool for retrospective dosimetry, it is preferable to use materials that are close to man, such as daily used items. Many different materials have been suggested for this purpose such as bricks from buildings, memory chips from different origins (telephones, ID-cards, cash and credit cards), ceramics from electronic devices, tooth enamel, house and workplace chemicals [Göksu and Bailiff, 2006; Correcher et al., 2009; Göksu, 2003; Bassinet et al., 2010; Inrig E et al., 2008; Dewitt R et al., 2010; Thomsen et al., 2002]. The focus in this thesis will be on ordinary household salt, which mainly consists of sodium chloride (NaCl), **Papers I, II and IV**.

### 3.2.1.1 Thermoluminescent dosimeters (TLDs)

The thermoluminescent effect has been thoroughly investigated and was first suggested as tool for dosimetry in the 1950s [Daniels et al., 1953]. Some of the suitable features are their small size, low price and absence of any need for electric supply or service in the field. There are many materials that emit luminescence during thermal heating, but only a few have properties suitable for dosimetry. It is desirable to have a linear dose response over the range of absorbed doses of interest, a high signal per unit absorbed dose, a stable signal (low fading) and low variation in signal per unit absorbed dose for various energies on the energy of the incident radiation. Although the requirements are not fulfilled by all TL dosimeters there is a wide variety of thermoluminescent dosimeters (TLDs) for use as a standard for occupational and environmental monitoring. Some of the key TL materials and their dosimetric properties can be found in e.g. Kortov (2007).

TL dosimeters are natural or synthetic materials. The historically most commonly used are crystalline materials of lithium fluoride (LiF) and calcium fluoride (CaF<sub>2</sub>), doped with different activators to improve the dosimetric properties. The LiF dosimeters have an effective atomic number ( $Z_{\text{eff}}$  of 8.14) which is close to air ( $Z_{\text{eff}} = 7.64$ ) and tissue ( $Z_{\text{eff}} = 7.42$ ). Hence, these dosimeters are often used for personal dosimetry purposes. LiF dosimeters are available with a variety of dopants, e.g. LiF:Mg,Ti and LiF:Mg,Cu,P and also by enrichment of <sup>6</sup>Li isotopes (thus being sensitive to neutrons). During the early phase of the long-term monitoring program in the Bryansk region, TLDs based on CaF<sub>2</sub> were used, with an at that time 10-30 times higher sensitivity compared to TLDs based on LiF:Mg,Ti. Recent development using LiF:Mg,Cu,P have increased the sensitivity of the LiF based dosimeters 10-30 times compared to LiF:Mg,Ti [Sáez Vergara 1999], thus now being as sensitive as the CaF<sub>2</sub> dosimeters but with the additional advantage of a better matching energy response compared to tissue. In the **Papers II and IV**, dosimeters of LiF:Mg,Ti (Harshaw TLD-100) and LiF:Mg,Cu,P (TLD Poland MCP-N) have been used. The LiF:Mg,Ti dosimeters were the same as in the previous surveys in the Russian villages [e.g. Thornberg et al. (2005)]. However, in the early period of the project (first three years) measurements were carried out with dosimeters of CaF<sub>2</sub> from the Norwegian Radiation Protection Authority [Wøhni 1995] and dosimeters based on LiF from Institute of Radiation Hygiene, St. Petersburg. An intercomparison between the dosimeters from the different laboratories showed a maximal difference of 4% of the mean value [Wøhni et al., 1991].

The LiF chips had the dimensions of  $3.2 \times 3.2 \times 0.9 \text{ mm}^3$  and were always put in pairs of two chips in special dosimeter holders of polymethyl methacrylate (PMMA) that consisted of two identical  $27 \times 58 \times 4 \text{ mm}^3$  plates. These holders were designed to be carried by the participants in a string around the neck, like a necklace, for a period of about one to two months.

### 3.2.1.2 Optically stimulated luminescence using NaCl

The OSL technique was initially applied in dating applications of quartz from sedimentary samples [Huntley et al. 1985]. Later the technique gained an increasing attention in areas of personal, environmental and retrospective dosimetry as well as dosimetry in radiation therapy [Bøtter-Jensen et al., 2003]. The principles are similar to that of TL, with the difference that the stored signal in the OSL material is released when the material is illuminated instead of heated as in TL.

In **Papers I, II and IV** the optically stimulated luminescence from ordinary household salt was studied and the methods were applied to retrospectively estimate the varying radiation exposure in the Russian and Belarusian villages. The dosimetric properties of five different brands of salt were investigated in the laboratory and the read-out sequence optimised. Four of the salts were common household salts obtainable in most of the groceries in Sweden and were sold in light-tight packages. Analytically pure (pro analysi) NaCl was also studied. All five salts were (visually) similar in terms of size and shape distribution of the salt grains, but differed somewhat in terms of additives and origin (sea salt, mine salt). Table 2 shows the physical properties of the five salts investigated.

*Table 2. General characteristics of the five salts investigated.*

Salt #	Name	Type	Additives	Grain size <sup>a</sup> [μm]	Package <sup>b</sup>
1.	NaCl	Pro analysi	None	450-300	Plastic
2.	Falksalt	Sea salt	Anti-caking agents	500-300	Plastic
3.	Jozo	Sea salt	Anti-caking agents	>500-300	Paper
4.	Falksalt	Mine salt	Anti-caking agents, potassium iodine (0.005‰)	450-300	Paper
5.	Falksalt	Mine salt	None	>500-300	Paper

a) The given size corresponds to the average grain size in a given package after sieving the salt. No sieve larger than  $500 \mu\text{m}$  was available but no individual grains were visually larger than about  $600 \mu\text{m}$ .

b) Other packages are available for some of the salts, but the ones used in the present study are shown in the table.

The salt was read in a TL/OSL reader (TL/OSL-DA-15; Risø National Laboratory, Roskilde, Denmark). This reader was also used for the TL read-outs of some of the TLDs used in **Paper II**. The TL-OSL reader system is based on three units: the read-out unit, a control unit (miniSYS) and a PC. The miniSYS controls the reader-out unit and allows for the communication of a regular PC to the reader-unit. The reader is in turn composed of a few main components: calibration source, stimulation light source, light detection system, a sample carousel and a heater. A schematic illustration of the reader is shown in Fig. 4.

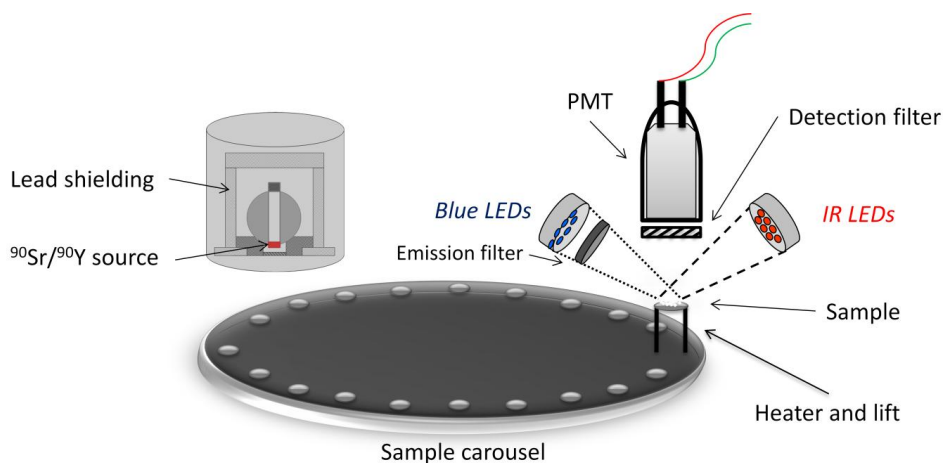


Figure 4. The Risø TL/OSL-DA-15 reader and the main components; PM-tube, light stimulation source, heating element,  $^{90}\text{Sr}/^{90}\text{Y}$  calibration source and the sample carousel.

On the sample carousel there are 48 positions where sample material can be put on special aluminium discs/cups ( $\text{Ø} = 9.7 \text{ mm}$ ). A sensitive analytical scale (ADAM PW254, ADAM Equipment Co Ltd, United Kingdom) was used to portion an amount of  $5.0 \pm 0.5 \text{ mg}$  of salt on each cup. This amount of specimen (hereafter referred to as an aliquot) was found to be appropriate as it covers a single layer of about half of the cup. In **Paper I**, 24 individual aliquots were used for each individual measurement whereas five individual aliquots were used in the later *in situ* measurements presented in **Papers II** and **III**. A study on the variability among equally treated aliquots showed that 24 aliquots were required to obtain a mean value of the response with an uncertainty less than 5% (standard error of the mean value, SEM). This amount of aliquots per sample was too high to be practically achievable when the dosimeter kits were used *in situ*. Therefore, a smaller amount of five individual aliquots were used to represent one dosimeter kit. This increased the uncertainty (SEM) of each assessed dose to slightly below 15%.

On the sample carousel each cup is transported to the position of the read-out, where a lift elevates the sample to distance of 55 mm from the photomultiplier tube (PMT) cathode. The lift includes a heating element made of Kanthal to provide efficient heating to the sample. Thermal heating is feasible up to  $700 \text{ }^\circ\text{C}$  at linear rates from  $0.1$  to  $30 \text{ }^\circ\text{C s}^{-1}$ , thereby also making TL-read-outs possible. For OSL-purposes the heating element is used to pre-heat the sample in order to reduce luminescence from unstable traps (which might recombine at room temperature and generate an unpredictable signal contribution during the read-out). This also has the advantage of possibly increasing the OSL signal by thermal transfer<sup>3</sup>. An elevated temperature is also used during the read-out phase, in combination with the light stimulation. The read-out parameters were adjusted for the purpose of estimating low absorbed doses, on

<sup>3</sup> Thermal transfer is in this sense a process where trapped charges are transferred from light-insensitive traps into OSL traps.

the basis of own measurements (**Paper I**) and others [Bailey et al. 2000; Thomsen et al. 2002; Zhang et al. 2005; Christensen 2007].

The optical stimulation was carried out by 28 blue LEDs (NISHIA, Japan) arranged in four clusters between the sample and the PMT. The wavelength of the stimulation light is  $\lambda=470\pm30$  nm and the total power density of the 28 LEDs is  $>40$  mW cm<sup>2</sup> at full power. A green long pass filter (GG-420) is used in front of the LEDs to reduce the intensity of the spectrum tail in the detection window (centred at  $\lambda=365$  nm). Similarly, another detection filter (Hoya U-340), which has a peak transmission around  $\lambda=340$  nm, is used in front of the PMT in order to reduce scattered stimulation light from reaching the PMT.

The reader unit was originally equipped with a 1.48 GBq <sup>90</sup>Sr/<sup>90</sup>Y calibration source giving a dose rate of 0.094 Gy s<sup>-1</sup> at the sample irradiation position. To obtain a reproducible absorbed dose to the sample an irradiation of at least 10 s was suggested [Christiansen 2007] corresponding to a relatively high absorbed dose. To be able to study the dose response in NaCl at doses down to a few mGy, a <sup>60</sup>Co-beam at Skåne University Hospital, Malmö, was utilized. The same unit was also used for the calibration of the personal TLDs used at the hospital and hence, had a traceable calibration to the Swedish Radiation Safety Authority. This unit allowed for the calibration with dose rates from about 20 µGy s<sup>-1</sup>. Consequently the low dose calibrations were carried out using the external <sup>60</sup>Co beam, and at higher absorbed doses the internal <sup>90</sup>Sr/<sup>90</sup>Y source was used. After a given absorbed dose to the salt, it was kept in darkness for 24 h until read-out in order to minimize fluctuations in the signal due to recombination of electrons in unstable traps.

When an OSL material is stimulated by monoenergetic light, luminescence with a different wavelength (usually higher than the stimulation light) is emitted. Different sources with different wavelengths of the stimulation light can be used and the stimulation is normally carried out in one of three different modes: continuous-wave OSL (CW-OSL), linearly modulated OSL (LM-OSL) and pulsed OSL (POSL). The CW-OSL stimulation mode, was adopted for all OSL measurements in the present papers. During CW-OSL the sample is stimulated with a constant light intensity and the emitted luminescence is registered during the whole stimulation duration. As the stimulation light is about 10<sup>8</sup> orders of magnitude higher than the luminescence from the sample [Thomsen 2004], this stimulation mode requires special filters to discriminate between luminescence and stimulation light. The emitted light decrease with stimulation time, as the OSL traps become depleted, and a characteristic OSL-decay curve is obtained. In Figure 5, CW-OSL and LM-OSL decay curves are plotted from two samples of NaCl of the same brand and package.



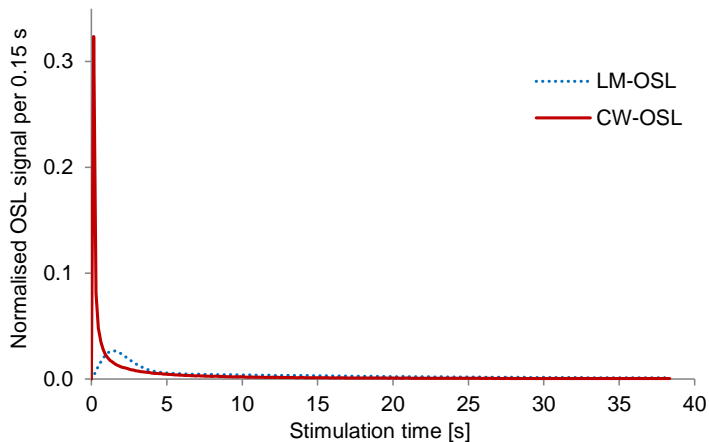


Figure 5. OSL signal (normalised to the 40 s integrated signal) for CW- and LM-OSL decay curves of salt (5.5 mg) from the same brand and package (salt #5 in table 2) after 160 mGy  $^{90}\text{Sr}/^{90}\text{Y}$  irradiation.

In LM-OSL the power of the stimulation source is linearly increasing, from zero up to a specified level of the total power. In this mode the intensity of the OSL signal will increase with increasing power up to a certain level, after which the intensity will decrease as the OSL traps becomes depleted. One of the advantages with this stimulation mode is that it is possible to identify OSL originating from different traps, since traps with different photo-ionisation cross-section will appear at different stimulation intensities. The third stimulation mode, POSL, is similar to CW-OSL with the difference that during POSL the stimulation source is switched off when the luminescence is collected. As this technique is based on time discrimination between stimulation and emission light there is no need for optical filters when using POSL.

In **Paper I**, the read out parameters were optimised in terms of stimulation mode, power of stimulation source, temperature during pre-heat and stimulation as well as the heating rate between these. The parameters were adjusted to a combination that achieved the highest luminescence per unit absorbed dose. With the optimised parameters the sensitivity of the salts were studied in the absorbed dose range from 1 mGy to 9 Gy. During these calibrations, the integration of the OSL-decay curve was relatively wide (40 s) to include also the slow component at high absorbed doses. In **Papers II** and **IV** the expected doses were much smaller than the comparable high calibration doses used in **Paper I** and therefore, a smaller integration of 20 s was chosen to make the read-out faster.

The main signal of the OSL decay function is here referred to as  $S_{\text{gross}}$  (counts). For the calculation of the absorbed dose to the salt, different approaches have been used in **Paper I** compared to **Papers II** and **IV** as the dosimetric conditions were slightly different. Throughout the laboratory calibrations, salts were taken from its original jars, standing inside a cupboard. The samples were always treated in darkness. Therefore, a background signal,

$S_{bkg}$ , was always present within these salts, corresponding to the absorbed dose since the salt was last exposed to light i.e. when it was purchased or packed at the factory. Hence, this background signal was determined for each aliquot of unirradiated salt,  $S_{bkg}$ , and subtracted from the total signal,  $S_{gross}$ . The difference refers to the net signal stemming from an exposure, e.g. from a given calibration dose in the laboratory or from exposures *in situ* in light-sealed packages. To reduce variations among equally treated samples, an average value of (normally) 24 aliquots were used for a given calibration dose and the signals were normalised to the sample mass according to Eq. 2 to get the net OSL signal,  $S_{net}$  (counts  $\text{mg}^{-1}$ ).

$$S_{net} = \bar{S}_{peak} - \bar{S}_{bkg} = \frac{\sum_{i=1...n} \left( \frac{S_{gross}}{m} \right)_i}{n} - \frac{\sum_{j=1...h} \left( \frac{S_{bkg}}{m} \right)_j}{h} \quad \text{Eq. 2}$$

where the first term is the average gross signal over the OSL-peak  $\bar{S}_{peak}$ , normalized to the weight,  $m$ , of each sample  $i$ . The sum of the weight normalized signals is averaged by the number of measured aliquots  $n$ . Similarly, an average background signal,  $\bar{S}_{bkg}$ , from  $h$  aliquots, is also determined and subtracted from  $\bar{S}_{peak}$  to obtain the net OSL signal,  $S_{net}$ .

The signal to dose response was determined for five different brands of household salt, including analytical grade salt (**Paper I**). Average signals at doses from 1 mGy up to 9 Gy were determined for all salts. The dose-response relationship, for absorbed doses  $D$ , was described in terms of specific luminescence,  $c_{specific}$  (counts  $\text{mg}^{-1} \text{mGy}^{-1}$ ):

$$c_{specific} = \frac{\partial S}{\partial D} \quad \text{Eq. 3}$$

The specific luminescence and the theoretical minimum detectable dose (MDD) were used as the main parameters for comparing the dosimetric properties of the salts investigated. An estimate of the MDD (mGy) of the five salts was calculated based on the variance of the natural background signal (Eq. 4).

$$MDD = 3 \cdot \sigma(S_{net,bkg}) \cdot c_{specific}^{-1} \quad \text{Eq. 4}$$

This theoretical estimate is applicable to the salts used in **Paper I** and is considered as an optimistic estimate since no other uncertainties have been considered that may potentially increase the MDD. The major uncertainties to the OSL-signal are related to the sample weight and variations between equally treated samples (i.e. luminescent properties due to variations in grain size and shape). Others uncertainty contributions are variations in luminescent properties between salt from different packages of the same brand on the market (i.e. storage conditions and duration).

A slightly different approach was used for assessing the OSL signal after the *in situ* measurements, as the conditions were optimised to be as beneficial as possible for low dose assessments. Two factors were changed from the laboratory situation: 1) the salt was completely bleached (zeroed) before the measurements were initiated and 2) the salt was kept in special dosimeter kits (sealed from ambient light and humidity). During these conditions,

$S_{gross}$  was estimated as during the laboratory conditions although a smaller region of interest (ROI) of the OSL-decay curve was used to define the OSL signal. The net signal was then determined as  $S_{gross}$  minus a background signal corresponding to the average area under two equally large ROIs over the tail of the OSL-decay curve.

### 3.2.2 Calibration of TL and OSL dosimeters

Both the LiF and NaCl dosimeters were calibrated in a similar manner using  $^{60}\text{Co}$  beams at either Sahlgrenska University hospital in Gothenburg or Skåne University hospital in Malmö. Both sources produced an air kerma rate traceable to the secondary standard laboratory at the Swedish radiation protection authority (SSM). The LiF-chips were individually calibrated in terms of absorbed dose to tissue in a  $20 \times 20 \times 7 \text{ cm}^3$  polymethyl methacrylate (PMMA) phantom. Another, but similar phantom was used for the calibration of the NaCl. The NaCl was positioned in an even layer of a few mm, in the dedicated phantom yielding an identical irradiation geometry to the one used for the LiF-chips.

### 3.2.3 Uncertainty estimate

When assessing the effective doses by TL dosimeters (TLDs) or OSL dosimeters (OSLDs) long time after the fallout was dispersed, and in distant areas, there are a number of uncertainties associated with the estimates. The major one is the background signal contributions to the total dosimeter reading.

OSLD: When the salt is used in pre-manufactured dosimeter kits and is taken from the same jar of salt, the situation is quite different from the one expected in an emergency situation when samples of household salt must be collected in kitchens of affected residents. However, given the situation in our study where a known salt brand with a pre-calibrated dose response is used, a considerable uncertainty of the estimated signal is still prevalent. This uncertainty depends on several components; the weight of the samples/aliquots was determined by the uncertainty of the analytical scale with an accuracy of 0.2 mg, corresponding to 4% of the typical weight of the aliquot. The aforementioned variability in the signal response between equally treated aliquots was in terms of standard error of the mean of five aliquots, less than 15%. The gross OSL signal was reduced by a background signal, to account for the dose received during transportation and storage. This dose contribution was estimated based on measurements at a limited number of positions but the magnitude is assumed to vary at different positions with about 20%. A further uncertainty is introduced if one would aim to determine the signal contribution from the deposition of artificial radionuclides *in-situ*. To do so, the signal stemming from natural background radiation and other components which do not originate from the deposition must be corrected for, and this correction introduces an additional uncertainty of about 15%.

For the brand and package of salt used in the field studies, the individual calibration coefficient,  $c_{specific}$ , was determined with an estimated uncertainty of 10% (including the uncertainty of the  $^{60}\text{Co}$  calibration). The final effective dose calculation is also associated with an uncertainty in the chosen conversion coefficients, which was estimated to 5% [Wøhni et al. 1995]. An unknown error is however introduced if applying this coefficient to an OSL signal

which has not been completely corrected for the signal contributions of the natural background radiation acquired *in-situ*, since the coefficient by Wøhni et al. (1995) refers to a specific *in-situ* deposition geometry mainly associated with radiocaesium fallout. If the signal contribution from the Chernobyl fallout is significant in proportion to the remaining signal contributions this error is assumed to be less than 20%.

**TLD:** The mean relative difference in the signals from the two TL-chips of the same dosimeter holder was 5% for both the LiF:Mg,Ti chips and the LiF:Mg,Cu,P chips. The uncertainty of the  $^{60}\text{Co}$  calibration was 3%. For the personal TLDs two background corrections were carried out, one for the pre-Chernobyl background and one for the transportation background dose. An uncertainty of 15% was used for the pre-Chernobyl background and a slightly higher, 20% uncertainty, was assumed for the transportation background. The measured signals in the TLDs were converted by the same means as the OSLDs and hence introduced an additional uncertainty of 5%.

Thus, by square summation of the components into a total uncertainty of the estimated effective dose to adults in the Belarusian village, one arrives at 37% and 22% when using the stationary OSLDs and TLDs, respectively. The corresponding uncertainty of the personal dosimeters used in the Russian villages is 26%.

As an example of the personal dosimeters in the Russian villages, a typical dose reading corresponded to 320  $\mu\text{Gy}$  over a total measuring period of 131 days, which includes storage and transportation during 64 days. The estimated total dose during transportation and storage during this period was 100  $\mu\text{Gy}$ , and with an assumed pre-Chernobyl background of 45  $\mu\text{Sv month}^{-1}$  ( $\times 2.1$  months), this gives an estimate of the effective dose of  $220 \times 0.92-95 = 107 \pm 28$   $\mu\text{Sv}$ .

### 3.3 External effective dose estimations

#### 3.3.1 Area monitoring using TL and OSL dosimeters in a rural village in Belarus

External effective dose estimates were carried out in the village of Svetilovichi (Belarus) using OSLDs and TLDs as well as by measurements of the ambient dose rate using a high pressure ionisation chamber and model calculations. The dosimeter measurements were carried out in three successive years 2008-2010, during about 2.5 month. The external doses were studied in de-contaminated and non-decontaminated areas as the village had partly been subjected to a series of decontamination programs. In 1991 a systematic decontamination was carried out in the village which included cleaning of roads and outside public buildings, i.e. the kindergarten and the hospital. Thereafter, in 1999, and as part of the IAEA regional project *RER/9/059* (Andersson et al. 2001), Svetilovichi was selected as a village for practicing and improving the existing techniques of decontamination. Up till 2006, about 50 houses and gardens have been decontaminated according to the program and today, all of the most heavily contaminated houses have been decontaminated.

The dosimeter kits used during the three years were treated similar to the method described above for the personal TLDs. As there are no direct conversions from estimated air kerma to effective dose,  $E$ , for OSLDs in salt, an approximate calculation was carried out by using the same conversion as for TLDs. This was considered the best achievable method for assessing a comparable effective dose of the dosimeters. The linear attenuation of LiF and NaCl are similar, with a difference of less than 5% in the photon energy range from about 100 keV to 2 MeV (National Institute of Standards and Technology). Hence, the same approach as described in *section 3.3.3* was applied for these dosimeters as well.

The dosimeter kits used in Svetilovichi consisted of the same holders as the ones used for the personal dosimeters in the Russian villages but they were light-sealed and put in plastic bags filled with silica gel. The dosimeter kits were put together during the evening before they were put in position in the village. To minimize unwanted signal before the dosimeter kits were distributed the TL-chips were kept in a lead jar (22 mm thick walls) and the salt was exposed to sunlight (during several hours). On transportation back to Sweden it was not possible to store the dosimeter kits shielded from ambient radiation and therefore, ambient survey meters (SRV2000 and GR110, see *section 3.3.2* below) were used along the travel route to estimate the dose during transportation.

### *3.3.2 Radiation protection survey instruments*

In recent years there has been a rapid development in the area of dedicated equipment for radiation protection and so called homeland security purposes. This is a development that has been nourished by the increasing awareness and fear of nuclear and radiological threats such as nuclear accidents/incidents and terrorist actions with improvised nuclear devices (dirty bombs). In **Papers II-IV** a selection of radiation protection instruments (provided from the Swedish Radiation Safety Authority (SSM)) has been used. These detectors, apart from the ionisation chamber, are today a part of the Swedish radiation emergency response organisations, which includes seven laboratories distributed all over Sweden. Below is a summary of the key features of the detectors used. The instruments were either calibrated or verified with test sources before the *in situ* measurements.

*GR110*: The most frequently used among the selected portable instruments in the survey was the GR110 instrument (SAIC, Exploranium, Canada). The GR110 was used during all surveys and at all positions of the stationary dosimeters. This instrument is based on a 75 cm<sup>3</sup> NaI(Tl)-detector which measures the photon fluence in the energy range from ca. 45 keV to 3 MeV. This instrument was used for its rapid response to changes in the radiation environment and long operational usage without any peripheral devices.

*RSS-131ER*: A high pressure ionisation chamber HPIC was utilized as reference and calibration instrument in the village of Svetilovichi in October, 2010. It is a Reuter Stokes model RSS-131ER (General Electric Energy, USA) which operates with 8 litres of argon gas at a pressure of 25 atmospheres in a spherical volume yielding a 4 $\pi$ -geometry with less than 2% reduction in the signal on the upper side of the detector. Ion-pairs are produced in the gas when ionising radiation interacts in the chamber. The ion pairs are then collected at the electrodes due to an electrical potential. A current is produced and the signal converted to

ambient dose equivalent  $H^*$ . The energy response is relatively flat in the energy range from 0.2 MeV to 4 MeV, but the ability to measure gamma falls off below 60 keV due to attenuation by the ion chamber walls and its external casing. The HPIC was positioned at different locations on a tripod using a height of 1 m above the ground. Also the heights 0.5 and 1.5 m were used as well at some reference locations to detect the vertical gradient in gamma intensity. The integration time of the ionisation chamber signal was 240 s and was repeated during 20 min to achieve a variation of the mean signal of about 2%.

*SRV2000*: A regular dose rate meter, based on two energy compensated Geiger Müller (GM) tubes. The SRV2000 (Mirron Technologies (RADOS), Finland) was mainly used during transportation and storage of the dosimeters, when they were not positioned or worn in the villages. This instrument is calibrated to display the signal as ambient dose equivalent and has also the ability to accumulate absorbed doses over the time it is in operation, thereby yielding an estimate of the transportation and storage dose to the dosimeters.

*GR135*: A multipurpose instrument that uses a GM-tube for dose rate estimates when the instrument is operated in search mode. The GR135 (SAIC, Exploranium, Canada) has the ability to separate gamma radiation by energy with a 65 cm<sup>3</sup> NaI(Tl) detector. The energy resolution (FWHM) for this detector is less than 9.0% at 661.6 keV (primary photon energy of <sup>137</sup>Cs). The instrument was used in the village of Svetilovichi to get a better understanding of the variation in the radiation environment at different positions in the village. The spectroscopic information was used to compare the gamma energy fluence between disturbed and undisturbed areas, and between indoor and outdoor surfaces in houses.

*Model calculations*: Effective dose estimates to population groups based on individual TLDs and/or OSLDs are time consuming, expensive and dependent on the desire and possibility of the participants to wear the dosimeters during the measuring period. Alternative to such individual measurements are therefore needed. In an attempt to model population doses and to compare to estimates based on TLD and OSLD measurements the model described in Eq. 5 [IAEA 2006] was used in **Paper II**. This model incorporates variations in the radiation field and differences in human behaviour in those fields, as well as a conversion from measurable values to health related parameters i.e. external effective dose,  $E_k$ .

$$E_k = \sum_i OF_{i,k} \cdot CF_{i,k} \int \dot{D}(t) \cdot LF_i(t) \cdot dt \quad \text{Eq. 5}$$

where  $OF_{i,k}$  is the occupancy factor for location  $i$  which incorporates the relative amount of time spent on location  $i$  by individuals of the  $k$ th population,  $CF_{i,k}$  is a dose conversion factor from measurable to risk quantities,  $\dot{D}(t)$  is the dose rate over an undisturbed reference area and  $LF_i$  is a location factor for area  $i$  which accounts for the dose rate reduction in the area compared to the reference area. In this model the effective doses for persons of different ages, occupations and living conditions may be estimated based on a series of reference measurements. Naturally, there is an uncertainty in the choice of the location and occupation factors which is difficult to estimate. In an attempt to overcome this, Golikov et al. (2002) carried out a survey in a few rural villages of in the Bryansk area where the inhabitants estimated the relative time spent in different locations. In **Paper II** these coefficients were

adopted together with *in situ* measurements in different locations using the HPIC and a conversion coefficient from  $H^*$  to  $E$  of  $0.7 \text{ Sv Gy}^{-1}$  [ICRP 1996]. The model calculations were generalised to an adult population living in wooden or brick houses, respectively, with indoor or outdoor work, respectively.

The model estimates were compared to the stationary dosimeters of NaCl (OSLDs) and LiF (TLDs) positioned inside and outside different houses in the village. The relative occupancy inside and outside was the same as those determined by Golikov et al. (2002), although restricted to the positions of the dosimeters. The calculated annual effective doses were also adjusted for the shielding effect from snow cover during the winter months, by applying a dose rate reduction factor of 0.72, taken from Ramzaev et al. (2006), during the months with snow cover.

### 3.3.3 Personal TL and OSL dosimeters in some villages in Bryansk

The external effective dose estimates previously carried out in the Russian villages were based on individual TLDs. Two TL-chips in each dosimeter holder were used to establish an average value for the individual dosimeter. In **Paper IV**, the same dosimeter holders and principles for assessing the effective dose was used: when the dosimeters are carried by the participants they will measure the absorbed dose to the body surface,  $D_{\text{surface}}$ . Hence a conversion to effective whole-body dose,  $E$ , has been estimated from the personal dosimeters according to Eq. 6.

$$E = D_{\text{surface}} \cdot (K_{\text{air}}/D_{\text{surface}}) \cdot (E/K_{\text{air}}) \quad \text{Eq. 6}$$

where  $K_{\text{air}}$  is the air kerma. The two conversion factors  $K_{\text{air}}/D_{\text{surface}}$  and  $E/K_{\text{air}}$  have been kept the same as during previous years, in view of that the product of these two are relatively stable [Wøhni, 1995]. Hence, a value of  $1.11 \text{ Gy/Gy}$  was used for the  $K_{\text{air}}/D_{\text{surface}}$  ratio [Jacob et al. 1988] and  $0.83 \text{ Sv/Gy}$  for the  $E/K_{\text{air}}$  ratio [Jacob et al. 1986] for the adults ( $>15 \text{ y}$ ) wearing dosimeters. As a part of the Russian-Nordic project the  $E/D_{\text{surface}}$  ratio was estimated *in situ*, using phantom measurements, yielding a value of  $0.92$  [Golikov et al. 2007] for adults (confirming the product of the two above mentioned conversion factors). A ratio of  $0.95 \text{ Sv/Gy}$  was used for the  $E/D_{\text{surface}}$  ratio for school children, based on measurements by Golikov et al. (1999).

Before the dosimeters were put together the evening before they were distributed to the participants, the TL-chips were stored in a lead jar. After the measuring period and until read-out, the dosimeters were stored in Russia for a few weeks or up to months (in 2008). During this storage and transportation, a background signal was added to the dosimeters,  $D_b$ . This additional dose was calculated according to Eq. 7.

$$D_b = \frac{t}{T} (D_{\text{TL}} - D_f) + \dot{D}_{\text{tr}} \cdot (T - t) \quad \text{Eq. 7}$$

where  $D_{\text{TL}}$  is the total dose to the reference background dosimeters,  $D_f$  is the dose accumulated during air transportation and  $\dot{D}_{\text{tr}}$  is the average dose rate during transportation outside the lead jar. The time denoted by  $t$  is the time the TL-chips were inside the lead jar

whereas the total time between calibration and read-out is denoted by  $T$ . A background correction for pre-Chernobyl background was also used. A dose rate for this component of  $45 \mu\text{Sv month}^{-1}$  (terrestrial and cosmic radiation) was adopted based on the estimates by Fogh et al. (1999). To estimate the annual effective dose from the dosimeters the shielding effect from snow during winter months was accounted for by multiplying the effective dose by a factor of 0.94 (the same factor as in Thornberg 2000).

### 3.4 Variability of the contamination level

Much effort have been put into the field of explaining and mapping the vertical migration of radionuclides in the environment e.g. [Mattsson et al. 1975; Finck 1992; Isaksson and Erlandsson, 1995] and the horizontal distribution e.g. [Finck 1992; Haugen 1992; Golosov et al. 1999; Izrael 2007; Farfán et al. 2011]. The horizontal distribution have mainly been mapped and investigated on large scales with the purpose of identifying contaminated areas for the future remediation and people protection.

A substantial amount of the deposited fallout from Chernobyl was released from the passing plume in combination with rainout. It is evident from maps, such as the one in Fig. 1 that the contamination level became unevenly distributed over Europe and smaller areas such as the Gomel-Bryansk spot. In an attempt to confirm the variability on an even smaller scale, a scale of a few meters to centimetres, a major mapping of some key village areas was conducted in parallel to the visits of the Russian and Belarusian villages. Included in this survey were measurements inside and outside almost fifty houses and gardens (dwellings, schools, administration buildings and hospitals) as well as in different relevant areas between these locations. Apart from the stationary dosimeter kits and the individual (mobile) TLDs, these measurements were carried out by the radiation protection instruments described in section 3.3.2. Measurements with the same GR110-instrument were carried out at all position where the other detectors, or dosimeters, had been positioned as well as other positions to get a denser sampling of measuring position. Hence these measurements serve as a relative reference to the other measurements carried out during the study period. In 2010, a calibration curve was therefore developed for this instrument by measurements *in situ* (in Belarus) with a high pressure ionisation chamber. As the measurements used for this cross-calibration was carried out in the same radiation environment as the other measurements, the calibration of the GR110 to the ionisation chamber can be assumed to be valid for all of the GR110 measurements in the area.

### 3.5 Internal effective dose estimations

Internal effective dose estimates were carried out in the Russian villages presented in Table 1. These estimates were conducted in 2006 and in 2008, like the external effective dose assessments, as a continuation of the long-term monitoring program previously (1990-2000) carried out in the Bryansk villages. The internal effective doses of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were estimated by *in vivo* measurements using portable  $63 \text{ mm}(\varnothing) \times 63 \text{ mm NaI(Tl)}$  detectors (ORTEC) coupled to a digiBASE (ORTEC). After 1994, mainly  $^{137}\text{Cs}$  was measured as the contribution from  $^{134}\text{Cs}$  then was less than 3% due to its much shorter half-life ( $t_{1/2} = 2.06 \text{ y}$ ).



Inhabitants of all ages and sexes were invited to get their body concentration of caesium estimated. The measurements were done by placing the NaI(Tl) detector in the knee towards the abdomen in a so-called Palmer geometry [Palmer 1966]. The measured individuals were representative for the rural population as the villages were all located within a few km from each other. In each village, an in-field laboratory was established in one of the public buildings (i.e. school or administration building) with a good shielding from the outside radiation.

The measured activity of caesium in the body,  $Q_{Cs}$  (Bq), can be expressed as in Eq. 8.

$$Q_{Cs} = (N - k_b \cdot N_b) \cdot K \quad \text{Eq. 8}$$

where  $N$  is count rate from  $^{137}\text{Cs}$  when the person is measured,  $N_b$  is the count rate in the absence of a person (free in air), and  $k_b$  is a shielding factor correcting  $N_b$  for the shielding effect of the body. The last term,  $K$ , is a calibration factor for the detector which is linearly related to body dimension and mass of the measured person. During these conditions, Thornberg (2000) estimated the minimum detectable body burden to 1 – 2 kBq.

Based on the measured level of caesium,  $Q_{Cs}$  (kBq), the individual effective dose from external exposure,  $E_{int}$ , was calculated. For the calculation of  $E_{int}$ , the age dependent metabolic and dosimetric parameters from the ICRP publications 56 and 67 [ICRP 1989, 1993] were used according to Eq. 9 [Zvonova et al. 1995].

$$E_{int} = r(m) \cdot Q_{Cs}/m \quad \text{Eq. 9}$$

where  $r$  ( $\text{mSv kg kBq}^{-1} \text{ y}^{-1}$ ) is a dose rate coefficient for the total content of  $^{137}\text{Cs}$  in a person with body mass  $m$  (kg).

## 4. Results and discussion

### 4.1 Optimisation of OSL read-out sequences for the use of NaCl as a retrospective dosimeter (**Paper I**)

The read-out sequence for the optically stimulated luminescence in NaCl, using blue LEDs in CW-OSL mode, was optimised for various types of commercially available salts and analytical grade NaCl. The stimulation mode and power of the light source was optimised to a combination giving the highest luminescence per unit absorbed dose for a given amount of salt. A moderate stimulation power corresponding to 40% of maximum power density of the LEDs ( $20 \text{ mW cm}^{-2}$ ) was found to be the most appropriate setting for the stimulation source. At this power the highest luminescence yield per sample mass was achieved [Christiansson et al. 2008]. The choice of the pre-heat temperature was done based on the measured OSL- and TL-peaks of the salt and by other similar studies on NaCl [Bailey et al. 2000; Thomsen et al. 2002; Zhang et al. 2005; Christiansen 2007]. The proposed protocol to use for achieving the highest luminescence per unit absorbed dose, from the various salts, is shown below.

1. Collect samples of salt that has been exposed to background radiation (and/or radiation from an accident) or a calibration dose.
2. Pre-heat to  $220^\circ\text{C}$ , using a temperature increase rate of  $2^\circ\text{C s}^{-1}$ , and then keep the max temperature fixed for 10 s.
3. Irradiate with blue LEDs with  $20 \text{ mW cm}^{-2}$  for 40 s at a temperature of  $100^\circ\text{C}$ .
4. *Apply a previously done calibration or continue the process by applying a calibration dose from the internal source of the set-up and then repeat the above process for different calibration doses (in step 4).*

The four investigated brands of commercially available salt, as well as the chemically pure NaCl, exhibited slightly different TL and OSL properties. An example of the OSL and TL curves of salt is shown in Fig. 6. All salts except *Falksalt fint bergsalt* (the one later used during the *in situ* measurements) had a notable TL-peak at about  $100^\circ\text{C}$ . This peak was emptied (and partly photo transferred) during the  $220^\circ\text{C}$  pre-heat before the OSL was carried out in these salts. The shape of the OSL decay curve varied with absorbed dose and type of salt. At high absorbed doses the decay of the OSL-curve is extended and the shape becomes a function of two components: one initial fast and thereafter a slow component. At very low doses the slow component is not possible to discern from the background signal from electronics and ambient noise and hence has the shape of one defined peak, emptied within  $\sim 1$  s. It was also observed that the OSL-decay curves were slightly different for the different brands of salt, in terms of decay rates. This effect was however not possible to discern at low absorbed doses (less than about 100 mGy) when the fast component was dominating.

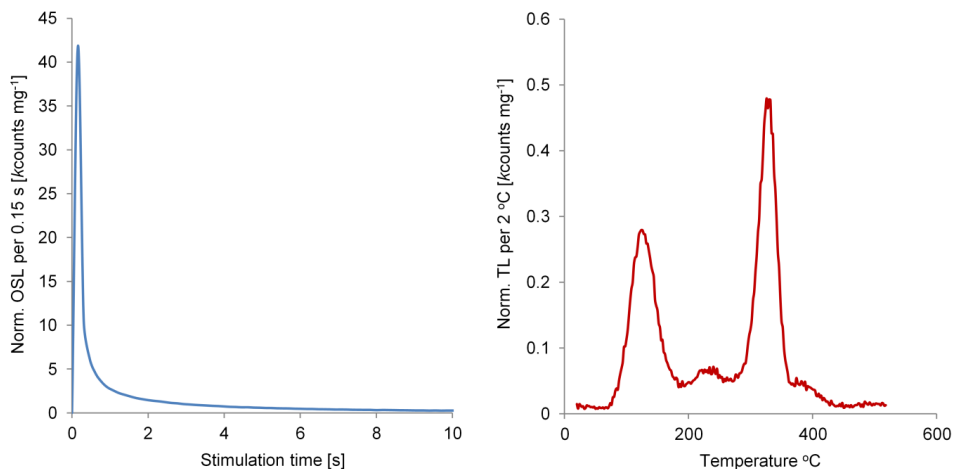


Figure 6. Weight normalised CW-OSL (left) and TL (right) curves ( $\text{kcounts mg}^{-1}$ ) for two different aliquots of the same brand of NaCl. The CW-OSL curve shows only the first 10 s of the read-out to illustrate the fast and the slow components of the decay function, although stimulation during 40 s and 20 s was used in **Paper I** and **Papers II, IV**, respectively. The signals in the curves are registered in intervals of 0.15 s and 2 °C, respectively.

The observed TL-glow peaks and the OSL-decay curves are influenced by the type of salt and given dose, as well as by the amount of salt stimulated to obtain the signal. Twice the amount of salt (up to a certain sample amount) gives rise to a doubling of the signal (at a given dose of about 0.2 Gy). The same proportionality was not seen for the TL-signal although a higher signal was achieved by increasing the amount of sample material. The other factor influencing the TL- and OSL-signals is the properties of the individual salt grains which physically differs regarding to size and shape which indirect, also changes the luminescent properties. Sample homogeneity and grain to grain variations can be visually observed (see table 2 in section 3.2.1.2), although taken from the same package. As the grains used on each cup are not identical, and to minimize the possible effect due to the grain distribution, a large amount of grains were used on each cup to average out any variations within an aliquot. The resulting signal was then normalised to the sample weight. Other methods for reducing the variability in the signal due to sample-to-sample variations have been proposed, e.g. the single aliquot regenerated-dose protocol (SAR) first proposed by Murray and Wintle (2000). The advantages and disadvantages of the different approaches will be discussed in section 4.2.

The fading of the signal will determine the magnitude of the signal in relation to the time between exposure and read-out. It has previously been reported that NaCl exhibits a fading at ambient temperature of only a few percentage during the first 24 h and thereafter much slower [Bailey et al. 2000; Thomsen 2004]. This is in agreement with the preliminary studies carried out during the present project where a small (<10%) initial fading was observed during the first 24 h. It is also our experience that the signals from dosimeters read during the first 24 h

varied more than for dosimeters read during the following days and weeks. All salts were therefore read after, at least, 24 h after irradiation.

The dose response relationship was studied for different salts, at low (1 mGy to 43 mGy) and high (130 mGy to 9.9 Gy) absorbed doses (Fig. 7). For this calibration curve to be valid it is assumed that the weight normalisation evens out any possible variations between the aliquots. Additionally, to minimise variations between equally treated aliquots a total number of twenty-four aliquots were used at each given calibration dose.

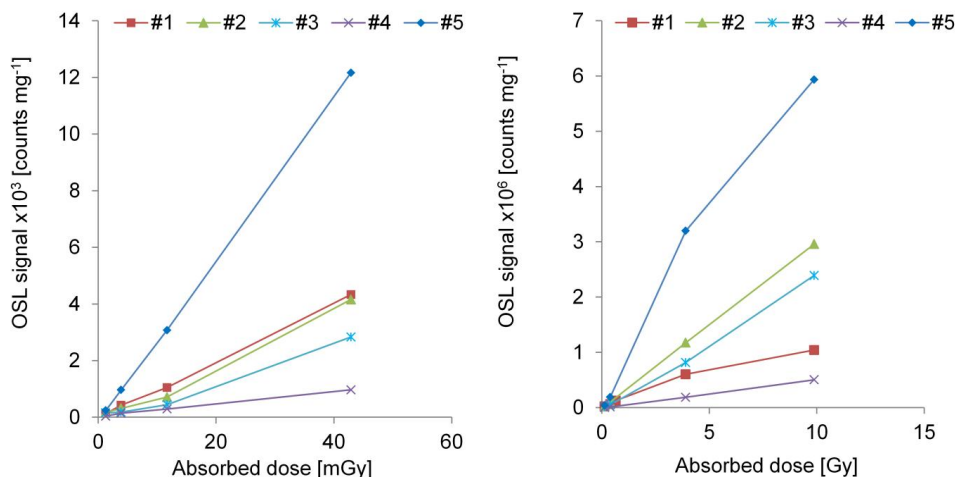


Figure 7. Dose response relationship (lin-lin) for the five salts investigated separated in two graphs: to the left the low dose region (1-43 mGy) and to the right the high dose region (0.13 Gy-9.9 Gy). The salt numbers referred to in the table legend are the same as in Paper I (1-Chemical NaCl; 2-Sea salt; 3-Sea salt; 4-Mine salt; 5-Mine salt). N.B. the unit on the abscissas are different, mGy to the left and Gy to the right.

In the range from 1 mGy to 100 mGy, the relationship was approximated by a Taylor expansion of the first order. According to this a calibration coefficient, in terms of specific luminescence, was deduced for all five salts in that dose range. The magnitude of  $c_{\text{specific}}$  varied from 31 counts mg<sup>-1</sup> mGy<sup>-1</sup> (salt #4) to 313 counts mg<sup>-1</sup> mGy<sup>-1</sup> (salt #5). These results demonstrate a large difference among the salts and thus indicate the importance of sensitivity calibrations for various batches of salt. The theoretical detection limit in terms of MDD varied from 0.1 mGy (salt #1) to 1.0 mGy (salt #4). The range in the observed MDD for the salt brands is similar to the one observed for the sensitivity due to the dominating influence of  $c_{\text{specific}}$ . These values for the MDDs should be viewed as optimistic estimates, as MDD also is affected by many other factors (e.g. type of salt and package, storage time since purchase, time of production, homogeneity within the salt). The values indicated here are from sporadically bought salts in Swedish markets and thus give a representative figure of the MDD for these salts.

Laboratory studies showed that all five investigated salts had properties suitable for dosimetry, e.g. both a linear dose response for  $^{60}\text{Co}$  exposures over a wide range of absorbed doses as well as a low detection limit. The dosimetric properties of the salt brands were rated based on the specific luminescence,  $C_{\text{specific}}$ , and the minimum detectable dose, MDD. On the basis of these two parameters, one particular salt that was found to be the most appropriate for retrospective dosimetry (salt #5, *Falksalt fint bergsalt*). Another good property of salt #5 was that it was relatively insensitive to clustering of the grains. Salt from this brand was therefore later used during the *in situ* comparisons to conventional dosimetry techniques (**Paper II, IV**).

## 4.2 Testing the NaCl dosimeters *in situ* (**Paper II**)

Dosemeters based on the OSL in NaCl were tested *in situ* and compared to TLDs, radiation protection instruments as well as model calculations (based on HPIC measurements). The measurements were repeated in three successive years (starting in 2008) during the summer months (end of May to mid-August). Table 3 summarises the measurements carried out and the equipment used in each year. Over these years, different improvements (and other factors) led to slightly changed measuring conditions in terms of dosimeters and equipment used. In spite of that, there was a moderate or strong correlation to the results measured by the salt and to the other radiation protection instruments and to the TLDs.

*Table 3. Surveys in Svetilovichi during the years 2008-2010 and the equipment used for the dose assessments. Each year, two visits were carried out when distributing and collecting the dosimeters in May-August. In 2010 a third expedition was carried out in October (2010b) to make reference measurements with the high pressure ionisation chamber (HPIC).*

	Dosemeter kits		Detectors	Number of kits (2×LiF and 5×NaCl)
	TLD	OSLD		
<b>2008</b>	LiF:Mg, Ti	Falksalt fint bergsalt '06	GR110, SRV2000	56 dosimeter kits
<b>2009</b>	LiF:Mg, Ti	Falksalt fint bergsalt '09	GR110, SRV2000	69 dosimeter kits
<b>2010a</b>	LiF:Mg, Cu, P	Falksalt fint bergsalt '09	GR110	99 dosimeter kits, salt sieved to < 400 µm
<b>2010b</b>	-	-	GR110, HPIC	-

As can be seen from Table 3 the dosimeter kits were not completely identical in terms of the two dosimeter materials, the salt stemming from two different packages of the same brand, and a change of LiF-chips in the last year of the survey. However, the same radiation survey meter (GR110) was used in all years and at all locations where the dosimeter kits were positioned. The variation (in terms of 1 SD of the mean readings,  $C_v$ ) in the GR110 reading when the dosimeters were put in position and collected, was determined to be 3.5%. A relationship between the readings of the HPIC and GR110 was determined in the dose rate

interval from  $0.10 \mu\text{Sv h}^{-1}$  to  $1.40 \mu\text{Sv h}^{-1}$  at various positions in Svetilovichi with outskirts, encompassing a varying radiation environment. The relationship between  $\mu\text{Sv h}^{-1}$  (HPIC reading) and cps (NaI(Tl)-detector of the GR110-instrument) was determined to follow the expression in Eq. 10.

$$\text{NaI(Tl)} = 2612 \cdot \text{HPIC}^{0.73} - 306 \quad \text{Eq. 10}$$

Hence, this relationship can be used to get a reasonable estimate of the ambient dose rate from the GR110-readings presented and vice versa.

During the three occasions of the *in situ* measurements in Svetilovichi, a total number of 250 dosimeter kits were distributed. Of these 250, 224 kits were determined as usable as 26 kits were insufficiently packed in 2010 and had to be excluded from the comparison due to a significant signal loss of the OSL signal (a different group of staff had prepared these specific dosimeters). As the dosimeter materials in the dosimeter kits were changed during the course of the study it is important to be aware of these changes when comparing the results from the different years (Table 3). The TLDs were kept the same in 2008 and 2009 but a new series of TLDs were introduced in 2010. For the OSLDs the same brand of salt was used in all three years but the salt portions embedded into the dosimeter kits were taken from different packages in 2008 compared to 2009 and 2010. A consequence of the change in dosimeter set-up is seen in the linear regressions of the dosimeter readings to the (non-converted) GR110 measurement at the same position (Eq. 11).

$$\left\{ \begin{array}{ll} \text{TLD}_{2008} = 0.24 \cdot \text{NaI(Tl)} + 0.05 & \text{OSLD}_{2008} = 0.28 \cdot \text{NaI(Tl)} + 0.04 \\ \text{TLD}_{2009} = 0.23 \cdot \text{NaI(Tl)} + 0.07 & \text{OSLD}_{2009} = 0.46 \cdot \text{NaI(Tl)} + 0.10 \\ \text{TLD}_{2010} = 0.26 \cdot \text{NaI(Tl)} + 0.03 & \text{OSLD}_{2010} = 0.47 \cdot \text{NaI(Tl)} + 0.06 \end{array} \right\} \quad \text{Eq. 11}$$

All three TLD regressions are similar in terms of their slope and intercept. This demonstrates a good agreement between the new and the old TLDs as well as between the dosimeters and the NaI(Tl)-detector. There is however a varying goodness of fit in terms of the mean absolute percentage uncertainty (MAPU) of 32%, 17% and 13% in 2008, 2009 and 2010, respectively. This indicates a smaller spread in the dose response relative to the NaI(Tl)-detector among the new TLDs. The difference between the 2008 and 2009 series is however difficult to explain. There is a more obvious difference in the regressions of the OSLDs (Eq. 11). Unaware at the time of the varying dosimetric properties between different packages of salt from the same brand, a new package of salt was bought for the measurements in 2009. As can be seen in Eq. 11 the slope of the regression (related to the specific luminescence) for the OSLDs in 2008 and 2009 differs by a factor of about 1.6, but the regression lines for the OSLD measurements in 2009 and 2010 are almost parallel. This shows that salt of the same brand, but from different packages, have different dosimetric properties. A possible explanation for this is that the salt is extracted from different layers within the same, or different, mines and thereafter possibly mixed with salt from other deposits before it is packed and distributed to the markets. Due to the higher sensitivity of the salt used in 2009, salt from the same package was also used in 2010 with the ambition to carry out individual calibrations after each read-out (see below).

The above mentioned situation addresses one of the main issues when using salt as a dosimeter; namely how to correctly assess and interpret the OSL-signal from the salt. By using a general calibration for a specific brand (and type) of salt it is possible to rapidly get a retrospective estimate of the absorbed dose for affected populations in an area subjected to radioactive fallout. However, as this calibration factor is sensitive to variations between individual salt samples, it should be carried out not only for salts of different brands, but also for different packages of the same brand and optimally, also for individual aliquots. A dose regenerative read-out protocol of the OSLDs, such as the SAR-method, is an alternative approach to overcome this problem. It has, however, the disadvantage of being too time consuming to be used in a scenario when many samples need to be evaluated rapidly in order to avoid fading and unwanted signal contributions. An improved version of the SAR-protocol, with a faster and more optimized calibration sequence for NaCl at low absorbed doses, is currently being developed [Christiansson et al. 2011]. However, this protocol was not possible to implement at the time of the read-out of the dosimeter kits from 2010. The previously determined calibration was therefore used in 2010 as well. The sensitivity changes of the salt used in 2009 and 2010 was adjusted by a factor of 0.60 compared to the calibration of the salt used in 2008 and hence, the discovery of the different sensitivities of the salts has been corrected for in the following evaluation of doses.

Altogether there is a good agreement in the annual effective dose estimates with the different dosimeters and model calculations (Fig. 8) although the model generally overestimates the doses measured by the different dosimeters slightly. Based on the conversion factor from the HPIC, the GR110 estimates agree within 15% of the model estimates. It should be noted that in the model, the average dose was based on measurements from several locations (in 2010) whereas the dosimeters are confined to the residence. This might also explain some of the deviations found for the OSLDs and TLDs compared to the calculated effective doses using the model. However, after the sensitivity correction of the OSLDs used after 2008, the estimates made by the OSLDs and TLDs are within 5%. The OSLDs and the TLDs are generally within 30% of the model estimates and they both consequently underestimate the model. This further suggests that the transportation background corrections ( $0.1 \mu\text{Sv h}^{-1}$ ) of the dosimeters were slightly overestimated considering the storage conditions and possible long term fading.

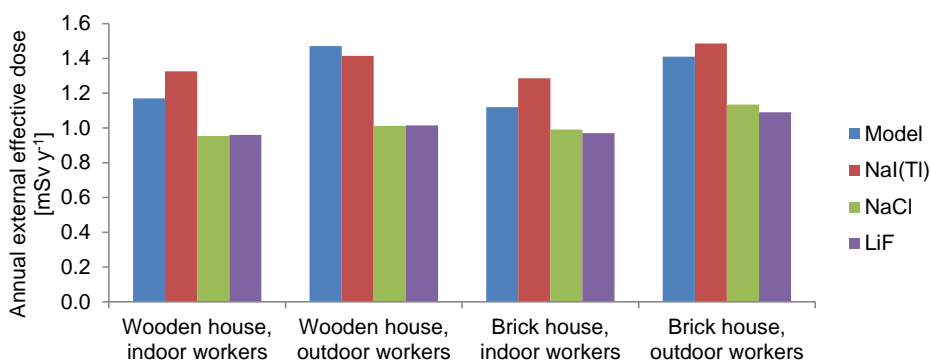


Figure 8. Annual external effective dose to the inhabitants of Svetilovichi estimated by model calculations, measurements with the GR110-instrument (NaI(Tl)) and luminescent dosimeters of NaCl and LiF. The estimates are grouped after house type (wood or brick) and if the person is working indoor or outdoor (no consideration to countermeasures have been taken).

The observed variations among the different methods used for assessing the external exposure are slightly higher for the outdoor measurements. Outside the houses the dose rate and its variations are generally higher than inside the houses (see section 4.3). It is however surprising to note that the effective doses estimated for indoor workers in wooden and brick houses are similar. This is not expected as the shielding from the larger and thicker brick walls generally reduces the radiation level inside from the contamination outside.

The annual external effective dose estimates ranged from about 1 mSv y<sup>-1</sup> to 1.5 mSv y<sup>-1</sup> depending on the assessment method used. The estimates are associated with an uncertainty related to the number of sampling positions, background dose correction and the conversion factors used from measured value to effective dose. The above mentioned levels refer to the total effective dose in Svetilovichi, as no background correction was carried out for the pre-Chernobyl background as was done for the personal dosimeters in Russia. The magnitude of the pre-Chernobyl dose rate, used in **Paper IV**, was 45 µSv month<sup>-1</sup> [Fogh et al. 1999]. As the dosimeters were on average in position during 2.5 months this contribution adds up to 0.11 mSv, for the measuring period. The external annual effective dose from Chernobyl <sup>137</sup>Cs is thus, roughly, in the range from 0.9 mSv y<sup>-1</sup> to 1.4 mSv y<sup>-1</sup>. This is similar to the 1.0 mSv y<sup>-1</sup> that was estimated by in 2004 by Fesenko et al. (2010), in the same village.

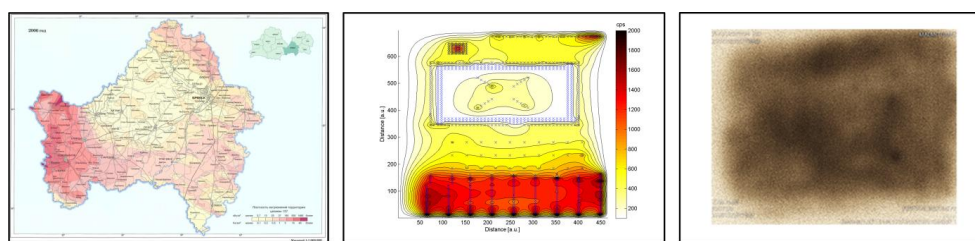
### 4.3 Variability in surface deposition and dose rate over the ground (Paper III)

There are large differences in deposition pattern depending on the characteristics of the source. The radioactive products from the CNPP were injected into the atmosphere at 1-2 km height (radioisotopes of iodine and caesium were even discovered at heights of 6 to 9 km) during a 10 day period from April 26 to May 5 1986 [Buzulukov and Dobrynin 1993]. The deposition pattern was very inhomogeneous through the northern hemisphere and to a high



extent influenced by rain during the passage of the plume from Chernobyl. The deposition pattern for Chernobyl debris differed much from that from the fallout from nuclear weapons tests, which was much more uniformly distributed and took place for almost two decades. This fallout was continuously deposited all over the year, preferentially as wet deposition during rain and snow with a maximum in April-July [Mattsson and Moberg 1991].

Compared to the fallout from nuclear weapons test, the Chernobyl fallout was released and deposited in a single event. The variability in the deposition of Chernobyl fallout over Europe has shown to be very high [Borzilov and Klepikova 1993; Izrael 2007; Atlas 2009], due to different weathering conditions in combination with the passing plume. Different decontamination measures, e.g. removing the top soil, have also contributed to the inhomogeneous radionuclide distribution in the horizontal layers in such areas. On smaller scales the inhomogeneous pattern persist (Fig. 9), down to at least a few centimetres.



*Figure 9. Deposition maps illustrating the radiation level on three different scales (hundreds of km, tens of m and down to a few cm). Left frame: Deposition of  $^{137}\text{Cs}$  from Chernobyl over the Bryansk region as of 2006 [Atlas 2009]. Middle frame: Contamination pattern around a decontaminated school in the village of Yalovka, Klinty region as measured in October 2008. Right frame: Scan of a photostimulable film ( $30 \times 24 \text{ cm}^2$ ) after exposure during one week of an evenly thick (6 mm) layer of soil taken from the village of Dementia, Novozybkov region in 2008. Note that the units in the different frames are not comparable.*

As the measured quantity is different in the images of Fig. 9, the absolute numbers should not be compared. Nevertheless, albeit related but not identical quantities are plotted in the images of Fig. 9, it illustrates the varying radiation level on a scale of several hundreds of km to part of a centimetre. The inhomogeneous pattern evidently persists even on the small scale.

The same type of inhomogeneous contamination pattern is seen within and between the villages and also within individual gardens. A specific example is shown in Fig. 10, where two wooden houses of similar construction and their gardens were surveyed. In both of these houses the magnitude of the indoor measurements are fairly similar, with a slightly higher level measured inside the non-decontaminated house. A considerably larger difference is found when comparing the outdoor levels in the gardens of the two houses. On the front side (side 1 in Fig. 10), both houses have a small flower bed in which the soil has been cultivated repeatedly during the years and the dose rate level above ground is similar. A specific area on the same side of the decontaminated house (indicated with a tree in the figure) showed a highly varying dose rate level. As it was not possible to remove the tree during the decontamination process, it was difficult to completely remove the soil at that position. This

left a spot with untouched soil, with levels of  $0.85 \mu\text{Sv h}^{-1}$  (at 0 m above ground),  $0.40 \mu\text{Sv h}^{-1}$  (at 1 m above ground) and  $0.23 \mu\text{Sv h}^{-1}$  (at 2 m above ground). Similar spots were also observed in other decontaminated gardens with large trees and root systems covering the ground.

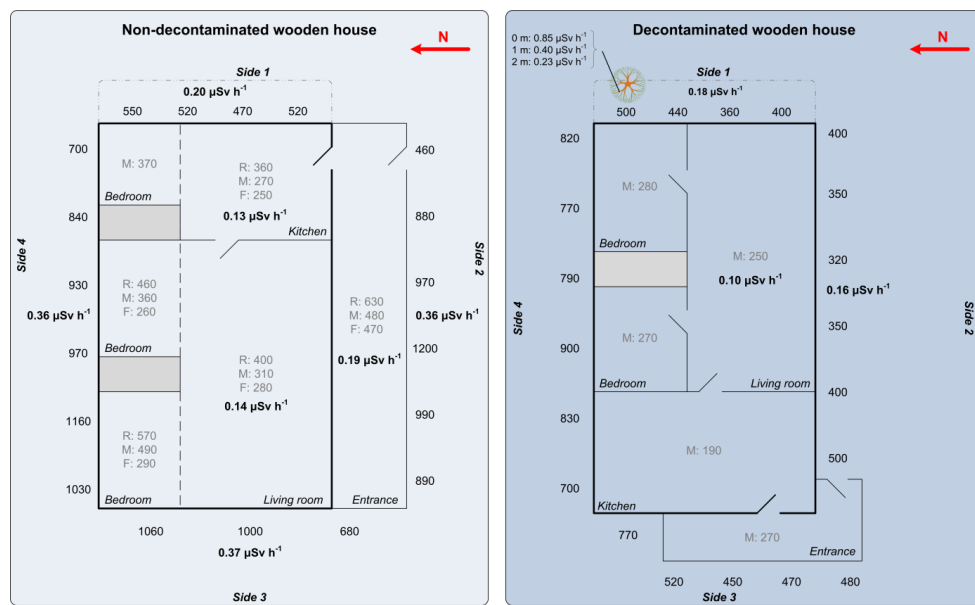
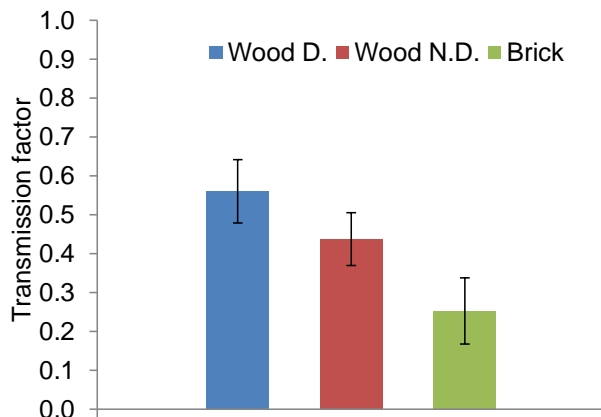


Figure 10. Illustration of two wooden houses and their gardens (seen from above), in the village of Svetilovichi surveyed by the GR110-instrument (cps) and the HPIC ( $\mu\text{Sv h}^{-1}$ ) at 1 m above ground. The notation R, M and F for the indoor measurements refer to the measuring height R=just under the inner roof, M=middle (1 m above the floor) and F=on the floor. The arrow indicates north.

The non-decontaminated house demonstrates a relatively homogenous radiation level at the three other sides (sides 2-4), with some minor changes in the corners of the house. The decontaminated house has a relatively uniform radiation level on three sides (sides 1-3) including the cultivated front side. However, on the side numbered 4 in Fig. 10, the measured dose rates at 1 m above ground are about twice as high as on the other sides. This particular side is generally not used by the residents and the radionuclide contamination is thus only affected by natural processes. The results indicate that less effort were done to reduce the radiation level on this side (side 4), than on the other sides of the house. This situation was also observed in other similar gardens and houses.

The residents of the contaminated villages are continuously exposed to external radiation from Chernobyl  $^{137}\text{Cs}$ . The exposure is lower inside the houses due to the shielding effect of the building. The influence of the building on the indoor radiation environment has been investigated in terms of the dose transmission factor (TF), defined as the average dose level indoors to that outdoors. In Fig. 11 are average dose transmission factors for 36 houses

surveyed in the Bryansk-Gomel spot of both wooden (decontaminated and non-decontaminated) and brick construction.



*Figure 11. Dose transmission factors for wooden houses (D: decontaminated; N.D: non-decontaminated) and non-decontaminated brick houses ( $\pm 1$  S.D.) based on GR110 measurements.*

A transmission factor close to 1 indicates a poor shielding and from a radiation protection point of view, it is thus desirable to have a TF as small as possible. For a given outdoor contamination level the best shielding is provided by a brick house due to its higher photon attenuation than a wooden house. When comparing the wooden houses (all are of similar construction and design), the decontaminated houses seem to have a higher TF. The reason for this is that the decontamination measures carried out around the houses have reduce the radiation level over those surfaces, but not proportionally so inside. The house-to-house variation of the indoor radiation level is small (often negligible) compared to the one observed in the gardens. Therefore, Fig. 11 shows that the countermeasures carried out managed to reduce the radiation level in the gardens to a magnitude that is today, lower than in the non-decontaminated gardens. Other factors that can influence the indoor radiation level are e.g. roof contamination, the use of contaminated wood for reparations, washing and maintenance of the indoor environment.

#### 4.4 External and internal effective doses in the Bryansk villages (Paper IV)

Assessments of the external and internal effective doses in a number of villages in the Novozybkov-Klintsy area have been carried out between 1990 and 2000 [e.g. Thornberg et al. 2005]. It was therefore of high interest for the assessment of the long-term consequences of the contamination, to repeat these measurements. This was done in 2006 and 2008. As in earlier years it was not possible to visit all the same villages every year but similar nearby

villages were chosen instead with a composition of the participants that was aimed to represent the general population in each village visited. Thus, inhabitants of a wide range of ages and of both sexes participated.

#### *4.4.1 External effective dose*

In 2008 the read-out of the luminescent dosimeters (TLDs and OSLDs) was slightly delayed due to transportation complications from Russia to Sweden. No dosimeters were returned from the village of Demenka. As an alternative to those dosimeters, the average external effective dose to the inhabitants in Demenka in 2008 was estimate based on TLD measurements in 2006 and scaled by the 2008 to 2006 ratio of surveys by handheld radiation protection instruments in the village, carried out at the same positions. Based on these measurements the average dose rate reduction from 2006-2008 was estimated to 15%. This figure was used as a conservative estimate as it is based on outdoor measurements only. Due to lack of TLDs, only OSLDs were used in Veprin in 2008. A conversion coefficient, based on measurements with dosimeter kits with both TLDs and OSLDs in the other villages visited in 2008, was determined to  $0.35 \mu\text{Sv } \mu\text{Gy}^{-1}$ . However, since the salt used in these dosimeters were not from the same package of salt as during the calibration (see *section 4.2*) an adjusted value of  $0.59 \mu\text{Sv } \mu\text{Gy}^{-1}$  was used for assessing the average effective dose for the inhabitants of Veprin.

Based on the above mentioned assessments of the dose in Demenka and Veprin 2008, and the measurements carried out in 2006 and 2008, the annual external effective doses as an average for all villages were determined to  $0.4 \text{ mSv y}^{-1}$  and  $0.3 \text{ mSv y}^{-1}$ , respectively. As this decrease corresponds to about 25% in just 2 years it is not in relation to the expected yearly decrease from physical decay (2%) and further vertical migration down in the soil which is expected to be in the same order of magnitude [Hille 1996; Hille et al. 2000]. An explanation for the observed rapid decrease is that the group of participants in 2006 and 2008 were slightly different in terms of occupation, spare time habits and the possibility or desire to wear the dosimeters throughout the measuring period. Another explanation is that the significant background signal, received in 2008 due to the prolonged storage before read-out, obscured the net contribution of the signal acquired during the measuring period.

#### *4.4.2 Internal effective dose*

The body burden of caesium was determined in 316 persons in 2006 and 324 persons in 2008 (plus 57 (2006) and 13 (2008) persons in the town of Novozybkov<sup>4</sup>). In both occasions there were slightly more young persons (<18 y) than adults, except in the village of Demenka in 2006. The whole body concentration (WBC) was found to be similar between persons of different ages, an observation also noted during previous years (1991-2000). In some years, however, deviations from this general trend have been observed. There are a number of factors that will influence body burdens of humans in a given area e.g. amount of forest food (mushrooms, berries, game) in the diet, soil type which determine the transfer factor of

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<sup>4</sup> The body burden of caesium in persons from the town of Novozybkov was measured for reference purposes and no TLDs were distributed to this group. All these persons were adults.

ground deposition to crops and pasture, attitude to governmental recommendations, economic situations among the residents. In years with rich mushroom harvest, as in 1998, the internal effective dose component dominated the total effective dose, an effect that was more pronounced in adults than children. A possible explanation can be that the adults generally eat more mushrooms than children. No significant correlation between the  $^{137}\text{Cs}$  deposition in the villages and the WBC of  $^{137}\text{Cs}$  has been observed, neither in the years 2006 and 2008, nor during earlier years [Thornberg 2000].

In Fig. 12 are given the average internal effective doses from  $^{137}\text{Cs}$  in 2006 and 2008 to the residents of the different settlements (including the town of Novozybkov), separated into adults and adolescents/children. In both years the average internal effective dose to the adults was twice as high as for the group of adolescents/children. The observed range of the effective dose estimates is normally wide (highest individual values are  $1.3 \text{ mSv y}^{-1}$  and  $1.9 \text{ mSv y}^{-1}$  for children and adults, respectively). The distribution of the individual effective doses is log-normal with some individuals exceeding the average values by a factor of 2-5. In this sense the measured persons from the town of Novozybkov are a more homogenous group where the distribution of the individual effective dose values is more Gaussian shaped.

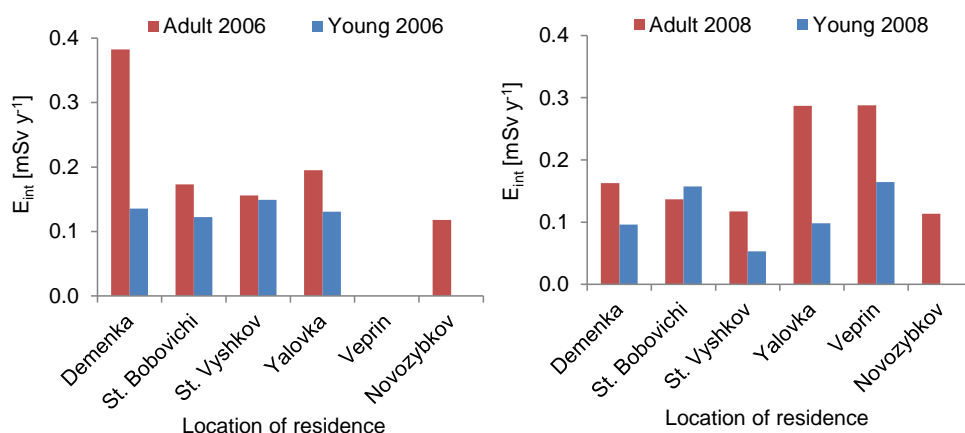


Figure 12. Estimates of average effective doses from internal  $^{137}\text{Cs}$  contamination to the inhabitants in the Novozybkov-Klintsy area, 2006 and 2008, for adults ( $>18 \text{ y}$ ) and young persons ( $<18 \text{ y}$ ). Adults from the town of Novozybkov are also represented in the graphs.

In general, a small but steady decline between the years 2006 and 2008, in the average effective doses is observed for most villages. One deviation from this decrease is found in the village of Yalovka. The slight increase observed in this village is mainly attributed to the adults, which may in turn be related to slightly different food habits of the measured individuals in 2006 and 2008. It is commonly known that the intake of forest food (mushrooms, berries, game), which can have much higher concentrations of caesium than locally produced milk and meat, will considerably increase the caesium concentration of persons that consume large quantities [Balonov 1999; Fesenko et al. 2000; Travnikova et al. 2001]. Persons that live in larger towns, as in Novozybkov, have access to food imported

from areas with less contamination and are generally not as dependent on locally produced food. Hence, these persons will have a comparably lower internal effective dose which will not be subjected to large year-to-year variations as observed in the villages (Fig. 12). As previously mentioned the year-to-year variations of the WBC and internal effective dose within and between the surveyed villages is complexly related to several individual factors such as economics, if the person is following the food recommendations and how they prepare their food. Another factor is the season of the year when the body burdens is determined. Autumns are associated with the abundance of forest food and hence, humans consuming these foodstuffs will take a large part of their diet from the forest during the autumn. This may result in an increase of the radiocaesium activity concentration (60-70% mean increase) between spring and autumn [Skuterud et al. 1997; Jesko et al. 2000]. In lack of body burden data in residents during springtime no correction for an increased autumn concentration has been carried out.

#### 4.4.3 Total effective dose to rural inhabitants in the Bryansk region

The Russian colleagues in the project started their monitoring program of the external and internal effective doses in the area already in 1986 [Ramzaev et al. 1996]. These measurements overlapped with the beginning of the present long-term program. An intercomparison during the years of the overlap showed a difference of less than 4% between the TLD dose estimates [Wøhni 1995]. The effective dose estimates by Ramzaev et al. (1996) is shown in Fig. 13 (1986-1990) together with the estimates from 1991-2008.

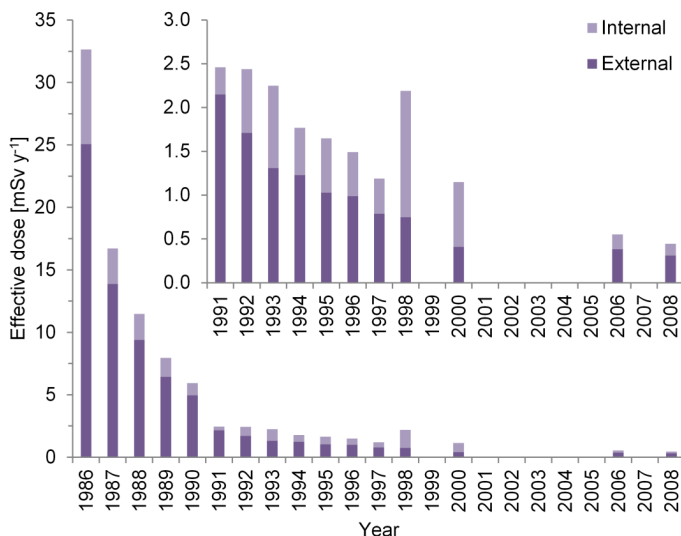


Figure 13. Average effective dose (internal and external) from Chernobyl fallout. Each bar represents the annual effective dose contribution averaged over all inhabitants that participated each year. Data for the period 1986-1990 are from Ramzaev et al. (1996).

At the time of the most recent survey (2006 and 2008), the internal and external effective doses to the inhabitants were low. The contribution from Chernobyl caesium to the total effective dose was determined to an average value of  $0.5 \text{ mSv y}^{-1}$  and  $0.4 \text{ mSv y}^{-1}$  in 2006 and 2008, respectively. This is comparable to the annual external effective dose from cosmic radiation and the internal effective dose from  $^{40}\text{K}$  in the human body (about  $0.5 \text{ mSv y}^{-1}$ ).

The decrease of the yearly effective dose has now levelled out. During the period from 1986 to 1990 the yearly effective dose was reduced by 80%. In the period after that, in the years 1991-1993, the external effective dose decreased with about  $20\% \text{ y}^{-1}$ , followed by a slower decrease of  $12\% \text{ y}^{-1}$  between 1994 and 1998. Thereafter and up to 2008 the rate of decrease was about  $9\% \text{ y}^{-1}$ , and about  $3\% \text{ y}^{-1}$  if excluding 1998. The internal effective dose is also reducing by time, but not with same temporal behaviour as the external effective dose. The relative contribution from internal exposure to the total effective dose ( $E_{\text{int}}/(E_{\text{int}}+E_{\text{ext}})$ ) has been fluctuating around a value of 0.3. Initially (1986-1990) the same ratio was lower (about 0.2). The ratio appears to be relatively stable at 0.3, except in some years (1998 and 2000) when the ratio has exceeded 0.6. The reduction in the sum of the two components are explained by the cumulated effects of the factors described earlier; The wide-scale countermeasures (including restrictions) and natural processes were the major factors during the first years after the accident. In the years from 1991 to 2008 the main variation in this ratio is attributed to changes in  $E_{\text{int}}$ , while  $E_{\text{ext}}$  have a comparably steadier decrease over time. The observed variations in recent years of  $E_{\text{int}}$  are combinations of human behaviour (towards the food recommendations) as well as natural processes. In future years, the internal effective dose will be of increasing influence of the total effective dose. Persons that consume large amounts of forest food may have up to 1.5-3 times higher internal effective dose than the external effective dose [Balonov et al. 1999]. Hence, the individual attitude towards food recommendations will thus be the most important factor in the years to come.

Although the measurements in 2006 and 2008 does not significantly influence the total accumulated does, they demonstrate an indication of the decrease rate which can be used to assess a prognosis for the 70 y accumulated effective dose. The average accumulated effective dose (from internal and external exposure) to the inhabitants in the Novozybkov-Klitsy villages during the study period from 1991-2008 was estimated to 24 mSv by using interpolation with a decrease rate of  $9\% \text{ y}^{-1}$  between the years where data is missing. This is the same as the  $25 \text{ mSv y}^{-1}$  estimated by Ramzaev et al. (1996) for the external effective dose in 1986 (the internal effective dose the same year was 7.6 mSv). For the period 1987-1990, the total effective dose is estimated to 42 mSv, which means in total 99 mSv for the period 1986-2008 as an average. By using the same decrease rate of the total effective dose up to 2056, the total cumulated effective dose ends at 104 mSv. By using the alternative rate of decrease ( $3\% \text{ y}^{-1}$ ) the cumulated effective dose over 70 y becomes a little higher, 111 mSv. This can be compared to the 70-100 mSv estimated by Thornberg (2000) using different decrease rates of the internal and external exposures and another set of data for the period 1986-1989. Persons that consume large quantities of forest products and spend much time outside in e.g. the forest, are however, likely to receive higher doses.

## 5. Concluding remarks

Nuclear and radiological events that are ranked higher than INES 5 on the IAEA event scale can be expected to result in absorbed doses of a few hundreds of mGy to a limited part of the affected population. In the initial stage after the event, retrospective dosimetry methods based on physical and biological indicators are highly needed as a complement to ordinary radiation protection instruments, both for providing urgent medical care in extreme cases, as well as to support the expertise with data in decision making processes and communication to the affected people. For the majority of the people, however, much lower doses than 100 mGy will prevail. Nevertheless, these individuals will be concerned regarding their risks for later health effects and they will more or less indirectly require accurate estimates of their individual risks. One of the challenges for many retrospective methods is that the detection limits many times exceed the individual absorbed doses of these individuals. In addition to alleviating people's concern regarding the radiation environments in their living areas, long-term follow-up using dosimeters *in situ* may be important also for epidemiological studies.

Since the years after the Chernobyl accident the combined effects of migration of radionuclides down into the soil, human processes (including actions to reduce the exposure), and physical decay have resulted in the observed dose rate reduction of about 3%  $y^{-1}$ . It is also evident that the inhomogeneous contamination patterns persist even a quarter of a century after the event, both on large as well as on small scales. The effect of this is averaged out when considering the exposure to the inhabitants in the various spots in the area, since it depends on the time spent at different locations. The result in terms of average effective dose from Chernobyl radionuclides to the population in the villages of the Novozybkov-Klitsy area was estimated to 100 mSv during the first 25 years. For the additional period 2012-2056 the additional dose contribution depends on the reduction rate of the effective dose, and is estimated to be between 3-10 mSv using the observed reduction rates of 9%  $y^{-1}$  and 3%  $y^{-1}$ , respectively. This value depends also on individual behavior, occupancy and age and may indeed, for individuals, vary above and below this. During the recent years the average annual effective dose in the villages in the Novozybkov-Klitsy area from Chernobyl radionuclides (now totally dominated by  $^{137}\text{Cs}$ ) was less than 0.5 mSv  $y^{-1}$ , of which 2/3 is estimated to come from external exposure and 1/3 from internal exposure. The effective dose rate from Chernobyl radionuclides to the inhabitants in the area have now been reduced by a factor of more than 60 since 1986, and the magnitude is now comparable to the effective doses from the natural background radiation. However, the present day low levels of effective doses in the Russian villages are becoming increasingly difficult to determine accurately. This will especially happen when, as in our case, there are large distances between *in situ* measurements and read-out facilities of the dosimeters, since cumbersome logistics prolong the proportion of time spent outside the measuring area and thereby necessitate uncertain background corrections. Thus, the associated uncertainty in the effective dose estimates are increasing as the dose rate decreases. One trend that is clear is that the relative importance of the internal component of the effective dose among the rural inhabitants in the Bryansk-Gomel spot is increasing, and will continue to do so in the future. The internal effective dose



Contribution from Chernobyl (now solely  $^{137}\text{Cs}$  in food) can however be estimated with high accuracy.

Optically stimulated luminescence in ordinary household salt has shown several properties that are useful for dosimetry. It has a high sensitivity to ionising radiation and a small (predictable) signal fading after exposure. Salt can be found everywhere and is commonly found in most households. The abundance of the salt makes it suitable as both an environmental indicator of ionising radiation and a possible tool for making estimates of individual absorbed doses to humans in the vicinity of the sampling location. One of the most important findings, however, is the potentially very low detection limit ( $<1\text{ mGy}$ ) of salt. Such low detection limit is useful for determining individual absorbed doses in the late period after a major radiological or nuclear event. The prospective *in situ* measurements, using salt dosimeters, have proven the salt to be comparable to both ordinary TLDs as well as model calculations, even at an extended period of time in ambient conditions and at absorbed dose rates down to  $0.1\text{ }\mu\text{Gy h}^{-1}$ . Similar to many other scintillators and luminescent materials, salt has the limitation of being sensitive to visible light, and can only function as a dosimeter when shielded from light. In this study the salt dosimeters were tested both under laboratory conditions as well as *in situ*, in pre-arranged dosimeter kits that were light-sealed. The situation will be different after an unexpected event, when samples of salt must be searched for, to retrospectively determine individual absorbed doses. In view of the results of this study, it is therefore of high interest to continue to investigate the characteristics of salt, to understand and improve the various factors influencing the accuracy of the dose assessments from samplings *in situ*. Such factors include the wavelength specific light transmission through various types of packages, dose distribution within a specific salt package, sampling technique within a given package. Other factors that also need to be investigated are the signal fading over several months to years during different conditions (humidity etc.), the conversion from absorbed dose in salt to effective dose, and rapid methods for combined read-out and calibration of individual samples.

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