A case study on possible radiological contamination in the Lo Uttaro landfill site (Caserta, Italy)

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Abstract. The prevention of radioactive pollution is a primary objective of environmental protection codes in the operation of solid waste landfill activities. Several studies have demonstrated the effectiveness of radiological monitoring of the main constituents of urban landfill waste. The present contribution reports on an investigation plan carried out to evaluate the possible radiological contamination in the municipal landfill Lo Uttaro, district of Caserta (Italy). The investigation focused primarily on the perimeter area of the landfill in order to assess the possible impact on the surrounding population. The results of measurements of the equivalent dose rate along the perimeter of the landfill show average values lower than the population dose limit due to natural background radiation. Several samples of soil, groundwater and leachate representative of the subsoil of the study area were collected and the radionuclides were measured by gamma spectrometry. The results of these measurements show the absence of artificial radionuclides, except for small amounts of $^{137}$Cs due to nuclear disasters occurring in the last 50 years on Earth, and the mere presence of $^{40}$K and other natural radionuclides belonging to natural radioactive chains of $^{238}$U, $^{235}$U and $^{232}$Th.

1. Introduction

The natural terrestrial gamma radiation is an important contribution to the average equivalent dose rate received by the World’s population. In the year 2000, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) estimated the worldwide average of the equivalent dose rate due to the natural background gamma radiation to be 2.4 mSv/y, currently considered as limit for the population (http://www.unscear.org/docs/publications/2000/UNSCEAR_2000_Report_Vol1.pdf) [1,2]. Human natural radiation exposure can be due to either external sources from $^{40}$K, $^{235}$U, $^{238}$U and $^{232}$Th in soil or internal sources from inhalation of Radon and its daughters in dust and of fumes from waste disposal sites [3-7]. Estimation of this radiation distribution is useful to assess the health risk for the population and to document pollution of environmental radioactivity due to anthropogenic activities. In recent years, the interest in such radioactive pollution study coming from different typologies of urban disposal sites is increasing [3,8,9]. Several works have mainly shown the effectiveness of the monitoring the constituents of urban landfill solid wastes [10-13]. To provide information on any possible radiological risk that can arise from waste generation and disposal to human health and environment is one of the primary objective of environmental protection codes. The present paper describes an investigation plan carried out to evaluate, for the first time, the possible radiological contamination coming from the municipal landfill Lo Uttaro, in the district of Caserta (Italy). Measurements of radionuclide activity concentration in samples of different matrices and of the
equivalent dose rate along the landfill perimeter were performed using appropriate and sensitive devices. The work was performed by the ‘Environmental Radioactivity Laboratory’ of the University of Campania “Luigi Vanvitelli” able to carry out qualitative and quantitative analyses by high resolution and low-background γ-ray (as well as X-ray, α, β) spectroscopy [14-20], together with the Toma Abele Trivellazioni S.r.l. that realized the geognostic, geotechnical and environmental characterization services of the Lo Uttaro landfill area.

2. The monitored landfill site
The investigated landfill Lo Uttaro (41°02’53”N, 14°21’11”E) is situated in the homonymous locality in the Municipality of Caserta (Campania, Southern Italy), located in the south-east of the city. Fig. 1 shows the perimeter of the studied area of about 1770 m and with an area of about 2·10⁵ m². The municipal landfill is located within the ‘Litorale Domitio Flegreo and Agro Aversano’ area, identified by the Italian law 426/98: ‘Reclamation interventions and environmental restoration of polluted sites’. The present investigation is approved by Campania Region (Directorial Decree prot. 4557/QdV/DI/B). The work was carried out during the year 2015. Currently, the landfill is dismissed.

![Figure 1. Map of the Lo Uttaro landfill (Caserta, Italy), bordered by a thick black line. The gray numbers (n°) denote the points where 48 measurements of equivalent dose rate were carried out along the perimeter of landfill (Sec. 3). Full black and half black circles denote the geognostic techniques used to extract the samples representative of the area (Sec. 3).](image)

3. Materials and methods
The investigation focused primarily on the perimeter area of the landfill in order to assess the possible impact on the population living or working in the surroundings, through measurements of equivalent dose rate. 48 measurements were performed every 35 m along the perimeter (Fig. 1) by means of a
portable proportional counter LB 123 D-H10 (BERTHOLD) (Fig. 2). The measurement principle is based on the detection of the charge produced by the ionization of a gas molecule caused by the passage of the radioactive particles by means of a gaseous ionization detector. The charge is proportional to the incident radiation energy of the particles. The main technical specifications of the detector are: 50 mm diameter x 275 mm length; 0.46 kg weight; measuring range 50 nSv/h-10mSv/h; energy range 30 keV-1.3 MeV; intrinsic background <0.1 cps; calibration factor 0.214 μSv/h per cps (www.berthold-us.com/images/RadPro/Dose_Rate/LB123D.pdf).

The second part of the investigation focused on the spectrometric analysis of several soil samples collected at the landfill site, in order to characterize quantitatively the radioactivity of the area. Soil cores, of 0.1 m diameter, from the surface up to 35 m of depth, containing waste representative of the subsoil in the landfill area were collected. The cores have been placed in boxes with five 1 m long lodgings. The coring of the soil samples was carried out through driving, pushing and rotating a core drill in normal boreholes (20-100 cm) and in piezometers that are small-diameter boreholes (5-10 cm) with the lower end in contact with the groundwater (Fig. 1) [21]. The soil samples were analysed by a portable high resolution HPGe γ-ray spectrometer Trans-SPEC-DX-100T (ORTEC) (Fig. 2). The High-Purity Germanium spectrometer detects the energy spectra of γ-rays by capturing all energy of the incident γ-rays from photoelectric effect within the detector volume. The main technical specifications of the detector are: 65 mm diameter x 50 mm length; 11.1 kg weight; relative efficiency >40% up to in excess of 2 MeV; resolution ≤1.6 keV at 122 keV and ≤2.3 keV at 1332 keV. The detector is equipped with a cryostat with miniature Stirling-cycle cooler capable of ~1 W of heat lift at 100 K and draws less than 25 W when operating (www.ortec-online.com/-/media/ametekortec/brochures/trans-spec-dx-100t.pdf).

The third part of the investigation concerned the spectrometric analysis of several groundwater and leachate samples collected in boreholes and piezometers at the landfill area, in order to determine quantitatively the radioactive state of the area. 18 samples of groundwater and 4 samples of leachate were collected in high density polyethylene and glass containers. The samples were measured by a P-type coaxial HPGe high resolution γ-ray spectrometer (ORTEC) (Fig. 2), contained within a lead shield that ensure a very low background. The main technical specifications of the detector are: 82 mm diameter x 150 mm length; efficiency of 70%; resolution of 1 keV at 122 keV and 1.9 keV at 1332 keV. The detector is equipped with a standard liquid nitrogen (LN2) cooling Dewar of 30 l capacity (https://www.ortec-online.com/-/media/ametekortec/brochures/gem.pdf).

All the instruments are equipped with electronics to process signals produced by the detector with an associated amplifiers, and a data processor to generate, display, and store the results of the analysis.

4. Results

The results of the measurements of the equivalent dose rate, 10 min each, along the perimeter of the landfill are displayed in Fig. 3. This quantity expressed in Sv/h [J/kg/h=m²/s²/h] represents the effects and the damage caused by radiation on an organism. The equivalent dose rate values range between...
0.079 and 0.268 μSv/h (equivalent to 0.69 and 2.35 mSv/y), with a mean value and standard deviation of 0.162 ±0.052 μSv/h equivalent to 1.42 ±0.46 mSv/y. The mean value (as well as every single value) is lower than the population equivalent dose rate limit due to the natural background radiation of 2.4 mSv/y (Sec. 1).

Figure 3. Results of equivalent dose rate measurements (μSv/h) carried out in 48 points along the perimeter of the Lo Uttaro landfill site.

The γ-ray spectrometric measurements carried out on the soil core samples collected at the landfill at different depth were performed using a homemade structure, shielded with several lead blocks, to reduce the influence of surrounding environment radioactivity. Moreover, the measurements were performed at a fixed distance between the detector and the sample, in order to keep the same geometric efficiency. The measurement time was 10 h for each sample to obtain a sufficiently high counting statistic. An environmental background γ-ray spectrum was subtracted by each sample spectrum. The results of these measurements are shown in Fig. 4, where for all the 48 boxes, containing 5 core samples each, the composition of the radionuclides present, expressed as a measure of the detection rate of ionization events per second (rate of counts per second−cps), is reported. The results show the mere presence of natural radioisotopes, which are 40K and those of the natural radioactive chains of 238U, 235U and 232Th. Only 5 boxes (1,7,11,15,22), containing core samples from 1 m to 5 m, have small amounts of the 137Cs radionuclide of anthropogenic origin.

The γ-ray spectrometric measurements carried out on groundwater and leachate samples collected at the landfill site were performed using 2.5 l Marinelli beakers; these are containers having a specific geometry (with a hole in the middle) to fit the Germanium detector shape, in order to have a better detection efficiency. The measurement time was 24 h for each sample. The γ-ray background was always subtracted. The quantity of revealed radionuclides is expressed as the specific activity in terms of the mass units of the sample (Bq/kg). The results are reported in Fig. 5, where two box-plot graphs show the statistical distribution of the radionuclides present in the overall collection of the 18 groundwater samples and of the 4 leachate samples. The results show the mere presence of natural radionuclides, those from the 238U, 235U, 232Th chains and also 40K, and the presence of small amounts of 137Cs anthropogenic radionuclide (within the ranges of 0.13-1.25 Bq/kg in groundwater samples and of 0.6-0.65 Bq/kg in leachate samples). These values are in agreement with the results obtained on soil core samples and are largely lower than the general reference value of 1 Bq/g which allows us to consider the matrix not subject to radiation protection measures [1-3].

5. Discussion
The overall results obtained from the gamma spectrometric measurements of soil cores, groundwater and leachate show the presence of natural radionuclides coming from 40K and the natural radioactive chains of 238U, 235U, 232Th, which represent the main external source of irradiation to the human body. The different concentrations of these radionuclides in soil are determined by the radioactivity of the rock and also the nature of the process of the formation of the soils [22,23]. Therefore, radionuclides in soil generate a significant component of the background radiation exposure to the population [24]; for this reason a variability of the results of equivalent dose rate measurements performed around the
landfill perimeter has been found. The higher values of Thorium radioisotopes coming from the measurements results in groundwater and leachate samples, collected at the landfill site, are likely due to the fact that the Thorium radioisotopes exist in plus four valence state (Th$^{4+}$) and form complex ions that are very soluble [25]. Thorium radioisotopes can be leached from primary source rock under proper conditions of acidity (pH) and oxidation potential. The small amounts of $^{137}$Cs in some of the samples collected at the landfill survey area are due to atmospheric fallout from the nuclear disasters occurring in the last 50 years on Earth, as papers in literature give evidence [26-29].

Figure 4. The radionuclides composition (expressed in counts per second) recorded in the 48 boxes containing samples of soil cores, collected at the Lo Uttaro site, is shown using a stacked bar diagram. The range of the soil depth from which were collected the 5 parts of the soil cores is also reported. The grayscale of the legend denotes the different gamma emitting radionuclides.

Figure 5. Specific activity (Bq/kg) of radionuclides detected in the 18 groundwater (a) and 4 leachate (b) samples from the Lo Uttaro site.
6. Conclusions
The aim of this work was to assess the possible radiological contamination coming from the Lo Uttaro landfill (Caserta-Italy). The work addressed several types of measurements to provide information in different environmental areas and matrices: (i) the equivalent dose rate along the landfill perimeter, using a portable proportional counter detector; (ii) the gamma spectroscopy on several representative samples of soil cores, groundwater and leachate collected in boreholes and piezometers in the landfill area, using HPGe detectors. The analyses results showed: (i) a mean value of equivalent dose rate equal to 1.42 ±0.46 mSv/y, lower than the population equivalent dose rate limit due to the natural background radiation of 2.4 mSv/y; (ii) the presence of natural gamma emitting radioisotopes (from the natural radioactive chains of $^{238}$U, $^{235}$U, $^{232}$Th and also $^{40}$K) in the collected samples, with small amounts of the $^{137}$Cs anthropogenic radionuclide due to atmospheric fallout from nuclear disaster in the last 50 years. The survey evidences that there is no radioactive contamination at the Lo Uttaro landfill site and that there is no radiological impact in the area surrounding the perimeter of the site.

References
[1] Tso MY and Leung JK 2000 *Health Phys.* 8 555-78.
[2] Rani A and Singh S 2005 *Atmospheric Environ.* 39 6306-14.
[3] Usikalu MR, Ola OO, Achuka JA, Babarimisa IO and Ayara WA 2017 *J. Phys.: Conf. Ser.* 852 012045.
[4] Sabbarese C, Ambrosino F, De Cicco F, Pugliese M, Quarto M and Roca V 2017 *Radiat. Prot. Dosim.* 177(1-2) 202-6.
[5] La Verde G, Roca V, Sabbarese C, Ambrosino F and Pugliese M 2018 *Nuovo Cimento C* 41(6) 218.
[6] La Verde G, Roca V, Sabbarese C, Ambrosino F and Pugliese M 2018 *Nuovo Cimento C* 41(6) 219.
[7] Ambrosino F, Pugliese M, Roca V and Sabbarese C 2018 *Nuovo Cimento C* 41(6) 223.
[8] Bachama YD, Ahmed AL, Lawal KM and Arabi AS 2017 *J. Afr. Earth Sci.* 130 269-73.
[9] Obed RI, Farai IP and Jibiri NN 2005 *J. Radioanal. Prot.* 201 305-12.
[10] Avwiri GO, Nte FU and Olanrewaju AI 2001 *Scienitia Africana* 10(1) 46-57.
[11] Budihardjo MA, Noveandra K and Samadikun BP 2018 *J. Phys.: Conf. Ser.* 1022 012030.
[12] Irhammi, Pandia S, Purba E and Hasan W 2018 *J. Phys.: Conf. Ser.* 1116 042014.
[13] Poškas P, Kilda R, Šimonis A, Jouhara H and Poškas R 2019 *Sci. Total Environ.* 667 464-74.
[14] Ambrosino F, Ambrosino F, Buompane R, Pugliese M and Roca V 2017 *Appl. Radiat. Isot.* 122 180-5.
[15] Ambrosino F, Buompane R, Pugliese M, Roca V and Sabbarese C, 2018 *Nuovo Cimento C* 41(6) 222.
[16] Brocchieri J, Scialla E, Ambrosino F, Terrasi F and Sabbarese C 2018 *Nuovo Cimento C* 41(6) 224.
[17] Ambrosino F, De Cesare W, Roca V and Sabbarese C 2019 *J. Phys.: Conf. Ser.* 1226(1) 012025.
[18] Ambrosino F, Thínová L, Briestenský M and Sabbarese C 2019 *Geol. Geodyn.* 10(6) 455-9.
[19] Ambrosino F, Thínová L, Briestenský M and Sabbarese C 2019 *Radiat. Prot. Dosim.* nc245.
[20] Ambrosino F 2020 *Appl. Radiat. Isot.* 159 109090.
[21] MacEwan R, Dahlhaus P and Fawcett J 2012 *Hydropedology* ed H. Lin (Amsterdam: Elsevier) chapter 14 pp 449-81.
[22] Orabi H, Al-Shareaf A and El Galefi M 2006 *J. Radioanal. Nucl. Chem.* 269 99-102.
[23] Al-Jundia J, Al-Bataina BA, Abu-Rukah Y and Shehadeh HM 2003 *Radiat. Meas.* 36 555-60.
[24] Goddard CC 2002 *Health Phys.* 82 869-74.
[25] Lauria DC, Almeida RMR and Sracek O 2004 *Environ. Geol.* 41(1) 11-9.
[26] Steinhauser G and Saey PRJ 2016 *J. Radioanal. Nucl. Chem.* 307 1801-6.
[27] Sprem N, Bubic I, Barisic D and Barisic D 2013 *J. Radioanal. Nucl. Chem.* 298 513-7.
[28] Gulakova AV 2014 *J. Environ. Radioact.* 127 171-5.
[29] Pratama HA, Yoneda M, Shimada Y, Satoshi F and Ikegami M 2019 *J. Phys.: Conf. Ser.* 1198 022026.