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Implementation of dose calculation methods for NORM by-products in building materials in the circular economy framework

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Introduction: Risk assessment of exposure to indoor pollutants plays an increasingly important role in human protection, and one of the main sources of indoor pollutants is building materials (BMs). In addition, production processes, including those related to BMs, are also involved in economic transition: the use of by-products from other industrial sectors as raw materials for the production processes in compliance with environmental sustainability is evaluated.

Methods: In this work, we evaluate not only the radiation protection of BMs but also the possibility of adopting the circular economy principles. The two main objectives of this study were 1) radiometric characterization and calculation of Index I of pozzolan from Altavilla Irpina (Avellino) in Italy, used as a natural igneous additive for concrete, using gamma spectroscopy, and 2) comparison of different methodologies for calculating the annual effective dose of BMs (CEN/TR 17113: 2017, RESRAD-BUILD software, and a previously developed experimental method). The same approach was extended to the possibility of reusing fly ash—a naturally occurring radioactive material (NORM) by-product of coal combustion in thermal power plants—for the production of concrete.

Results and Discussion: The study aligns with the principles linked to the circular economy to extend the life cycle of materials by reducing the need for natural resources, suggesting a possible positive compromise between radioprotection and preservation of environmental heritage.

KEYWORDS

radiation protection, circular economy, NORM residues, building materials, RESRAD-BUILD, environmental sustainability, indoor pollution, risk assessment

1 Introduction

Gamma emissions from building materials (BMs) are of particular interest for radiation protection since the interaction between ionizing radiation and biological tissues can produce harmful effects such as damage to the genetic material and, therefore, the occurrence of neoplastic diseases (UNSCEAR, 2000; BEIR VII, 2006; ICRP, 2007).

These biological effects and health impacts require the presence and continuous updating of international guidelines and reports (such as the International Commission on Radiological Protection [ICRP] reports), which are used as a scientific basis for drafting regulations.

Therefore, in Europe, a specific directive on exposure to ionizing radiation was published in 2013 (European Union, 2013), which was transposed in Italy, in 2020, with the Legislative Decree (D.lgs.) 101/20 (Repubblica Italiana, 2020).

These documents have changed the approach to radiation protection risk management both from a methodological point of view (such as the adoption of graded approaches) and from an operational point of view (such as screening tools that are useful for decision-making purposes and establishing reference levels) (Trevisi et al., 2023).

One of the areas where the legislation is applied is external exposure to BMs (art. 29; Repubblica Italiana, 2020). It is also important to remember that only since 1999, with the RP112 (European Commission, 1999), BMs became a part of the regulatory system for radiation protection.

The reference level for external exposure to indoor gamma radiation emitted by BMs, in addition to outdoor exposure, is set at 1 mSv/y for a representative person. The first approach to adopt is to use a screening tool, Index I (Annex II of Repubblica Italiana, 2020), that allows the identification of, in the first place, BMs from a radioprotection point of view. In the same Annex II, there is a further indication relating to the identification of BMs to be investigated. In fact, there is a list of BMs classified as 1) natural materials (alum shale and BMs or additives of natural igneous origin such as granitoids, porphyry, tuff, pozzolan, lava, and derivatives of zirconiferous sands) and 2) materials incorporating residues from naturally occurring radioactive material (NORM) industries (fly ash, phosphogypsum, phosphorus slag, tin slag, copper slag, red mud, and residues from steel production).

Considering this, knowledge of the radiological characteristics of these materials is applied to develop high-performance methodologies for the calculation and evaluation of the exposure dose.

This work, therefore, has several aims:

- To determine the radiometric measurements of pozzolan using gamma spectrometry. Pozzolan is used not as a BM but as an additive of natural igneous origin, as reported in Annex II (Repubblica Italiana, 2020);
- To perform a comparative analysis of different methods for determining the annual effective dose using tools such as the technical report CEN/TR 17113:2017 (European Committee For Standardization, 2017), RESRAD-BUILD software (Yu C. et al., 2022), and an experimentally developed method (Nuccetelli et al., 2015), in particular through an analysis of the application of pozzolan as an additive in BMs. Simultaneously, a study was conducted on fly ash, a residue resulting from coal combustion in a thermoelectric power plant, a NORM industry (Labrincha et al., 2017).

Investigating the potential reuse of these types of residues as secondary raw materials for the composition of BMs, and more, allows for comprehensive radioprotection monitoring and extends the material's lifecycle in alignment with the principles of the circular economy (CE) (British Standards Institution, 2017; Kirchherr et al., 2017).

In the past, the industrial production process was organized following a linear model based on "take, make, and dispose" (Ghisellini et al., 2016; British Standards Institution, 2017; Kirchherr et al., 2017). However, today, Europe is increasingly engaged in implementing measures that promote CE (European Commission, 2022). The benefits of the linear model are evident both in the economic sphere (e.g., reduction of raw material costs and creation of new industrial markets for waste treatment and/or reuse) and, more importantly, in the environmental and, consequently, public health domains.

A correct treatment and reuse of by-products or residues based on their characteristics have contributed to the widespread use of the CE concept in various industrial sectors, including the NORM industries (Liden et al., 2018; Andavan and Pagadala, 2020; Bituh et al., 2021; Oliveira et al., 2023) and the BM industry (Nuccetelli et al., 2017b; Labrincha et al., 2017; Nasir et al., 2017; Schroeyers et al., 2018; Sanjuán, 2022).

Therefore, this work also aims to assess the feasibility of using residues from NORM industries (such as fly ash) as a replacement for natural inert additives like pozzolan in concrete production. Moreover, in line with the CE principles, this would reduce the environmental impact in terms of landscape preservation and the economic burden associated with pozzolan extraction.

2 Materials and methods

2.1 Sampling

The Campania region (southern Italy) is characterized by a diversified geology mainly because of the volcanic origin of its soils and formations from different geological eras (Guarino et al., 2022). This results in a different distribution of the natural radioactivity content in the soils (Ambrosino et al., 2023). For radiometric research and characterization activities, pozzolan from Altavilla Irpina (Avellino) was sampled.

For sample preparation, UNI EN ISO 18589-2:2015 (Measurement of radioactivity in the environment—soil—Part 2: Guidance for the selection of the sampling strategy, sampling, and pre-treatment of samples) (ISO 18589-2:2015) was applied as this protocol guarantees homogeneity and uniformity of the samples. The samples were sieved, oven-dried (DIGITRONIC Selecta 2005141) at 105°C for 2 h, and subsequently homogenized to form a powder. The final product was weighed and sealed in a Marinelli beaker for 4 weeks to allow Ra-226 and its daughters to reach secular equilibrium. More details are reported in La Verde et al. (2020, 2021a). The quantity of analyzed samples was sufficient to ensure statistical significance.

2.2 Gamma spectrometry

The samples were measured by high-resolution gamma-ray spectrometry with a coaxial high-purity germanium (HPGe ORTEC®) detector (model GMX-45P4ST). The characteristics of the detector are described in detail in La Verde et al. (2021b). The energy performance of the detector is defined by a relative efficiency of 48% and an energy resolution, measured as full width at half maximum (FWHM), of 2.16 keV at 1.33 MeV. The minimum detectable activity (MDA) of the system was estimated with 95% confidence level (Currie, 1968).

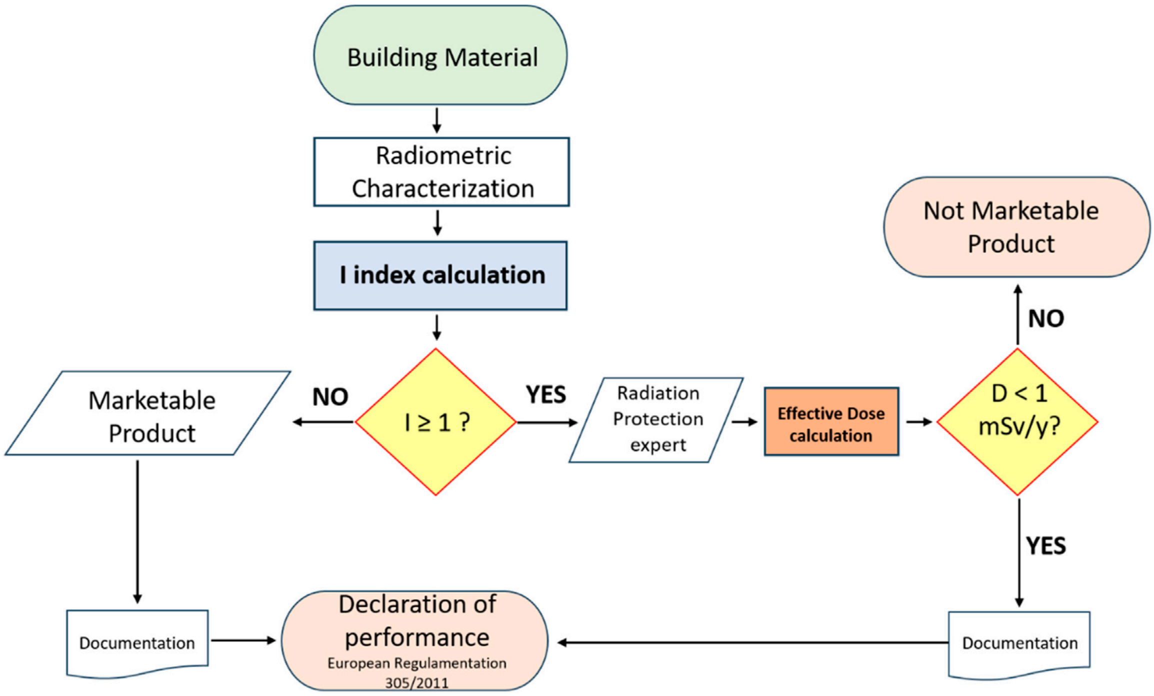

FIGURE 1
Risk management in radiation protection for BMs.

The spectra were acquired by Ortec DSPEC-LF unit plus MCA Emulator software and analyzed with GammaVision Spectrum Analysis software.

The background spectra were also acquired and subtracted to provide the measure without the additional background found. In order to collect enough counts for a significant statistic, approximately 172,800 s (i.e., 48 h) of counting time was set for samples and 259,200 s (i.e., 72 h) for the background.

The gamma-ray spectra of each sample were analyzed, considering transition energies of interest from the U-238 and Th-232 decay chains, and also from K-40.

2.3 Index I

In this work, Index I reported in Annex II of Repubblica Italiana (2020) was considered and is defined by Eq. 1:

$$I = \frac{C_{Ra-226}}{300} + \frac{C_{Th-232}}{200} + \frac{C_{K-40}}{3000}, \tag{1}$$

where C_{Ra-226}, C_{Th-232}, and C_{K-40} are the activity concentrations of the corresponding radionuclides (in Bq/kg) in the BM.

For more information on the parameter values in Eq. 1, i.e., 300, 200, and 3,000, see Markkanen (1995).

Products with Index I values less than or equal to 1 ($I \leq 1$) can be freely used; for products with Index I values greater than 1 ($I > 1$), a more accurate gamma dose estimate could be determined; and products with Index I values exceeding the reference level (1 mSv/y) may not be used for civil engineering buildings, such as dwellings and buildings with a high occupancy factor.

Risk management follows the flow chart shown in Figure 1.

In specific cases, e.g., when the product of the density (ρ) and thickness (d) of the BMs is greater than 470 kg/m^2, Index I needs to be remodulated, as demonstrated in Nuccetelli et al. (2015).

2.4 Dose assessment

Three different methods were used to calculate the annual effective dose (D) for a representative person:

1. Technical Report CEN/TR 17113:2017 described in European Committee for Standardization (2017), where a room model has the dimension 4 m × 3 m × 2.5 m;
2. RESRAD-BUILD (Yu C. et al., 2022); and
3. The ISS method described in Nuccetelli et al. (2015) and elaborated with the ISS room model of dimension 5 m × 4 m × 2.8 m.

The occupancy time was supposed to be 7,000 h per year (corresponding to an occupancy factor of 0.8). For dose assessment, two other parameters are required: the density (ρ) of the BM and thickness (d) of the material used. For each ideal concrete, the density is equal to ρ = 2,400 kg/m^3, and its use in a reference room configuration with thickness equal to d = 0.2 m was considered.

As described in Pepin (2018), the method used in the CEN technical report for calculating the external dose from BMs is based on the approach applied in RP112 (European Commission, 1999). It consists of a point-kernel method that uses the Berger approximation for the build-up factors (Pepin, 2018). This approach is similar to the one used in RESRAD-BUILD to calculate the external dose.

The RESRAD-BUILD code is part of the RESRAD family of codes developed at the Argonne National Laboratory (USA) to

TABLE 1 Inputs used in the calculations with RESRAD-BUILD.

Exposure duration (days)	Occupancy factor	DCF	Room dimension (m)	ρ [kg/m^3]	d [m]
365	0.8	ICRP 144, adult	4 × 3 × 2.5	2,400	0.2

TABLE 2 Mean activity concentrations of Ra-226, Th-232, and K-40 in pozzolan sourced from Altavilla Irpina (Avellino), determined using gamma spectrometry for five different samples.

Ra-226 [Bq/kg]	Th-232 [Bq/kg]	K-40 [Bq/kg]
61 ± 3	69 ± 3	1,117 ± 55

analyze potential radiation exposure of human and non-human biota resulting from environmental contamination of residual radioactive materials. In particular, RESRAD-BUILD has been designed to evaluate the exposure dose of an individual who works or lives in a building contaminated with radioactive materials (Yu C. et al., 2022).

The CEN method and ISS method are based on two different room models, but as already shown in Risica et al. (2001), the room dimension does not have a significant impact on the exposure. In this work, RESRAD-BUILD software was used with the room model of the CEN guide.

The calculation of external exposure dose by RESRAD-BUILD allows the user to use different dose conversion factors (DCFs). In this work, the most recent DCFs updated by ICRP 144 (ICRP, 2020) were used, and additional information is provided in detail in Venoso et al. (2024). The exposure dose for the adult individual in the population has been taken into consideration. Table 1 presents various inputs employed in RESRAD-BUILD ; all other pathways (ingestion and breathing) were set to 0, as only external exposure was exclusively considered.

3 Results

Five samples of pozzolan were analyzed through gamma spectrometry. The mean activity concentrations of Ra-226, Th-232, and K-40 are illustrated in Table 2.

In this study, an "ideal" concrete, obtained by following the ACI Standard 211.1-91 (American Concrete Institute, 2009), was considered. The ACI standard defines concrete as a composite consisting of the following mass percentages: 15% cement, 14% water, 26% sand, and 45% gravel.

Based on the nature of the inert components, cement is further classified according to the UNI EN 197-1: 2011 standard (UNI EN 197-1, 2011) into Portland cement (CEM I), composite Portland cement (CEM II), blast furnace cement (CEM III), pozzolanic cement (CEM IV), and composite cement (CEM V). It can be observed that pozzolan and fly ash can be employed as a component in CEM II (mass percentages ranging from 6% to 35%), CEM IV (mass percentages ranging from 11% to 55%), and CEM V (mass percentages ranging from 18% to 49%). Therefore, it is possible to use mass concentrations of pozzolan or fly ash ranging from 6% to 55% in cement. The investigation focused on cement, considering different concentrations of pozzolan and fly ash within the aforementioned range (6%–55%). The remaining part of cement consists of clinker.

For the study of an ideal concrete, with respect to the characteristics of individual components, literature information was used, except for one type of pozzolan, which was sampled and experimentally measured.

Therefore, the final configurations are as follows: 1) concrete composed of two types of pozzolan (natural material), one of which was sampled from Altavilla Irpina in Campania, and 2) concrete composed of two types of fly ash (NORM residue).

For the calculation, it is necessary to know the radiometric data of concrete, in addition to the mass percentage of each component of the concrete. For pozzolan, literature data from the Vesuvius volcano (Sabbarese et al., 2021) and radiometric data obtained experimentally in this work, through the measurement of Ra-226, Th-232, and K-40, were used. For fly ash, literature data were used: one relating to a low radiological content and in use in Italy (Nuccetelli et al., 2017a) and one with a high radiological content from Brazil (Flues et al., 2007).

Index I is usually applied to BMs and not to its components; however, in this work, Index I was also calculated for the individual components exclusively to give an idea of the radiological content in these matrices. Table 3 presents the radiometric data with Index I for the pozzolans and fly ashes used.

For the activity concentrations of the other components, the weighted averages of the values reported in the ISTISAN Report 17/36 (Nuccetelli et al., 2017a) in the Italy section were considered. Water has a non-zero but low radionuclide content that does not have an impact on dose assessment; therefore, activity concentration values were set at 0.

The following four scenarios were considered:

- an ideal concrete consisting of different percentages of pozzolan from Altavilla Irpina (I = 0.92);
- an ideal concrete consisting of different percentages of pozzolan from the Vesuvius volcano (I = 9.90) (Sabbarese et al., 2021);
- an ideal concrete consisting of by-products from NORM industries, in particular, different percentages of fly ash from Italy (I = 1.40) (Nuccetelli et al., 2017a);
- an ideal concrete consisting of different percentages of fly ash from Brazil (I = 10.65) (Flues et al., 2007).

Total D is the result of the dose contribution from each component based on the mass percentage of each component. Therefore, for each of the four concrete configurations, mass percentage ranges within the minimum and maximum range of use of the inert component, i.e., pozzolan or fly ash, were considered. An additional 10% was added to the maximum percentage for a more comprehensive radioprotection assessment.

The calculations assume that the population-weighted values have an average absorbed dose rate in air outdoors from terrestrial gamma radiation of 50 nGy/h (European Commission, 1999). Considering a conversion factor of 0.7 Sv/Gy and an occupancy time of 7,000 h, it

TABLE 3 Activity concentration and Index I in pozzolans and fly ashes used.

Sample	Ra-226 [Bq/kg]	Th-232 [Bq/kg]	K-40 [Bq/kg]	Index I
Pozzolan from Altavilla Irpina (experimental)	61	69	1,117	0.92
Pozzolan from the Vesuvius volcano (Sabbarese et al., 2021)	713	1,048	6,846	9.90
Fly ash from Italy (Nuccetelli et al., 2017a)	170	140	400	1.40
Fly ash from Brazil (Flues et al., 2007)	3024	73	621	10.65

TABLE 4 Index I as mass percentages of pozzolan/fly ash in cement varies for the four study cases.

Percentage of pozzolan/fly ash (%)	Index I concrete_% pozzolan from Altavilla Irpina	Index I concrete_% pozzolan from Vesuvius volcano	Index I concrete_% fly ash from Italy	Index I concrete_% fly ash from Brazil
#1_6	0.28	0.36	0.28	0.37
#2_20	0.28	0.55	0.30	0.58
#3_35	0.29	0.76	0.32	0.80
#4_45	0.30	0.90	0.33	0.95
#5_55	0.30	1.04	0.34	1.10
#6_65	0.30	1.18	0.35	1.25

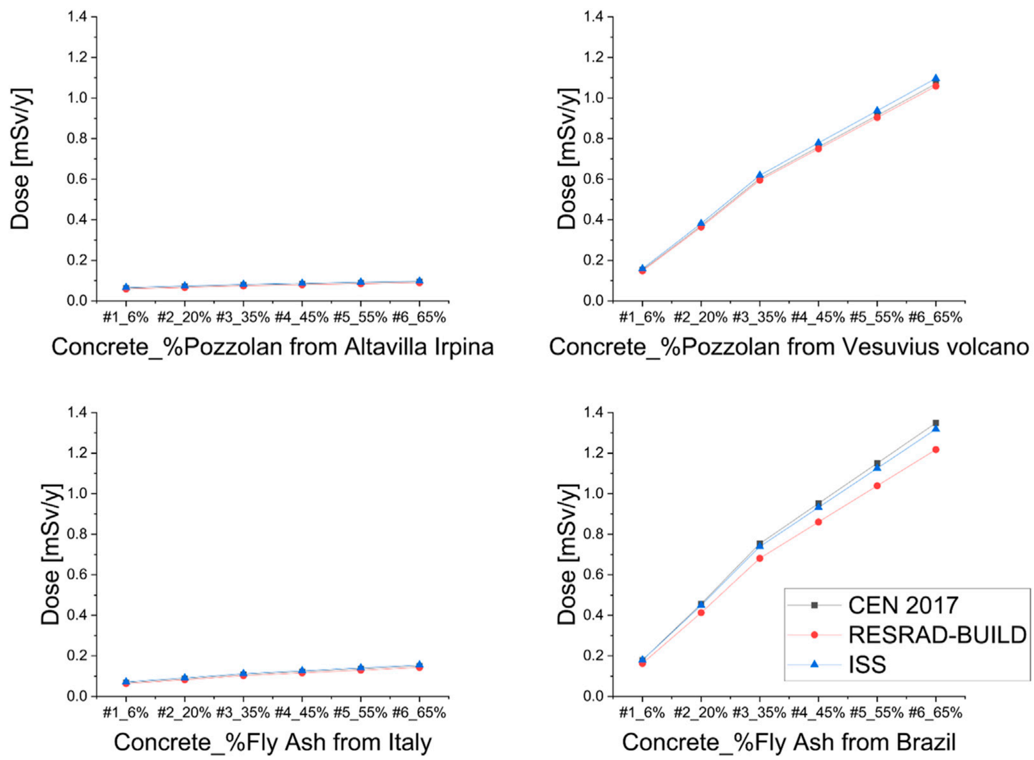

FIGURE 2
Dose as mass percentages of pozzolan/fly ash in cement vary for the four study cases.

corresponds to a background dose rate of 0.25 mSv/y. The Directive 2013/59/EURATOM (European Union, 2013) establishes 1 mSv/y as the reference value, in addition to the natural background dose (e.g., 0.25 mSv/y). Consequently, the total annual dose indoors, comprising contributions from BMs and background radiation, may exceed 1 mSv, reaching, for example, 1.25 mSv. RP 112 (European Commission, 1999) adopts the perspective that BMs shield against the entirety of natural background radiation. To account for this, the total exposure induced by the building is calculated; subsequently, the exposure resulting from terrestrial background gamma radiation is subtracted from this total, yielding the excess exposure.

In addition to calculating D using the three different methods, Index I was also computed across various configurations, and the results are shown in Table 4.

Figure 2 shows D calculation results obtained using the three different implemented models as the mass percentages of pozzolan/fly ash vary in cement. Detailed data are provided in Supplementary Tables S1–S4.

4 Discussion

The radiometric results obtained through gamma spectrometry for pozzolan from Altavilla Irpina do not raise specific radiological concerns, as it exhibits radiological content significantly lower than that of other pozzolans (Sabbarese et al., 2021).

Figure 2 shows that the estimates of D obtained by the three different methods are perfectly overlapping, except in the fourth scenario where RESRAD-BUILD, implemented with the most recent DCFs (ICRP 144), takes a less conservative approach. It should be noted, however, that since RESRAD-BUILD was employed with the latest DCF, it also represents the most accurate estimation.

It can be observed that Index I serves as an excellent screening tool: in all four scenarios, when the Index I value is below 1, a corresponding dose lower than 1 mSv/year is consistently observed.

From Figure 2, it can be observed that for low-radioactivity pozzolan and fly ash, any percentage of inert material ensures radioprotection for individuals in the population. This holds true even for mass percentages of up to 65% of inert material, representing an excess compared to the standard formulation (UNI EN 197-1, 2011). When comparing the results obtained, it is essential to consider the different radiological contents in terms of Index I of the constituents, particularly the lower values that differ by 52% (pozzolan I = 0.92 and fly ash I = 1.40). However, these matrices represent values that are likely available on the market; therefore, the assessment pertains to the possibility of substituting fly ash with pozzolan while maintaining equivalent mechanical performance.

Contrary to the results observed previously, in the case of high-radioactivity pozzolan and fly ash, it is evident that the choice of the mass percentage of pozzolan and fly ash can determine the marketability of the resulting concrete. For high Index I values of the constituents, the mass percentage difference is 8% (pozzolan I = 9.90 and fly ash I = 10.65), and this is evident also in the dose results (Figure 2).

In conclusion, the results from pozzolan/fly ash assessments encourage the use of NORM by-products in concrete preparation from both radiation and socioeconomic points of view since the approach embodies the principles of the circular economy, i.e., to create materials that can guarantee sustainability and efficiency standards while saving on the exploitation of the territory's natural resources. While not fully adhering to one of the principles of radioprotection, i.e., dose limitation, the use of NORM by-products nonetheless represents a promising compromise between radioprotection and conservation in future natural resource management.

It is always necessary to pay particular attention to mass percentages while using fly ash with a high radiological content and, consequently, a high Index I. In this regard, Index I is usually applied to BMs and not to their components; however, calculating Index I for individual components can provide an idea of the radiological content in these matrices.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material; further inquiries can be directed to the corresponding author.

Author contributions

GL: Conceptualization, data curation, investigation, methodology, visualization, and writing–original draft, review, and editing. GG: Formal analysis, investigation, methodology, software, visualization, and writing–original draft, review, and editing. FA: Writing–review and editing. MP: Conceptualization, funding acquisition, project administration, resources, supervision, validation, and writing–review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fbuil.2024.1385680/full#supplementary-material

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