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ORIGINAL ARTICLE

# Quantifying annual internal effective $^{137}$Cesium dose utilizing direct body-burden measurement and ecological dose modeling

Benjamin A. Jelin[1], Wenjie Sun[1], Alexandra Kravets[2], Maryna Naboka[3], Eugenia I. Stepanova[4], Vitaliy Y. Vdovenko[4], Wilfried J. Karmaus[5], Alex Lichosherstov[3] and Erik R. Svendsen[1]

The Chernobyl Nuclear Power Plant (CNPP) accident represents one of the most significant civilian releases of $^{137}$Cesium ($^{137}$Cs, radiocesium) in human history. In the Chernobyl-affected region, radiocesium is considered to be the greatest on-going environmental hazard to human health by radiobiologists and public health scientists. The goal of this study was to characterize dosimetric patterns and predictive factors for whole-body count (WBC)-derived radiocesium internal dose estimations in a CNPP-affected children's cohort, and cross-validate these estimations with a soil-based ecological dose estimation model. WBC data were used to estimate the internal effective dose using the International Commission on Radiological Protection (ICRP) 67 dose conversion coefficient for $^{137}$Cs and MONDAL Version 3.01 software. Geometric mean dose estimates from each model were compared utilizing paired *t*-tests and intra-class correlation coefficients. Additionally, we developed predictive models for WBC-derived dose estimation in order to determine the appropriateness of EMARC to estimate dose for this population. The two WBC-derived dose predictive models identified $^{137}$Cs soil concentration ($P < 0.0001$) as the strongest predictor of annual internal effective dose from radiocesium validating the use of the soil-based EMARC model. The geometric mean internal effective dose estimate of the EMARC model (0.183 mSv/y) was the highest followed by the ICRP 67 dose estimates (0.165 mSv/y) and the MONDAL model estimates (0.149 mSv/y). All three models yielded significantly different geometric mean dose ($P < 0.05$) estimates for this cohort when stratified by sex, age at time of exam and season of exam, except for the mean MONDAL and EMARC estimates for 15- and 16-year olds and mean ICRP and MONDAL estimates for children examined in Winter. Further prospective and retrospective radio-epidemiological studies utilizing refined WBC measurements and ecological model dose estimations, in conjunction with findings from animal toxicological studies, should help elucidate possible deterministic radiogenic health effects associated with chronic low-dose internal exposure to $^{137}$Cs.

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## INTRODUCTION

Radiocesium ($^{137}$Cesium, $^{137}$Cs) is a common fallout component of an atmospheric nuclear explosion,[1] a catastrophic failure at a nuclear power plant (such as the recent Fukushima Daiichi accident)[1] or an attack utilizing a radiological dispersal device.[2] Areas that are contaminated by $^{137}$Cesium fallout are of significant concern to governmental public health authorities because land-use and occupancy will likely need to be altered to mitigate risks to human health for many years. The Chernobyl Nuclear Power Plant (CNPP) accident represents one of the most significant civilian releases of $^{137}$Cs in human history. Environmental contamination from this event has and will continue to provide important insight into the refinement of risk assessments in the event of another such significant $^{137}$Cs release.

The CNPP accident occurred on 26 April 1986 and released 85 PBq of $^{137}$Cs into the Earth's atmosphere, which was deposited over an area greater than 200,000 km$^2$.[3] Approximately 71% of the total $^{137}$Cs released was deposited in the former Soviet countries of Belarus, the Russian Federation and Ukraine.[3] In the Chernobyl-affected region, $^{137}$Cs is considered to be the greatest on-going environmental hazard to human health by radiobiologists and public health scientists.[4,5]

Exposure to $^{137}$Cs, a long-lived gamma and beta emitter ($t_{1/2} = 30.2$ years), is of concern to human health because of its environmental persistence, biological availability in soil and food, and rapid incorporation into biological systems as a potassium congener.[3,6] Studies have shown that $^{137}$Cs deposited in soil and water can be transferred readily to edible plants and animals in contaminated areas.[4] Once ingested, $^{137}$Cs is spread homogeneously throughout human tissue and has an average biological half-life of 100 days.[7] Overall, ingestion of $^{137}$Cs leads to a relatively uniform absorbed equivalent dose to almost all tissues in the human body.[8]

In CNPP-contaminated areas where the population is dependent on locally grown food and forest products,$^{137}$Cs is considered by most experts to be the most significant driver of internal

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[1]Department of Global Environmental Health Sciences: Tulane University School of Public Health and Tropical Medicine, New Orleans, Louisiana, USA; [2]Institue of Cell Biology and Genetic Engineering, National Academy of Sciences, Kyiv, Ukraine; [3]Radioecological Center, Ukrainian National Academy of Sciences, Kyiv, Ukraine; [4]Research Center for Radiation Medicine, Academy of Medical Sciences of Ukraine, Kyiv, Ukraine and [5]Division of Epidemiology, Biostatistics and Environmental Health, School of Public Health, University of Memphis, Memphis, Tennessee, USA. Correspondence: BA Jelin, Tulane University School of Public Health and Tropical Medicine, Department of Global Environmental Health Sciences, 1440 Canal Street St. 2100, New Orleans, LA 70112, USA.
Tel.: +1 917 282 3184. Fax: +1 504 988 1726.
E-mail: bjelin@tulane.edu
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radiation dose.[9,10] Radioecological studies have shown that $^{137}$Cs in soil has exhibited an effective ecological half-life that is shorter than physical decay alone as the radionuclides have vertically migrated further into the soil.[3,11] However, the rapid decrease in $^{137}$Cs soil activity is not reflected in the rate at which $^{137}$Cs activities have decreased in animals and non-agricultural food products, such as mushrooms and berries.[5,11] These findings suggest that the vertically mobile fraction of $^{137}$Cs in soil resides in the rooting zone of plants[3] and fungi.[6,7,9] Radionuclide intake in $^{137}$Cs-contaminated areas, like those affected by the CNPP accident, can be exacerbated by poor or worsening socioeconomic situations because populations are forced to rely on such contaminated non-agricultural food products.[9]

There is significant ambiguity in the literature about the correlation between soil concentration and directly measured $^{137}$Cs body burden.[5,6,9,12,13] These inconsistent findings can be attributed to the heterogeneity of $^{137}$Cs behavior in contaminated soil[3,14] and/or the countermeasures used by the exposed population to reduce dietary exposure.[7] However, additional studies have shown that soil type, agricultural practice (including chemical and physical countermeasures) and human activity are better predictors of the behavior of $^{137}$Cs in the food chain, and, therefore, of human exposure.[14] Generally, the dose attributed to the ingestion of $^{137}$Cs in CNPP-affected regions is considered to be low, but exposure is ongoing and chronic.[8]

This study assessed a dynamic cohort of children living in the Narodichi area of Zhitomir Oblast, Ukraine that received significant $^{137}$Cs fallout from the CNPP accident and where contamination is still present. Whole-body counts (WBC) taken from this cohort, measuring individual subjects' body burden, were converted to dose estimates utilizing standardized dose coefficients developed by ICRP and MONDAL dose models.[15] The cohort's annual internal effective dose from $^{137}$Cs was also assessed through the soil-based ecological model for assessment of radiological consequences of agricultural lands contamination (EMARC), which takes into account the interaction of $^{137}$Cs with different soil types, as well as food chain interaction and contaminated food consumption. We investigated predictors of WBC-derived internal dose to see whether soil, the main determinant of dose estimation for the EMARC model, was able to predict the internal dose. Estimations from the WBC method and the EMARC method were then compared for agreement. The goal of this study was to characterize dosimetric patterns and predictive factors for WBC-derived $^{137}$Cs internal dose estimations in an exposed population, and compare these estimations with the soil-based EMARC dose estimation model. We hypothesize that (i) soil $^{137}$Cs contamination is a significant predictor of internal effective radiation dose and (ii) that ecological dose modeling will provide an appropriate means of assessing $^{137}$Cs exposure in this cohort.

## METHODS

### Study Population

The Narodichi region (Zhitomir Oblast, Ukraine), located approximately 80 km from the CNPP site, experienced significant fallout from the CNPP accident.[4] The population in this region experiences little migration and relies on homegrown food and forest products for subsistence.[7] As of 2008, there were 579 school-age children in this region, of whom 518 (95%) were healthy enough to participate in a cross-sectional study. This cohort included children of school age who were recruited with the permission of their parent to participate in a single health survey related to radiation exposure.[7] The 61 school age children who did not participate were acutely ill during the time of this study. All testing occurred at the Central Hospital of Narodichi during the 2008–2010 school years.[7]

### Whole-Body Counting (WBC)

Subjects were assessed utilizing a "SCRINNNER-3M" whole-body gamma-spectrometer equipped with a Pb collimator (Institute of Human Ecology, Kiev, Ukraine) with a thickness of 50 mm and a coaxial scintillation detector NaI(Tl) Ø150 × 100 mm. The spectrometer was incorporated into a standard chair that has lead shielding to reduce background radiation. The collimator thickness was 50 mm and the detector was a coaxial scintillation detector NaI(Tl) Ø150 × 100 mm. Calibration was accomplished using six taurpalin phantoms representing six different age groups, which were filled with dried peas of a known radionuclide content. The limit of detection for the "SCRINNNER-3M" on a 70 kg adult phantom for a 3-min measurement was 340 Bq, which allows for a maximum error of 30% (30% limit under the standards of the International Commission on Radiological Protection (ICRP)[7]). Children were measured for 3–5 min in order to reach the required 30% error limit. Body burdens were measured according to the recommendations of the Research Center of Radiation Medicine of the Academy of Medical Sciences of Ukraine, which are detailed elsewhere.[16,17]

### Annual Internal Effective Dose Calculation Utilizing WBC Method

All WBC measurements were converted into weight-normalized body-burden (Bq/kg) in order to use the internal effect dose by the ICRP dose conversion coefficient for $^{137}$Cs of 0.0025 mSv/y/Bq/kg.[15] Additionally, internal effective dose was assessed utilizing MONDAL Version 3.01 software (National Institute of Radiological Sciences, Japan) which incorporates non-weight-normalized body burden (Bq) and the age-dependent parameters dictated by ICRP publications.[15,18–20] The $^{137}$Cs mode of intake was set to chronic with a period of intake of 365 days with measurement occurring one day after last intake (f1 set to 1.0 based on ICRP 30 GI tract model[19]).

### Soil, Milk and Potato Activity

Average $^{137}$Cs soil activity for 2008 was obtained through a public report from the Ukrainian Ministry of Health based on a decay calculation initiated from soil measurements taken in 1992.[21] Milk and potato radiation activities for each affected region were determined annually through measurements made by the Ukrainian Ministry of Emergency Situations and Population Protection from the Consequences of the Chernobyl Catastrophe and the Ukrainian Ministry of Health according to Ukrainian Approved Emergency Order 27.07.2011 No. 764.

### EMARC: Mechanistic Ecological Model

This dosimetric model was developed according to the guidelines proposed by the National Commission of Radiation Protection of the Population and Ministry of Ukraine on Emergency Affairs and Protection of the Population from the Consequences of the Chernobyl Catastrophe in order to empirically quantify the ecological and societal factors that determine internal $^{137}$Cs exposure. The model first determines the rates of natural attenuation for different agricultural soil matrix compositions by empirically determining the rate of fixation and release of $^{137}$Cs, diffusion rate and $^{137}$Cs solubility. Secondly, the model accounts for changes in soil-to-plant transfer factors over time depending upon the $^{137}$Cs availability in rooting layers and solubility in the specific soil matrix type. Lastly, the model utilizes average age-dependent reference diets to estimate an individual's intake of $^{137}$Cs to determine an individual's internal contamination. Internal activity estimates are multiplied by the ICRP 72 age-dependent dose coefficients to estimate dose over a specific time interval.[20] In this study, we determined that the soil in the Narodichi region was relatively homogenous and, therefore, was classified as "sod-podzolic weakly clayed" (Figure 1).[22] Our study utilized $^{137}$Cs soil measurements from village of residence, which has been shown to be significantly correlated with WBC measurements in children.[23] Initial soil radioactivity was derived utilizing the radioactive decay function which was necessary in order to use the EMARC method. This derivation did not include soil matrix interaction which could lead to an underestimation of dose. All coefficients, equations and justifications are provided in detail elsewhere.[14]

### Statistical Analysis

The analysis done for this paper utilized Statistical Analysis Software (SAS 9.1, Cary, NC, USA) and IBM Statistical Package for Social Sciences (SPSS 18, Chicago, IL, USA). The MONDAL Version 3.01, ICRP 67 and EMARC dose data were compared using the paired *t*-test, simple correlation and Two-Way Mixed-Effect Intra-class Correlation Coefficient (ICC) in order to determine agreement. WBC derived dose data were determined to be log-normally distributed and, subsequently, log-transformed. A linear regression was performed on the log-transformed ICRP and MONDAL dose estimates including the predictive covariates of season and $^{137}$Cs activity

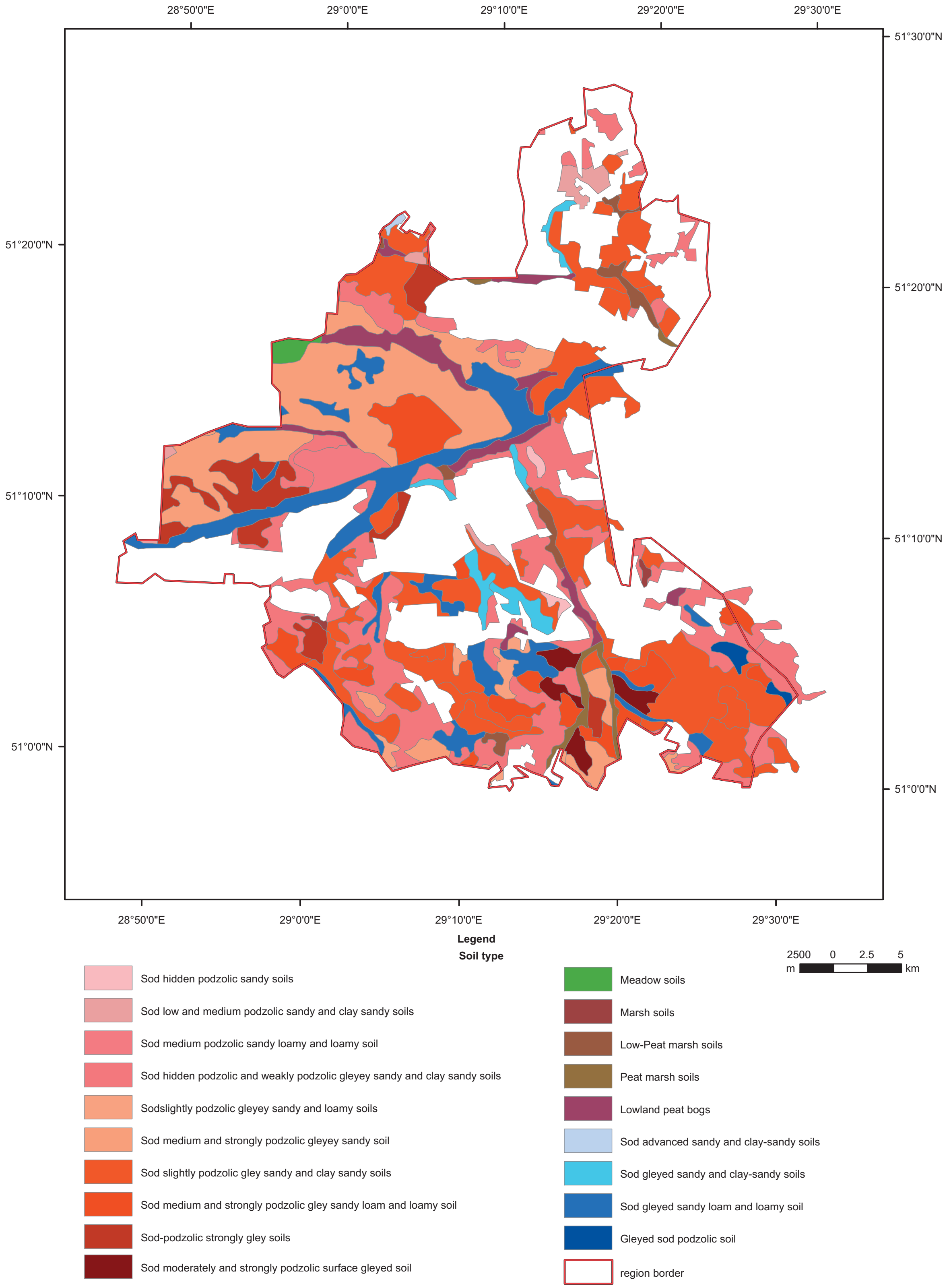

**Figure 1.** Narodichi Regional Soil Profile.

concentrations in soil, milk and potatoes. Season was analyzed by the seasonal categories Spring (March 1st–May 31st), Summer (June 1st–August 31st), Autumn (September 1st–November 30th), with Winter (December 1st–February 28th) as the reference season. Significant covariates identified in the predictive model determined the appropriateness of using the soil-based EMARC dosimetric model to assess the Narodichi cohort. ICRP and MONDAL-converted dose data were compared with EMARC using simple linear regression. The chosen statistical significance level was alpha = 0.05.

**Table 1.** Descriptive statistics and geometric mean annual internal effective $^{137}$Cs dose.

| | *Frequency* | *Percent* | *ICRP annual geometric mean $^{137}$Cs internal dose mSv/y (95% CI)* | *MONDAL annual geometric mean $^{137}$Cs internal dose mSv/y (95% CI)* | *EMARC annual geometric mean $^{137}$Cs internal dose mSv/y (95% CI)* |
|---|---|---|---|---|---|
| *Sex* | | | | | |
| Male[a,b,c] | 275 | 53.1% | 0.1651 (0.1470–0.1854) | 0.1507 (0.1349–0.1683) | 0.1717 (0.1604–0.1839) |
| Female[a,b,c] | 243 | 46.9% | 0.1647 (0.1467–0.1849) | 0.1479 (0.1324–0.1653) | 0.1958 (0.1826–0.2099) |
| *Age at time of exam* | | | | | |
| 8 years[a,b,c] | 20 | 3.9% | 0.2109 (0.1459–0.3048) | 0.1529 (0.1064–0.2120) | 0.1296 (0.1047–0.1605) |
| 9 years[a,b,c] | 23 | 4.4% | 0.1927 (0.1578–0.2353) | 0.1644 (0.1332–0.2029) | 0.1097 (0.0889–0.1354) |
| 10 years[a,c] | 21 | 4.1% | 0.1427 (0.0967–0.2092) | 0.1257 (0.0885–0.1786) | 0.1054 (0.0797–0.1393) |
| 11 years[a,b] | 51 | 9.8% | 0.2629 (0.2171–0.3185) | 0.2731 (0.2252–0.3312) | 0.1511 (0.1367–0.1670) |
| 12 years[a,b,c] | 65 | 12.5% | 0.2274 (0.1883–0.2746) | 0.1647 (0.1373–0.1975) | 0.1321 (0.1153–0.1512) |
| 13 years[a,b,c] | 75 | 14.5% | 0.1952 (0.1615–0.2361) | 0.1584 (0.1323–0.1898) | 0.2587 (0.2346–0.2853) |
| 14 years[a,b,c] | 78* | 15.1% | 0.1438 (0.1110–0.1862) | 0.1345 (0.1057–0.1711) | 0.2217 (0.1971–0.2495) |
| 15 years[a,b] | 86 | 16.6% | 0.1340 (0.1110–0.1620) | 0.1348 (0.1120–0.1623) | 0.2004 (0.1794–0.2238) |
| 16 years[a,b] | 80 | 15.4% | 0.1165 (0.0921–0.1473) | 0.1209 (0.0958–0.1526) | 0.1963 (0.1741–0.2214) |
| 17 years[a,b,c] | 19 | 3.7% | 0.1176 (0.0766–0.1807) | 0.1054 (0.0708–0.1569) | 0.2499 (0.2083–0.2998) |
| *Season of exam* | | | | | |
| Spring[a] | 257 | 49.6% | 0.2221 (0.2003–0.2462) | 0.1968 (0.1777–0.2179) | N/A |
| Summer[a] | 14 | 2.7% | 0.1205 (0.0650–0.2232) | 0.1000 (0.0526–0.1901) | N/A |
| Fall[a] | 191 | 36.9% | 0.1592 (0.1424–0.1780) | 0.1451 (0.1309–0.1609) | N/A |
| Winter | 56 | 10.8% | 0.0513 (0.0311–0.0849) | 0.0514 (0.0322–0.0822) | N/A |
| Total | 518** | 100% | 0.1649 (0.1519–0.1790) | 0.1494 (0.1381–0.1612) | 0.1826 (0.1738–0.1918) |

Abbreviation: N/A, not applicable. *$n = 77$ for EMARC because of non-reported data. **$n = 517$ for EMARC because of non-reported data. [a]Significantly different ICRP *vs* MONDAL across row ($P < 0.05$). [b]Significantly different ICRP *vs* EMARC across row ($P < 0.05$). [c]Significantly different MONDAL *vs* EMARC across row ($P < 0.05$).

### Human Subjects Protection

This cross-sectional study was approved by both the Committee on Bioethics of the Research Center for Radiation Medicine, Academy of Medical Sciences of Ukraine and the Office of Research Compliance at the University of South Carolina which received initial funding for this study. The study's human subject protection protocol was also approved by the Tulane University Human Research Protection Program.

## RESULTS

This study included 518 children from the Narodichi region, 53.1% were male and 46.9% female. The age distribution at the time of the medical exam can be seen in Table 1. Spring was the season with the highest participation (49.6%) followed by Fall (36.9%), Winter (10.8%) and Summer (2.7%) (Table 1).

The estimated $^{137}$Cs internal effective dose for the studied population was log-normally distributed across all three dose models. The geometric mean of the EMARC model estimate (0.183 mSv/y, 95% CI: 0.1738–0.1819 mSv/y) was the highest followed by the ICRP 67 dose estimates (0.165 mSv/y 95% CI: 0.1519–0.1790 mSv/y) and the MONDAL model estimates (0.149 mSv/y, 95% CI: 0.1381–0.11516 mSv/y) (Table 1). All three models yielded significantly different geometric mean dose ($P < 0.05$) estimates for this cohort when stratified by sex, age at time of exam and season of exam, except for the mean MONDAL and EMARC estimates for 15- and 16-year olds and mean ICRP and MONDAL estimates for children examined in the Winter (Table 1). None of the EMARC estimates were above the ICRP dose limit to the general public (1 mSv/y).[24] The ICRP 67 and MONDAL models yielded 1.9% and 2.1%, respectively, at or above the same ICRP prescribed limit of 1 mSv/y.[24] Estimates from the ICRP 67 and MONDAL models were well correlated ($r = 0.971$, $P < 0.0001$, Cronbach's Alpha = 0.985, ICC = 0.964) (Table 2), but a paired samples *t*-test showed that the means were significantly different ($t = 9.81$, $P < 0.0001$).

The log-transformed ICRP 67 (F = 5.27, $P < 0.0001$) (Table 2) and log-transformed MONDAL dose (F = 5.25, $P < 0.0001$; Tables 3 and 4) overall predictive models identified $^{137}$Cs soil concentration ($P < 0.0001$) as the strongest predictor of annual internal effective dose from $^{137}$Cs. Additionally, the ICRP 67 predictive model identified Spring WBC measurement as a significant predictor of annual dose (F = 4.14, $P = 0.04$), whereas the MONDAL did so marginally (F = 3.84, $P = 0.05$). Overall, the predictive models only explained approximately 8% of the total variability (ICRP 67: $r^2 = 0.078$ and MONDAL: $r^2 = 0.077$), showing that the models

**Table 2.** Measures of intraclass correlation between dose models.[a]

| *Model* | *ICRP 67 model* | *MONDAL model* | *EMARC model* |
|---|---|---|---|
| ICRP 67 model | 1.000 | 0.964 | 0.349 |
| MONDAL model | | 1.000 | 0.361 |
| EMARC model | | | 1.000 |

[a]This analysis was a Two-Way Mixed-Effect ICC focusing on Absolute Agreement.

**Table 3.** Predictive model results for log-transformed ICRP 67 data.

| *Parameter* | *Estimate* | *Standard error* | *t value* | *P value* |
|---|---|---|---|---|
| Intercept | − 1.13818 | 0.145287 | − 7.83 | < 0.0001 |
| Milk activity (Bq/l) | − 0.00054 | 0.000903 | − 0.6 | 0.5501 |
| Potato activity (Bq/kg) | − 0.07103 | 0.040643 | − 1.75 | 0.0813 |
| Soil activity 2008 (Bq/m$^2$) | 0.002751 | 0.000563 | 4.88 | < 0.0001 |
| Fall measurement | 0.162006 | 0.108269 | 1.5 | 0.1354 |
| Summer measurement | 0.01148 | 0.129954 | 0.09 | 0.9297 |
| Spring measurement | 0.188674 | 0.092778 | 2.03 | 0.0427 |

$r^2 = 0.078$. Winter was used as the dummy variable for this analysis.

**Table 4.** Predictive model results for log-transformed MONDAL data.

| *Parameter* | *Estimate* | *Standard error* | t *value* | P *value* |
|---|---|---|---|---|
| Intercept | − 1.2243 | 0.142887 | − 8.57 | < 0.0001 |
| Milk activity (Bq/l) | − 0.00017 | 0.000888 | − 0.2 | 0.8441 |
| Potato activity (Bq/kg) | − 0.06587 | 0.039972 | − 1.65 | 0.1002 |
| Soil activity 2008 (Bq/m$^2$) | 0.002637 | 0.000554 | 4.76 | < 0.0001 |
| Fall measurement | 0.190522 | 0.106481 | 1.79 | 0.0744 |
| Summer measurement | − 0.00757 | 0.127808 | − 0.06 | 0.9528 |
| Spring measurement | 0.178728 | 0.091246 | 1.96 | 0.0509 |

$r^2 = 0.077$. Winter was used as the dummy variable for this analysis.

did not capture a significant portion of the variability in the study population's internal exposure to $^{137}$Cs. However, the finding that proximal residential $^{137}$Cs soil activity is a significant predictor of internal effective dose validates the use of a soil concentration indicator or soil-based ecological model, like the EMARC model, to estimate annual dose for the study population because their food is often locally grown.

The mean EMARC dose estimate was significantly higher than both the ICRP ($t = 2.42$, $P = 0.02$) and MONDAL ($t = 5.23$, $P < 0.0001$) mean dose estimates. Log-transformed EMARC estimates were significantly correlated ($P < 0.0001$) with log-transformed dose estimates from ICRP ($r = 0.395$) and MONDAL ($r = 0.411$) models. Additionally, log-transformed EMARC dose estimates and log-transformed dose estimates from MONDAL and ICRP models showed fair correlation and absolute agreement (ICC = 0.361 and ICC = 0.349 respectively).

## DISCUSSION

Overall, the annual internal $^{137}$Cs effective dose estimates for the study population were low compared with the ICRP annual limit of 1 mSv/year to a member of the public.[24] Approximately 2% of the WBC-derived dose estimates from this study exceeded the ICRP limit, which is much greater than the approximately 0.01% found in a larger WBC study of the entire Zhitomir Oblast in 2008.[6] This divergence is most likely due to regional and sub-regional differences in diet composition, socioeconomic characteristics and proximal environmental contamination. Handl et al.[25] conducted a year-long modified total diet survey (1998–1999) of 13 residents of Christinovka, a town located in the Narodichi region, which measured total annual $^{137}$Cs intake and dose. Geometric means of our study's $^{137}$Cs dose estimations for the study population were lower, but in the same order of magnitude as the geometric mean found by Handl et al.[25] of 0.3 mSv/y from 10 years earlier. Lower dose estimations would be expected because of the physical decay and vertical migration of radionuclides in soil matrices over the difference in study periods.

The WBC method is generally considered among health physicists to be the "first priority" among those methods for internal $^{137}$Cs dose estimation.[26] Although, our results showed that ICRP 67 and MONDAL $^{137}$Cs internal effective dose estimates for members of the study population were strongly correlated ($r^2 = 0.971$), the means were significantly different ($P < 0.0001$). WBC methods provide a heavily assumptions-based cross-sectional dose approximation and differences in model assumptions and subject measurement times could account for this significant difference found between the methods in our study. The annual internal effective dose estimates derived from WBC measurements were significantly predicted by proximal soil $^{137}$Cs activity ($P < 0.0001$) and Spring WBC measurement ($P \approx 0.04$ utilizing ICRP 67), but the statistical model only explained approximately 8% of the total variability. The small amount of variability explained by our predictive model indicates that there are other significant covariates, most likely dietary predictors and individual nutritional status, which could capture more of the variability in the study population's WBC-derived dose. Studies, such as that of Likhtarev et al.,[10] have identified milk as the main contributor to internal $^{137}$Cs dose in CNPP-affected Oblasts, including Zhitomir. The assessment by Bouville et al.[5] of internal effective dose estimation methods also indicated that $^{137}$Cs milk activity is helpful in estimating whole-body doses over time and geography. Additionally, according to consumption data from 1995, milk is the most consumed foodstuff (1 kg fresh weight per day) that is significantly contaminated (54 Bq/kg fresh weight) in the Narodichi regional diet.[25] Milk and its consumption can, therefore, be reasonably assumed to be a significant contributor to internal $^{137}$Cs dose. However, we found that milk is not a significant predictor of internal effective dose in the study population (ICRP 67: $t = -0.60$, $P = 0.5501$, MONDAL: $t = -0.20$, $P = 0.8441$).

The findings of Travnikova et al.[27] in a population of inhabitants in Veprin (Bryansk, Russia) showed that milk was a reliable predictor of $^{137}$Cs body burden initially after the CNPP accident, but over time, changed to forest mushrooms and other wildly grown or harvested food products. Therefore, wild mushrooms in addition to other forest products, such as wild berries, represent possible additional $^{137}$Cs sources that could be significant predictors of WBC-derived $^{137}$Cs internal dose. According to Handl et al.,[25] even though mushrooms and berries represent a small proportion of the population's diet in the town of Christinovka (located within the Narodichi region), the consumption of these foodstuffs contribute 95% of the population's $^{137}$Cs internal effective dose.[25] It is likely that the lack of inclusion of these two forest products as covariates in our study's predictive model could explain a significant portion of the currently unexplained variability. The designation of mushrooms and berries as significant drivers and predictors of internal $^{137}$Cs dose is not universal in the literature. Consumption rates of mushrooms and berries have historically been low compared with other foodstuffs in CNPP-affected areas[5] and rates in the Narodichi area have been measured at 0.010 kg Fresh Weight per day and 0.011 kg Fresh Weight per day, respectively.[25] However, their significant contribution to internal $^{137}$Cs dose could be explained by their extremely high mean $^{137}$Cs activities; 2,600 Bq/kg fresh weight for berries and 200,000 Bq/kg fresh weight for mushrooms (measured in Christinovka between July and October 1998), which have been shown to be multiple orders of magnitude greater than activities measured in consumed milk.[25] Therefore, we recommend that these forest products be included in future dose estimation studies for this population.

The seasonal effect found in this study's WBC-derived dose predictive models was representative of the indeterminate statistical error generally seen in WBC measurement and dose estimation. As $^{137}$Cs body burden is dependent upon $^{137}$Cs intake, the WBC measurements will follow the patterns of consumption of $^{137}$Cs-contaminated food.[12] The predictive model for log-transformed WBC-derived dose identified Spring WBC measurement as a significant predictor of dose, which is inconsistent with the findings of similar studies of other populations in Zhitomir, as well as Oblasts in Belarus and the Russian Federation.[6,28] A study of Korosten City, Zhitomir (a lesser contaminated area compared with Narodichi) utilizing the same WBC method indicated that Autumn WBC measurements were significantly higher than other seasons.[6] The study by Seiketani et al.[28] of the Bryansk region (a more highly $^{137}$Cs contaminated region) also identified Autumn WBC as having the highest measured activities for most years.

Residents of the Narodichi region have tended to consume wild berries year round, whereas wild mushrooms are consumed during a defined 3–4-month period[25] in early Summer and Autumn (July–October) when fungal growth is optimal.[29] WBC measurements of this regional population in periods of higher mushroom consumption would therefore lead to higher WBC measurements during and immediately after these periods. Furthermore, another seasonal aspect of WBC-derived dose estimates for the children in this study is the two supplemented uncontaminated meals a day provided to the children during the school year.[7] WBC measurement of these children during the Spring (49.6%) could have led to an underestimation of the annual internal effective dose because these direct measurements would not capture the unsupplemented Summer months of the year or the increased consumption of contaminated wild mushrooms in early Summer and Autumn. Therefore, our finding of Spring WBC measurement as a significant predictor of dose and the season with the statistically highest dose is perplexing and unexpected. The unexplained variability is likely due to modulations in diet and activity that were not captured by this analysis.

The uncertainty of the WBC-derived $^{137}$Cs dose data and the relative ease of collection and availability of residential $^{137}$Cs soil activity data facilitated the use and comparison of an alternate ecological dose estimation model, EMARC. The mechanistic EMARC model was able to incorporate both of the significant covariates identified in the WBC-derived predictive model: residential $^{137}$Cs soil activity and seasonal changes in the consumption of contaminated food throughout the year. Our findings show a strong correlation between WBC methods (ICC = 0.964) and fair correlation between both WBC models and EMARC (ICC≈0.355) suggesting that the EMARC dose estimation model is incorporating more risk factors. This supposition is supported by our finding that residential soil is a significant predictor of annual $^{137}$Cs whole-body dose.

The log-transformed EMARC dose estimation model provides a more precise estimation of dose ($\mu_{\text{geo}} = 0.1826$ mSv/y, 95% CI: 0.1738–0.1819 mSv/y) when compared with both ICRP 67 ($\mu_{\text{geo}} = 0.1649$ mSv/y, 95% CI: 0.1519–0.1790 mSv/y) and MONDAL ($\mu_{\text{geo}} = 0.1494$ mSv/y, 95% CI: 0.1381–0.11516 mSv/y) dose models for the same population. Additionally, EMARC estimations of $^{137}$Cs internal effective dose for the study population, however, were significantly higher than estimates from WBC-derived dose estimates (MONDAL: $P < 0.001$, ICRP: $P = 0.0157$). This finding suggests that WBC-derived models may underestimate annual internal effective dose when compared with the EMARC for this population. In general, ecological models have been shown to produce higher dose estimates than direct WBC measurement because the ecological methods are not able to take into account countermeasures and food product avoidance.[10] Our findings show that the EMARC geometric mean is 11% greater than ICRP dose estimates and 22% greater than the MONDAL estimates for the study population. The higher EMARC dose estimations, however, are somewhat inconsistent with the findings of Kravets et al.,[14] which show the EMARC model produced $^{137}$Cs internal dose estimates that were 60–90% higher than WBC-derived dose estimations for study populations in the Kiev and Zhitomir regions during 2001. Kravets et al.[14] suggest that changes in economic consequences can lead to deviations from the reference diet utilized by the model, and, therefore, cause fluctuations in the agreement of these EMARC and WBC-derived estimates which could explain the difference.

Although there are significant differences in the estimated doses from each model used in this study, they are on the same order of magnitude and very low compared with health-effect-based regulatory standards for radiation exposure. In general, epidemiological monitoring programs in CNPP-affected regions have mainly focused on the cancer, as well as mortality.[8] There was a noticeable increase in the pediatric thyroid cancer incidence due to radioiodine exposure from the CNPP accident[6,8,30,31] owing to radioiodine's thyroid-seeking behavior and rapid deposition of radioactive energy in a relatively small organ.[31] Radioiodine exposure, however, occurred over a relatively short duration, owing to its extremely short half-life ($t_{1/2} = 8$ days).[30] For longer-lived radionuclides, like $^{137}$Cs, stochastic outcomes have rarely been seen in the literature because the disease state takes a long time to develop and occurrences are hard to attribute to a single exposure.[31] According to ICRP 60 and NCRP Report No. 115, the estimated increased lifetime risk for fatal and non-fatal cancer from a 1 mSv (ICRP dose limit) $^{137}$Cs whole-body dose (Quality Factor = 1) is 0.005% and 0.006%, respectively, for a member of the general public.[24,32] Most of the estimated doses from the study population were below ICRP dose limit, and therefore the added probability for stochastic outcomes would be significantly low. According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), "there has been no persuasive evidence of any other health effect [other than childhood thyroid cancer and leukemia in emergency workers] in the general population that can be attributed to radiation exposure."[31]

Therefore, relatively little is known about the non-cancer health effects of chronic low-dose $^{137}$Cs exposure as few studies on the subject are published in English.[4] According to Ukrainian and Russian studies' findings, indices of general morbidity by some classes of disease in children of Belarus living in territories with soil $^{137}$Cs activities of 555–1480 kBq/m$^2$ exceeded those of children living in "pure" territories and territories with soil activities of 185–555 kBq/m$^2$.[33–36] Further, some studies published in English have shown that long-term chronic $^{137}$Cs exposure is associated with immune and hematopoietic modulation as well as pulmonary malfunction and malformation in children living in CNPP-contaminated areas.[4,7,37–39] Additionally, some animal metabolomic studies and hormonal assays have shown that chronic $^{137}$Cs ingestion alters levels of plasma and urinary metabolites of steroidogenesis as well as certain circulating steroid concentrations,[40,41] which are critical to fetal and childhood development.[42] The epidemiological and radiobiological study of deterministic radiogenic morbidity for those exposed to chronic low-doses of $^{137}$Cs is ongoing and is expected to further elucidate the probable health consequences from such exposures.[31] We found that the EMARC methodology represents an epidemiologically suitable source of $^{137}$Cs dose data applicable for the study of $^{137}$Cs deterministic effects in the study population because the necessary inputs are readily available and the model allows for both retrospective and prospective dose estimation.

### Limitations

Our study has some limitations. Soil activity used in our predictive model was measured in 1992 and was forecasted to 2008 levels utilizing a simple decay scheme for $^{137}$Cs, which did not include the characteristics of its interaction with soil matrices and could alter the forecast results. Furthermore, the EMARC model requires $^{137}$Cs soil activity directly after deposition as an input in order to calculate internal dose. For our study, we utilized the 2008 soil activity data and did a simple reverse decay scheme to 1987 (considered directly after deposition by the EMARC model). This transformation also did not take into account $^{137}$Cs characteristic soil behavior; however, WBC doses estimations were in the same order of magnitude, suggesting this was an appropriate way to assess the study population's $^{137}$Cs dose. However, this study's inability to incorporate pre-1992 soil matrix interactions may also be responsible for the discrepancy found between the WBC and EMARC model estimations. This study was also limited by its cross-sectional nature, as WBC's were only taken once per subject during this study period. Multiple measurements over a multiyear period and across seasons would allow us to assess annual dose more longitudinally and compare it more effectively to EMARC estimations.

## CONCLUSION

Children living in the CNPP-affected areas have been and continue to be exposed to elevated environmentally persistent $^{137}$Cs levels at doses that are below regulatory health guidelines. However, the non-stochasic health effects from such an exposure have yet to be fully understood. Further prospective and retrospective radio-epidemiological studies utilizing refined WBC measurements and ecological model dose estimations, in conjunction with findings from animal toxicological studies, should help elucidate possible deterministic radiogenic health effects associated with chronic low-dose internal exposure to $^{137}$Cs.

## CONFLICT OF INTEREST

The authors declare no conflict of interest.

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