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REVIEW

Medical effects of internal contamination with actinides: further controversy on depleted uranium and radioactive warfare

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Abstract The Nuclear Age began in 1945 with testing in New Mexico, USA, and the subsequent bombings of Hiroshima and Nagasaki. Regardless of attempts to limit the development of nuclear weapons, the current world arsenal has reached the staggering dimensions and presents a significant concern for the biosphere and mankind. In an explosion of a nuclear weapon, over 400 radioactive isotopes are released into the biosphere, 40 of which pose potential dangers including iodine, cesium, alkaline earths, and actinides. The immediate health effects of nuclear explosions include thermal, mechanical, and acute radiation syndrome. Long-term effects include radioactive fallout, internal contamination, and long-term genotoxicity. The current controversial concern over depleted uranium's somatic and genetic toxicity is still a subject of worldwide sustained research. The host of data generated in the past decades has demonstrated conflicting findings, with the most recent evidence showing that its genotoxicity is greater than previously considered. Of particular concern are the osteotropic properties of uranium isotopes due to their final retention in the crystals of exchangeable and nonexchangeable bone as well as their proximity to pluripotent stem cells. Depleted uranium remains an unresolved issue in both warfare and the search for alternative energy sources.

Keywords Radioactive warfare · Depleted uranium · Organotropic radioisotopes · Nuclear proliferation · Internal contamination with radionuclides

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The immediate and long-term effects of exposure to radiation and radioactivity have been extensively studied in the past several decades. The development of the worldwide nuclear arsenal has been continuous since the Trinity Test in New Mexico in July 1945 [1]. The subsequent use of a uranium bomb over Hiroshima on August 6, 1945, killed over 80,000 people immediately with tens of thousands more killed by radiation exposure in the following months [2], and the plutonium bomb over the city of Nagasaki 3 days later resulted in an initial death toll of over 74,000 [3]. Albert Einstein described it as nuclear weapons changing everything except our way of thinking.

While those bombs were in the kiloton range, testing of nuclear weapons and delivery systems has unceasingly continued with the constant improvement of the lethal power until today's megaton range bombs and intercontinental delivery systems. The current nuclear arsenal has already reached beyond apocalyptic dimensions regardless of the Non-Proliferation Treaty (NPT) [4]. By 2014, Russia had approximately 2000 nuclear warheads and about 2000 tactical warheads with the intention to dismantle 3500 warheads not designated for use anymore. Great Britain's arsenal contained 120 strategic nuclear warheads, while the United States had 1600 strategic warheads with a total number of 4800 warheads, not to mention many other countries with nuclear programs.

In a nuclear explosion, over 400 radioactive isotopes are released into the biosphere, of which approximately 40 pose potential dangers to mankind [5]. Those with organotropic qualities and long half-lives present the danger of irreversible tissue damage or the induction of malignant alterations. Organospecific radioisotopes also present immediate danger to their natural target organ. In the event of an internal contamination, the most important hazard is plutonium, which is osteotropic. It is deposited in the

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nonexchangeable bone crystals, where it can cause irreparable chromosomal damage and aberrations, genotoxicity, malignant alterations, and cellular death [6]. It stays in the body for decades, continuously exposing the internal environment to radiation [7].

Another key radioactive isotope is cesium with 21 radioisotopes, the most important of which is cesium 137 (^{137}C), a product of nuclear fission that has been studied extensively as a significant component of radioactive fallout [8]. It is a metabolic homolog of potassium. Iodine is present in the form of 10 radioactive isotopes produced during a nuclear explosion. Iodine 131 (^{131}I) is the most significant because of its beta emission and half-life of 8 days. In the event of a nuclear explosion, iodine is a major cause of concern for internal hazard because of its volatility and ability to enter the body via inhalational pathways [9]. Strontium 90 (^{90}Sr) is a product of nuclear fission and is among the most hazardous radioisotopes for internal contamination. The routes of entry for strontium are predominantly ingestion and inhalation, with a rapid organotropic response when absorbed in the body through skin lesions. Uranium has three isotopes that are potential hazards of internal contamination (^{234}U, ^{235}U, ^{238}U), which are predominantly alpha emitters with long half-lives. Nuclear weapons are made of highly enriched uranium 235 (over 80 %) with the half-life of 7.04×10^8 years.

The immediate health effects of nuclear explosions include acute radiation syndrome and combined injury of blast, heat, and external radiation. Acute radiation syndrome (ARS) is a severe illness resulting from very high levels of radiation exposure during a short period of time with the immediate effects on the hematopoietic system and radiosensitive undifferentiated cells [10]. These are the primary targets along with the gastrointestinal system with the dominant symptoms of nausea, diarrhea and vomiting. The most resistant tissues are highly sensitive undifferentiated cellular population of the central nervous system, which, however, are also affected with the syndrome of early transient incapacitation [11]. In the event of a nuclear explosion, a majority of the casualties would suffer from combined radiation injury (CRI), referring to those with both conventional trauma and radiation injury [12].

Long-term effects of radiation as a result of radioactive fallout and internal contamination have been studied in both war scenarios and industrial accidents. They include both somatic and genetic adverse effects and can be found on the worldwide scale because of stratospheric and tropospheric deposition of radioactivity carried by the high winds around the globe. This was first described as the black rain following Hiroshima and Nagasaki. The thousands of studies on widespread contamination by radioactive fallout include current detection in the Mediterranean basin [13] and in the grasslands of Siberia [14] as a result

of Fukushima [15, 16]. This demonstrates the potential irreversible consequences of chronic internal radiation exposure on the genetic pool of earth's population.

These already formidable potential effects of internal contamination with radioactivity are further enhanced by the military use of RDDs (radiological dispersal device) or dirty bombs. RDD is any device that causes the purposeful dissemination of radioactive material without a nuclear detonation [17]. An example of this is the potential use of plutonium 239 (^{239}Pu), the most toxic substance known to man, which according to some sources is capable of causing genotoxic and malignant alterations to the entire population of the earth. According to some information, one pound of plutonium could cause 8 billion cancers [18]. Of particular interest for the modern and future radioactive battlefields is the potential terrorist use of RDDs.

A major current concern of the internal contamination is depleted uranium (DU), which remains poorly understood. Although its name suggests that it is less harmful than naturally occurring uranium, the lack of consensus among the thousands of studies on its effects on the biosphere raises questions. Uranium, which is a heavy pyrophoric metal and one of the primordial elements of the physical world, has 27 known isotopes, all of which are radioactive. The three most prominent isotopes (^{234}U, ^{235}U, ^{238}U) occur in nature, while ^{236}U is manmade. They have a highly ionizing capacity of alpha corpuscular disintegration, and their half-lives range from a few nanoseconds to billions of years [19]. Uranium 234, with a half-life of 2.44×10^5 years, is least frequently cited as a most important radiation hazard, yet is 17 times more radioactive than ^{235}U, a fact that was described over 60 years ago, but never taken as a serious radiation concern because it is difficult to detect and is dispersed during nuclear explosions with long-term effects [20].

Depleted uranium is largely a misnomer for the byproduct of the enrichment process of ^{235}U, a fission fuel of nuclear reactors and nuclear weapons [21]. It has no clear single physical identity. Depleted uranium is used for military purposes, as it is a very efficient material for the armor penetrators in tank battlefields. There are unforeseeable biosphere and adverse health consequences when it is released into the environment predominantly in the form of radioactive aerosols that are deposited in the internal environment of the organism mostly via respiratory pathways. Enrichment of ^{235}U is accomplished by separating ^{238}U, which has a low radiation hazard because of its long half-life, but is still an alpha emitter with high ionization capacity. Its radiotoxic risk is enhanced by its decay isotopes, such as thorium 234 (^{234}Th) and protactinium 234 (^{234}Pa), also an alpha emitter with the half-life of 2.47×10^5 years. In addition, depleted uranium contains other actinides, such as plutonium.

The ultimate concern of the life sciences is focused on the metabolic pathways and uranium effects in the target organs and the tissues of their final retention and incorporation. The effects of uranium on the human being are somatic [22] and genetic [23] with the capacity of alteration of cellular structure and function ranging from morphological through transgenerational genetic effects. Although the penetrating power of uranium in the tissue is low, usually not passing the thickness of the skin, once deposited in the vicinity of an undifferentiated cell population, it may exert a host of adverse morphological and functional consequences [24]. Previously, the predominant opinion was that uranium was not a potent carcinogenic agent [25]. However, more recent evidence demonstrates that its genotoxicity is greater than previously considered, specifically in the erythropoietic and reproductive systems, as well as its teratogenic effects with mutagenesis and chromosomal aberrations [26].

The osteotropic properties of uranium isotopes are of particular concern due to their final retention in the crystals of either exchangeable $CaHPO_4$ or nonexchangeable $3Ca_3(PO4)_2\ Ca(OH)_2$ bone, where they become an integral part of the bone structure, modeled by their prototype of fluoroapatite [27]. This is either by isoionic or heteroionic exchange or by the apposition to the surface of the crystals, where they are further buried by other elements of the internal environment. They alter the natural structure of the bone tissue by substituting other osteotropic elements such as strontium, uranium, radium, and other bone seeking nuclides. Once incorporated, they become a part of nonexchangeable bone in which they produce somatic and genetic alterations by both their corpuscular emissions and long half-lives. This bone retention provides a close environment for the interaction with highly undifferentiated and highly radiosensitive pluripotent stem cells, easily affected by the mutagenic and carcinogenic properties resulting in cellular death. In addition to osteotoxicity, some uranium isotopes are also highly nephrotoxic, as well as capable of producing myeloproliferative disorders in the hematopoietic system. Once incorporated in the bone minerals, they cannot be routinely removed except by the theoretical de-corporation mechanisms of osteoclastic activity, which is not of practical use as a therapeutic alternative.

The research data implications for biological and clinical manifestations, in particular regarding the carcinogenic effects of DU exposure in war veterans and civilians, are still expanding. Evidence from the recent battlefields in the Balkans demonstrates an increase in lymphomas among the peacekeeper and soldier veterans of Bosnia and Kosovo, particularly Hodgkin's lymphoma [28]. This agrees with a Croatian study on testicular germ cell cancer that concluded a higher incidence of testicular cancer in eastern Croatia as a possible result of the presence of depleted uranium [29]. However, other reports on leukemia and overall cancer incidence in Dutch [30] and Norwegian peacekeepers deployed in Kosovo between 1999 and 2011 have been inconclusive [31]. This evidence demonstrates the further need for studies of the effects of micro particles of DU in the human body [32].

Similarly, the data from Iraq reports mixed results, with the more recent evidence of the carcinogenic effects of DU on human beings clearly showing adverse effects. Contrary to earlier data regarding veterans of Gulf War I, genotoxicity and carcinogenesis was reported in 1998 at the International Atomic Conference with the genomic disease increase being significantly higher in the population of Iraq after internal contamination with uranium. Among the civilian population in Iraq, the overall incidence of lung and breast cancer, leukemia and lymphoma was recently found to be elevated in the Iraqi province of Mosul [33]. Likewise, a recent study in Fallujah, Iraq, on close to 5000 members of the exposed population suggests genetic damage and a higher risk of cancer, including leukemia in children [34]. This is further proven by the increased mutation frequency in the offspring of DU exposed fathers [35]. It was also found that depleted uranium causes chromosomal instability within cancerous lung epithelial cells [36] and in DU induced leukemia [37]. Low doses of depleted uranium may act as an important mediating agent in the mutagenic effects and death of microphages [38]. Also, studies recently published in China, referencing three consecutive ten-year periods (1980–2010), show that as a result of the Iran–Iraq war, incidences of leukemia in the Iraqi province of Ninawa have increased, but there are inconclusive results about overall cancer incidences [39]. In contrast, some studies suggest that long-term exposure to DU in Gulf War veterans did not cause chromosomal alterations [40]. While controversy remains, the literature review suggests that DU presents a long-term hazard as a major toxic and mutagenic agent with conclusive evidence of the elevated incidence of leukemia in the Balkans and sarcomas in Iraq. These findings indicate a need for further studies [41].

In addition to structural changes, uranium causes complex functional alterations [42]. Although the gastrointestinal pathway of contamination with actinides is negligible, it is quite dramatically opposite to the inhalational patterns with significant pathological changes of the respiratory system, including chronic obstructive pulmonary disease (COPD). The recent evidence of depressed immunological function and necrotic changes in the peritoneal macrophages as well as in the killer cells of the spleen with decreased function of the T cells is conclusive of the immunological alterations in rodents exposed to different doses of ingested uranium when compared to the

control group [43]. DNA strand breaks have been observed after exposure to uranyl acetate and ultraviolet radiation in the recent studies on macromolecules [44]. Further, studies of the renal mitochondrial function recently suggested both structural and functional adverse changes in the mitochondria of the rat kidney with damaged membrane integrity, uranium being the causative agent in this nephrotoxicity [45]. In addition, other cellular studies have recently determined that the toxicity of uranium depends on compartmentalization in the macromolecules, tissues and cells, and is directly related to the type of cell.

These cellular studies are enhanced by the current reports on embryological toxic effects of depleted uranium with the observation of anomalies and decreased weight of the embryological and fetal development as well as teratogenicity of DU of pregnant mice [46]. The radius of the particles and the total intake of uranium oxides impact the extent of the irradiated tissues [47]. The respiratory tract burden resulting from a prolonged inhalational exposure to aerosols of DU is still currently being evaluated [48]. DU contamination has also recently been associated with the substantial alterations and environmental hazards 30 years after the contamination with DU [49].

This is further verified by studies not previously documented in the experimental research of the effects of embedded uranium fragments in the multi-organ systems, with the conclusion that the renal and skeletal systems are adversely functionally changed after being imbedded with fragments and having chronic exposure to depleted uranium [50]. Recent studies in southwestern Iran on the effects of DU contamination from the military conflicts and exposure to the radioactive dust that originated in Iraq show, however, a lack of evidence of radiotoxicity [51]. A comprehensive article on exposure to DU by the inhalational pathway studied by the mass spectrometry at the DU testing site in New York did not show conclusive correlation between exposure to DU and adverse effects, but the results showed increased urinary excretion of uranium up to 20 years after the inhalational exposure [52]. A Russian article on uranium fragments embedded in the human tissues compared lead and depleted uranium, and suggested that depleted uranium may be less dangerous than lead because of the short range of alpha particles [53]. This report, however, neglected to compare penetrating power in the skin with mutagenic capacity in undifferentiated cells.

Notably, recent studies indicate that conventionally considered radioresistant tissues are also vulnerable to uranium exposure with the evidence of decreased brain function in uranium contaminated patients from the Gulf War. French studies suggest increased uranium concentration in the central nervous system of rats exposed to uranium inhalation [54]. Experimental evidence also demonstrates neurophysiological alterations in rats after only 2 weeks of exposure to 150 mg/L DU in drinking water [42]. Cellular radiobiology studies demonstrated myelotoxic changes in rodents with imbedded DU particles and oncogenic transformation of the osteoblasts exposed to DU. Long and comprehensive recent experimental evidence suggest mutagenic DNA effects and immune system toxicity [55]. Some of the conclusions of the abundant experimental evidence indicate a positive correlation between even a very small internal radiation exposure and both morphological and functional integrity compromised by the internal contamination with uranium isotopes, which warrants a sustained continuation of the research in the area of internal contamination with actinides [56].

Despite numerous and sometimes contradictory reports on the effects of depleted uranium in military conflicts, it can be safely concluded that DU is still far from being sufficiently and uniformly understood. However, it has been strongly suggested that it is a contributing factor for a number of illnesses, including leukemia, by the updated methodology including radiochemistry and alpha spectrometry [57]. This extends to the studies of perinatal mortality and birth defects in uranium contaminated populations, a largely under explored area of experimental and clinical research with a clearly warranted need of continuing studies on the etiological causes of subcellular, tissue and organic pathology in DU contaminated subjects. The consequences of the Gulf War internal contamination by DU suggests the correlation between DU environmental exposure and birth defects, enhanced by the contributing evidence from the Iraq Al-Anbar province, with over 11,000 cases of birth defects and stillbirths recorded in almost 3 years of observation (2000–2002) [58]. However, other studies in Iraq, the United Arab Emirates, Turkey, and Iran show no conclusive evidence of a correlation between environmental DU and birth defects in the areas of the Gulf War, although they have a higher rate of birth defects than the Western world.

Congenital anomalies as a consequence of DU have been extensively studied. There is a strong suggestion of the incidence of cancer in populations of the children of British Gulf War veterans and exposed female veterans through contamination by inhalational pathways by aerosolized particles of uranium oxides, corpuscular disintegration and long half-lives [59]. According to NATO sources, this is related to the testing of uranium weapons and is well correlated with genetic anomalies in southern Iraq. This is also critically evaluated in light of infant leukemia rates as a consequence of the Chernobyl fallout, as reported by the research studies in Germany, Scotland, Belarus, Greece and Wales [60]. The overall conclusions of the effects of DU exposure on the Gulf War and Balkan veterans appear to demonstrate the adverse health effects as

compared with the control population, including birth defects, leukemia [61] and a positive correlation of uranium effects in the DU exposed lung epithelial cells [62]. Regardless of the obvious risks, depleted uranium has become part of the standard operating procedures in the fields of warfare and energy production.

In contrast to the apocalyptic specter of World War III, there is undeniable potential benefit from peaceful use of nuclear energy for the sustained needs of the ever-contracting and ever-more energy demanding world. New initiatives searching for alternate sources of energy have been underway in the past two decades, including the CERN Project in Switzerland and other, yet undisclosed projects of utilization of nuclear fusion, which are still in the rudimentary phase of development. By the recent evidence of the Nuclear Atomic Energy Agency, this peaceful use of nuclear fuel sustains the industrial world, but it has created a legacy of adverse effects of nuclear waste. The nuclear power plant accident in Fukushima in 2011, and the one earlier in Chernobyl in 1986 most recently upset the balance of complacency.

The medical concerns of the Nuclear Age are not only focused on the immediate fatality from nuclear war. Long-term adversity for the biosphere is projected on the transgenerational scope, altering the very essence of life for eons of evolutional history of the Earth. This includes genotoxicity of the long-term radioactive fallout with modifications and mutations of the genetic code resulting in the totally altered present concepts of life. Regardless of the evidence of the adverse effects of radioactive waste over the last four decades, conclusive uniformity is still lacking and not always because of insufficient scientific evidence. As Arthur Schopenhauer wrote, "When a book hits a head and the hollow sound is heard, it is not always the book's fault" [63].

Uranium use in warfare and energy is an example of the compromise between the Nuclear Age and the Age of Reason. Solid scientific familiarity of the risk of exposure, etiology, pathogenesis, treatment and prevention of exposure to ionizing radiation can contribute to global awareness of new risks of living with the modern technology. While benefits of nuclear energy are known and undeniable, the effects of DU on the biosphere are far from being conclusive with many unresolved contradictions. All this data contributes to a challenging field still under investigation. This ongoing area of controversy reaches far beyond basic and applied science, extending to an arena further enhanced by the agenda of various political agencies and interest groups, not infrequently interfering with the objective science [64]. In conclusion, the term "depleted uranium" is a semantic attempt to reduce awareness of the significance of its hazard to the biosphere.

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