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Reflections on the future developments of research in retrospective physical dosimetry

Paola Fattibene [a,*], Francois Trompier [b], Celine Bassinet [b], Bartłomiej Ciesielski [c], Michael Discher [d], Jonathan Eakins [e], Chryzel Angelica B. Gonzales [f], Christelle Huet [b], Alexander Romanyukha [g], Clemens Woda [h], Małgorzata Juniewicz [c], Hyoungtaek Kim [i], Jungil Lee [i], Agnieszka Marciniak [c], Sergey Sholom [j], Hiroshi Yasuda [f]

[a] Istituto Superiore di Sanità, Core Facilities, Viale Regina Elena 299, 00161, Rome, Italy
[b] Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-SANTE/SDOS/LDRI, F-92260, Fontenay-aux-Roses, France
[c] Department of Physics and Biophysics, Medical University of Gdansk, Dębinki 1, 80-211, Gdańsk, Poland
[d] Paris-Lodron-University of Salzburg, Department of Environment and Biodiversity, 5020, Salzburg, Austria
[e] United Kingdom Health Security Agency (UKHSA) RCE, Chilton, Didcot, Oxfordshire, OX11 0RQ, UK
[f] Research Institute for Radiation Biology and Medicine (RIRBM), Hiroshima University, Kasumi 1-2-3, Minami-ku, Hiroshima, 734-8551, Japan
[g] Naval Dosimetry Center, 8950 Brown Drive, Bethesda, MD, 20889, USA
[h] Institute of Radiation Medicine, Helmholtz Zentrum München, Neuherberg, Germany
[i] Radiation Safety Management Division, Korea Atomic Energy Research Institute, 989 Daeduk-ro, Daejeon, 34057, Republic of Korea
[j] Department of Physics, Oklahoma State University, Stillwater, OK, 74078, USA

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ABSTRACT

Electron paramagnetic resonance, thermoluminescence, and optically stimulated luminescence, with biological tissues and inert materials are well established physical methods for retrospective dosimetry in acute accidental exposures. The objective of this article is to provide a view of the questions still open, the current challenges and the needed solutions. As research on emergency response methods is encountering increasing difficulties in terms of financial and human resources in many countries, it is essential to identify the research priorities and pay attention to cost-effective research paths. The intention of the paper is to stimulate discussion in the scientific community and to encourage collaboration among laboratories toward goals that address the real needs in retrospective dosimetry for acute exposures.

1. Introduction

1.1. What is physical retrospective dosimetry?

The term "retrospective dosimetry" (RD) refers to a class of methods for the determination of dose after the occurrence of a radiation event, in situations where assessment of doses to exposed individuals is necessary or required, but conventional dosimeters were not available or were insufficient [1,2]. These methods use materials found in everyday objects or biological tissues, taking advantage of the possibility provided by specific physical and biological techniques to measure radiation-induced changes in these materials. The adjective "physical" is added to distinguish measurement techniques based on physical principles, in particular the electron paramagnetic resonance (EPR) spectroscopy and the luminescence techniques (thermoluminescence, TL, and optically stimulated luminescence, OSL), from those which make use of biological approaches, such as the cytogenetics techniques [3]. Neutron activation based on the measurement of beta- or gamma-rays from activated radionuclides is also a RD physical method [1], but it can be used only after neutron irradiation and will not be dealt with in this paper. Another subclassification of physical RD is the one between "biophysical" and "fortuitous", with the former referring to the use of human biological tissues (tooth enamel, bone, fingernails), and the latter to inert materials contained in personal items or artefacts (so called fortuitous dosimeters). RD finds application in assessing individual doses as part of risk analysis studies and to estimate stochastic

Abbreviations: EPR, Electron paramagnetic resonance; TL, Termoluminescence; OSL, Optically stimulated luminescence.

* Corresponding author.
E-mail address: paola.fattibene@iss.it (P. Fattibene).

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effects, months or years after prolonged or acute exposures [2], or to support diagnosis or treatment choice in medical management or reassurance assistance in the short term after acute exposures [1].

1.2. Focus of this paper

The rationale of this paper is to provide suggestions for priorities, avenues and unexplored paths of research in physical RD, seeking a balance between needs, challenges and resources. Indeed, in recent years, the number of laboratories working in the field has declined and many have been converted, resulting in a reduction in human resources. More generally, like all research aimed at emergencies, RD suffers from a discontinuity in financial and human investment by research institutions in non-emergency times. Many laboratories thus find themselves having to choose which line of research to continue and which to cut. The idea that guided this work was to seek, within a group of researchers, as much consensus as possible on research goals and a roadmap that can help research groups target some key research questions. The overarching questions can thus be summarized as: which key research questions should at the minimum be answered in the next years to bring RD to a level of practical operability and through which approaches?

The paper focuses on methods that provide a dose, which can be directly related to the dose to the individual, a short time after a potential exposure. This paper is not intended to provide recommendations on which method to use in specific scenarios (see for instance Refs. [4,5], nor to deeply review the methods, for which the reader is invited to read more focused publications [6–11] and especially the extensive review by ICRU [1]. There are some recent interesting applications of RD that deserve to be mentioned and followed, but will not be treated here because they are out of scope of the paper. Among these may be included applications in radiation protection of the environment through dose estimation in animals, such as EPR in wild animal tooth enamel [12,13], or nuclear forensics and non-proliferation applications, such as retrospective characterization of nuclear material in space and time [14].

Section 2 will discuss the historical evolution of physical RD methods and techniques. Section 3 will provide a brief description of each assay type, focusing on strengths, weaknesses and challenges with solutions currently proposed and under development. Section 4 presents the Authors' personal views about research lines suggested to address these challenges.

2. Evolution of retrospective dosimetry

2.1. A brief history of research in RD

The history of research in RD has been a sequence of accelerating and decelerating phases, from which lessons can be learned. The use of a ubiquitous or readily available personal item or small sample of biological tissue as a dosimeter for the estimation of individual dose has been a goal for decades. In the 1990s, the US-DOE Low Dose Radiation Research program [15] and the European Commission Framework Programs [16] developed epidemiological programs in cohorts who had received prolonged exposures in the past, with the ultimate goal of evaluating the long-term stochastic risk estimation. The radiation markers for dosimetry were required to be stable over the years and apparent at doses relevant for the stochastic effects, i.e., of the order of hundreds of mGy. Tooth enamel with EPR and construction bricks with TL/OSL, having these characteristics, were used to assess, respectively, the individual and environmental dose in survivors of atomic bomb attacks in Japan [17], in populations of villages and workers of the Mayak nuclear plant in the former Soviet Union [18,19], Chernobyl liquidators [20,21], and in nuclear testing sites at Semipalatinsk [22,23].

In parallel and since the early days, EPR and to a lesser extent TL have been applied in so-called small-scale events (International Atomic Energy Agency (IAEA) [24–29], with small number of individuals affected and short range release, which are actually those classified as IAEA INES levels 1–4 [30]. Experience has shown that these events are commonly characterized by a lack of detail about the radiation field and circumstances of occurrence, they are often associated with doses above the deterministic effects threshold and involve heterogeneous whole-body or localized irradiation [1]. RD was used for dose mapping on the body to identify the organs at greatest risk and enable timely medical management of injuries [31]. The main criterion of choice is the actual availability of the specimen on the subject's body. In fact, tooth enamel and bone were used in four and twelve events, respectively, despite their invasiveness, while the most sensitive sugar in only three cases because it is rarely carried by a subject [1].

After the attacks of September 11, 2001, awareness of a radiological threat from terrorists or from malicious acts drew attention towards events that may involve large numbers of people in the civilian population, possibly up to thousands of individuals, along with the corresponding uncertainty regarding the homogeneity or heterogeneity of the exposure and dose levels. This drew attention to the need for assays suitable for such events [5]. RD can offer support in three phases [32]: initial dose estimation for dosimetric triage (phase I), to identify individuals with doses above the threshold for acute radiation syndrome (ARS), i.e., about 1 Gy [33]; dose assessment (phase II) for the purpose of medical management of individuals identified as irradiated during phase I; and a retrospective dose assessment (Phase III) for the psychological reassurance of individuals, especially potentially vulnerable groups (children, pregnant women, or affected people), who had been identified in previous phases as unexposed or receiving low exposures. This was followed by a change in research funding, as also mentioned in Ref. [34]; which was reflected in topics of publications: the number of papers on EPR dosimetry with dental enamel began to decline whilst that on other methods, more suitable for acute exposures, increased (Fig. 1).

2.2. The route towards the development of a multi-assay approach

The landscape of techniques and materials that can be used for the purposes outlined above is broad, and each has different characteristics in terms of sensitivity, specificity to radiation, stability of the radiation-induced signal (RIS) over time, response to environmental conditions, and invasiveness. Consequently, each combination of a technique and material, which we will call an assay, may be suitable for a certain scenario, but not for another. The most appropriate way to look at RD then is as a toolbox [32] from which to take the right tool (assay) when

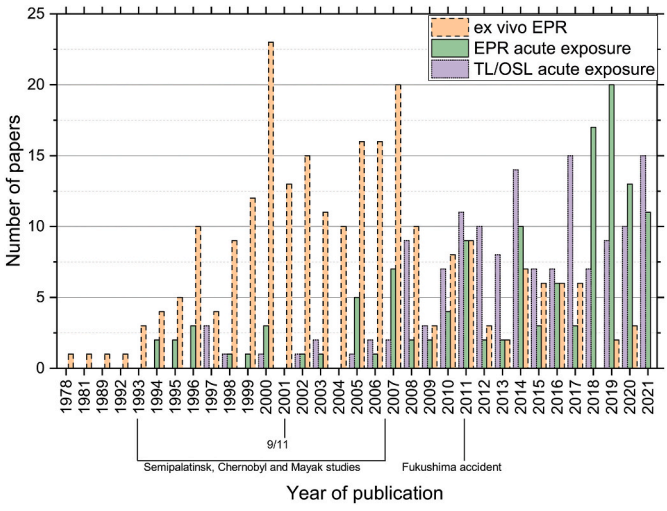

Fig. 1. Trend of peer-reviewed publications between 1992 and 2022 on tooth enamel by *ex vivo* EPR (orange - dashed line), fortuitous materials by TL/OSL after acute exposure (purple - solid line), and EPR after acute exposure (green - solid line).

needed, depending sometimes simply on the availability of samples. A single method is usually not sufficient to provide the necessary information, and a multi-assay approach combining the available methods has shown its strength in many complex cases, as will be described below.

Reviews in the early years [35–37] had already identified as sufficiently promising the same assays used predominantly today. First [38] and then [39] identified gaps in harmonization and standardization of protocols, appropriate calibrations at secondary calibration laboratories (SSDLs), and quality control [39] were the firsts to mention the potential use of RD in terrorism-related emergencies, albeit on a small-scale, recommending the establishment of a network of EPR and TL/OSL laboratories. These gaps remained unfilled until, with the threat of terrorism after 2001 and the urgency of having an RD tool in a mass event with a high rapid response capability, it became clear that no "golden method" would be available in the short term and a multiple-assay strategy seemed to be the only viable strategy. The first agreement on this multi-assay approach is found in the consensus paper by Ref. [40]; followed by reports from several projects [8,9,32]. The need to harmonize the results of different assays and laboratories has prompted the creation of networks. Networks are needed to assist in radiological scenarios involving the measurement of a number of samples that exceeds the capacity of a single laboratory or a single country, whether this is of units or of hundreds of samples. Networks are also essential to organize inter-laboratory comparison (ILC) programs and field testing to maintain operational capacities and capabilities in non-emergency times, which is a challenging task for a single laboratory, as the COVID-19 experience has shown [41]. Several networks exist nowadays. RENEB ("Running the European Network in Biodosimetry", www.reneb.net) is an operational network of biological and physical laboratories [42]. Networks dedicated to research are the European Radiation Dosimetry Group, EURADOS (eurados.org), the Asian Radiation Dosimetry Group, ARADOS (https://www.nirs.qst.go.jp/usr/ARADOS) and the Korea Retrospective Dosimetry Network, KREDOS [43]. EURADOS published a Strategic Research Agenda [44] and a 5-year Roadmap [45], which include strategic actions for RD aimed at improvement, consolidation, and standardization of existing radiation markers; and identification and development of new markers. An ICRU-EURADOS joint report was published on methods for initial phase assessment of individual doses following acute exposure to ionizing radiation [1]. With the goal of encouraging a multidisciplinary approach, the report reviewed biological and physical methods, as well as *in vivo* bioassays, neutron activation analysis, and radiation field mapping, examples of many case studies and applications, and ILC comparisons. The report assessed the maturity of the methods, defining whether each can be considered to be established, in development, or experimental.

It is interesting to note that RD has also been included in the 2018 Plan of the IAEA Response Assistance Network (RANET) [46], a global network that acts as an intermediate between countries when one determines that an effective response to nuclear or radiological emergencies requires resources beyond its national capacity. EPR and luminescence fortuitous and biophysical RD are offered as resources for localized exposure, using materials close to the exposed part of the body: head (with EPR/teeth), extremities (EPR/nail), other localized sites (EPR/bone or EPR and TL/OSL with personal items), for small-scale events. Only *in vivo* tooth enamel assay is suggested for dosimetric triage.

When planning new research, a key issue is the amount and the continuity of funds. Programs for radiation protection research are available in almost all countries [34]. In Europe, in the framework of the Euratom Research and Training Programme, which covers nuclear research and innovation, the entire radiation protection research is managed by the Pianoforte Partnership (2021–2027) (https://pianoforte-partnership.eu/). U.S. programs have been leaders in supporting retrospective dosimetry for years, foremost among them the ongoing NIH Centres for Medical Countermeasures Against Radiation Consortium, which has among its goals the development of new techniques and devices to measure radiation exposure in the human body, but explicitly addresses as areas of non interest the non-biologically-based dosimetric methods such as fortuitous dosimeters (https://cmcrcniaid.org/content/instructions, last access on 21 October 2022). The Japanese government increased the size of the budget for overall studies related to the follow-up to the Fukushima Daiichi accident in March 2011, but the size of the budget has gradually decreased over the past 10 years. Most of these funds focus mainly on effective measures to protect or improve the health (including mental health) of people affected by nuclear/radiological accidents, rather than basic scientific issues [34]; H. Yasuda, personal communication). In the Republic of Korea (South Korea), there is no major research project conducted exclusively on the topic of retrospective dosimetry (RD) due to reduced funding in the field (J. Lee, personal communication). No surprise then that in the scientific community there is a shared feeling that RD research funding is challenging. One of the reasons could be the impression of a field which has not been able to innovate much in the last years, which has stimulated this paper.

3. Description of the RD assays

3.1. Features of the RD assays

Out of the many RD assays with EPR and TL/OSL suitable for the dosimetry of individuals, a few use biological tissues (tooth enamel, bones and fingernails), whereas the number of inert materials is wider and ranges from smartphone components to pharmaceuticals, from dust silicates lodged on objects, to salt and sugar contained in food. The features of the above described assays, the factors limiting the operational status, the open and debated topics are summarized in Table 1. For each limiting factor, the current directions of research are outlined and will be further commented in Section 4.

Tooth enamel is carbonated hydroxyapatite (CHA) that in healthy teeth has no structural change or turnover with time. The paramagnetic carbonate ion CO_2^- is the radiation sensitive marker. Three approaches exist for the EPR assay with teeth (Fig. 2). The assay of *ex vivo* extracted or exfoliated teeth [2,6] has clear limitations in the initial estimate of acute exposures and will not be considered in this paper. Alternatively, Q-band *ex vivo* [49] or L- or X-band *in vivo* have been respectively used or proposed [52,115]. The same paramagnetic carbonate ion CO_2^- is the radiation-induced marker in irradiated bone [64], but with continuous renewal of the CHA matrix and low sensitivity to radiation (about 10–20 times lower than tooth enamel [65]. Despite it, bone has been used in small-scale events where the levels of dose are typically so high to lead to local amputation or to justify invasive bone biopsies because of actual or expected soft tissue or bone necrosis (>40 Gy) [69–72].

Nails are made up of the protein α-keratin, where ionizing radiation generates free radicals, which are the sources of EPR signals. The growing body of literature that appeared in the last decade (e.g., see Refs. [1,116] and references therein) demonstrates a wide interest in this assay because of the non-invasive nature of sampling and because no other assay is suitable for localized exposures of extremities.

The ubiquity of mobile and electronic devices and especially of cell phones makes their components potentially powerful fortuitous dosimeters: the glass of the touchscreen displays and screen protectors; the aluminium or other metal oxides in the so-called surface mount devices (SMD), which include surface mount resistors (SMR), inductors (IND), and capacitors (CAP); and the material (filler) used to encapsulate microchips within an integrated circuit (IC) package [117]. Radiation sensitive filler material is also used to encapsulate chips found inside credit cards, electronic identity cards, and SIM cards [88,97,118]. Fig. 3 shows a diagram of the assays deriving from electronic devices, which are currently proposed for RD.

Among the many other materials whose dosimetric properties have been studied, worth mentioning for their sensitivity and signal stability

Table 1
Illustration of the assay features which are considered more relevant, the factors limiting the operational status and the possible directions of research.

Assay		Main features of the assay	Factors limiting operational status and debated topics	Possible directions of research
Material	Technique			
Tooth enamel biopsy	EPR	The sample is collected by a biopsy of tooth enamel. No further sample preparation is required. The RIS is stable over time and well characterized (IAEA, 2002; ICRU, 2002 [6]; ISO, 2020). Biopsies collection from the tooth lingual side avoids confounding EPR signals induced by medical radiographs [47] and sunlight [48]. Timely dose assessment after EPR measurement due to rapid signal analysis [49]. Detection limit <0.4 Gy [49]. Applied in two accidents [29,50].	The amount of tooth enamel needed (2–10 mg) is comparable to what is typically lost in professional dental cleaning. This makes the assay invasive, although minimally: practicable and ethical protocols for the chip removal from the tooth should be set up. The assay has currently been used by only a single laboratory [51], which has proposed a protocol with strong potential but not yet validated. Organizing an ILC is made difficult by the low number of laboratories with Q-band EPR spectrometers and the high training and expertise required.	Develop and test a standard protocol for tooth biopsy collection and eventual restoration, if needed. Validate the method by: a) blind tests comparing Q-band and X-band measurements in the same tooth biopsy; b) performing ILCs with laboratories having Q-band spectrometers, with prior training.
in vivo tooth enamel	EPR	Non-invasive assay [52]. The measurement is taken on anterior teeth exposed to sunlight, which produces a bias in dose equivalent to about 0.33 Gy for a 50-year exposure [48]. Prototype spectrometers have been able to measure 2 ± 1 Gy in one subject having received total body irradiation for bone marrow ablation [52]. The assay has been applied in people and cows potentially affected by the Fukushima accident [53,54].	Existing deployable *in vivo* spectrometers have been developed under the leadership of one single research group [55]. Independent validation tests are lacking. The *in vivo* EPR response is sample dependent [56] and requires procedures for dose response calibration [57]. The detection limit is about 2 Gy because the spectral resolution of the currently used L-band spectrometers does not allow to distinguish RIS and BGS [52].	Validate the assay by an ILC or blind dose test with samples irradiated at independent facilities. Calibrate EPR response for differing enamel thickness and shape, for instance with head phantoms containing extracted human teeth. Explore the feasibility of developing *in vivo* spectrometers using higher microwave frequencies (X- or S-band) having higher field resolution and signal to noise ratios (Guo et al., 2021; Ikea and Ishi, 1989).
Finger- and toenails	EPR	Easy sample collection. Ubiquitous. Fingernail assay has been applied in four radiological events [58,59] Complex EPR spectrum with RIS, BGS, and mechanical-induced signals.	The existence of a stable component of RIS is still a debated topic [58,60,61]. The absence of a stable RIS makes nail dosimetry not applicable. Chemical treatments have been proposed to restore the initial unstable RIS intensity, but it is unlikely that these could be used immediately after an accidental exposure. The mechanisms of radical formation, and the cause of unwanted EPR signals not induced by radiation, are not well understood. Water, heating and exposure to light influence the stability of the RIS and the BGS [62,63]. These effects have an impact on sample preparation. The BGS overlaps RIS and have the same line-shape, so it cannot be subtracted [58].	The first research priority is to confirm or not the existence of a stable RIS. Whatever the existence or not of a stable RIS, the possibility to restore, by chemical treatment, the unstable component of RIS is also of high priority. Improve understanding of the mechanism of radical(s) formation. Evaluate the influence of water, light and temperature separately so as to avoid cross-bias. Perform standardization of sample preparation and blind dose tests between laboratories to test single aspects of the procedure, such as the irradiation-to-measurement time interval and the storage conditions. Explore different models for signal analysis to separate and quantify radicals by: optimization of the spectral parameters using Q-band EPR; use of chemical treatments such as antioxidant reagents; study of a relatively large number of individuals of different age, gender, lifestyle factors, health-related diseases, or previous medical exposures.
Bone	EPR	Extremely invasive assay, requires a bone biopsy. Radiation induced radical is the same as in tooth enamel, with well characterized dosimetric properties [64]. Bone is 10–20 times less sensitive than tooth enamel because of smaller mineral density [65]. The RIS fades during bone-life because bone is a metabolically active organ [66], but it is stable after biopsy [67,68]. Despite the invasiveness and the low sensitivity, bone assay was applied in accidents with amputations/biopsies and dose higher than 40 Gy [69–72].	As bone is a metabolically active organ undergoing continuous remodelling throughout life, the remodelling impacts on dose from past exposures requires investigation. The small mass of bone biopsies currently limits the minimum detectable dose to tens of Gy when measured by X-band EPR [51]. The procedures for dose assessment were successfully applied in small scale accidents, localized irradiation and high doses, but require validation and standardization.	Evaluate/quantify bone remodelling and corrections for potential dose underestimation. Investigate Q-band measurement of RIS and BGS components to obtain higher signal to noise ratios. Explore the use of bone masses as small as achievable to determine the dose gradient along/within a bone fragment. Standardize experimental protocols conforming to the ISO (2013) and test by independent laboratories as part of an ILC.

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Table 1 (*continued*)

Assay		Main features of the assay	Factors limiting operational status and debated topics	Possible directions of research
Material	Technique			
Mineral glass	EPR	Present in smartphone displays and other items and so nearly ubiquitous. Detection limit is higher than 1.0 Gy in older models of displays and under laboratory conditions [73]. An ILC performed, and one in progress, under RENEB (see 3.2). Resistance by owners to hand over electronic device to rescuers.	The evaluation of the glass RIS intensity is time consuming because the EPR spectrum is composed of several RIS components, a BGS, and a UV and visible light-induced signal (LIS) [74]. The spectrum evaluation is complicated by the different fading rate observed in different RIS components [10, 75]. BGS overlaps RIS, but cannot be subtracted from the EPR spectrum of irradiated samples because its line shape varies with glass type (i.e., the smartphone model) [76–78]. EPR measurements performed by the currently used X-band in vitro spectrometers requires destruction of the glass components. Rescuers meet resistance from owners to give own device.	Investigate the characteristics and properties of LIS to unambiguously indicate its presence in the EPR spectrum, the effects of light on RIS stability. Enhance computational methods aiming at spectral decomposition [74]. Explore procedures to thermally bleach the RIS and the LIS to extract the BGS from irradiated samples. Develop X-band resonators for the measurement of intact display glass of the smartphone [79,80] and if possible *in situ*.
Mineral glass	TL, OSL, TA-OSL, PTTL	Destructive assay. High luminescence sensitivity to radiation. The "pre-bleached TL protocol" [81] of display glass was validated in an ILC and a field test (see 3.2). Size of display glass of modern smartphones has progressively increased over time. Resistance by owners to hand over electronic device to rescuers.	New protocols (i.e., PTTL and TA-OSL) have been developed [82–84] to improve parameters such as the BGS intensity and the degree of signal fading, but limited testing of the protocols so far. Laborious sample preparation for touchscreen and display glass. Intrinsic signals currently limit the detection limit and produce outliers in dose evaluation. In particular, BGS overlaps RIS and leads to overestimation of the assessed dose. Intrinsic signals are reduced by mechanical or etching procedures carried out during sample preparation, but it is labour-intensive, requiring special equipment [85,147]. Non-destructive approaches to measure the rear glass of a mobile phone have been developed in recent studies [86]. These require custom-made readers, which are not commercially available. Determination of absorbed dose by measurement of the red TL emission observed for a specific type of display glass lacks a standardized measurement protocol and knowledge of the dosimetric properties is currently limited.	Investigate PTTL and TA-OSL properties on different brands of mobile phones and trial irradiations of intact phones. Final validation by ILC. Search for fast and simple separation of different glass layers, with the option to replace the display component extracted for testing. Development of new measurement protocols and tools for data analysis to minimize the impact of intrinsic signals and improve the detection limit, avoiding the need of chemical sample treatments if possible. A wider range of testing of this procedure with different brands/models of mobile phones, followed by an ILC to validate. This prerequisites the development of commercial readers, which can accommodate and measure intact phones. Further development of the measurement protocols and dosimetric characterization of the glass type.
SMD (Resistors, inductors)	OSL, TL	High sensitivity to ionizing radiation and low BGS for OSL, but variable BGS for TL (ICRU, 2019). Phone is destroyed by sampling. Protocol for rapid and more accurate dose measurement developed for OSL with SMRs [87] and validated in an ILC and field test [87, 88]. Uncertainty budget from OSL using SMRs has been obtained [89]. Dose assessment with TL possible after OSL measurement, giving two independent dose results [90]. TL method enlarges the capacity for dose assessment in a network as pure TL readers in laboratories and national radiation protection agencies could then be used as well. Resistance by owners to hand over electronic device to rescuers.	For SMRs a dependence of fading rate on dose has been suggested for OSL [91] and a variability in fading rate observed for both resistors and inductors [88,89,92]. For TL an irreversible sensitivity change occurs after the first readout, and variable BGS are present for resistors [93,94]. Correction factors were suggested, but these were assessed on older mobile phone samples. Variability of fading rate for TL on resistors from different phones is currently not known. A photo-transferred TL (PTTL) study has demonstrated the measurement of deep and more stable traps, with detection limits between 100 and 200 mGy, but for large resistors, which are not found in modern smartphones [95]. The size and number of SMDs in devices is expected to decrease more and more, leading to reduction of sensitivity of the OSL/TL signal. On the other hand, increase in stimulation power of the OSL reader has improved measurement sensitivity and this could continue in the future.	Multi-laboratory assessment of the dosimetric properties on a larger and more up-to-date sample set. Development of a standardized protocol for TL and validation by an ILC. Independent validation of the results and assessment of the detection limit of the PTTL method in modern smartphones Continuous update of the achievable detection limit with modern equipment and smartphones. Research into the use of other detection windows using TL that promise higher sensitivity [94].

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Table 1 (*continued*)

Assay		Main features of the assay	Factors limiting operational status and debated topics	Possible directions of research
Material	Technique			
Filler in encapsulations (chip cards)	TL, OSL	Item of low replacement cost, therefore could be donated more easily than smartphones in case of accident. Chip cards with translucent encapsulations do not require cleaning or chemical treatment, and can be prepared very quickly using punching tools. Detection limits as low as 10 mGy immediately after irradiation were reported with OSL [146]. Chip cards with molded encapsulations are less sensitive and require chemical extraction of the filler [96].	Some chip cards are not radiation sensitive using TL [97]. Detection limits (OSL) can vary significantly between different types of chip cards [88]. Signal stability is poorer than that of other fortuitous dosimeter materials (<20% of initial signal remaining after 10 days) [146]. The uncertainty introduced by fading correction is unknown. A variation in OSL fading rate was observed between different types of chip cards [88]. Preheating in OSL or use of TL have been attempted to obtain a more stable signal, but high BGS (up to tens of Gy), signal recuperation and sensitivity change after the first reading were observed [97,146]. Chip cards with molded encapsulations have so far been investigated in a single laboratory only [96]. Fading correction factors seemed to be insufficient for dose reconstruction.	Comprehensive overview of the ratio of non-radiation to radiation sensitive chip cards and of the variation in detection limits. Assess the variability in fading rates on a large sample set and develop methods of dose uncertainty assessment due to fading. Develop criteria to identify in which fading category a sample falls into (OSL). Explore new protocols for isolating more stable signals and for minimizing the side effect of thermal treatment. Independent confirmation or modification of the protocol by other researchers and development methods for successful fading correction. Validation of a harmonized protocol in an ILC.
Filler in encapsulations (IC chips on PCB)	TL, OSL	Material is expected to still exist in the future unlike other SMDs (surface mount devices), such as resistors, inductors, capacitors, that have continuously decreased in number and size in modern mobile phones. Possibility for non-destructive measurement by future development of an in-situ measurement approach. Resistance to giving the mobile phone from the owners.	The influence on dose reconstruction of heat generated by an IC chip during operation has not been evaluated. Sample preparation is labour-intensive, more than for SMDs. Special equipment is needed to extract and cut IC chips without damage [98]. The lack of a deeper understanding of the TL/OSL mechanisms has led to the suggestions of different measurement approaches for dose assessment [99,100]. High BGS and low sensitivity to radiation in TL [99,100].	Investigation of the effect of phone operation on the evaluation of dose. The development of a standardized simple sample preparation protocol. Development of a standardized protocol and its validation by means of an ILC. Measurements of TL emission spectra and exploration of new detection windows to potentially improve TL characteristics
Other assays	TL/OSL/ PSL/EPR	The application of EPR and luminescence techniques to several new materials for RD measurements have been proposed. Examples are: Silicate/TL: found in dust, which is ubiquitous (e.g., in any item and in tobacco); low detection limit [101]. Salt/OSL [102], TL [103] and PSL [104]: Easy sample preparation. PSL is a low cost and deployable equipment; low detection limit [105]. Sugar/EPR: very well characterized by many studies. Used in very few cases of accident (Hütt et al., 1996; [106]. Dental Ceramics with OSL/TL [107,108] Drug and pharmaceuticals [109,110]: Fabrics, including face masks with TL [111, 112].	Common challenges: most assays have been proposed by one laboratory, there is a lack of studies and poor characterization	The investigation of new materials should follow, where possible, available reference standards. In particular, EPR assays should be tested following the ISO [113,114]. It is important that successful candidates are subjected to ILCs. A standard for OSL/TL on the minimum criteria for TL/OSL RD should be foreseen

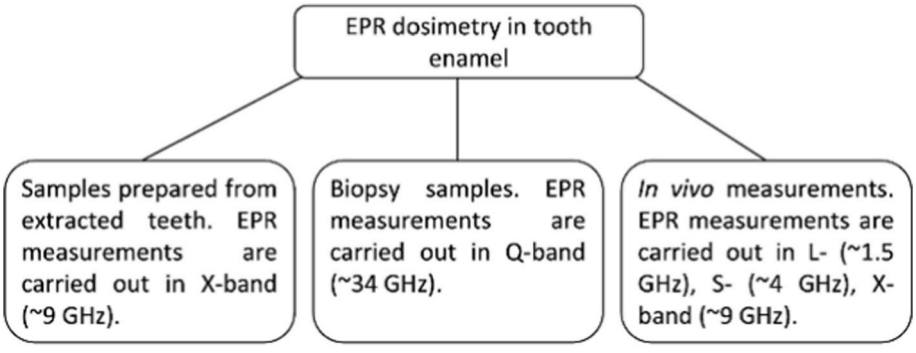

Fig. 2. Approaches for sample collection and EPR signal measurement in EPR dosimetry of tooth enamel.

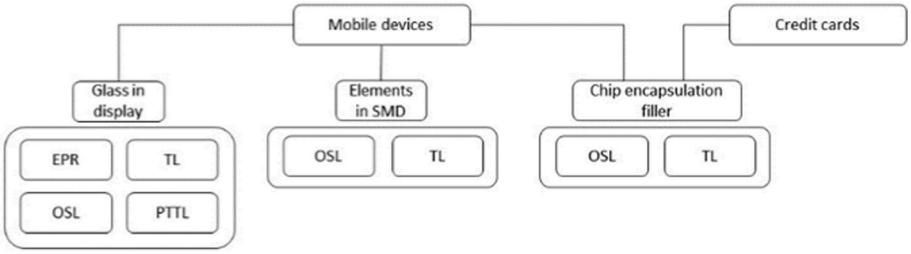

Fig. 3. Personal item/material/technique combinations proposed for RD.

characteristics are sugar, salt and other substances that could be present in the form of sweets, snacks or medicines. These materials, however, have rarely been found on the body of an exposed subject in the accidents occurred so far. On the other hand, definitely ubiquitous are the silicates present in dust: these are found in almost every item, from clothing pockets to the inside of smartphones to the cigarette tobacco. Finally, interesting is the study of fillers in polymer-fiber materials of surgical face masks [111], which have been definitively a ubiquitous object in recent years.

3.2. Inter-laboratory comparisons (ILC)

Several ILCs on TL/OSL and EPR assays were organized over the past decade including: EPR assay on touchscreen glass [73], OSL on SMR from mobile phones [87], and TL on the display glass from mobile phone touchscreens [9]. Details are reported in Table 2. These ILCs were carried out under controlled laboratory conditions. The participants were invited to use a shared procedure and had the possibility to attend a training session, especially on sample preparation, before starting the measurements. Although, in general, the agreement between estimated and nominal doses were reported to be quite satisfactory, some outliers were observed. For the ILC of EPR with glass, higher variability was obtained when the samples were stored in uncontrolled sunlight exposure. In the OSL ILC [87], reported misidentification of electronic components as a source of error for a participant who did not attend the training. For the protocol using TL on display glass, the existence of a pronounced intrinsic background signal in some glass samples was suggested as a cause of dose overestimation in Ref. [9].

Field tests exercises (Table 2) were also carried out (e.g., Refs. [43,88,119,120]; here, irradiations were made in more realistic conditions, with mobile phones and other materials placed on anthropomorphic phantoms. The participants were required to use the protocols used in Refs. [9,87]. In most of these exercises, the doses estimated by the participants were compared with those of reference dosimeters. For the protocol using TL on display glass, the dose overestimation reported in Ref. [9] was confirmed in Refs. [88,120]. However, the reasons for under- or overestimation of doses were not systematically explained. The field tests confirmed the importance of using a multi-dosimeter approach in a complex radiation field. The results by Ref. [119] demonstrated that the organization of a field test is very demanding in terms of logistics and time management.

The continuation of ILCs was encouraged by ICRU [1] and EURADOS [45]. A RENEB ILC, which included EPR and luminescence methods among other techniques, was recently organized and a EURADOS ILC using EPR on tooth enamel is in progress.

3.3. Computational approaches

Computational approaches have been proposed in different ways for small-scale and large-scale accidents.

3.3.1. Organ dose mapping

To help physicians make a diagnosis and define the therapeutic strategy, an assessment of the heterogeneity of the dose in the body, in particular at the bone marrow, is more important clinically than the overall average dose [31]. Computational approaches have been used to map organ doses, with the help of biological or fortuitous dosimetry measurements, when irradiation modalities (e.g., time, distance, energy) are unknown. By using Monte Carlo (MC) codes together with personalized numerical anthropomorphic phantoms constructed from medical images of the victims, one can model several exposure scenarios to test different hypotheses, which can be validated or refuted by comparing with data from RD assays and biological dosimetry. Application of this approach in past accidents involving a few people, for both global and localized exposures, has shown success (see for instance Refs. [29,121,122].

3.3.2. Dose assessment to the individual

The approach described above is time-consuming and can be applied only in case of few exposed subjects. Alternative approaches have been devised to relate the dose in the fortuitous dosimeter to the dose received by the person. The fortuitous nature of the dosimeter means that it and the individual can receive very different exposures, depending on the radiation field, the individual's orientation, the dosimeter's location on the body, and the difference in energy response of the fortuitous dosimeter material compared to soft tissue. The difference between the 'true' individual dose and that reported using the personal item may be acceptable in some circumstances, but has been estimated as $>10\times$ for photon irradiation in some other cases [123], and even more for neutron irradiation, and may dwarf the uncertainty associated with the physical measurement itself.

In RD, there is no agreed dose quantity or concept for assessing the level of exposure of the individual and their immediate medical care needs [124]; ICRU, 2019). Attempts have been made to explore quantities to estimate whole-body dose [125], to derive factors that correlate them with doses as a function of exposure conditions [123,126,127], and to produce pre-tabulated databases of conversion coefficients (personal communication by J. Eakins) [128]. produced an operational tool based on calculated dose conversion coefficients from EPR fortuitous dosimeter to organ- and whole-body for photons and neutrons in several accident exposures (source at 1 m, in a pocket, in a hand, and contaminated floor). Interestingly, this tool has never been used in accidents, where dedicated MC simulations have always been preferred due to their (thankfully) small-scale natures.

Because in many practical circumstances the precise details of exposure are likely partially or completely unknown, ICRU [1] suggested that conversion of dose in the dosimeter to dose to the body might be appropriate only in specific small-scale scenarios. These could be those in which relatively few individuals are exposed and conditions could be reconstructed in greater detail than in large-scale events, although this may not always be true in all contexts, or in those cases where scenario complexity hinders timely simulation, as in the case of neutron irradiation in criticality accidents [129]. Instead, the ICRU [1] report recommended that the dose to be used for early-stage dose assessment after a large-scale radiological event should be the absorbed dose in the fortuitous dosimeter material, without any conversion factor applied. The report also called for further research and development on this issue. The above citations have raised some discussion points, but

Table 2
Main features of the international ILCs.

Reference (year of the ILC)	Objectives	Lab/Field test *(No. of participants)*	*Assay*/Protocol	Test dose	Reference dosimeter (Yes/No)	Lessons learned/Open questions
[87] (2012–2013[a])	Validation of a protocol by participants. Aim of establishing a laboratory network to increase Europe's capacity to respond to a mass radiological emergency.	Laboratory test *(12)*	*SMR/OSL* Same protocols for all participants: "full-mode" and "fast-mode". Fading correction, with an analysis template provided to the participants.	Three mobile phones per participant, irradiated with the same ^{60}Co source. Blind doses of 0.3, 1.7, and 3.3 Gy (air kerma).	No	Prior training of the participants before the exercise resulted in successful avoidance of misidentification of components and consequent outliers.
[73] (2012)	Evaluation of the detection limit and the ability of the method to assess correct dose categories (<2 Gy and >2 Gy).	Lab test *(13)*	*Touchscreen glass/EPR* Same preparation procedure, measurement protocol and EPR signal software package for all participants. Two sets of samples: A) samples from a homogeneous batch of glass, stored in the same setting; B) samples from different smartphones and stored in different light and temperature settings.	Calibration samples irradiated at 0, 0.8, 2, 4, and 10 Gy. Blind doses at 0, 0.9, 1.3, and 3.3 Gy.	No	Dose <2 Gy and >2 Gy were distinguished for samples of group (A), but not for (B). Detection limits and uncertainties were affected by sample storage conditions. Training offered to all participants before the ILC proved to be a key factor for success.
[9] (2015[a])	To test the performance of a TL method on glass.	Lab test *(11)*	*Display glass/TL and other assays* All participants applied the same protocol [81, 147], using a single dose calibration point at 1 Gy. Two preparation protocols applied.	Blind doses of 0.6, 1.5, and 2.5 Gy	Yes[a]	Assessed dose within 95% confidence interval of the test doses. In a few cases, overestimation of the assessed dose was possibly due to a pronounced intrinsic BGS.
[88] (2014)	Assess and evaluate physical RD methodologies using objects of daily life in a simulation of a radiological event (lost source in a bus).	Field test *(12)*	*SMR/OSL and other assays*$^{\infty}$ Same protocols for all participants: "full-mode" and "fast-mode" protocols on SMR [87].	Dose range 0–2 Gy depending on the distance of the mobile phones from the source. Two radiation sources used.	Yes (Luxel)	The reasons for dose under- or overestimation from outliers should be investigated and identified because they could be a valuable source of information.
[119] (2019)	Simulation of an event with anthropomorphic phantoms. To reconstruct doses in fortuitous materials. To relate estimated doses to organ doses in various exposure geometries. To reconstruct the exposure geometry by combining the dose estimates from various items.	Field test *(15)*	*SMR/OSL, display glass/TL, other assays*$^{\$}$ The participants were given the choice of using either the measurement protocols of previous ILCs or use their own developed protocols	Dose range 0–2 Gy depending on the distance of the mobile phones from the source.	Yes (TLDs, Luxel, glass)	Key lessons learned during and after this field test related to logistics and time management (e.g. transporting the anthropomorphic phantoms to and from the test site was both time consuming and expensive, on site each task took longer than expected …).
[43,120] (2019)	To verify the reliability of TL and OSL RD assays with mobile phones placed on anthropomorphic phantoms. To confirm the harmonization of the results between the different fortuitous materials and between the participating laboratories. To test the geometrical response of the applied protocols.	Field test (3)	*Resistors/OSL, display glass/TL, chip cards/OSL for 1 lab* Same model of mobile phone, same protocols: pre-bleached TL protocol [81,147] for the display glass samples, and "full-mode" and "fast-mode" protocols [87] for the resistors.	Dose >1 Sv for the phantom located closer to the source, and <1 Sv for the phantom located further away. 16 mobile phones placed at eight positions.	Yes (LiF:Mg, Cu,Si TL detectors)	Outliers were observed. Unoptimized use of the protocols was one of the main sources of uncertainty. An unexpected but systematic over-estimation of reconstructed doses requires re-examination.

[a] Personal communication from the Authors, ∞ Display glass/TL (pre-bleached TL protocol [81], SMI/OSL and TL, chip cards/OSL, household salt/OSL, dental ceramics/OSL and TL, $ OSL or TL assays with salty snacks, cigarettes, Kleenex, chip cards, dental ceramics, textile bags, etc; EPR assays with tooth enamel, sucrose, ascorbic acid, xylitol, stevia sweeteners, etc. Only one EPR laboratory participate.

there is still no consensus on the conclusions, as noted by ICRU.

Computational tools have been tested and validated against experiments under well-known source terms and exposure conditions for application in radiological accident dosimetry (e.g., Ref. [130]. Comparisons of computed and TLD measured organ doses in anthropomorphic phantoms in a well-defined radiation field were carried out (Lemosquet et al., 2004; Huet et al., 2009; Huet et al., 2019; [127,131], as well comparison with physical RD assays [119,127]. In the field tests described in 3.2, the parameters of the simulation were tuned by comparing computed doses to doses measured with reference passive dosimeters placed in and on phantoms and together with samples used as retrospective dosimeters. As a matter of fact, uncertainties in distance, source activity and problems in simulating the actual source induced some errors. Nevertheless, such an approach was possible in those controlled field tests because the exposure conditions and source term were well known, as well as the doses at some points. But it would be less applicable in more typical scenarios, especially in large scale accidents with insidious exposure, which are likely to be more ill-defined: the 'where', 'when' and 'how' circumstances of such events would often be unknown.

4. Future research directions

Table 1 has summarized suggested lines of research that can be found in the scientific literature. In this section we will try to group them into five macro-categories of actions with the hope that they will facilitate collaborative research projects, paying attention especially to those directions that are expected to lead effective results in a short term or that can lead to novel paths.

4.1. Expanding the knowledge base for theoretical understanding

Despite the research effort and the significant amount of published literature, it appears that most assays seem to have reached a situation of a stalemate, as seen by a not increasing number of publications, especially in the field of EPR dosimetry (Fig. 1). A game changer could be a shift of research towards the understanding of the formation mechanisms, the precursors of the spectrum components and the physico-chemical properties of the paramagnetic centres that generate the EPR signals, rather than phenomenological studies about the shape and properties of EPR signals, which have been the main focus of investigations so far. Learning more deeply about the origin of the native signals and their physico-chemical properties can guide the sample preparation, the spectrum analysis, and especially lead to a better standardization of the methods and an uncertainty reduction. This approach is perhaps more useful in EPR than in TL/OSL because an EPR spectrum is composed of several signals, many of which are non radiation induced.

The following list highlights specific research areas that would enrich knowledge.

4.1.1. Use of a multi-technique approach

A wealth of information can be derived from comparative analysis studies of the same material carried out with different techniques to investigate different chemical-physics phenomena and mechanisms. The most obvious comparative analysis seems to be the integration of EPR, TL, and OSL performed on the same material, where EPR analysis can help to identify electrons or holes or charge transfers. This approach has a solid background in dating, but has been explored in very few cases for RD [78,132]. Infrared or Raman spectroscopy could be used to analyze molecular bond breaks as a result of radiation or sample preparation steps, on paramagnetic centres, as well as on defects inactive for EPR [133,134]. In the context of EPR techniques, a multi-frequency approach may help: from the most common comparison of Q-band and X-band signals (exploiting the resolution power of the EPR Q-band to distinguish signals), to the sophisticated technique of electron-nuclear double resonance (ENDOR), which allows identification of the nuclei near the defects, as e.g., done with tooth enamel [135].

4.1.2. Quantum chemistry calculation

Quantum chemistry approaches make it possible to predict the properties of a given molecule and how it will undergo chemical reactions by studying its electronic structure, and how these electrons interact with those of other molecules or atoms, through the Schrödinger equation, without requiring any experimental data. This approach is widely used in EPR spectrometry studies, but to our knowledge, in RD EPR has only been preliminarily used for different types of sites in nails [136,137] and sucrose [138]. It has also been performed for TL/OSL dosimetry (e.g., Ref. [139].

4.1.3. Understanding the influence of individual sample characteristics on the signal

Within a same class of personal items, dosimetric properties vary with the specific type of the materials under study; for example, the fading rate changes with the model of chip card [88,97], and different glass types exhibit different line shape and stability of the background signal (BGS) in EPR [51]. It would be useful to associate the properties of paramagnetic and luminescent centres with the chemical composition and manufacturing processes of the material type under study. To support comparisons of results from different research groups, it would be useful to accompany EPR and TL/OSL studies with chemical analytical methods, electron microscopy techniques (TEM, SEM), or various spectroscopic methods (e.g., XRD, FTIR NMR), whenever possible. Similarly, as much data as possible on age, sex, lifestyle factors, medical conditions, current therapies, or previous medical exposures should be provided in the studies of biophysical assays.

4.2. Further development of methods and optimization of the procedures

For phases I and II of large-scale events (see 2.1), the response must be so rapid that even the networks are not a valid solution, requiring the additional time for shipping samples between laboratories. Phase III does not require the same level of rapidity, but the dose to be measured is lower than 1 Gy with significant delay since exposure. Most RD methods do not fulfil these requirements (Table 1). Bottlenecks are met during the phases of sample collection, preparation and measurements.

Sample collection presents obstacles related to logistics, such as the time required to collect samples or the difficulties in convincing potential victims to donate personal items, in particular the smartphones. Current research on RD assays sometimes does not take into account these specific problems expected to be faced in real incidents. Dialogue and knowledge exchange between research institutes and organizations responsible for crisis management should be fostered to optimally target research and development in light of the real needs of crisis management.

Sample preparation time is often another bottleneck. Because of this, a single laboratory can measure daily only some units of samples, in the most optimistic case (as the field tests have shown). Optimization of sample preparation and measurement protocols, finalized to reduce the dose assessment time, would be a turning point for many methods. Alternatively, the research of assays that do not require sample preparation could be pursued, such as for instance PSL with silicate dust [101] or the non-invasive methods described in 4.3.

A main recurrent source of uncertainty with all EPR (except tooth enamel) and TL/OSL assays is the instability in time of the RIS, with the fading rate depending on several environmental factors, such as light, UV, temperature and humidity, during different phases, e.g., before the irradiation, during collection, shipping, sample preparation, and measurement. Because the time of irradiation may not be accurately known, as for example for an insidious large-scale exposure scenario (such as a radioactive source in a city subway), the limitations of assays with significant signal fading need to be assessed clearly. The future

applicability of these assays will depend on finding mitigation solutions. One could be the standardization of the controlled conditions for storage and shipping (ISO, 2013), which will contribute to reduce the sources of uncertainties associated to the correction factors that are currently employed. Another solution could be the minimization of the sampling-to-measurement time using field deployable instruments that might be distributed to multiple sites at the same time. Devices to be favoured should require no sample preparation, for the above explained reasons, and minimum training of operators because the lack of human resources is always a bottleneck. The non-invasive methods (see subsection 4.3) are in principle candidate for deployable dosimetry, but the current prototypes are very sophisticated and require highly skilled operators.

Signal fading could also turn in an advantage in some situations. One strategy that has been little explored is to combine the different fading rates of signals in the same assay or of different assays to trace back the time when the accident occurred, for instance comparing the dose from different assays of the same item (e.g., EPR in glass and OSL in electronic components), or different signals in the same assay.

4.3. Tools and technologies for non-invasive assays

Many of the described RD assays ask for invasive sampling (e.g., bone) or damage of the item collected (e.g., smartphone). For items considered valuable by the exposed person, as a cell phone certainly is, destruction is a deterrent, which could be justified only by a high reliability of the assay, i.e., a virtual absence of false positives or false negatives. The breakthrough could be the development of non-invasive or minimally invasive methods.

4.3.1. Development of non-invasive assays

Particularly exciting is the development of equipment that can measure teeth *in vivo* (by L- or X-band) and EPR [52,79,115]; Guo et al., 2021) or OSL direct measurement of personal objects without prior destruction [86]. This research avenue requires the continuous development of new technological approaches, and inevitably demands large human, financial and time resources, as the ongoing problems with producing an *in vivo* L-band EPR reader for teeth have demonstrated [55]. Despite these difficulties, this research path is worth pursuing, if funding is adequate. However, efforts should not continue for assays whose properties (e.g., fading or BGS) are still unclear: for example, one could wonder whether *in vivo* measurements of fingernails should be pursued until the presence of a stable EPR RIS component has been confirmed (Table 1).

4.3.2. Development of semi-invasive methods

An alternative approach is the development of semi-invasive methods, i.e., application of invasive assays to samples with masses sufficiently small to not significantly or irreversibly damage the item. This category includes, for instance, the EPR Q-band measurements of tooth enamel biopsies described above [49] and the non-destructive OSL with shavings from PCBs in electronic watches, which requires minimal sample preparation [140]. Semi-invasive assays typically make use of the same instrumental techniques as used for the invasive methods, and do not require the development of innovative instrumentation. They are therefore more likely to be achievable in the medium term than the non-invasive assays described above. These methods should be tested by independent laboratories and in future ILC programs.

4.4. Standardization of procedures and method validation

One valuable aspect in designing a cost-effective strategy for research is to share and integrate research results among laboratories. The existing laboratory networks can be an invaluable support for this. To this goal, it is necessary to make results as comparable as possible through the standardization of the procedures and the harmonization of data. A valuable tool is the ISO standard for EPR assays [113], which proposes a methodological frame and recommends the minimum criteria for setting-up procedures, at all steps: from sample collection to the relevant issues of the calibration source, to dose reporting, and for evaluating the assay performance parameters.

Method validation is carried out with ILCs. According to the ISO 17025 reference standard [141] the objective of ILCs depends on the level of maturity of the considered method. If the method is still under development, ILCs are essential to assess, the degree of similarity of specific performance characteristics of an assay by different laboratories. Factors known to influence results should be held as constant and uniform across participants as possible, or varied if one wants to confirm the presence of biases. For already developed methods, ILCs are typically used to assess the random variation or the systematic differences in the

Table 3
Goals, research questions and directions of research of computational modeling in RD.

Research questions	End goal	Possible directions of research
How to ease and improve the dose reconstruction?	Dose assessment and dose mapping for small-scale events	Develop libraries of phantoms, sources, shielding, etc. Develop implementable computational tools [131] Develop means to perform calculations 'on the fly', to generate conversion coefficients immediately after events rather than relying on pre-tabulated databases
How to harmonize? Which dose quantity is needed?	Dose assessment for large-scale and small-scale events and dose mapping for small-scale events	Train modellers in computational techniques Support the benchmarking of application through the setup of intercomparison exercises Support collaboration between different experts in RD, and between experts in RD, crisis management units and clinical staff.
In which circumstances are the exposure conditions known with sufficient detail to allow dose conversion coefficients to be accurately computed and applied in large-scale scenarios?	Dose assessment for large- and small-scale events	Analyze the types of situations in which ignorance of the precise exposure conditions may either be minimized or have minimal impact, within an acceptable level of agreement also to be established.
Under what sets of situations might the dose to the fortuitous dosemeter be a sufficiently reliable indicator of the dose to the individual?	Dose assessment for large- and small-scale events	Develop consensus on what level of discrepancy and/or conservatism is acceptable for triage dosimetry seeking collaboration among involved figures, such as experts in RD, crisis management units and medical experts.. Generate a database of such conditions and the associated limitations/ uncertainties of reporting dosimeter doses 'as is'.
How best to provide dose harmonization from multiple materials (fortuitous dosimeters, radiation monitors, …), either co-located or spatially distributed?	Dose assessment for large- and small-scale events	Develop methods to inter-relate such doses, both to each other and to the individual.

measurement results among laboratories or towards an accepted reference value. A well-designed ILC should be constructed to answer only one or two clear questions. The results from past ILCs (see 3.2) were constructive, but future ILCs should be planned only after new knowledge has been produced, and after reasons have been identified for the random variations or systematic deviations observed in previous ILCs. This is especially true because the ILCs are demanding exercises in terms of financial, human and time resources.

An important element for minimizing biases is the training of novice participants before the exercise, as was also demonstrated in two ILCs (see 3.2). An additional long-term merit of training sections is dissemination of the method and good practice, building the basis for a trained network [9].

4.5. Testing applications

4.5.1. Design of field tests with RD assays

Field-testing is useful in assessing the true applicability of a method in the actual context in which it will be used, especially the interaction between laboratories and the organization in response to an event. In principle, field-testing should be applied to assays that have reached a level of maturity, with procedures robust enough to rule out uncertainties due to different sources. In practice, instead, this is often done whenever the opportunity (i.e., mainly funding) arises, because the organization of a field test is a complex exercise. This was the case for the field testing described in 3.2, which left unexplained the reasons for the deviations between the laboratories' results and from the reference dose. The field tests were certainly useful to gather lessons on logistics and time management of such large exercises, which will be helpful to consider in the organization of future field tests [119]. Again as for ILC, in the future the design of field tests will have to be elaborated with clear and limited objectives, to avoid potential misinterpretation caused by many uncontrolled or unfixed parameters.

4.5.2. Analysis of the results of dose assessment in previous radiological events

As mentioned in subsection 4.2, a dialogue between the operational units in charge of handling the incidental events and the research laboratories would be helpful to define the real needs for RD, for example, for medical care or reassurance of the population. In the absence of this, help may come from a study of the lessons learned in past radiological events, to understand which RD solutions were useful and which could have worked better, thus identifying gaps to be filled for the types of incidents already encountered, and to extrapolate the specific needs of large-scale incidents based on the different phases of handling these types of events. For example, a non-exhaustive analysis of some past incidents on the use of validated or developing techniques was conducted in ICRU [1], which allowed some preconceived ideas to be challenged and some gaps to be better defined. An enormous source of detail can be provided by the IAEA reports, a list of which is given in the references. It is about achieving as much consensus as possible on these issues and a clear road map to guide research. It is about changing working methods starting with needs and not, for example, the search for funding. Approaches that may be disconnected from the realities of the field should be avoided. For example, taking into account operational constraints on recovery of biological or non-biological samples, which may occur weeks after irradiation, it seems appropriate to focus on materials with low fading signals or that do not change if they come in contact with water during decontamination (see nails).

4.5.3. Investigation of the response to radiation energy and radiation quality

In view of applications to accidents, it is essential to study the response to different radiation quality (particularly low-energy photons, β-rays, and neutrons). However, these studies should be conducted on assays that have already been fully characterized with reference gamma radiation, so as to avoid confounding effects due to properties not yet well known.

4.6. Future research of computational modelling in RD

A separate discussion is deserved for the perspective of computational modelling. Table 3 lists some of the research questions of relevance to these topics, and suggests some potential ways forward.

A novel path would be the development of libraries, templates, and implementable computational tools [131] that could be rapidly distributed to emergency response teams, to permit calculations to be performed 'on the fly' following small-scale incidents (Table 3, line 1). Direct interaction with victims and witnesses will allow a more reliable description of the exposure scenario to be constructed, which may in turn lead to more accurate modelling of exposure conditions. The possibility could be explored to use MC alone when geometries of exposure and/or source parameters are known.

A key focus for dose assessment (large- and small-scale) within the computational approach surrounds the problems highlighted by ICRU 94 (Table 3, lines 2−4). ICRU 94 recommends using the fortuitous dosimeter dose (in Gy) from personal items as a rough indication of the initial-phase dose assessment for triage purposes only; obviously, this quantity cannot be used alone for further medical decision making. The scope, impacts, and limitations of this approximation need to be further explored. Part of this work stream could reframe some of the previous efforts in computational dosimetry, with results being used not to improve accuracy, but rather indirectly to estimate the uncertainties resultant from either applying or not applying conversion factors, thereby ruling in/out classes of exposure for which RD provides sufficiently reliable dose estimates. Closer collaborations between physical dosimetrists and stakeholders such as clinical staff will be important to achieve this end. More in general a consensus on the circumstances and the acceptable level of approximations and uncertainties should be sought among different figures, including the expert in emergency radiation protection and the crisis management units, as already mentioned in 4.2, which is an extremely challenging task in all types of emergencies.

It is possible that, in the future, RD could employ a combination of two or more different fortuitous dosimeters located about an individual, thereby giving a rough dose distribution and insights that may help with field reconstruction and the uncertainties associated with point-of-test dosimetry using single personal items (Table 3, line 5). The questions would then be: how can results from a range of fortuitous dosimeters be combined to provide estimates of the overall dose to the individual; and to what extent might the uncertainties from single dosimeters be mitigated by multiple input methods? Another potential task for computational approaches could therefore be to derive means for inter-relating such doses, both to each other and to the individual, when a variety of different techniques are applied in concert leading to a range of spatially distributed dose estimates. This is particularly important given the recommendations of ICRU 94 that the absorbed dose to the dosimeter material is the result to be used in initial-phase dose assessment: when more than one fortuitous dosimeter is used, leading to a range of results due to their different locations and/or materials and/or techniques etc., harmonization is needed to crystallize a single 'dosimeter dose' from their disparate set of values in order to agree the outcome of the assessment.

The ever-increasing CPU power and abilities of Monte Carlo codes, the advent of GPU computing, and the development of advanced phantoms, including personalized or deformable phantoms that may be positioned in almost any posture of interest [142], can only enhance the potential of computational dosimetry and improve the speed with which dose assessment and mapping procedures may be accomplished. Together, these advances could vastly increase the rate at which different scenarios may be modelled, and hence greatly extend the variety of configurations that may feasibly be considered for RD. Finally, a

look at the future of research cannot ignore the role of the increasing opportunities offered by the field of Artificial Intelligence. For instance, deep learning and computer vision are increasingly being used in many sectors of dosimetry and radiation protection [143] and a few preliminary experiences in EPR and thermoluminescence dosimetry [144, 145]. From these future scenarios, it is clear that computation will become more and more relevant to experimental dosimetry. There will be an increasing need to learn skills in computational techniques and training will be an essential part of this development.

5. Conclusions

Building upon the wealth of research findings described in the scientific literature and the overall understanding of the current status, the authors have presented their views on aspects that merit further research, with an eye to effective use of resources. There is indeed a need to find a balance between the number of questions that should be answered to have an optimum system, and the scarcity of funding and resources, which is endogenous to emergency-addressed research. The presented conclusions are totally personal, by no means exhaustive and do not preclude further explorations.

One valuable aspect in designing a cost-effective strategy for research is to share and integrate results among laboratories. This can be achieved by promoting consolidation and standardization of the procedures, when possible through the laboratory networks. Hopefully the still-debated topics could find faster resolutions from collaborative works.

There has been a lively debate in the last years about the most appropriate use of RD, i.e., whether in small- or large-scale events, or for homogeneous or heterogeneous exposures. One answer should come from the development of time-effective protocols, able to provide responses in timescales far shorter than those possible at present.

There is a clear need to improve understanding of the various physical mechanisms that generate the signals in fortuitous dosimeters. The outcomes might explain some effects that are not currently understood, and could also form the basis for opening new applications of these physical techniques.

While looking at the future, one should also learn from the past: analysing the solutions found in previous accidents will avoid repetitions of errors and the following of routes that are not practically suitable in real events.

Finally, horizon scanning will always be critical. As the current retrospective dosimeters and techniques are developed, so too must the parallel task of computational modelling, with an eye to innovative or smart technology and Artificial Intelligence solutions.

Disclaimers

The views expressed in this article are those of the authors and do not necessarily reflect the official policy or position of the Department of the Navy, Department of Defense, nor the U. S. Government.

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CRediT authorship contribution statement

Paola Fattibene: Conceptualization, Writing - Draft preparation, Reviewing and Editing, Supervision. **Francois Trompier:** Conceptualization, Writing – Draft preparation, Reviewing and Editing. **Celine Bassinet:** Writing – Draft preparation on Interlaboratory comparisons, Reviewing and Editing. **Bartłomiej Ciesielski:** Writing – Draft preparation on EPR dosimetry with mineral glass, Reviewing and Editing. **Michael Discher:** Writing – Draft preparation on Luminescent dosimetry, Reviewing and Editing. **Jonathan Eakins:** Writing – Draft preparation on Computational dosimetry, Reviewing and Editing. **Chryzel Angelica B. Gonzales:** Writing - Draft preparation on EPR dosimetry with fingernail, Reviewing and Editing. **Christelle Huet:** Writing – Draft preparation on Computational dosimetry, Reviewing. **Alexander Romanyukha:** Conceptualization, Writing – Draft preparation on EPR dosimetry with tooth enamel, Reviewing and Editing. **Clemens Woda:** Writing – Draft preparation on Luminescent dosimetry, Reviewing and Editing. **Małgorzata Juniewicz:**Writing – Draft preparation on EPR dosimetry with mineral glass, Editing. **Hyoungtaek Kim:** Writing – Draft preparation on Luminescent dosimetry with filler in encapsulations. **Jungil Lee:** Writing – Draft preparation on Luminescent dosimetry with filler in encapsulations. **Agnieszka Marciniak:** Writing – Draft preparation on EPR dosimetry with mineral glass, Editing. **Sergey Sholom:** Writing – Draft preparation on Luminescent dosimetry with surface mounted devices. **Hiroshi Yasuda:** Writing - Reviewing and Editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Francois Trompier has patent #WO 2016/055315 A1 issued to International Patent. Paola Fattibene is serving in an editorial capacity for Radiation Measurements.

Data availability

No data was used for the research described in the article.

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