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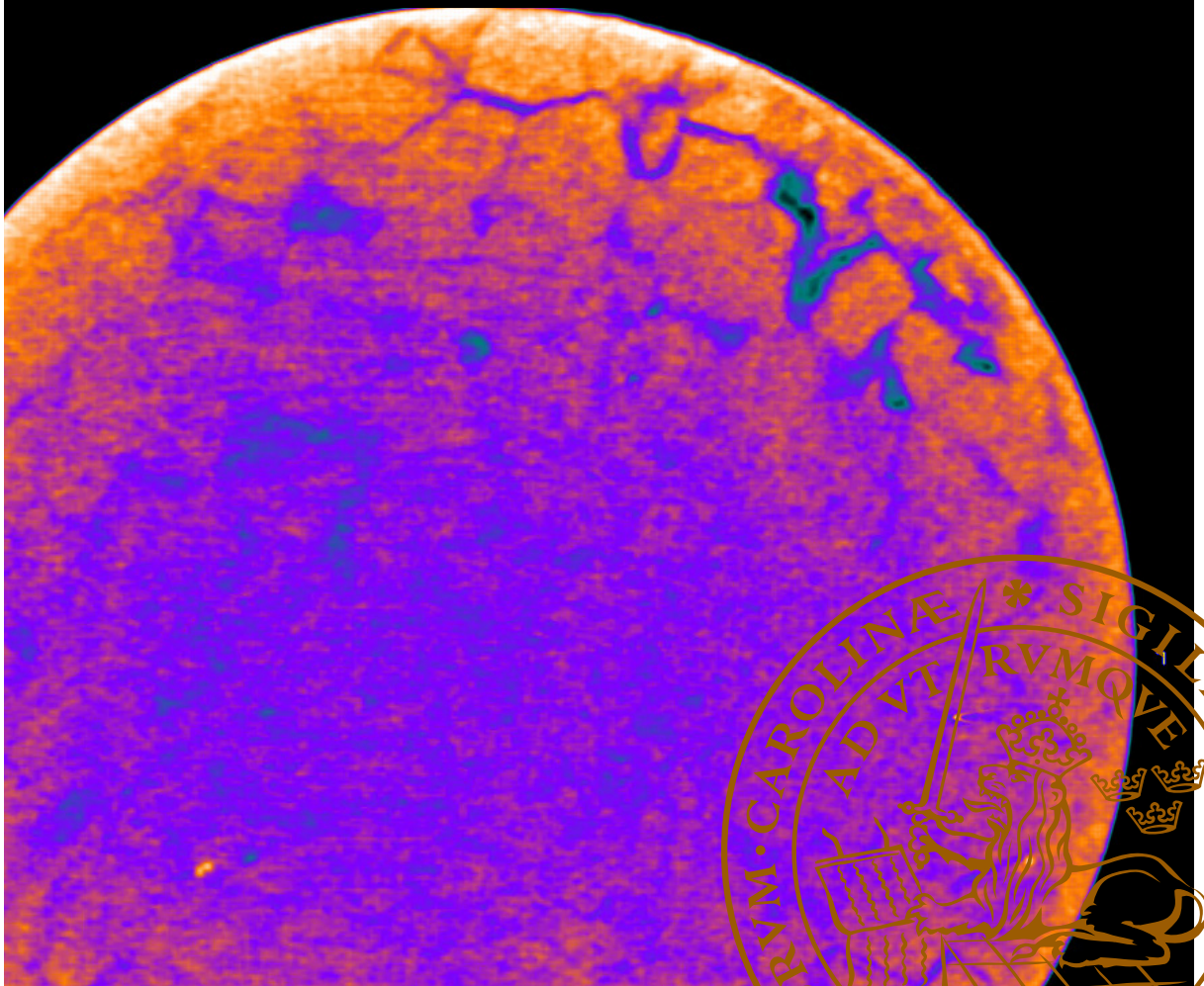
# Optically Stimulated Luminescence Dosimetry with NaCl Pellets

Dosimetry for prospective applications

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LOVISA WALDNER

DEPARTMENT OF TRANSLATIONAL MEDICINE | LUND UNIVERSITY





## Optically Stimulated Luminescence Dosimetry with NaCl Pellets



# Optically Stimulated Luminescence Dosimetry with NaCl Pellets

Dosimetry for prospective applications

Lovisa Waldner



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DOCTORAL DISSERTATION

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<b>Abstract</b> <p>The use of ionising radiation in medical diagnostics and therapies, for energy production, and in research, require tools for radiation dosimetry. To follow the principles of justification, optimisation and dose limits both retrospective and prospective radiation exposure assessments are needed. In many workplaces, where ionising radiation is used, there is a limited supply of dosimeters for other applications than occupational dosimetry, either because of centralised or outsourced dosimetry services, or economic reasons that limit the dosimetry in general. A dosimeter based on NaCl pellets is a potential complement to commercial dosimeters in many situations, e.g. where many cost-effective detectors are needed, or when there are no alternatives available. NaCl is a luminescent material that has been thoroughly studied for retrospective dosimetry. However, NaCl may be further utilised also in prospective dosimetry. The basic luminescence and dosimetric properties of the NaCl make it suitable for applications both in personal and area dosimetry. NaCl compressed to pellets is especially useful for dosimetry as the handling is simplified and the dosimetric properties are improved compared to grains of salt. Using NaCl, assessments of low radiation doses are possible, with detection limits around 20 µGy. Furthermore, the signal to dose response is linear between at least 1 and 300 mGy and in the same dose range, absorbed doses are well estimated using a single calibration dose. The OSL signal is stable over at least one month, enabling long term measurements of chronic radiation exposure as well as acute exposures evaluated after some days or weeks. Due to the large signal over response in NaCl compared to tissue for low energy photons, a badge with appropriate filtering is required to measure the dose quantities of interest, such as personal and ambient dose equivalent (<math>H_p(10)</math> and <math>H'(10)</math>, respectively). When investigating the response to different radiation qualities, the signal yield seems to depend on the LET of the radiation and no response is indicated for neutrons. For all 102 salts investigated, there are variations in dosimetric properties, larger than a factor of 10 for some properties, but all salts may still be used for prospective OSL dosimetry. A dedicated readout unit would be beneficial to further increase the usability of the approach. Preliminary implementations of the NaCl for dosimetry in radiation protection and optimisation applications show satisfactory results, in many areas with dose estimations in agreement with other detector systems as well as theoretical calculations. Further implementation of the NaCl pellet dosimetry as a complement to commercial dosimeters would enable radiation protection assessments previously not possible due to limited access to alternative detectors.</p>		
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# Optically Stimulated Luminescence dosimetry with NaCl pellets

Dosimetry for prospective applications

Lovisa Waldner



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# Abstract

The use of ionising radiation in medical diagnostics and therapies, for energy production, and in research, require tools for radiation dosimetry. To follow the principles of justification, optimisation and dose limits, both retrospective and prospective radiation exposure assessments are needed. In many workplaces, where ionising radiation is used, there is a limited supply of dosimeters for other applications than occupational dosimetry, either because of centralised or outsourced dosimetry services, or economic reasons that limit the dosimetry in general. A dosimeter based on NaCl pellets is a potential complement to commercial dosimeters in many situations, e.g. where many cost-effective detectors are needed, or when there are no alternatives available. NaCl is a luminescent material that has been thoroughly studied for retrospective dosimetry. However, NaCl may be further utilised also in prospective dosimetry. The basic luminescence and dosimetric properties of the NaCl make it suitable for applications both in personal and area dosimetry. NaCl compressed to pellets is especially useful for dosimetry as the handling is simplified and the dosimetric properties are improved compared to grains of salt. Using NaCl pellets, assessments of low radiation doses are possible, with detection limits around 20  $\mu\text{Gy}$ . Furthermore, the signal to dose response is linear between at least 1 and 300 mGy and in the same dose range, absorbed doses are well estimated using a single calibration dose. The OSL signal is stable over at least one month, enabling long term measurements of chronic radiation exposure as well as acute exposures evaluated after some days or weeks. Due to the large signal over response in NaCl compared to tissue for low energy photons, a badge with appropriate filtering is required to measure the dose quantities of interest, such as personal and ambient dose equivalent ( $H_p(10)$  and  $H^*(10)$ , respectively). When investigating the response to different radiation qualities, the signal yield seems to depend on the LET of the radiation and no response is indicated for neutrons. For all 102 salts investigated, there are variations in dosimetric properties, larger than a factor of 10 for some properties, but all salts may still be used for prospective OSL dosimetry. A dedicated readout unit would be beneficial to further increase the usability of the approach. Preliminary implementations of the NaCl for dosimetry in radiation protection and optimisation applications show satisfactory results, in many areas with dose estimations in agreement with other detector systems as well as theoretical calculations. Further implementation of the NaCl pellet dosimetry as a complement to commercial dosimeters would enable radiation protection assessments previously not possible due to limited access to alternative detectors.

# Populärvetenskaplig sammanfattning

All exponering av joniserande strålning ökar risken för framtida hälsoproblem (t ex. cancer). För yrkesverksamma med joniserande strålning t ex. på sjukhus och inom kärnkraftsindustrin finns ett behov av strålskydd och stråldosmätningar, för att undvika sådana hälsoeffekter. Därför övervakas all personal som arbetar med strålning med dosimetrar, för att kontrollera att deras exponering är under lagstadgade dosgränser. I många situationer där strålning används är tillgången på dosimetrar begränsad, antingen för att dosimetritjänster har centraliserats eller hanteras av privata företag, eller pga. ekonomiska skäl där dosimetri inte prioriteras, eller är begränsad av andra skäl.

Vanligt hushållsalt, som består av natriumklorid, NaCl, kan användas för att detektera joniserande strålning då NaCl samlar på sig en signal som kan läsas ut med en teknik som kallas optisk stimulerad luminiscens, OSL. Denna OSL signal kan sedan översättas till en stråldos.

För att underlätta hanteringen och potentiellt förbättra de dosimetriska egenskaperna, föreslås här att saltet pressas samman till pellets. De undersökta salten har detektionsgränser kring 20  $\mu\text{Gy}$  (motsvarar ungefär två tandröntgen) vilket gör det möjligt att mäta mycket låga stråldoser med NaCl som pellets. Dessa NaCl pellets har också ett linjärt samband mellan signal och dos mellan 1 till 300 mGy, vilket är ett dosintervall relevant för persondosimetri. OSL-signalen är stabil i minst en månad vilket möjliggör långtidsmätningar eller dagar/veckor mellan bestrålning och utläsning av signalen. Vi föreslår därför att NaCl pellets kan användas som ett komplement till de kommersiella dosimetrar som används idag, speciellt när stora mängder detektorer behövs men även då det inte finns några tillgängliga alternativ. Grundliga undersökningar av de dosimetriska egenskaperna för NaCl visar att alla salt lämpar sig för både persondosimetri och punktmätningar av strålningen i omgivningen. En enkel utläsningsenhet för NaCl pellets skulle ytterligare underlätta vidare implementering och öka användbarheten. Hittills har de föreslagna NaCl pelleten visat lovande resultat som stämmer väl överens med tillgängliga metoder, t ex. i olika sjukhusmiljöer med joniserande strålning. En dosimeter baserad på pellets av NaCl, som ett komplement till kommersiella dosimetrar, skulle möjliggöra strålskyddstillämpningar som tidigare inte varit möjliga på grund av begränsad tillgång på andra detektorer.

# Abbreviations

CW – Continuous wave

IR – Infra red

LED – Light emitting diode

LET – Linear energy transfer

MCNP – Monte Carlo N-Particle Transport Code

OSL – Optically stimulated luminescence

PMMA – Polymethyl methacrylate

PMT – Photo multiplier tube

TL – Thermoluminescence

UV – Ultra violet

MDD – Minimum detectable dose

MMD – Minimum measurable dose

CPE – Charged particle Equilibrium

SAR – Single Aliquot Regeneration

# Original papers

This thesis is based on the following four publications. They will be referred to by their Roman numerals.

- I. Waldner L, Bernhardsson C. Physical and dosimetric properties of NaCl pellets made in-house for the use in prospective optically stimulated luminescence dosimetry applications. *Radiat Meas.* 2018;119:52–7.
- II. Waldner L, Rääf C, Hinrichsen Y, Herrnsdorf L, Bernhardsson C. Experimentally determined and Monte Carlo–calculated energy dependence of NaCl pellets read by optically stimulated luminescence for photon beams in the energy range 30 keV to 1.25 MeV. *Radiol. Prot.* 2020;40:1321
- III. Waldner L, Rääf C, Bernhardsson C. NaCl pellets for prospective dosimetry using optically stimulated luminescence: Signal integrity and long-term versus short-term exposure. *Radiat Environ Biophys.* 2020;59(4):693-702
- IV. Waldner L, Bernhardsson C. Dosimetric properties of NaCl pellets made in-house from 102 different types of NaCl collected from 47 countries around the world and their suitability for prospective optically stimulated luminescence dosimetry. Submitted to *Rad. Mes.*

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# Related publications

Bernhardsson C, Waldner L, Vodovatov A. Advancements in prospective dosimetry with NaCl read-out by optically stimulated luminescence. *Med Phys Balt states* 2017;13:26–30.

Waldner L, Bernhardsson C, Woda C, Trompier F, van Hoey O, Kulka U, et al. The 2019-2020 EURADOS WG10 and RENEB field test of retrospective dosimetry methods in a small-scale incident involving ionizing radiation. *Radiat Res.* 2021;195(3):253–64.

# Introduction

When using ionising radiation in medical diagnostics or therapies, for energy production, or in research, there is a need for different ways of dosimetry, both retrospective and prospective. The ICRP states dose limits as one of its three main principles for radiation protection [1]. To follow national laws and guidelines, occupational exposure need to be systematically monitored and practices optimised. The availability of detectors for dosimetry in e.g. hospitals is often limited to personal dosimetry, with some additional availability of dosimeters for eye lens or finger dose measurements. Hence, the dosimeters are often earmarked for specific measurement situations and when dosimetry services are centralised or outsourced to external companies, the possibilities for new applications is limited. In some places around the world there can also be economic factors that limit the access to detectors because the dosimetry is not prioritised. Furthermore, in emergency situations involving ionising radiation, it would be beneficial to produce and distribute large quantities of detectors at a low cost. A simple NaCl pellet dosimeter may be distributed to aid medical triage and to reassure the worried well, or as an alternative to justify evacuation in areas where the contamination and potential exposure to ionising radiation following a radio nuclear event is unknown.

A simple, low cost, dosimeter based on NaCl pellets is a useful complement to commercial dosimeters where a large numbers of detectors are needed or when there is a shortage of alternative dosimeters, thereby enabling radiation protection in applications previously not possible or limited. Combining commercial dosimeters with NaCl pellets enables improved dose mappings and phantom studies with larger coverage or with better spatial resolution of measuring points. The configuration of the suggested NaCl pellets allow for the use of already existing dosimetry phantoms intended for other types of detectors, e.g. anthropomorphic phantoms. The low cost and ready availability of NaCl make it possible to produce large quantities of detectors, for one-time use, and in combination with a simple readout unit, low cost dosimetry may be performed where dosimetry possibilities previously have been limited.

# Objectives

The general aim of this thesis was to develop a prospective passive OSL dosimeter made from NaCl pellets, for applications in e.g. medical clinics, nuclear industry and for environmental radiology. The specific objectives were:

- to find the optimal configuration of NaCl pellets in terms of mechanical stability and homogeneity, depending on grain size and compression force (Paper I),
- to investigate the basic material characteristics in terms of luminescence and dosimetric properties such as OSL signal yield, signal to dose linearity and detection limits (Paper I),
- to optimise a OSL readout protocol for the NaCl pellets (Papers I and III),
- to investigate the photon energy dependence of the NaCl pellets by determining how the magnitude of the OSL signal per given dose depends on photon energy (Paper II),
- to investigate the dosimetric properties affected by extended/chronic exposures, such as signal stability over time and signal yield over time (Paper III),
- to investigate how the dosimetric and luminescence properties of NaCl vary depending on additives and type of salt for a large set of salts collected from all over the world (Paper IV),
- to investigate if and how the OSL signal depends on the type of radiation during exposure, specifically by studying the OSL signal from exposure to neutrons, protons, electrons, photons and alpha particles. (manuscript in preparation)

# Background

Household salt, consisting of sodium chloride (NaCl), is sensitive to ionising radiation in that it may store a radiation induced signal that can be read by optically stimulated luminescence, OSL or thermoluminescence, TL. The TL and OSL properties of NaCl have been investigated for a number of different types of salt, for applications in both retrospective and accident dosimetry [2–9]. Because of the favourable dosimetric properties of NaCl, some of the focus has turned towards prospective dosimetry [10–14]. With prospective dosimetry, it is possible to make optimised use of the dosimetric properties of NaCl, with better control of the measurement conditions (as compared with NaCl for retrospective dosimetry). Due to the rather cumbersome handling of salt, when it is in grain form, it has been suggested to compress the NaCl grains to pellets [10,13–15], thereby simplifying the handling of the salt both during measurement and readout. Compressing pellets from NaCl of selected grain sizes also reduces the uncertainties arising from uneven heating of different shaped salt grains during TL or preheat [16], and the varying signal yield from different sized and/or irregular grains [17,18]. Most investigations of NaCl within retrospective applications focus on doses in the order of 1-10 Gy, or higher. For prospective personal and area dosimetry, much lower doses, in the order of milligray are of more importance as it is rather the stochastic effects of exposure to radiation that are central, than the deterministic effects.

In the development of a passive luminescence dosimeter with a new type of detector material, many things need to be considered; from the basic physical characteristics and luminescence properties of the material, to the radiation protection quantity when used in different applications. For personal dosimetry, the main purpose of attempting to accurately quantify external exposures to ionising radiation is to estimate the risk of late health effects of the radiation, e.g. cancer. One of the quantities for estimating these effects is the effective dose but it is a theoretical quantity based on epidemiological data that cannot be directly measured. Instead operational quantities such as personal dose equivalent,  $H_p(d)$ , are used in practice. To measure  $H_p(d)$ , a personal dosimeter is used. These quantities are defined at a certain depth in tissue ( $d$ ), and therefore, dosimeters for measuring these quantities should be calibrated in well-defined radiation fields in fixed geometries. For such calibration to provide accurate dose estimations, the material's dosimetric properties need to be well established and all conditions in relation to the cavity theory need to be fulfilled. For any material, the first thing to investigate is the material itself, to

determine fundamental properties; such as the stability of the luminescence signal, the signal yield and detection limits. A thorough understanding of the material's dosimetric properties is necessary before any further steps are taken towards an acceptably working personal dosimeter.

## Fundamental dosimetry

There are several quantities used for radiation protection and personal dosimetry [1,19,20]. Below is a short description of some of the physical, protective and operational quantities that are important in the development of a personal or area dosimeter.

### Dose Quantities

The physical quantities for radiation protection include fluence, kerma and absorbed dose and they can all be estimated with absolute measurements such as calorimetry or ionisation chamber methods.

The fluence,  $\phi$ , is defined as the number of photons (or other indirect ionising radiation),  $N$ , crossing an area of infinitesimal size,  $da$ , according to Equation 1.

$$\phi = \frac{dN}{da} [\text{m}^{-2}] \quad \text{Equation 1}$$

Taking into account the energy of the individual photons that cross the area  $da$  gives the energy fluence,  $\Psi$ , that can be defined according to Equation 2, where  $R$  is the total energy of the  $N$  photons that strike the infinitesimal area  $da$ .

$$\Psi = \frac{dR}{da} [\text{J} \cdot \text{m}^{-2}] \quad \text{Equation 2}$$

For indirect ionising radiation such as photons and neutrons, kerma (kinetic energy released per unit mass),  $K$ , is defined as the energy transferred,  $E_{tr}$ , to a small volume with mass  $m$ .  $E_{tr}$  is defined as the kinetic energy that is transferred to charged particles in a small volume, regardless of how the energy is spent after this transfer. Kerma is defined according to Equation 3.

$$K = \frac{dE_{tr}}{dm} = \Psi \cdot \frac{\mu_{tr}}{\rho} [\text{Gy}] \quad \text{Equation 3}$$

The mass energy transfer coefficient,  $\mu_{tr}/\rho$ , depends on the photon energy and the atomic number of the medium. It describes the fraction of kinetic energy transferred from photons to charged particles. The Kerma can be divided into two components,

collision kerma,  $K_{col}$  and radiative kerma,  $K_{rad}$ .  $K_{col}$  is the result of coulomb force interactions with orbital electrons and leads to local energy depositions in a material.  $K_{rad}$  includes bremsstrahlung from the slowing down of charge particles, and in-flight annihilation. Related to  $K_{col}$ , not taking into account radiative losses, is the mass energy absorption coefficient,  $\mu_{en}/\rho$ . This is defined according to Equation 4, where  $g$  is the fraction of kinetic energy which is lost in radiative energy loss processes. For low photon energies and low atomic numbers,  $g$  is usually small, and may be neglected.

$$\frac{\mu_{en}}{\rho} = \frac{\mu_{tr}}{\rho} (1 - g) \text{ [cm}^2/\text{g]} \quad \text{Equation 4}$$

The absorbed dose,  $D$ , is relevant to all types of ionising radiation, both direct and indirectly ionising. It relates to the energy imparted,  $\epsilon$ , that is the absorbed energy in the infinitesimal volume with mass  $m$ . The energy imparted is defined as the sum of all energy depositions in a volume element  $dV$ . Absorbed dose is defined according to Equation 5. Under conditions of charge particle equilibrium, CPE, the absorbed dose is equal to the collision kerma. In an environment with CPE, the number of charged particles leaving a small volume  $dV$  is equal to the number of charged particles entering the same volume.

$$D = \frac{d\epsilon}{dm} = \Psi \cdot \frac{\mu_{en}}{\rho} \text{ [Gy]} \quad \text{Equation 5}$$

Protection quantities, or risk related quantities, such as equivalent dose,  $H_T$  and effective dose,  $E$ , have been developed to account for the different biological effect of various tissues for different types of radiation. These quantities are related to late health effects such as cancer, for different tissues and organs [1]. The equivalent dose is described by Equation 6 where  $D_{T,R}$ , is the mean absorbed dose to an organ  $T$  for a given type of radiation  $R$ .  $w_R$  is a weighting factor that takes into account tissue effects by different types of radiation.

$$H_T = \sum_R w_R \cdot D_{T,R} \text{ [Sv]} \quad \text{Equation 6}$$

For photons and electrons,  $w_R$  is 1, for protons 5, and for alpha particles 20 [1]. For neutrons  $w_R$  is determined by a continuous function, depending on energy, ranging from about 2 to 21.

The effective dose, described by Equation 7, is the sum of the equivalent doses for each organ, multiplied with a weighting factor that takes into account the different organs and tissues radiation sensitivity as defined by the ICRP [1].

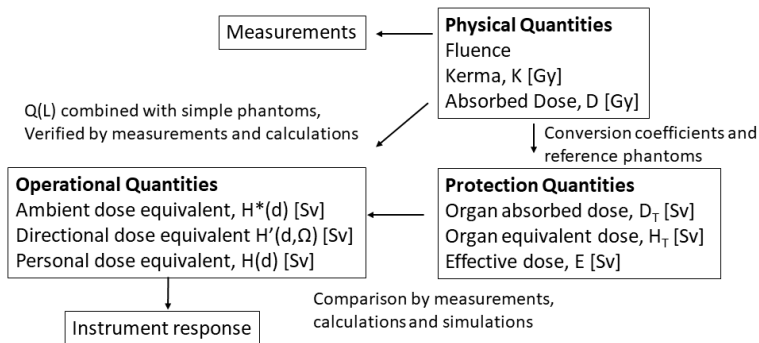
$$E = \sum_T w_T \cdot H_T \text{ [Sv]} \quad \text{Equation 7}$$

The weighting factors are derived from epidemiological studies of e.g. atomic bomb survivors [21]. This implies that  $w_T$  is an average for men and women, of varying age, and therefore does not take into account individual variations in radiation sensitivity.

Relating a radiation exposure to a future radiation detriment, e.g. risk for stochastic effects, is a common endpoint in radiation protection but as the effective dose is reliant on weighting factors based on epidemiologic studies, the main quantity cannot be directly measured. Instead, operational quantities have been developed for estimation of the effective dose by direct measurements. Personal dose equivalent,  $H_p(d)$ , is used for external exposure of the whole body, eye lens and skin whereas the ambient dose equivalent,  $H^*(10)$  is used for area monitoring. The quantities are developed by the ICRU and are based on the dose equivalent,  $H$ , which is the product of the absorbed dose and a LET dependent factor [22].

The ambient dose equivalent,  $H^*(10)$ , is independent of the incident direction of the radiation and measured at a 10 mm depth in the ICRU sphere (a 30 cm in diameter sphere made from tissue equivalent material) [23]. The angular dependent counterpart is the directional dose equivalent,  $H'(0.07)$ , defined as the dose equivalent at a 0.07 mm depth in the ICRU sphere. Both  $H^*(10)$ , and  $H'(0.07)$  can be calculated from absorbed dose measurements in air, using tabulated data (see e.g. [24]).

For individual monitoring, the quantity personal dose equivalent,  $H_p(d)$ , is used. This is the equivalent dose in soft tissue and for a whole body exposures it is defined at a depth of 10 mm,  $H_p(10)$ . For skin and extremities, the organ dose equivalent is measured at 0.07 mm,  $H_p(0.07)$ , and for the lens of the eye it is measured at 3 mm,  $H_p(3)$ .  $H_p(d)$  has a directional dependency and a personal dosimeter is usually calibrated in an anterior-posterior direction.



**Figure 1.** Relationship between the various quantities used in radiation protection. Adapted from ICRP 116 [19].

## Cavity theory

To determine an absorbed dose to a medium, a radiation sensitive detector may be inserted in the medium. The detector and medium are usually not made from the same material and to relate the dose to the detector to that of the medium, cavity theory is used (see e.g. Attix, 1986 [20]). Cavities are classified as small, intermediate or large in relation to the range of secondary charge particles produced by photons in the medium.

A small detector is defined as small enough not to disturb the fluence of charged particles in a medium. For this to stay true, CPE is necessary. Such type of detector is referred to as a Bragg-Grey cavity or an electron detector. For small detectors, the absorbed dose to a material,  $D_{med}$ , is determined by Equation 8 where  $D_{det}$  is the dose to the detector and  $S/\rho$  is the unrestricted stopping power of a given material for a specified electron energy. The stopping power is divided into two parts, one arising from interactions with atomic orbital electrons (collision stopping power), and one radiative part which arises from interactions with atomic nuclei (radiative stopping power). The total stopping power is the sum of the collision and radiative stopping power. The unrestricted stopping power differs from the restricted stopping power in that it considers all energy depositions from secondary electrons, not only the localised energy transfer.

$$D_{med} = D_{det} \cdot \left( \frac{S}{\rho} \right)_{med,det} \quad [\text{Gy}] \quad \text{Equation 8}$$

If the detector is large compared to the range of secondary particles in a material, the dose to the material will instead depend on the mass absorption coefficient ratio of the detector and target medium as the interaction is governed by the photon interactions. This kind of large detector is also referred to as a photon detector. For large detectors, the absorbed dose to a material is determined by Equation 9, where  $\mu_{en}/\rho$  is the mass absorption coefficient of a given material for a specified photon energy. Again, CPE is a requirement.

$$D_{med} = D_{det} \cdot \left( \frac{\mu_{en}}{\rho} \right)_{med,det} \quad [\text{Gy}] \quad \text{Equation 9}$$

Detectors of intermediate size where a mix of electron and photon interactions determine the absorbed dose in a material are called Burlin Cavities. In practice, many detectors, including the one described in this thesis, are Burlin cavities. As the range of the secondary particles are determined by the photon or electron energies of the primary radiation field, detectors will be considered small or large depending on photon energy [20].



# Luminescence

The most common types of passive dosimeters for assessing cumulative external doses are based on the luminescence phenomena, either on thermoluminescence, TL, or optically stimulated luminescence, OSL. These types of dosimeters are mainly used for occupational monitoring of radiation exposure in e.g. hospitals, the nuclear industry, and in research. The dosimeters are worn at all times in the workplace and are usually evaluated on a regular (e.g. monthly) basis. The basic principles for TL and OSL detectors are the same and only the method for obtaining a quantifiable signal differs.

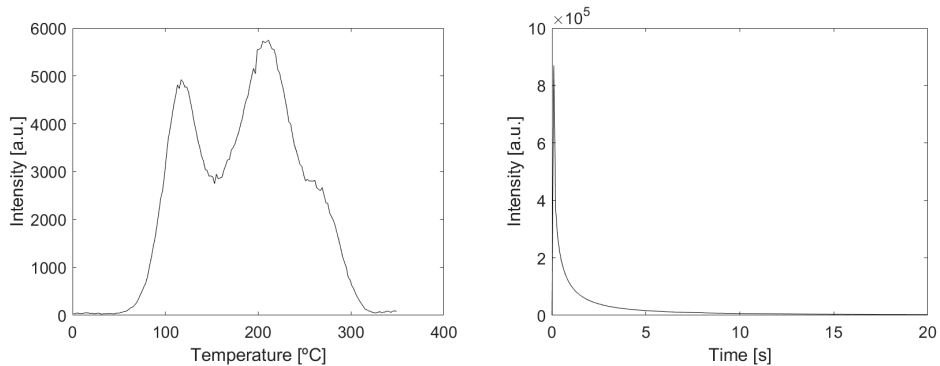
## Basic principle

Some crystalline materials, usually insulators and semiconductors, have the ability to store information about a previous exposure to ionising radiation in their crystal lattice [25]. In a perfect crystal, the energy levels of the solid consists of a valence band and a conduction band with a so called forbidden bandgap between. However, crystals are never perfect but contain impurities and defects which causes localised energy levels to form in the forbidden bandgap. Defects in the crystal lattice may be caused by displacements of atoms, missing or foreign atoms, or extra atoms in otherwise unoccupied positions. The localised energy levels are called electron or hole traps if they can capture a charge carrier, or recombination centres if it can capture charges of opposite sign. When the material is exposed to ionising radiation, electrons are excited from the valence band to the conduction band where they can roam freely. When they lose enough energy, they return to the valence band, but some electrons get trapped at localised energy levels. This results in a metastable state that can be stable for up to several hundred thousand years. By stimulating the material with light or heat, the electrons are re-excited to the conduction band and when they de-excite and combine with holes at recombination centres, luminescence photons are emitted. These luminescence photons may be detected as a signal by for example a photomultiplier tube, PMT. The intensity of the photon emission is proportional to the radiation exposure.

## Stimulated luminescence

The dosimeter materials are stimulated using either heat (TL) or light (OSL) to give trapped electrons enough energy to be released from the localised energy states in the crystal lattice. TL has been used for passive dosimetry since the 1950s [26]. When linearly heating the material and simultaneously counting the luminescence photons, a glow curve is produced as a function of temperature (Figure 2, left). OSL is a slightly newer technique, suggested [27,28] a long time before the usefulness in

dosimetry was realised [29]. Common illumination lights include blue or green LED's, IR, laser or UV light. For the continuous wave stimulation mode, (CW) OSL, the material is illuminated with a fix LED effect while simultaneously counting the emitted luminescence photons that results in a signal decay curve as a function of time (Figure 2, right).



**Figure 2.** Thermoluminescence glow curve (left) and optically stimulated luminescence decay curve (right) 1 hour after exposure to 1 Gy of beta radiation, for NaCl pellets made from Falksalt Finkornigt hushällssalt, Salinity AB, Sweden.

## The Risø TL/OSL reader

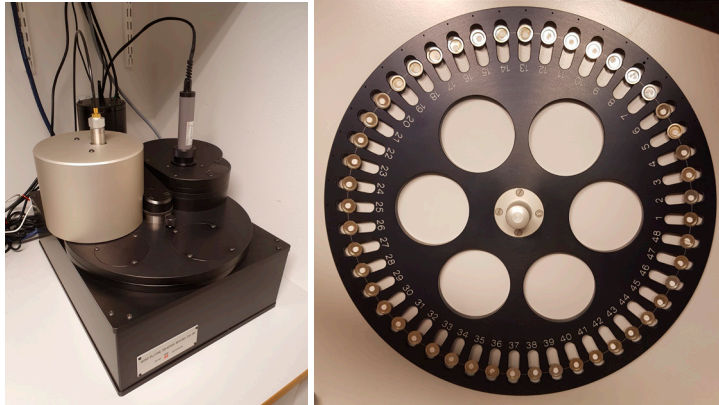
All TL and OSL signal readouts were performed using Risø TL/OSL readers (TL/OSL-DA-15 and DA-20, DTU Physics, Denmark). The main components of the Risø TL/OSL readers include: 1) a carousel for sample cups, 2) stimulation light sources, 3) a  $^{90}\text{Sr}/^{90}\text{Y}$  calibration source 4) a heating element, and 5) a photo multiplier tube, PMT [30,31]. The reader enables individual stimulation, heating and irradiation of 48 samples at the time.

Samples are placed on stainless steel cups which are situated on a detachable sample carousel that fits 48 cups. Each separate sample cup may be heated, irradiated and optically stimulated separately in the reader. The heating element also acts as a lift and when a sample is in the correct position it is lifted up towards the PMT for readout, to optimise the light collection geometry.

For TL stimulations, the temperature of the heater/lift is linearly increased while the PMT registers the luminescence photons. For OSL stimulation, LED lights are used to illuminate the sample while the luminescence signal is registered by the PMT. Depending on the need, the readers may be equipped with blue, green or IR LEDs for light stimulation. The PMT has a transmission window around 200-400 nm and is equipped with a Hoya U-340 filter (for blue stimulation) to filter out scattered light from the stimulation. For CW light stimulation and readout, it is important that

the wavelengths of the stimulation and emission light are well separated, or the PMT will not be able to distinguish between stimulation and emission photons.

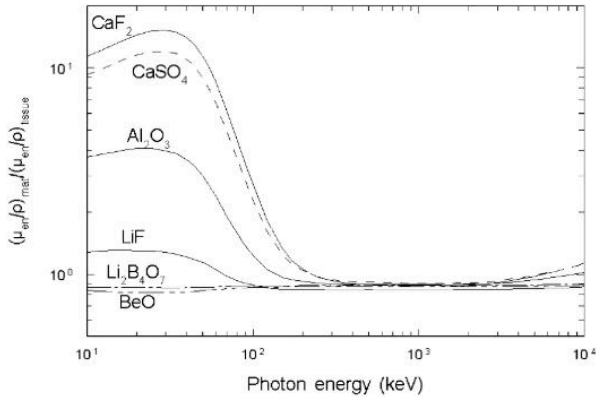
The internal  $^{90}\text{Sr}/^{90}\text{Y}$  calibration source emit beta particles with a maximum energy of 2.27 MeV. The activity of the source may be chosen to fit the intended area of application or research. As a standard, quartz is used to calibrate the radiation source [32] and to determine the current dose rate.



**Figure 3.** The Risø TL/OSL DA-20 reader (left) and the sample carousel fitting 48 samples at the time (right).

## Materials for luminescence dosimetry

One of the most common materials for TL dosimetry is lithium fluoride, LiF, usually doped with magnesium, titanium, copper or potassium. It was one of the first materials to be suggested for TL dosimetry and it has been used for this purpose since the 1950s [26]. The LiF dosimeter materials may be used to measure radiation exposures over several orders of magnitude, with a linear signal to dose response between 1 Gy and 10 Gy [33]. The thermal fading is limited to around 3-10% per year [34], and because the effective atomic number,  $Z_{eff}$  of LiF ( $Z_{eff}=8.4$ ) and tissue ( $Z_{eff}=7.35$ ) are similar, there is only a small over response to low energy photons as shown in Figure 4 [33].



**Figure 4.** The energy response curves for some dosimeter materials compared to tissue under conditions of CPE.  $Z_{eff}$  of tissue is 7.35 to be compared with  $Z_{eff}$  for LiF = 8.4,  $Al_2O_3$  = 11.3,  $CaF_2$  = 16.9,  $CaSO_4$  = 15.5 and NaCl at 15.2. Theoretically, the energy dependence for NaCl should be similar to that of  $CaSO_4$ . Bos 2001 [33].

For OSL dosimetry, aluminium oxide,  $Al_2O_3$ , doped with carbon, is the most commonly used material. Aluminium oxide was developed for TL dosimetry [35] but has been found to be more effective as a OSL dosimeter material [36,37]. The OSL signal yield is high, the signal to dose response is linear over several orders of magnitude, and the loss of signal to fading is less than 5% per year [38,39]. As all other OSL materials it needs to be shielded from light during and after irradiation, until signal readout, for the signal not to be depleted. Because of the difference in  $Z_{eff}$  between  $Al_2O_3$  ( $Z_{eff}$  = 11.3) and tissue, there is a larger signal over response to low energy photons compared to LiF.

Sodium chloride (NaCl) is a halide mineral that may be evaporated from oceans and mined from sedimentary rock and is accessible almost anywhere around the world. It is a crystalline insulator that consists of equal amounts sodium ions and chloride anions resulting in a  $Z_{eff}$  of 15.2. Because of the high  $Z_{eff}$  compared to tissue, a large signal over response is expected for low photon energies. The thermoluminescence from NaCl have been extensively studied (see [25] and references therein) and both the OSL and TL properties of NaCl have been investigated for applications in retrospective and accident dosimetry. The dosimetric properties, in combination with the accessibility and low cost, makes it an interesting material for simple and cost-effective dosimetry, both for retrospective and prospective applications.

## Dosimetric and luminescence properties

In the developing of a passive personal or area TL or OSL dosimeter, several dosimetric and luminescence properties are fundamental to investigate. The general

considerations for a new dosimeter material are described in this section whereas the specific methods for evaluating the dosimetric properties of NaCl pellets are described in the Method section.

### **Signal to dose linearity and dose reconstruction ability**

For accurate dose estimations, the relationship between the radiation induced signal and the exposure needs to be known. A simple relation between signal and dose is preferable as the conversion from signal to dose becomes straightforward. A material with a constant signal per given dose for any dose within a dose range exhibits a linear relationship. If the signal to dose ratio decreases with increasing doses, the relationship is sublinear and if the signal to dose ratio increases with increasing doses it is supralinear [40]. Sublinearity may be observed close to the saturation levels either where the radiation exposure has caused nearly all available electron traps to be filled, or the luminescence signal saturates the PMT.

The ability to correctly estimate the radiation dose from an unknown exposure is of importance for accurate dosimetry. It may be evaluated either as a dose recovery ratio where a given dose is compared to the estimated dose or as a linear equation fitted to a graph of estimated doses as a function of given doses. For dose recovery ratios, a ratio of 1 indicates perfect dose reconstruction ability and for the linear equation, a slope of 1 indicates the same.

### **Specific luminescence**

A well working dosimeter materials must exhibit a high signal yield. The quantity specific luminescence,  $c_{spec}$  (counts·mGy<sup>-1</sup>·mg<sup>-1</sup>), which is the absorbed dose and weight normalised OSL signal, is a useful measure for quickly determining a comparable quantity of the signal yield of a salt. A similar definition can be found in Thomsen et. al [6].

### **Detection limits**

For many radiation protection applications, the dose rates are close to the natural background levels, which means it is desirable that the dosimeters are able to estimate low doses (well below 100 mGy), in order to assess  $H_p(10)$  levels below e.g. 1 mGy per month to assure compliance with the dose limit of 20 mSv y<sup>-1</sup> for workers. Low detection limits also result in shorter measurement times. Low detection limits require low background signals from un-irradiated samples but impurities in the detector materials and electronic noise of the readout equipment may cause elevated background OSL signal.

## **Sensitisation**

The signal yield of some dosimeter materials changes when exposed to ionising radiation as more defects are created in the crystal lattice [41]. For materials where the radiation dose is quantified using a pre-established conversion factor or calibration curve, a change in sensitivity of the material may cause inaccuracies in dose estimations. If the material is damaged, e.g. a chipped detector, the pre-established curve cannot be used for dose estimations.

## **Reproducibility**

The properties of a material suitable for dosimetry need to be reproducible. For identical exposure scenarios, the results of the dose estimations should be the same. The variation among detectors handled in the same way need to be small enough to ensure accurate dose estimations. The properties of a multi-use detector are expected to remain the same for each new exposure but for detectors intended for one time use it is more important that the dosimetric properties are the same for each detector.

## **Fading**

The loss of signal in a dosimeter material comes from the depletion of certain traps in the crystal lattice. Room temperature is enough to empty the most shallow traps of some materials whereas light exposure may completely deplete the OSL signal in some materials. There is also the possibility of redistribution of trapped electrons in the lattice because of tunnelling effects [25,42].

A stable signal over time is of importance to be able to correctly estimate absorbed doses when readout does not immediately follow the exposure. A fading that is not known and correctly adjusted for at readout will lead to underestimations of the radiation exposure.

## **Signal yield over time**

Compressing a OSL sensitive crystalline material can affect the crystal lattice, possibly inducing new defects that can cause a shift among the impurity states, with a shift in emission wavelength as a consequence [43]. If this is the case, the change needs to be considered for accurate dose estimations.

## **Energy dependence**

An optimal dosimeter material for personal dosimetry shows a signal response similar to that of human tissue. Differences in effective atomic number,  $Z_{eff}$ , between

the dosimeter material and tissue will lead to a difference in signal response depending on photon energy. The difference will be largest at low photon energies where the photoelectric effect dominates and the cross section is related to the atomic number according to  $\sim Z^3$ . As mentioned previously, at CPE the energy response is related to the mass energy absorption coefficients of the dosimeter material and tissue (Eq. 9).

## **Particle response**




Other radiation qualities than photons and electrons can be of interest when estimating the absorbed dose from an unknown irradiation or exposure situation. Neutrons, protons and heavy ions may contribute to the dose depending on the dosimeter material and the exposure situation. A known response to these types of radiation is vital for correct dose estimations as their biological effect per unit absorbed dose may differ significantly from that of photons and electrons, as may the LET dependent signal induction in the dosimeter material.

# Materials and methods

## NaCl pellets

During the course of this project, over a hundred different salts were investigated to some extent. The most studied salts however are the three presented in Table 1. New packages of the most used brand of salt (*Falksalt Finkornigt hushållssalt*), were continuously purchased during the course of this study resulting in salt from different batches and possibly also of different origin. Other than sieving the salts into different size fractions, no preparation was carried out before compressing the salt to pellets. The NaCl pellets were stored in transparent plastic jars or bags under both natural and artificial light conditions. No additional bleaching of potential background signal was performed on the pellets after production.

**Table 1.** Name and packaging of the most commonly used salts in this thesis.

Name	Falksalt Finkornigt hushållssalt, Salinity AB, Sweden	Falksalt finkornigt medelhavssalt, Salinity AB, Sweden	Sodium Chloride, reagent grade, Scharlau, Scharlab, Spain
Salt #	1	2	3
Package			

The NaCl pellets in Papers I-III were produced using a specially-made tool (Promech lab, Sweden) and a hydraulic hand press (Hamron, Sweden), both shown in Figure 5, using a pressure of 0.8 tonnes per pellet. The optimal pressure and grains size fraction used to produce the most mechanically stable pellet was investigated



in Paper I. For the Promech tool a pressure of 3 tonnes produces five mechanically stable pellets (0.8 tonnes per pellet) and the optimal grain size fractions were determined to be either 100-250  $\mu\text{m}$  or 250-400  $\mu\text{m}$  depending on the salt. The produced pellets are 4 mm in diameter and  $0.8\pm 0.2$  mm thick, made from about 20 mg of NaCl each.

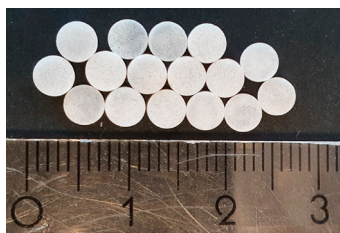
In Paper IV, a standard desktop tablet press tool (TDP 0 Desktop Tablet Press, LFA Machines Oxford Ltd), shown in Figure 6, was used to produce NaCl pellets of the same size, 4 mm in diameter and  $0.8\pm 0.2$  mm thick. The semiautomatic press produces one pellet per rotation of the lever, with the same dimensions using a similar pressure (about 0.5 tonnes) for each rotation ( $\sim 20$  pellets per minute).



**Figure 5.** The in-house developed press tool (left) and the hydraulic hand press (right) used for making the pellets in Papers I-III.



**Figure 6.** The standard desktop tablet press tool used for making the pellets in Paper IV (right). The left image shows a close-up view of the die cavity and the upper punch.



**Figure 7.** Photo of the NaCl pellets, as produced by the standard desktop press tool, next to a ruler (cm scale). The pellets are 4 mm in diameter and 0.8-1.0 mm thick.

## OSL and TL signal readouts

### Reader specifications

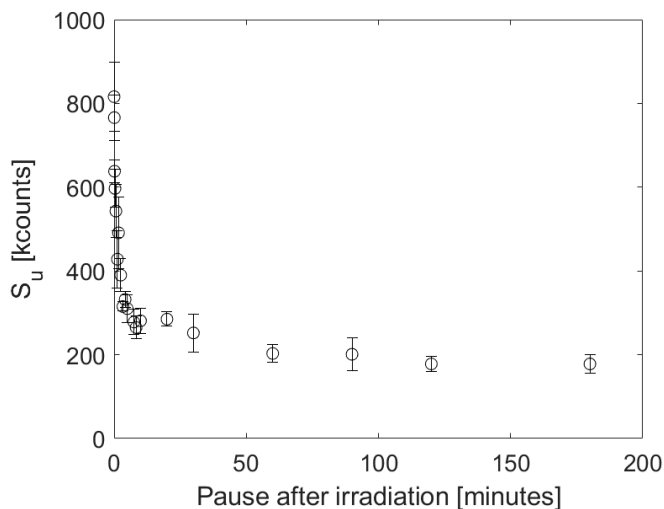
All TL and OSL signal readouts during this project were performed using two different Risø TL/OSL readers (TL/OSL-DA-15 and DA-20, DTU Physics, Denmark). The readers are equipped with internal  $^{90}\text{Sr}/^{90}\text{Y}$  radiation sources (20 MBq as of 9<sup>th</sup> of April 2009 and 100 MBq as of 22<sup>nd</sup> of October 2010) with absorbed dose rates of  $0.58 \pm 0.01 \text{ mGy s}^{-1}$  (as of 23<sup>rd</sup> of April 2021) and  $5.03 \pm 0.1 \text{ mGy/s}$  (as of 21<sup>st</sup> of June 2021) to quartz (calibration quartz, DTU Nutech, Batch 123 [32]), respectively. The activities of the sources were specifically chosen to enable irradiations resulting in absorbed doses relevant for external personal dosimetry. The dose rate to NaCl is calculated using a stopping power ratio of 0.938 [44] between NaCl and quartz ( $\text{SiO}_2$ ).

All readouts were performed using continuous wave, CW, stimulation mode by means of blue ( $\lambda=470 \text{ nm}$ ) LEDs.

### Readout protocol

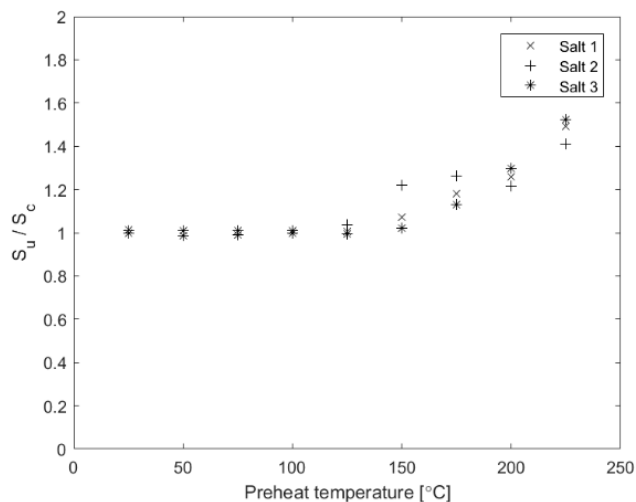
In many works where NaCl is used for dosimetry, mainly retrospective applications, the investigations show a linear signal to dose response and a graph of how the preheat temperature effects the signal yield [45–48]. Many researchers choose to use the single aliquot regeneration, SAR, protocol [49] for dose determination, a protocol that is commonly used for luminescence dating and retrospective dosimetry. After readout of the so-called natural dose (the dose which is to be quantified), several absorbed doses of increasing size are given to each sample, to establish an individual calibration curve for each sample [50]. The preheats in the SAR protocols are often based on the earlier mentioned temperature dependant signal yield investigations [3,45,51]. The SAR protocol works well for many materials but is quite time consuming. Using only one calibration dose rather than

several regenerative doses and test doses reduces the readout time significantly which is why a new readout protocol was developed for the NaCl pellets. However, it was found that the 220°C preheat previously used [3,47] for OSL signal readout of NaCl grains, alters the efficiency of the NaCl pellets [12]. When using a SAR protocol, the OSL signal from the natural dose, here referred to as the unknown dose, is read first, after a preheat to a set temperature. All irradiations that follow are thus performed on salt that has been heated and these irradiations compose the dose response curve from which the unknown dose is determined. If heating changes the efficiency of the salt, the natural dose and the SAR regenerative doses are not given under the same conditions which may cause erroneous dose estimations, something also reported by Polymeris et al. [7]. Because of the problem introduced with the 220°C preheat, this step was replaced by a pause for the signal to stabilise before readout (Paper I). Readout at different times after irradiation reveals a rapid initial decrease in signal after irradiation with a stabilisation after ~1 hour (Figure 8). Because of this finding, readout is performed at least 1 hour after exposure, both for the unknown dose and the calibration dose in the reader.



**Figure 8.** Initial signal fading after irradiation (22.5 mGy,  $^{90}\text{Sr}/^{90}\text{Y}$ ) of the NaCl pellets (Salt 1). Each data point represents the average signal of five individual pellets and the uncertainty bars correspond to 1 SD.

In some cases, a preheat is however preferable to a 1 h pause, e.g. for readout of only a few samples, and when the results are needed fast. After investigating at what temperature the efficiency of the NaCl pellets was altered (Figure 9), a preheat of 100°C was chosen for further measurements and found to be sufficient to obtain a stable signal for readout (Paper III), equivalent to the 1-hour pause.



**Figure 9.** Ratio between the measured blue light CW OSL signal ( $S_u$ ) and the calibration signal ( $S_c$ ) using different preheat temperatures before signal readout.  $D_u$  (the given/unknown dose) and  $D_c$  (the calibration dose) were fixed at the same value (21.6 mGy). The experimental uncertainties, in the order of 2% are smaller than the symbols used in the figure.

All OSL readouts using the Risø TL/OSL readers follow the protocol in Table 2 with either a pause or a 100°C preheat before readout. When the first, so called unknown, exposure takes place outside of the reader, no additional pause in the reader is required before the first readout.

**Table 2.** The protocol for OSL-signal readouts of NaCl pellets using the Risø TL/OSL reader. Either Steps 2.1 and 5.1 or Steps 2.2 and 5.2 were used for readout depending if a preheat or a pause was used.  $S_u$  – signal after exposure to an unknown dose;  $S_c$  – signal after exposure to a calibration dose.

Step	Operation and readout settings
1.	Administration of the unknown dose, $D_u$ , as accumulated during usage or by the internal $^{90}\text{Sr}/^{90}\text{Y}$ source
2.1	Preheat to a set temperature, $T$ , at a heating rate of 5°C/s and hold at $T$ for 10 seconds
2.2	Pause for 1 hour
3.	Readout of $S_u$ at ambient temperature, continuous OSL at 40% (~16 mW cm <sup>-2</sup> ) of maximum blue ( $\lambda=470\pm30$ nm) LED intensity during 20 seconds
4.	Administration of a calibration dose, $D_c$ , using the internal $^{90}\text{Sr}/^{90}\text{Y}$ source
5.1	Preheat to set temperature, $T$ , at a heating rate of 5°C/s and hold at $T$ for 10 seconds
5.2	Pause for 1 hour
6.	Readout of $S_c$ at ambient temperature, OSL at 40% of maximum blue LED intensity for 20 seconds

All TL readouts were performed with linear heating from room temperature to either 350°C or 500°C depending on the investigation. The heating rate was 2°C/s.

## Absorbed dose estimation

In this work, the absorbed dose to be estimated is referred to as the unknown dose, corresponding to the natural dose in luminescence dating. The OSL signal,  $S_u$ , from an unknown exposure of an absorbed dose  $D_u$  is defined as the integrated number of luminescence counts registered during the first 5 seconds of the 20 second OSL signal readout. The initial part of the OSL decay curve decreases rapidly during the first seconds of readout to a background that then decreases slowly. The integrated luminescence during the last 5 s of the recorded OSL decay curve is used as a background correction when further irradiations and readouts are performed on the same pellet. This is done to reduce any (potential) remaining induced signal from the former exposure and readout. The calibration signal,  $S_c$ , is thus defined as the integrated OSL signal between 0 and 5 s, corrected for the background of the previous OSL signal readout between 15 and 20 s. The absorbed dose to the NaCl pellets is then calculated according to Equation 10.

$$D_u = \frac{S_u \cdot D_c}{S_c} \text{ [mGy]} \quad \text{Equation 10}$$

As described previously, stopping power ratios and mass energy absorption coefficients are used to convert the dose to salt to the material of interest, usually air, water, or tissue.

The absorbed dose may also be estimated using a signal to dose calibration curve, pre-established by using pellets of the same batch of salt. In this case, only steps 1-3 in the protocol in Table 2 need be performed at readout. The obtained OSL signals are then compared to the calibration curve to determine the corresponding absorbed dose.

## Dosimetric and luminescence properties

The optimal dosimetric and luminescence properties of a luminescent dosimeter material are described in the background section. All of these properties have been extensively investigated in Papers I-IV to form a basis for introducing NaCl pellets as a complementary dosimeter for personal and area dosimetry applications.

### **Signal to dose linearity and dose reconstruction ability**

The signal to dose response was obtained by irradiating different samples of NaCl pellets with increasing absorbed doses,  $D_u$ , and reading the OSL signals,  $S_u$ , after one hour. The linearity in the signal to dose response was determined by fitting a

linear regression to the experimental data. The Pearson  $R^2$  value for these regressions provides an estimation of the signal to dose linearity correlation. Adding a calibration dose,  $D_c$ , to the pellet after reading the unknown signal,  $S_u$ , allows for estimation of the absorbed dose using the calibration signal,  $S_c$ , according to Equation 10. The derivative of the equation fitted to this response curve (given vs estimated  $D_u$ ) provides the NaCl's ability to correctly determine the given absorbed dose. A value of 1 indicate correctly determined doses.

## Specific luminescence

$c_{spec}$  was determined by dividing the weight normalised  $S_u$ 's by the given absorbed dose,  $D_u$ , for a number of NaCl pellets. The data from the signal to dose linearity was used to calculate a  $c_{spec}$  representative for the whole dose range between 1 and 300 mGy. A  $c_{spec}$  was calculated for each NaCl pellet and given absorbed dose, and a total  $c_{spec}$  was calculated as the arithmetic mean of the separate calculations.

## Detection limits

The detection limits were investigated (Paper I) and theoretically determined as the minimum detectable dose, MDD (mGy), and minimum measurable dose, MMD (mGy). MDD describes the dose at which a signal is large enough to be separated by the background and the MMD describes the dose level which can be quantified. The MDD is calculated as 3 times the standard deviation of the background signal of an un-irradiated sample and the MMD as 10 times the standard deviation of same background signal [52]. The signal is then converted to an absorbed dose using the specific luminescence.

## Sensitisation

The sensitisation [%] was investigated (Paper I) by repeatedly irradiating ( $D_u$ ) and reading the OSL signals,  $S_u$ , from the same sample 10 times. The sensitisation is determined by comparing the OSL signal from the first irradiation to the OSL signal of the last irradiation, for the same pellet. The irradiations and readouts were separated by about 1 hour and no effects of a decreasing signal yield with time was expected.

## Reproducibility

In terms of reproducibility, the main focus (Paper I) was put on the variation of the read signal or estimated absorbed dose among identically exposed samples. This was calculated as the coefficient of variation,  $C_v$  [%], according to Equation 11,

where  $\sigma S$  (or  $\sigma D$ ) is the standard deviation of the OSL signal,  $S$  (or estimated dose,  $D$ ).

$$C_v = 100 \cdot \frac{\sigma S}{S}, C_v = 100 \cdot \frac{\sigma D}{D} [\%] \quad \text{Equation 11}$$

## Fading

The fading (Papers I and III) was defined as the decrease in the read OSL signal,  $S_u$ , and estimated dose,  $D_u$ , over time after exposure. To investigate the fading, the OSL signal was read on multiple occasions, from sets of different pellets, after exposure to the same absorbed dose at  $t=0$ .

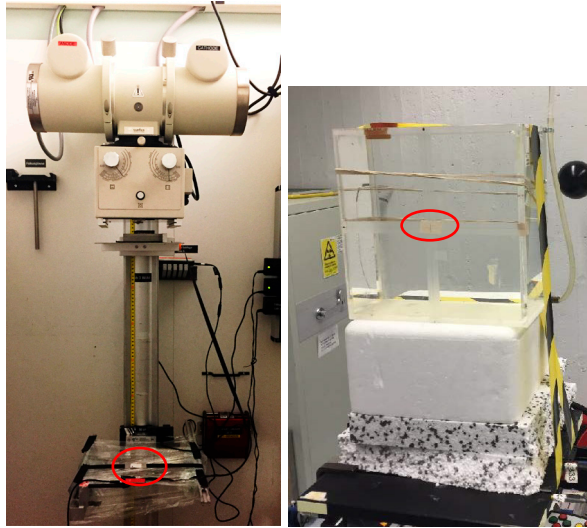
## Signal yield over time

The compressing of salt grains to pellets may affect the electron trap distribution in the NaCl crystals that may induce more short lived electron traps. Due to this, the OSL signal yield was investigated over time after pellet production (Paper III). NaCl pellets were irradiated, and the OSL signal read, on several occasions after production of the pellets, usually directly after production and up to several weeks or months.

## Energy dependence

The energy dependence of the NaCl pellets was investigated in two separate setups; free in air and with a backscattering medium (Paper II). A number of different X-ray spectra were used for low photon energy exposures with the addition of  $^{137}\text{Cs}$  (662 keV) and  $^{60}\text{Co}$  (1173 keV, 1332 keV) for higher photon energies. All irradiations took place at laboratories with well-defined calibrations, traceable to primary standard laboratories. The absorbed doses to salt were estimated using Equation 10 and normalised to the reference value measured or calculated at the time of exposure. For the setup with the NaCl pellet package suspended free in air (Figure 10, left) the estimated doses to the NaCl pellets were normalised to  $K_{air}$  (see section Dose quantities in the Background) at the point of exposure and for the geometry with the NaCl pellets on the surface of an ISO slab phantom [53] (Figure 10, right), the estimated doses were normalised to  $H_p(10)$ , at the point of exposure.

In addition to the experimental results, the mass energy absorption ratios and stopping power ratios were calculated for NaCl compared to air or tissue. The energy dependence was also simulated using MCNP [54] for an additional comparison. The energy deposition in a NaCl pellet volume was simulated and compared to reference  $K_{air}$  and  $H_p(10)$  calculated from the simulated fluence at the centre of the pellet volume location.



**Figure 10.** The two exposure geometries for investigating the energy dependence of the NaCl pellets. The red circles indicate the NaCl pellet packages. The left picture is taken at RTI group in Mölndal and the right picture is taken at the Swedish radiation safety authority in Stockholm.

### **Particle response (Preliminary)**

The OSL and TL responses in NaCl pellets were evaluated for alpha particles, neutrons, protons, photons and electrons. The pellets were exposed to alpha particles at DTU Risø using a Risø TL/OSL reader with an alpha irradiator. Eight different absorbed doses ranging between 130 and 1300 mGy (absorbed dose to water) were given to six NaCl pellets each. Additional NaCl pellets were irradiated with a sheet of paper covering the NaCl pellets to measure only the gamma contribution. The gamma contribution was determined to be lower than the standard deviation of the measured signals and therefore neglected in all dose estimations.

For proton exposure, the NaCl pellets were irradiated with 100 MeV protons from a linear accelerator at the Skandion Clinic in Uppsala. The NaCl pellets, packaged in light sealed aluminium tape, were positioned on a 10 cm thick slab phantom during irradiation. An ionisation chamber (calibration gives absorbed dose to water) was placed in the same position as the NaCl pellets to be used as a reference for each given dose. Eight different absorbed doses (ranging between 100 and 1000 mGy) were given to separate packages of NaCl pellets.

Photon irradiations were performed using a  $^{60}\text{Co}$  unit at the radiotherapy department at SUS Lund. The dose rate at the point of exposure, was 44.5 mGy/minute, determined by an ionisation chamber system as absorbed dose to water. 7 sets with 6 pellets each were given different absorbed doses between 45 and 890 mGy. The



NaCl was positioned on a slab phantom during irradiation, about 5 m from the  $^{60}\text{Co}$  source.

The electron exposures were performed using the internal  $^{90}\text{Sr}/^{90}\text{Y}$  source of the local Risø TL/OSL DA-20 reader. The dose rate was 4.66 mGy/s to NaCl.

For each type of radiation quality and for each absorbed dose, six NaCl pellets were irradiated. Three of them were read by OSL and three of them were read by TL. Because the alpha and proton irradiations took place in distant facilities from our lab, the time between the production of the pellets and irradiation, and the time between irradiation and readout, varied for the different exposures. All pellets were at least 7 days post production at the time of exposure which allowed for some stabilisation of the signal yield.

All given doses were converted to absorbed dose to water using stopping power ratios for quartz, NaCl, and water depending on the source calibration.

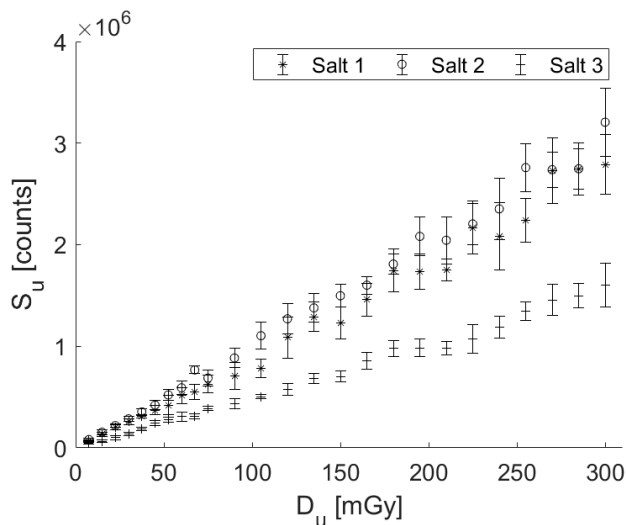
Neutron irradiations took place at the source testing facility at Lund university. NaCl pellets were placed in two different setups around a PuBe neutron source with both fast and slow neutrons as well as a wide range of gamma photons. For one of the setups, the gamma component was minimised which also resulted in a lower neutron flux. LiF chips (MCP-N), not sensitive to neutron exposure, were included in the NaCl packages. These were used to correct for the gamma contribution from the PuBe source and the surrounding materials.

# Results

## Dosimetric and luminescence material properties

### Signal to dose linearity and Dose reconstruction ability

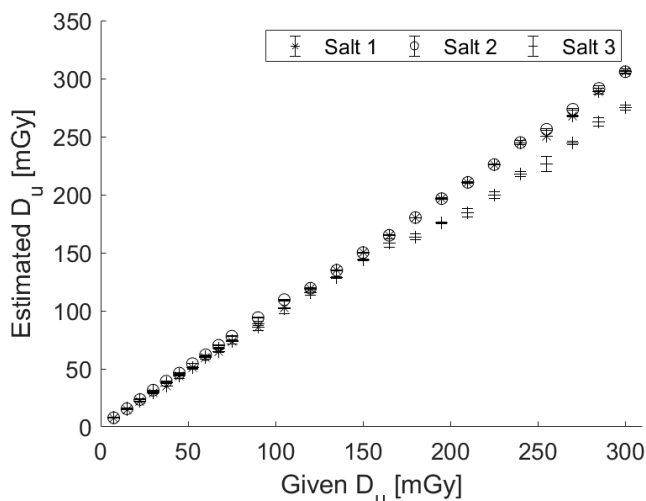
The NaCl pellets show a linear relationship between the OSL signal and the absorbed dose up to at least 300 mGy according to Figure 11. This was first shown in Paper I for the three different salts in Table 1. For doses larger than 300 mGy there is still a linear relationship (up to at least 6 Gy based on unpublished data) but emphasis was put on lower doses (<300 mGy) because of their relevance for external personal dosimetry.



**Figure 11.** The average OSL signal (counts) of the NaCl pellets as a function of the administered dose (mGy). Each readout was performed 1 hour after irradiation and at room temperature. The uncertainty bars correspond to 1 SD of ten NaCl pellets. (Adapted from Waldner & Bernhardsson 2018).

After individual calibration of each NaCl pellet and calculation of the absorbed dose using Equation 10, a linear relationship is shown also between the given and estimated absorbed doses according to Figure 12. In Figure 12, a calibration dose,

$D_c$ , twice the size of the experimental  $D_u$  was used for absorbed dose estimation as this was found to be the most optimal size for accurate dose estimations. In cases where  $D_u$  is unknown, a signal to dose calibration curve may be used to calculate the absorbed dose and thereby determine the optimal size of  $D_c$ .



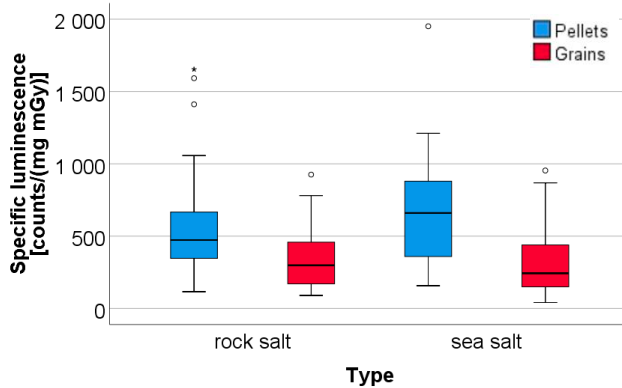
**Figure 12.** Dose estimations of  $D_u$ , with  $D_c$  twice the size of  $D_u$  for Salts 1-3 in Table 1. Each data point is the average of five pellets and the uncertainty bars represent 1 SD. (Adapted from Waldner & Bernhardsson 2018).

In Paper IV, the investigation of 102 different salts showed linear signal to dose responses up to at least 100 mGy for all salts when compressed to pellets. For the same 102 salts, but measured as grains, the doses were on average more poorly estimated.

### Specific luminescence

For Salt 1-3 (Table 1), investigated in Paper I,  $c_{spec}$  ( $\text{counts} \cdot \text{mg}^{-1} \cdot \text{mGy}^{-1}$ ) ranged from 259 to 576 with the lowest  $c_{spec}$  for the analytical salt. This is to be expected as it contains fewer impurities than the other salts.

Of the 102 salts investigated in Paper IV,  $c_{spec}$  ranged from 115 to 1950 with a median of 496. Figure 13 shows the  $c_{spec}$  depending on type of salt, for both NaCl grains and pellets. There is no statistically significant difference (independent t-test) in  $c_{spec}$  depending on type of salt, content of iodine or anti-caking agent. However, there is a statistically significant difference (paired t-test) when compressing the grains to pellets, with higher  $c_{spec}$  for the pellets.

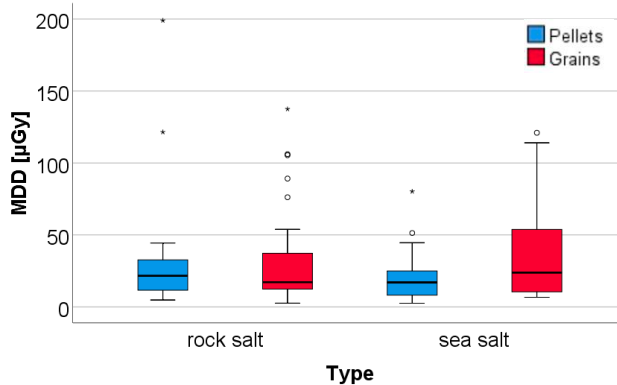


**Figure 13.** Boxplots of the specific luminescence for 102 different salts depending on type (rock or sea salt) for both NaCl grains and pellets.

## Detection limits

Due to the very low background signals from un-irradiated NaCl pellets, the detection limits are correspondingly low. In Paper I, detection limits in terms of MDD were found to be around 5-21  $\mu\text{Gy}$  for Salt 1-3 in Table 1.

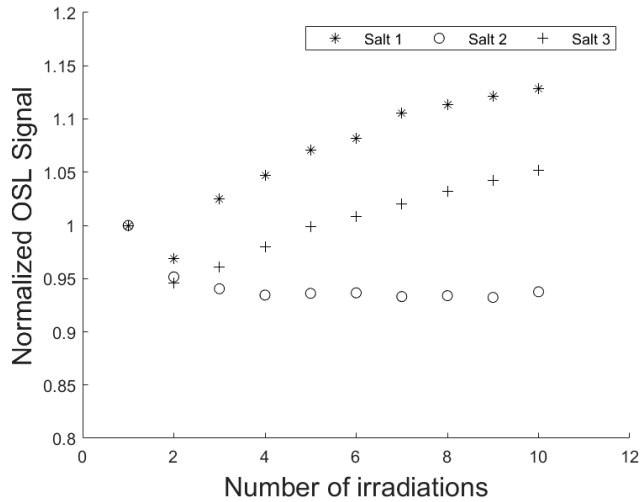
For the 102 different salts in Paper IV, the detection limits for the NaCl pellets vary between 2  $\mu\text{Gy}$  and 1036  $\mu\text{Gy}$  with a median MDD of 19  $\mu\text{Gy}$ . Boxplots comparing the detection limits depending on the type of salt, for grains and pellets, are presented in Figure 14. There are no statistically significant differences depending on type of salt or additives. Furthermore, the detection limits are not improved when compressing the grains to pellets. Signal readout before any exposure to ionising radiation gives low stable backgrounds for the majority of the investigated salts and the background signals are not further reduced if bleached. There is no sign of a OSL signal as cumulated from background radiation for any of the salts which can be explained by the fact that the salt is not shielded from light before they are exposed to ionising radiation i.e. when handled, and stored in the laboratory.



**Figure 14.** Boxplots of the detection limits, MDD, for 102 different salts depending on type (rock or sea salt) for both grains and pellets. The y-axis has been cut at 200  $\mu\text{Gy}$  not to distort the figure, five outliers are thereby excluded from the plots.

## Sensitisation

Irradiation of a material may lead to the induction of more defects in the lattice and thereby more trapping centres. As a consequence, the radiation sensitivity may change. In Figure 15, the sensitivity change is shown as the normalised OSL signal as a function of 10 consecutive irradiations (30 mGy each) and readouts. After 10 repetitions, Salt 1 shows an increase in sensitivity of about 13% compared with the first readout, Salt 2 shows a corresponding decrease of 6% and Salt 3 an increase of 5%. As part of the investigations in Paper IV the sensitisation was investigated again for the three salts. The repeated exposures were slightly smaller which resulted in less sensitisation. Just as in Figure 15, many of the salts investigated in Paper IV showed an initial decrease in signal after the second irradiation with a subsequent increase for the following irradiations.



**Figure 15.** The sensitivity change as a function of number of irradiations (30 mGy each) and readouts. Each data point corresponds to the average OSL signal of ten NaCl pellets normalised to the average OSL signal read after the first irradiation. (Adapted from Waldner & Bernhardsson 2018.) Salt 1-3 are presented in Table 1.

Even though it is clear from the results that there is a sensitisation of the NaCl pellets when they are exposed to ionising radiation, this has not led to problems when using the NaCl pellets for dose estimations in the experimental studies in Papers I-IV, in part because of the one-time use of the NaCl pellets.

## Reproducibility

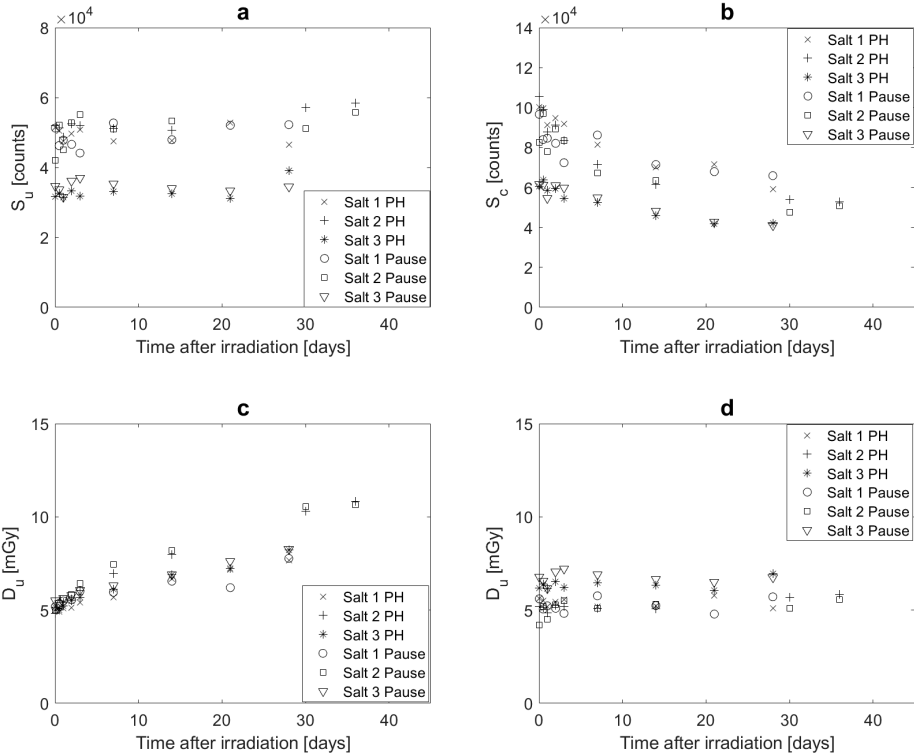
The alternative to a calibration dose to estimate the unknown absorbed dose to the NaCl pellets is to use a signal to dose calibration curve such as the ones in Figure 11, or similar ones where the signal is also normalised to the weight of the NaCl pellet. Table 3 shows the reproducibility, the variation in signal or dose estimation among a set of samples, expressed as the coefficient of variation, depending on how the signal is normalised. In terms of this measure, a signal weight normalisation shows a small improvement in terms of variation but normalisation with a calibration dose gives the smallest variation among the samples. Instead of the weight, the calibration dose ( $D_c$ ) takes into account the amount of OSL signal giving material of each pellet and this is not necessarily proportional to the weight.

**Table 3.** The signal and dose reproducibility expressed as a range of  $C_v$ , depending on  $D_u$ 's between 1 and 300 mGy, for Salts 1-3 (Table 1). Two different signal normalisation methods are used, as compared to only observing the OSL signal: to the weight of the pellet (mg) and to the given calibration dose (mGy).

Salt	None (counts)	Weight (counts mg <sup>-1</sup> )	Calibration dose (mGy <sup>1</sup> )
Salt 1	5.0-22.3%	3.8-18.8%	0.9-3.3%
Salt 2	5.0-15.9%	4.3-10.9%	0.3-1.8%
Salt 3	4.3-20.8%	4.0-19.9%	0.3-3.4%

## Fading

When investigating the signal stability over time in Paper I, an increase in the estimated dose over time was observed. Therefore, the signal stability over time was further investigated in Paper III. Looking only at  $S_u$  over time (Figure 16a) it remains stable over at least 30 days and there is no indication of inverse fading. However,  $S_c$ , resulting from the  $D_c$  at each time of readout, decreases with time (Figure 16b). Calculating  $D_u$  at each time point according to Equation 10 results in the graph in Figure 16c where an apparent inverse fading is observed. Even though there are cases of inverse fading reported [55,56], this is not what is shown in Figure 16c. The reason for the apparent inverse fading is the decreasing  $S_c$ 's, that is, the decrease in signal yield over time for the same  $D_c$ . When using the signal to dose calibration graph in Figure 11 to calculate the absorbed dose at each time point after irradiation, the estimated doses remain stable over time. To achieve a stable dose estimation at different time points after irradiation, the actual signal yield of the NaCl pellets over time needs to be considered. If the decrease in signal yield over time can be corrected for, the dose estimations using a calibration dose will be more reliable, and more reproducible, than using a signal to dose calibration curve.

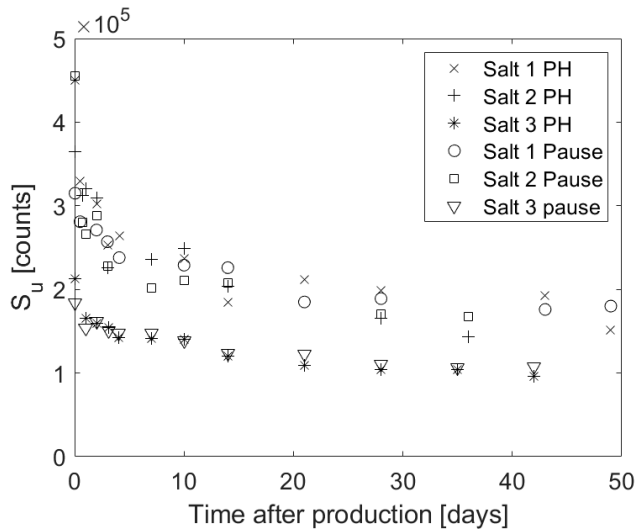


**Figure 16.** a) Unknown signal,  $S_u$ ; (b) calibration signal,  $S_c$ ; (c) absorbed doses calculated with calibration dose normalisation; and (d) absorbed doses calculated with the dose response curve from (Waldner and Bernhardtsson 2018), as a function of time after exposure (using either a pause “Pause”, or a preheat protocol “PH” for Salts 1–3 in Table 1). Measurements were performed between 1 hour and up to 36 days after irradiation, after a given dose ( $D_u$ ) of 5 mGy. The standard deviation for each data point is in the order of 10% for a,b,d and 2% for c. The pellets were produced 24 hours before exposure to  $D_u$ .  $D_u$  was administered simultaneously for all pellets.

## Signal yield over time

After the discovery of the decrease in calibration signal with time in the fading study, the signal yield over time was investigated for un-irradiated pellets (as compared to the fading study where the decreasing  $S_c$ 's are seen for previously irradiated pellets). Exposure to a  $D_u$  from the  $^{90}\text{Sr}/^{90}\text{Y}$  source with subsequent readout after 1 hour, at different time points after the production of the pellets, resulted in the graph in Figure 17. There is an initial fast decrease in signal yield during the first week, followed by some stabilisation in  $S_u$  with a slower decrease in signal yield. It has been suggested [43] that compressing the crystal lattice may shift the impurity states in the band gap, possibly changing the emission wavelength of the luminescence photons, and the decrease in signal yield may be a consequence of this.

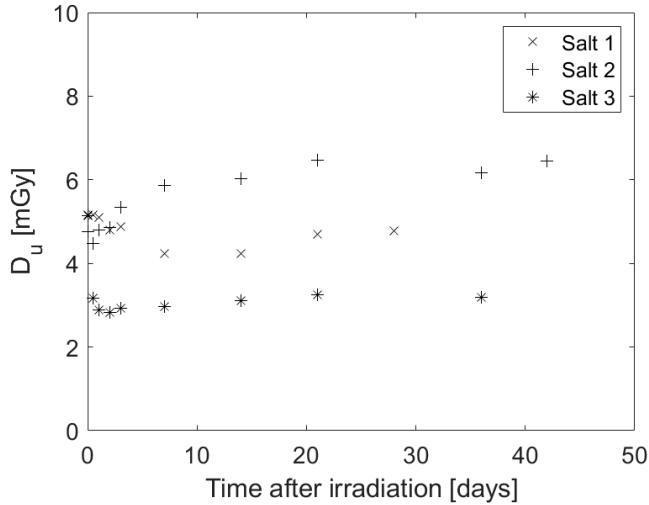




**Figure 17.** The measured OSL signal,  $S_u$ , after irradiation with an absorbed dose of 28.8 mGy at different times after production of the NaCl pellets (Table 1). The signals were read using the readout protocol in Table 2, with either a pause before readout or a 100°C preheat before readout. The uncertainty ( $\pm 1$  standard deviation) varies between 10% and 20% for each data point. Each data point is the arithmetic mean of  $S_u$  read from 5 pellets.

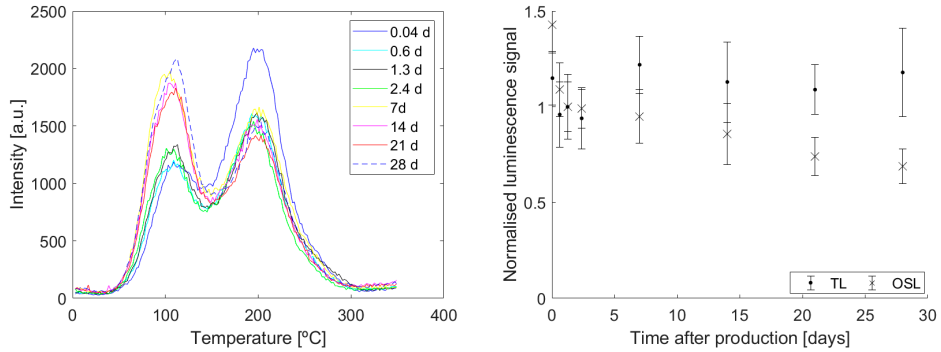
For exposures and readouts close together in time, the effects of the decrease in signal yield are not a problem. However, for the decrease in signal yield not to be a problem when the readout is performed a few days or weeks after the exposure, the decrease in signal yield either need to be corrected for or the pellets be allowed to stabilise before being used.

Figure 18 shows the same data as Figure 16c but with a correction of  $S_c$ , to account for the decrease in signal yield with time, at each readout after exposure. The correction comes from a fitted equation to the graphs in Figure 17. A slight apparent inverse fading is still observed but with a smaller increase over time than in Figure 16c. Further optimisation of the correction to  $S_c$  is needed for accurate dose estimations when exposure and readout are separated in time.



**Figure 18.** Estimated absorbed dose  $D_u$  from Figure 16c, with corrections for the decrease in efficiency of the OSL signal yield. OSL signals were read using the 1 hour pause protocol. The standard deviation for each data point is in the order of 2%.

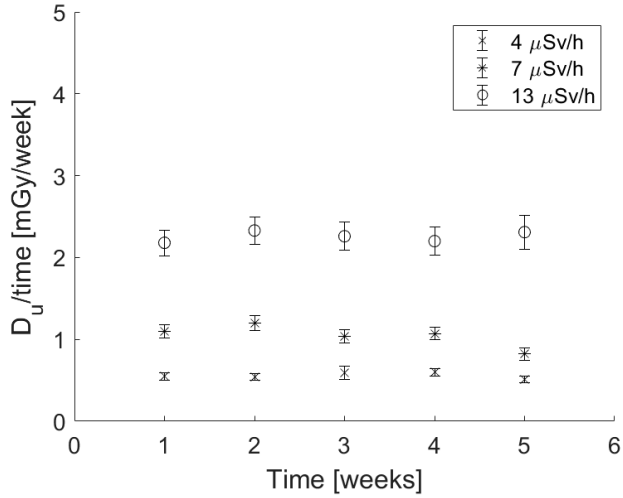
To further investigate the decrease in OSL signal yield with time, the TL curves were investigated (Figure 19, left) at different times after the pellets were produced. At a given time, the pellets were given an absorbed dose of 180 mGy and the TL curve was measured after a 1-hour pause. Some redistribution of the TL signal with time can be observed in the left graph in Figure 19. However, comparing the integrated TL and OSL signals (normalised to the 1-day measurement) at each time of exposure and readout, there is no clear decrease in the TL signal with time. It is possible that the OSL and TL signals originate from different localised energy levels in the crystal lattice, in which case the TL curves will not help explain why the OSL signal yield decreases with time. A first step however, is to investigate the TL curves in a wider range of emission wavelengths to determine if the energy levels change with time after compression.



**Figure 19.** Left: TL curves for Salt 1 in Table 1. Each curve represents readout 1 hour after an absorbed dose of 180 mGy, at different times after production of the pellets. Each curve is the mean signal from 3 NaCl pellets. Right: The integrated TL and OSL signal yield over time after exposure to an absorbed dose of 180 mGy, normalised to the 1-day data point. Each data point is the mean signal from 3 NaCl pellets and the uncertainty bars represent 1 SD.

## Chronic exposures

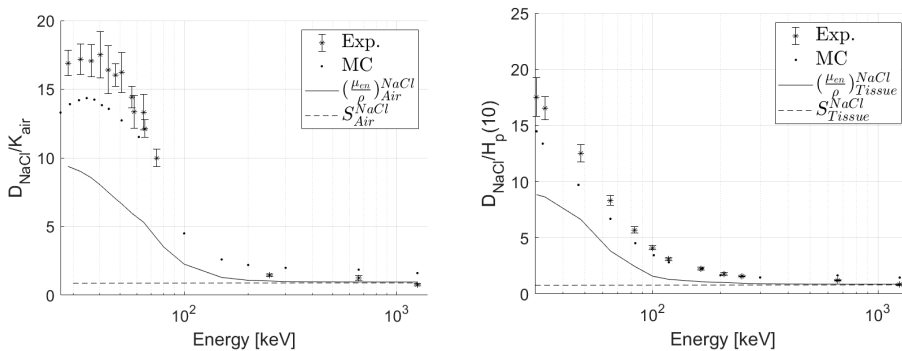
For many applications, the exposure of the NaCl pellets is extended over time and not acute as for the investigations in the TL/OSL reader in Papers I, III and IV. By exposing the NaCl pellets to low dose rates of an external point source of the gamma emitter  $^{137}\text{Cs}$  (20 MBq) for up to five weeks, and estimating the absorbed dose to 5 NaCl pellets each week, the effects of chronic exposure as well as the effect of the decreasing signal yield with time were investigated. The calibration dose at the time of readout was given promptly by the internal  $^{90}\text{Sr}/^{90}\text{Y}$  source of the OSL reader. Here, the decreasing signal yield with time is not observed and the absorbed doses, normalised to the exposure times, are constant as observed in Figure 20. Our conclusion is that the pellets age and the impurity states stabilise during the radiation exposure and because of this, the difference in signal yield between the first exposure and the calibration dose is not as prominent as in the cases with acute or prompt exposures.



**Figure 20.** Weekly absorbed dose,  $D_U/time$ , in NaCl pellets at three different dose rates as calculated by Eq. 1 from the signals in Fig. 8. The reference dose rates for the different data series are given as ambient dose equivalent as measured by a handheld detector.

## Energy dependence

Investigating the energy dependence of the NaCl pellets gives information about the signal response compared to that in air or tissue. As the dose to air or tissue is normally more relevant for personal dosimetry than the dose to NaCl (most often used for retrospective dosimetry), a simple relationship between the quantities is desirable for accurate prospective dosimetry.



**Figure 21.** Left: The NaCl pellet (Salt 1 in Table 1) energy dependence, defined as the ratio of  $D_{NaCl}$  and  $K_{air}$  at the measuring point free in air. Right: The ratio of  $D_{NaCl}$  and  $H_p(10)$  at the point of exposure of the NaCl pellets positioned on a backscatter phantom. The graphs also shows the Monte Carlo simulated energy dependencies as well as the theoretical mass energy absorption coefficients,  $\mu_{enp}$ , and stopping power ratios,  $S_{air}^{NaCl}$ , for NaCl and air or tissue. The uncertainty bars represent one standard deviation eight NaCl pellets at each energy.

The experimentally determined absorbed dose responses in NaCl as compared to the response in air or tissue, for a range of photon energies, are shown in Figure 21. The figure also shows the Monte Carlo simulated responses as well as the theoretical mass absorption coefficient ratios,  $\mu_{en}/\rho$ , taken from Hubball and Seltzer [57], of NaCl and air or tissue, along with the corresponding stopping power ratios.

As can be observed in Figure 21, there is a (seemingly systematically) discrepancy between the experimental and Monte Carlo simulated energy dependencies at low energies, and the theoretical  $\mu_{en}/\rho$  ratios over the entire range of energies. The studied range of the energy dependence can be divided into two components. The first part is mainly related to the difference in  $\mu_{en}/\rho$  coefficients, the absorbed dose energy dependence. Due to the large difference in effective atomic number,  $Z_{eff}$ , an over response in NaCl compared to air and tissue is expected for low photon energies where photo absorption is the dominant interaction. The second part of the energy dependency is the intrinsic energy dependence that arises from the luminescence process in the NaCl. If the luminescence process is energy dependant, this will not be taken into account by the simulations or other theoretical calculations.

For an occupational dosimeter made from NaCl, the badge/holder needs a build-up layer in front of the pellets that slightly flatten the energy response. For low energy photons the build-up layer will decrease the signal due to attenuation and for higher photon energies, the build-up layer will increase the signal. Combining different filters is necessary to flatten the energy dependence over the whole range of energies. Other filter combinations can be used to distinguish doses from different types of radiation as well as photon energies and to measure different dose quantities.

## Particle response (Preliminary)

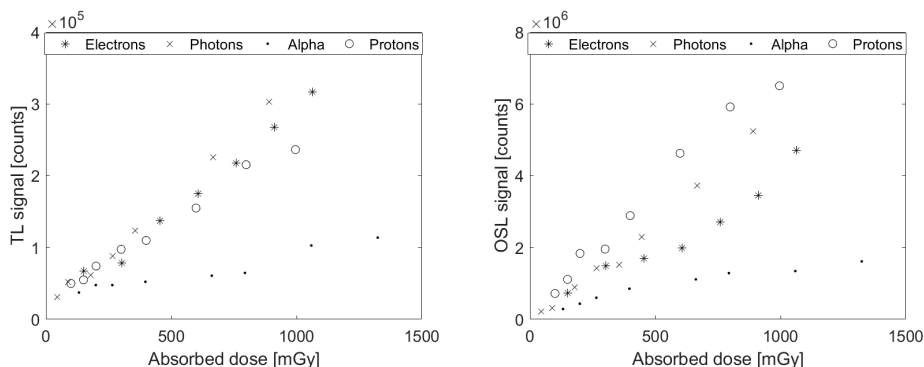
Figure 22 shows the TL curves read from NaCl pellets after exposure to photon, proton, electron and alpha irradiation. The curves have been normalised to the given absorbed dose to water for each type of exposure, to be able to compare the curves in a representative way. All exposures show resulting TL peaks at around 200°C but for the alpha exposures, there is no peak at around 100°C. The proton curve shows only a small peak at around 100°C and it is hard to distinguish from the 200°C peak. The peak at around 450°C seems to have some dependence on the LET of the particles. Many TL investigations on salt only heat the NaCl to around 350 °C but a peak around 450°C is consistent with findings by e.g. Bailey et al. [8]. This peak is often less pronounced for electrons and photons than in Figure 22 and sometimes it is not visible at all. In Timar-Gabor et al. [48] the TL curves stop at 350°C but for

one of the salts there is some indication that there is an increase in signal after 300°C indicating it could depend on the type of salt.



**Figure 22.** Dose normalised TL curves after proton, electron, photon and alpha exposure of NaCl pellets. Each curve is the mean signal from 3 NaCl pellets.

In the left graph of Figure 23, the TL signal to dose response curves show the lowest signal yield for the particles with the highest LET, consistent with other studies on e.g. LiF [58]. In the right graph of Figure 23 however, the proton irradiation shows the highest OSL signal yields, in the same order of magnitude as the photons.



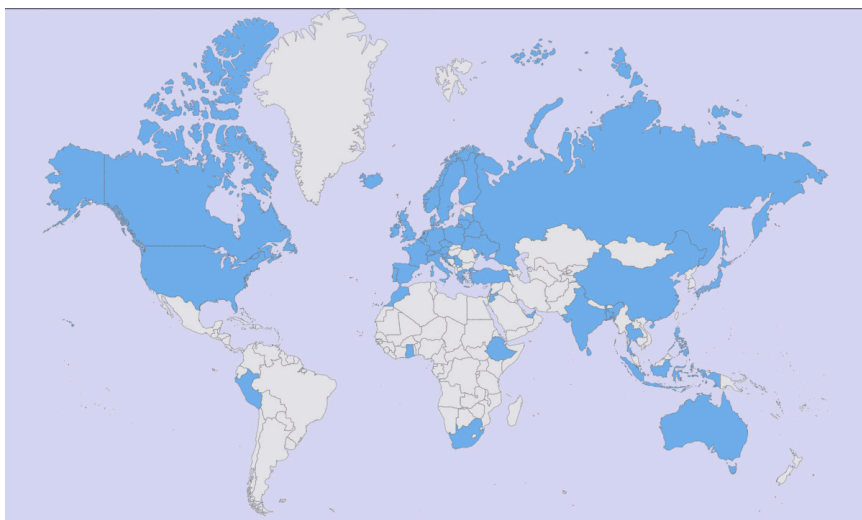
**Figure 23.** Left: The integrated TL signal as a function of absorbed dose after proton, electron, photon and alpha exposure of NaCl pellets. Each data point is the mean signal from 3 NaCl pellets. Right: The OSL signal as a function of absorbed dose after proton, electron, photon and alpha exposure of NaCl pellets. Each data point is the mean signal from 3 NaCl pellets.

Exposure to protons or alpha particles is not very common outside of proton therapy facilities or specific research laboratories, and they usually do not pose the same kind of radiation protection problem as for electron and photon exposures. It is however interesting to investigate how the LET of different types of ionising radiation affects the signal induction in NaCl, e.g. for applications in space dosimetry.

For the neutrons exposures, the NaCl packages with LiF chips (MCP-N) were positioned outside a water tank with a PuBe neutron source for two weeks. The  $H_p(10)$  from neutrons was close to 800  $\mu\text{Sv}/\text{day}$  (as measured by an electronic personal dosimeter, EPD-N2, Thermo Fisher) resulting in a total  $H_p(10)$  of around 11 mSv. The absorbed dose to the LiF chips and the NaCl pellets were determined and the photon contribution to the absorbed dose, estimated by the LiF chips, was subtracted from the absorbed dose estimated by the NaCl pellets. The mean energy of the photon component from the exposure is unknown but a wide range of different energy gamma rays are produced in the water surrounding the BePu neutron source. Taking into account a correction for an energy over response of at least a factor of 2 for the NaCl compared to LiF (or water/tissue) no signal is induced by neutrons in the NaCl pellets.

## Salt from around the world

The map in Figure 24 shows the 47 countries, marked in blue, from which 102 household salts have been collected during the last 5 years. Salt has been collected during conferences and holidays with the help from colleagues and friends. Both rock salts and sea salts were collected, with a variation of additives such as iodine and anti-caking agents. Focus was put on the country of purchase, whereas the specific mine or sea origin was not considered.



**Figure 24.** World map indicating every country (in blue) from which at least one household salt has been collected. Map courtesy of amCharts.com.

The luminescence and dosimetric properties of pellets made from the 102 salts were investigated in Paper IV. The mean and median values for all dosimetric properties investigated are presented in Table 4. Also presented in Table 4 are the results of paired samples t-test that show how the dosimetric properties are altered when compressing grains of salt to pellets; it was found that there is a statistically significant improvement in all dosimetric properties except detection limits.

Independent t-tests show that for grains of salt, there is significant difference in  $c_{spec}$  depending on the iodine content and in the reproducibility depending on type of salt. Such differences are not seen for the pellets. However, for the pellets, the reproducibility is better for salts containing iodine. All investigated salts may be used for prospective dosimetry as long as the expected doses for the intended applications are above the detection limits. A more detailed investigation into the dosimetric properties of a specific salt is warranted before using it for dosimetry applications.



**Table 4.** Numeric mean and median values for all dosimetric properties investigated in Paper IV. The mean difference values refer to the difference in mean between the grain and pellet values for each dosimetric property, and the significance values indicate if the difference is statistically significant or not.

	Mean		Median		Mean difference	Significance (p-value)
	Grains	Pellets	Grains	Pellets		
<b>MDD</b> [ $\mu\text{Gy}$ ]	43	38	20	19	5.7	>0.05
<b>c<sub>spec</sub></b> [ $\text{counts}\cdot\text{mg}^{-1}\cdot\text{mGy}^{-1}$ ]	327	586	291	496	-260	<0.001
<b>Reproducibility</b> [%]	0.87	0.45	0.74	0.36	0.415	<0.001
<b>Sensitisation</b> [%]	72	4.7	62	4.1	67.6	<0.001
<b>Signal to dose linearity</b>	0.983	0.996	0.988	0.998	-0.013	<0.001
<b>Dose reconstruction</b>	0.787	1.003	0.783	1.001	-0.216	<0.001

# General Discussion

For NaCl pellets produced according to the optimal configuration established in Paper I, the basic dosimetric properties all indicate that NaCl pellets may be used for OSL dosimetry, especially when using the readout protocol developed in Papers I and III. Although NaCl may still be useful for OSL dosimetry in its current form there are some areas that can be further optimised, such as improving the compensation for the energy over response (Paper II) and the difference in signal response depending on type of radiation. There are several applications for using NaCl pellets for OSL dosimetry as long as the mean energy of the radiation field is known to some extent, both for short term and prolonged exposures. In paper IV it is further shown that almost any type of household salt from around the world may be used for OSL dosimetry. A personal dosimeter made from NaCl pellets has shown convincing potential as a complement to commercial dosimeters for many radiation protection applications, especially when large numbers of detectors are needed.

## NaCl pellets as a dosimeter material

Household salt can be found almost anywhere in the world, making NaCl a promising material for accessible and low cost dosimetry. Compressing the NaCl to pellets simplifies handling of the material which reduces the preparation time before readout, as well as the risk for cross contamination of samples. When in pellet form, the samples do not need to be weight normalised which also simplifies the readout process and as the pellets are close to identical, the OSL signals become more reproducible as compared to grains of salt. In terms of dosimetry, it has here been shown that several of the luminescence and dosimetric properties of NaCl are improved for the pellets compared to grains and the only downside, although possible to account for, seems to be the change in signal yield over time. The loss of signal yield with time is a problem created by compressing the NaCl but the advantages of handling the NaCl as a pellet may outweigh the inconvenience of having to account for such change in sensitivity.

The comprehensive study of luminescence and dosimetric properties show that almost any household salt compressed to pellets can be utilised for OSL dosimetry

with only a few initial measurements before its use. Knowledge of the detection limits and a basic signal to dose response curve is enough for rough dose estimations but the signal stability over time and signal yield over time should be investigated beforehand for more accurate dose estimations. There are variations in  $c_{spec}$  up to a factor 20 for different brands of salt but variations up to a factor of 5 have also been observed for different batches of the same salt. When using salt from different batches the salt may either be mixed together and a common calibration graph created, or each batch will require its own signal to dose response. This is mainly important if such a calibration graph, e.g. like the one in Figure 11, is used to estimate the absorbed doses to the salt. If the recommended calibration dose of each pellet is used for dose estimation instead, the difference in  $c_{spec}$  depending on batch will not have as much of an impact.

The energy dependence of the NaCl pellets has only been investigated for one type of salt but as this mainly depends on  $Z_{eff}$  of NaCl compared to tissue, the type of salt should not matter as the different types of additives (iodine, anti-caking agent) cannot vary enough to affect  $Z_{eff}$ . As an example, the amount of iodine in Salt 1 in Table 1 is 50 ppm. As of now, there are no custom filters or badges for the NaCl pellets but the pellets may still be used for a wide range of photon energies and dosimetry applications. As long as the user has an idea of the mean photon energy of an exposure, an equation fitted to the NaCl specific energy dependence graph can be used to correct for the over response.

## NaCl pellet dosimetry in practice

With the press tools described in the Method section, up to 1000 NaCl pellets may be produced per hour. After exposure to ionising radiation, the absorbed dose may be roughly estimated (uncertainties ~20-30%) for 48 pellets in around 1 hour, or more accurately estimated (uncertainties <10%) in around 3 hours. The degree of accuracy may be chosen depending on the application. For some applications where results are needed fast, larger uncertainties may be acceptable. Such applications may include triage in emergency preparedness situations or environmental monitoring with large numbers of detectors. For personal or area dosimetry in the medical clinic or nuclear industry, more time may be given for measurement and evaluation, allowing for smaller uncertainties.

For all applications with NaCl pellets, the protection from light exposure is of importance to avoid depletion of the OSL signal. When using dosimetry phantoms in the medical clinics, the phantoms may need to be handled in the dark during and after exposure to ionising radiation. Although, smaller phantoms can usually be covered in light tight bags or tape during and after exposure. and many larger phantoms, such as the CIRS [59] and Rando [60] anthropomorphic phantoms, have

been found to, in themselves, shield the NaCl pellets sufficiently from light during and after irradiation.

Light tight encapsulation of one or several pellets per measuring point is possible but the packaging of the pellets increases the size of each unit. Using such packaging is not optimal for use in e.g. finger dose measurements, and is impossible to use with some existing dosimetry phantoms.

For personal dosimetry where occupational doses are required to be reported to relevant authorities, the dosimetry systems need to be legal and certified for this purpose. In its current state, the NaCl pellets may be used together with such existing dosimeters, for initial dose mapping of environments of interest to gain useful input for simulations of radiation doses and changes with time, or as a supplement to other dosimeters in situations where a large number of detectors are useful for covering large areas or give better spatial resolution of the exposure in an environment. By themselves the NaCl pellets are a good first indicator of where to perform more detailed measurements, or even where to perform initial counter measures, based on identified hotspots and other points of interest in an unknown environment. The NaCl have been utilised in this way with satisfactory results, in a number of practical projects, both for environmental monitoring (grains) [2,61], and at various clinics at the university hospitals in Malmö and Lund (pellets) [62,63].

## Future prospects

### **Emergency preparedness applications**

For emergency preparedness applications, pellets may be produced and distributed without any preparatory measurements. The basic properties may then be investigated in the laboratory while the dosimeters are in use, after which they are fast evaluated in the laboratory. Distributing NaCl based dosimeters to inhabitants of contaminated areas after e.g. a nuclear accident could be an alternative to immediate evacuation where the contamination and potential exposure to ionising radiation is unknown or not determined. Simple dose estimations with NaCl dosimeters may be used to make decision on whether evacuation is needed or not, potentially saving people from having to leave their homes [64–66]. A personal dosimeter may also help to calm worried individuals after a radio nuclear incident, as compared to generally estimated charts or maps of the dose distributions. Ideally, the emergency preparedness dosimeter would also have a badge with filters to correct for the NaCl energy dependence. If blueprints for such NaCl dosimeter badges were made available to the public, personal dosimeters could even be produced by anyone owning a 3D printer as household salt can be found in most homes, at least as grains. However, if the mean photon energy in the environment

is roughly known, personal dose equivalents may be estimated by energy dependence graphs if using simple packaging of the NaCl pellets (that may be made from materials that can be found in any household). Depending on how well the mean photon energies can be evaluated in an affected area, personal dose equivalents may be evaluated with reasonable uncertainties using NaCl and a dosimeter holder made from e.g. paper.

## **Badge and holder**

The next step towards a fully functioning personal dosimeter is a badge holder with appropriate filtering. The filtering may vary depending on application with higher demands on energy dependence corrections for e.g. radiology applications which mainly utilises photon energies below 100 keV compared to environmental monitoring where the photon energy may range from about 60 to 3000 keV, or the nuclear industry where  $^{60}\text{Co}$  energies (1250 keV) dominate. Combinations of filters may be used to discriminate certain photon energies, to measure beta radiation or to compensate the energy dependence. If no suitable filter combination can be created to flatten the energy dependence sufficiently, filters that alter the energy dependence, in combination with unfiltered pellets, can be used in combination with mathematical corrections to adjust the estimated dose.

Early results indicate a difference in the thermoluminescence curve depending on the type of radiation used to expose the NaCl pellets. For higher LET radiation, a peak between 400-500°C seems to be more dominant than for low LET radiation. A dependence on LET is also seen for the OSL signal. This signal response difference may also need to be considered when optimising a filter and badge design, depending on application.

## **Dedicated readout unit**

For the dosimetry with NaCl pellets to be fully applicable, OSL readers for dose evaluations need to be available. Some readers are available in research laboratories [67–70], like the one used in this project [30,31], and as the OSL dosimetry technique is further developed, more readers will hopefully be developed. There are, however, limitations on how these readers may be used and it is not all readers that allow for changes to the readout parameters to fit the NaCl pellets.

For emergency preparedness situations, OSL readers used for luminescence dating (both stationary and portable) may be utilised for readout with comparable results. Also, the TL signal from NaCl may be read using TL readers already existing in many workplaces (e.g. hospitals, nuclear power plants), if the absorbed doses are above the TL detection limits.

In many places around the world there are economic factors that limit the access to dosimeters or dosimetric tools for making justified decisions, for optimisation procedures, or for monitoring dose limitations with ionising radiation. The suggested NaCl pellets may help improve or encourage to develop the radiation protection and safety culture in these countries, both for personnel, caretakers and patients.

A dedicated reader for the NaCl pellets would ideally be small enough to fit in a briefcase, making it easy to transport and bring out *in-situ*. Using the protocol with a one-hour pause for OSL signal stabilisation instead of preheat means there is no need to include a generator for sample heating. For relative measurements, or dose estimations using a calibration curve established using external radiation sources an internal calibration source is not necessary. For applications where a calibration dose is needed, an attachment to the reader would be an option. With the development of more efficient SiPM detectors[71,72], using these for luminescence detection would make the reader more compact than one with a PMT. Ideally the reader would be connected to a smartphone and app controlled.

# Conclusions

In this thesis, NaCl compressed to pellets has been investigated as a potential complement to commercially available dosimeters, in passive prospective OSL dosimetry. A dosimeter made from NaCl, read by OSL, is a low cost alternative for fast, simple, and potentially very precise dose estimations within a range of applications in e.g. medical clinics and emergency preparedness. The main conclusions of this thesis are the following:

- The handling of household salt is simplified when compressing it to solid pellets. The most mechanically stable pellets are produced from salt grains between 100  $\mu\text{m}$  and 400  $\mu\text{m}$  using a compression force of 0.5-0.8 tons, depending on the press tool.
- The basic dosimetric properties of NaCl pellets, such as signal to dose response, detection limits, signal stability over time and dose reconstruction ability are all suitable for prospective OSL dosimetry applications.
- The energy dependence of NaCl pellets compared to  $K_{air}$  and  $H_p(10)$  shows a strong over response for low photon energies, mainly due to the difference in  $Z_{eff}$  between NaCl and tissue equivalent materials. This over response need to be considered in further development of a badge with filters to decrease the photon energy dependence.
- When using NaCl pellets without any dedicated badge/holder, the OSL signal from a specific NaCl pellet can be used together with the energy dependence graphs may be used to correct for the over response at low photon energies, as long as the mean photon energy of an exposure is known.
- The OSL signal from the NaCl pellet may be read after either a pause of at least 1 hour or a preheat of 100°C. The choice of using either a pause or a preheat for emptying of short-lived electron traps may be determined by the number of samples as a preheat will results in faster readout for small sample sizes.
- For radiation qualities other than beta/gamma the LET dependence of the OSL signal induction need to be corrected for to achieve accurate results.

- The NaCl pellet may be used as a complement to commercial dosimeters when there is a need for large numbers of detectors or the availability of other dosimeters is limited.
- To be able to utilise the full potential of the NaCl pellet in OSL dosimetry, a dedicated reader needs to be developed for fast and simple readouts and dose determinations.



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# Optically Stimulated Luminescence Dosimetry with NaCl Pellets

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This thesis describes the use of NaCl pellets for passive prospective dosimetry. A dosimeter based on NaCl pellets enables simple and cost effective dose measurements with good accuracy, a linear signal response, and low detection limits. The NaCl pellet dosimeter may be used as a complement to other types of dosimeters when large numbers of detectors are needed or when there is a shortage of alternative detectors.