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Evaluating radon loss from water during storage in standard PET, bio-based PET, and PLA bottles

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HIGHLIGHTS

- Radon loss from water during storage in polyethylene terephthalate (PET) and polylactic acid (PLA) bottles was evaluated.
- Surface/volume ratio and thickness of plastic materials were studied.
- A correction for dissolved radium concentration was applied to estimate gas loss.
- Proper corrections for degassing efficiency of aerators were developed.
- The interference of H_2O on radon daughter electrostatic collection was quantified.

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ABSTRACT

Polyethylene terephthalate (PET) and polylactic acid (PLA) bottles were tested to evaluate radon loss from water during 15 days of storage. PET bottles (lower surface/volume-ratio vials) lost 0.4–7.1% of initial radon, whereas PLA bottles lost 3.7% of it. PET bottles with volume of 0.5 L, lower surface/weight ratio, and hence higher thickness display proportionally reduced radon loss. Corrections for dissolved radium are needed during analyses. Formulas for calculating degassing efficiency and water interference on electrostatic collections are developed.

1. Introduction

At present, polyethylene terephthalate (PET) is widely used in the large-scale production of bottles to store soda drinks and other beverages, because it is light, hygienic, and maintain the fizzy taste of carbonated drinks for adequate periods. Although it is one of the safer plastics, PET is not intended for repeated use. Bottles made from this porous plastic are difficult to clean, and can harbor bacteria, particularly when used many times. In addition, studies suggest that repeated use of PET containers might release bis(2-ethylhexyl) phthalate (DEHP), an endocrine-disrupting compound and probable human carcinogen, as well as antimony, an eye, skin, and lung irritant at high doses (Shotyk et al., 2006; Sax, 2010). This plastic material (classified as 1, according to the Society of the Plastic Industry (SPI) resin identification coding system) is recyclable, but the quality degrades with each cycle. Therefore, PET is typically "downcycled" into products such as fleece apparel, carpet fibers, and plastic straps.

Although petroleum is the major source of PET, bio-based plastic products are also increasingly used currently for packaging, thereby reducing CO_2 emissions. Coca-Cola plantbottle™, a PET plastic partly made from plants, commercialized since 2009 in 28 countries all over the world, and Acqua Lilia plantbottle™ in Italy are some of the examples. Another example is the Bio Bottle made from Ingeo™ PLA, a polylactic biopolymer, used by Acqua S. Anna in Italy for Rebruant and Vinadio springs.

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Soda drinks and mineral water plastic bottles provide a global and virtually unlimited supply of water sample vials for the assay of radon in water, although glass is technically the best choice for that. However, glass is fragile and its transport often leads to breakage. Thus, the need for shatterproof bottles led us to test the performances of plastic bottles of different types, thicknesses, and surface/volume ratios to store samples of water for a period of 2 weeks, before radon measurement.

Some investigations are reported in the literature for 1.3-L PET, 2.5-L HDPE (high-density polyethylene), and 2-L LDPE (low-density polyethylene) bottles (Leaney and Herczeg, 2006). Radon losses during a 12-day storage are lower in PET (about 7% after 4 days), and higher in HDPE (about 15% after 4 days) and LDPE (27% after 4 days) bottles. In this study, radon losses are reported after 4 days, for comparison with other shorter records. Saito (1983) showed that 1.1-L HDPE bottles lose about 20% of initial radon after 4 days. The value is lower than that measured by Leaney and Herczeg (2006), probably because of lower surface/volume ratio. De Simone et al. (2015) tested 1-L HDPE bottles and found a radon loss of about 22% for a 4-day storage. This is the highest loss among those quoted for HDPE, which could be attributed to a corresponding higher surface/volume ratio. Finally, Tuccimei et al. (2015) tested 0.355- and 1.75-L PET bottles and demonstrated a negligible decrease of radon concentration after 15 days of storage.

These studies demonstrate that PET bottles show better performance than those of HDPE and LDPE in storing water for the assay of radon, with the lowest loss. It is also evident that the lower the surface/volume ratio of the bottle is, the better the performance, with other parameters being unchanged. In this study, Coca-Cola PET bottles (1.75, 1.25, and 0.5 L) and two bio-based plastic vials (1.5-L Acqua Lilia plantbottle™ and 1-L Acqua S. Anna Bio Bottle) were tested to evaluate radon loss during storage. In order to investigate the way in which this parameter influences gas loss, 0.5-L PET bottles (Acqua di Nepi mineral water) were also included in the second step of this test, with approximately the same surface/volume ratios as 0.5-L Coca–Coca vials, but different thicknesses of PET.

2. Materials and methods

2.1. PET and PLA bottles

PET and PLA bottles are manufactured in two steps: (i) preforms, including the thread or the mouthpiece for the cap of the finished bottle are produced by plastic injection into molds and (ii) the preforms are cast to their final shapes in a stretch blow molder. The weight of each bottle, regardless of its volume, depends on the preform characteristics; weights ranging from 15 to 40 g are commonly used. Hence, thickness will be affected, which needs to be investigated.

2.2. Natural groundwater enriched with ^{222}Rn

Groundwater from a 5-L/min discharge spring in Valle della Caffarella area (Roma, Italy, Fig. 1) was chosen for the experiments, because of its high radon content (236 ± 8 Bq/L, Pizzino, 2015) and the location of the area being only few kilometers from Roma Tre University and Istituto Nazionale di Geofisica e Vulcanologia (INGV) laboratories, where measurements were performed. Groundwater belongs to “Complesso delle Vulcaniti Indifferenziate” hydrogeological unit (Capelli et al., 2012), consisting of products from Colli Albani volcano (3 and 4 in Fig. 1b). Its composition is Ca–HCO_3, with abundant potassium and sodium (Pizzino, 2015), typical of groundwater in high-potassium volcanic areas of the Roman Comagmatic province (Conticelli and Peccerillo, 1992). The salinity of the source is about 740 mg/L (electrical conductivity at 25 °C is 865 μS/cm) and very constant. The effect of salinity on radon solubility can be considered negligible in our experiments, as reported in Leaney and Herczeg (2006), where much higher salinity solutions (NaCl = 80, 16,500, 35,000, and 53,000 mg L^{-1}) were tested.

Groundwater was sampled nine times from January to June 2015, and radon activity concentration was always measured using a RAD7 monitor with Big Bottle RAD H2O accessory and cross-checked using activated charcoal collectors counted by gamma spectrometry (Galli et al., 1999). This independent method shows radon activity concentration ranging from 236 to 240 Bq/L from

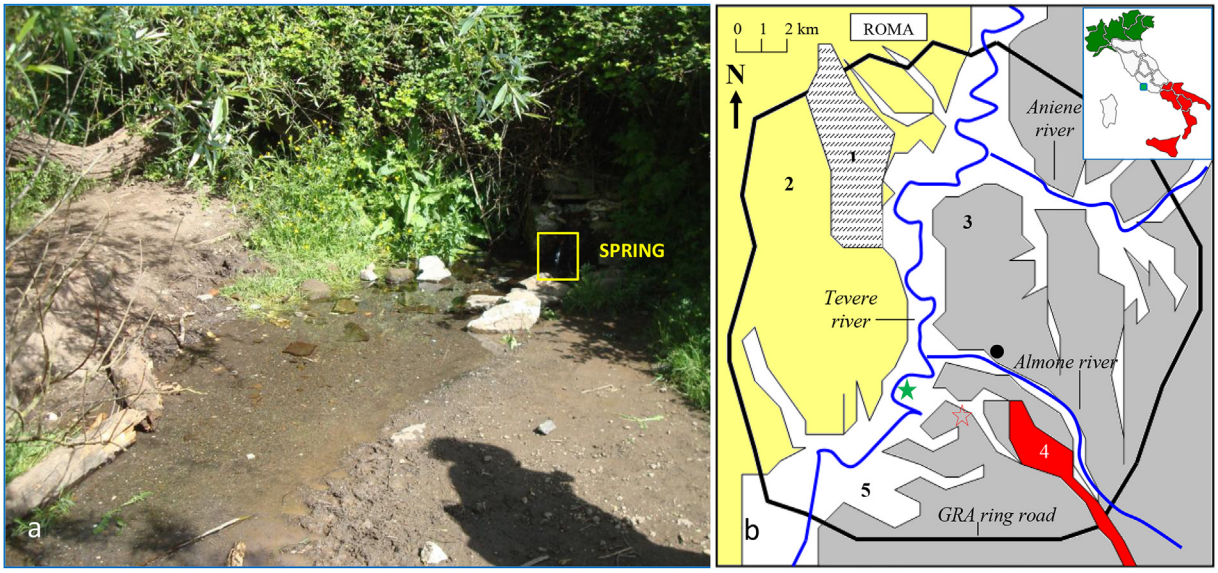

Fig. 1. Valle della Caffarella spring (a) is placed in Roma (Italy). Its location is shown by the closed circle in (b), where a simplified geological map of the city is reported. 1) Plio-Pleistocene marine to transitional deposits; 2) Sabatini district volcanoes; 3) Colli Albani district ignimbrites; 4) Colli Albani district lavas; 5) Alluvial sediments of River and its tributaries. Stars indicate Roma Tre University (closed green) and INGV (open red) laboratories. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

January to June 2015, also in agreement with data reported in Pizzino (2015). The variability of ^{222}Rn concentration (238 ± 2 Bq/L) is lower than the average analytical uncertainty of gamma spectrometry (238 ± 4 Bq/L). At least three different bottles of any investigated series were sampled every time: 1.75-L PET (Coca-Cola), 1.25-L PET (Coca-Cola), 0.5-L PET Coca-Cola, 0.5-L PET Acqua di Nepi, 1.5-L plantbottle™ (Acqua Lilia), and 1-L BioBottle™ (Acqua S. Anna), making water overflow the bottle to replenish the volume at least thrice. The first bottle of all the six types was measured in the following hours to have a zero time value (A_0), which could be used as a reference for no radon loss during storage. The other bottles were measured in the following days, following a scheduled program to complete the monitoring over 15 days of storage.

2.3. RAD7 monitor with Big Bottle RAD H2O accessory

The RAD7 monitor (Durridge Co., Inc.) is equipped with an electrostatic PIPS collector (passivated ion-implanted planar silicon detector) of alpha emitters and a spectrum analyzer, to select countings of different radon daughters. Mode "Sniff" allows us to use only the short-lived ^{218}Po to detect ^{222}Rn, which has the advantage of attaining equilibrium with the parent in just 15 min. Therefore, it is possible to set the cycle time at 15 min and repeat it for a minimum of seven times (and often up to 12 times). During the tests, the pump was on for the entire run to ensure equilibrium between dissolved and extracted radon. Air was extracted using a Teflon aerator, which consists of a single 23-cm-long vinyl tubing with an air stone fixed at its lower end, and a cap at the upper end, delivering incoming air from RAD7 via a check valve to the bottle and retransmitting it through the bubble trap to the desiccant (drierite). Dried air is then conveyed to RAD7 in a closed-loop circuit. A data logger records the temperature at the bottle–elastic clinching strap interface during the measurement for calculating the radon solubility coefficient. The experimental apparatus is shown in Fig. 2. Typical analytical uncertainties for radon concentration values of 200 Bq/L are about 5% (i.e., 200 ± 10 Bq/L).

2.4. Gamma-ray spectrometer

Radon measurements can be made by γ rays emitted by ^{214}Pb and ^{214}Bi, radon short-lived daughters, using a γ spectrometer when the secular equilibrium is reached. The low-background spectrometer available at INGV laboratories, Rome, consists of a shield made of lead, either casting or pellets, surrounding a NaI(Tl) scintillator (3 × 3 in.), optically coupled to a photomultiplier. The pulse shaping is performed by a preamplifier and an amplifier, and the counting of peaks at 295, 352, and 609 keV is done by a 4-k multichannel analyzer. The spectrometer response is verified daily by counting an activated charcoal canister containing a standard source of ^{226}Ra (376 ± 10 Bq).

2.5. Radium calculation

A Marinelli beaker (1.035 L) was filled with water from Valle della Caffarella spring, and analyzed 33 times with a γ spectrometer over 2 months to evaluate the radium content. The radon concentration plot (Fig. 3) results from the decay of the initial excess radon summed to the radon in equilibrium with radium; hence, the plot was interpolated with the following exponential function:

$$y = y_0 + Ae^{R0\ x} \quad (1)$$

where

y = radon concentration (Bq/L) at time t (min),

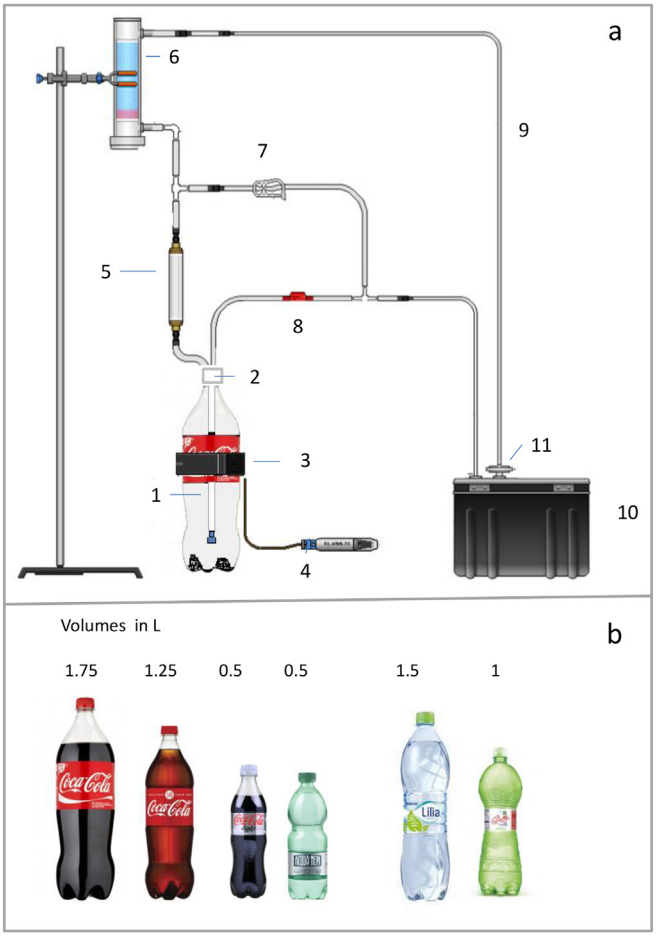

Fig. 2. Big Bottle RAD H2O configuration (a, modified from Big Bottle RAD H2O manual, Durridge Co., Inc., available at www.durridge.com). 1) Plastic soda bottle; 2) Screw-on Teflon aerator, with a single air stone; 3) Elastic clinching strap; 4) Temperature data logger; 5) Bubble trap; 6) Laboratory dryer; 7) Clip; 8) Check valve; 9) Vinyl tubing; 10) RAD7 radon detector; 11) Inlet filter. Plastic bottles used for the experiments (b). From left to right: 1.75-L PET Coca-Cola bottle, 1.25-L PET Coca-Cola bottle, 0.5-L PET Coca-Cola Light bottle, 0.5-L PET Acqua di Nepi mineral water bottle, 1.5-L Coca-Cola plantbottle™, commercialized in Italy by Acqua Lilia, 1-L "BioBottle" made from Ingeo™ PLA, used by Acqua S. Anna.

y_0 = radon concentration (Bq/L) in equilibrium with ^{226}Ra (C_{Ra} in Eq. (2)),
A = initial excess radon concentration (Bq/L),
R_0 = time constant (min^{-1}), and

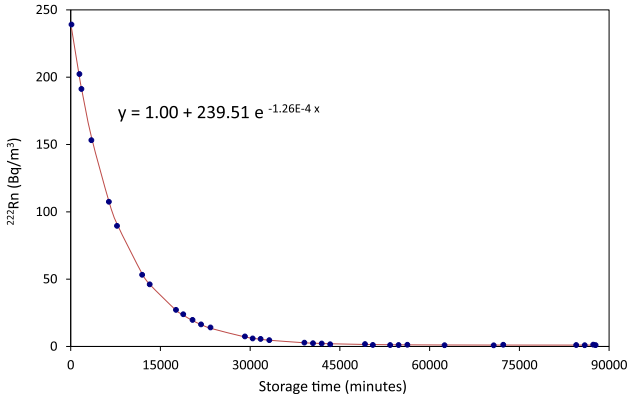

Fig. 3. Plot of radon measurements over 2 months for determination of dissolved ^{226}Ra. Errors (1 σ) are enclosed in the symbols.

Table 1
Volumes of bottles used for the experiments.

Bottle	Coca175 (mL)	Coca125 (mL)	Coca50 (mL)	Lilia150 (mL)	Ingeo100 (mL)	Nepi50 (mL)
A	1809	1282	524	1514	1111	517
B	1808	1284	524	1530	1117	513
C	1820	1286	524	1523	1113	520
D	1789	–	526	–	–	517
E	1809	–	–	–	–	515
F	1790	–	–	–	–	514

x = time elapsed from sampling (min).

2.6. Radon-in-water calculation

Radon activity concentration in water samples was calculated using the following equation (modified from De Simone et al., 2015), where background concentration in recirculating air is negligible:

$$C_w = (Ca_{FIT30}((V_a + \alpha(T)V_w) - V_h/\alpha(T)) - C_{Ra})/DF \times AF \qquad (2)$$

where

$V_a = V_{R7} + V_d + V_t + V_b$,
C_w = original radon concentration in the water, corrected for ^{226}Ra (Bq/m^3),
Ca_{FIT30} = radon concentration value at t = 30 min of an exponential fit of RAD7 data recorded during each 15-min run (30–120/180 min) (Bq/m^3),
T = temperature of water in the bottle (°C) (Fig. 2),
$\alpha(T) = 0.105 + 0.405\ e^{-0.0502\ T}$ = equilibrium coefficient from Fritz von Weigel equation (Weigel, 1978),
V_w = volume of water in bottle (see Table 1),
V_{R7} = internal volume of the RAD7 (0.768E-03 m^3),
V_d = equivalent desiccant column volume (0.673E-03 m^3),
V_t = volume of tubing and aerator (0.053 E–03 m^3),
V_b = volume of bubble trap (0.051E-03 m^3),
V_a = total volume of air in the system (1.545E-03 m^3),
$V_h/a(T)$ = radon loss in the head space of air above the water in the plastic bottle, where V_h is the head space volume,
C_{Ra} = ^{226}Ra concentration in the water (1.00 ± 0.09 Bq/L),
DF = Decay Factor (=$e^{-t/\tau_{Rn}}$, where t (min) is the time elapsed between water sampling and 30 min after the beginning of the run and τ_{Rn} (min) is the radon average life, 7938), and
AF = adjustment of instrument calibration factor (0.9966, in this case).

Radon concentration fit (Ca_{FIT30}) was obtained by interpolating a data set ranging from 30–45 to 135–180 min (depending on the available cycles). The choice of the first datum used for the interpolation, 30 or 45 min, derives from statistical test, as illustrated in the following. The 30-min datum is critical, because its value is influenced by factors such as (i) degassing efficiency related to the bottle size and the type of air stone, (ii) time required to attain equilibrium between radon concentration in the two phases (water and air), and (iii) radon activity in the closed loop. All the experiments were classified based on bottle characteristics (plastic type and size), air stone type (the original was replaced because of its rupture), radon concentration classes, and departure of the 30-min datum from the exponential fit (within 1 σ, between 1 and 2 σ, or beyond 2 σ, see Table 2).

The class with higher activity for each experimental setup (given by the Big Bottle configuration reported in Fig. 2 and a single bottle type) was used to evaluate its efficiency by checking the plot of 30-min datum of each run, i.e. within 1 σ, between 1 and 2 σ, or beyond 2 σ from the relative fit curve. If deviations from the fit are

Table 2
Classification of experiments on the basis of bottle characteristics (plastic type and size), air stone type, radon concentration classes, and departure of the 30-min datum from the exponential fit (within 1 σ, within 2 σ, or beyond 2 σ). N denotes the number of measurements in the highest activity class for each bottle type.

Bottle	N	>50 Bq/L			<50 Bq/L		
		<1 σ	1–2 σ	>2 σ	<1 σ	1–2 σ	>2 σ
Air stone included in the Durridge soda bottle aerator kit							
1.25 – PET	6	5	1	–	–	–	–
1.75 – PET	5	2	3	–	–	–	–
1.5 – BIO PET	5	4	1	–	–	–	–
1 – PLA	2	2	–	–	–	–	–
Air stone borrowed from the standard Durridge big bottle aerator kit							
0.5 – PET	8	5	3	–	6	4	–
1.25 – PET	6	6	–	–	4	2	1
1.75 – PET	8	6	1	1	7	2	–
1.5 – BIO PET	8	5	3	–	3	4	1
1 – PLA	10	10	–	–	6	–	1
0.5 – PET (Nepi)	4	3	1	–	6	1	–

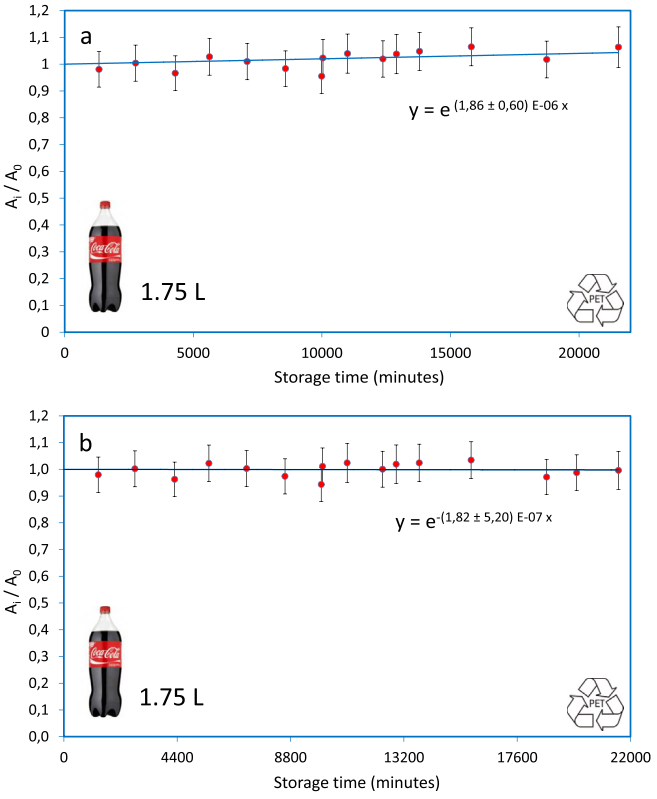

Fig. 4. Plot of A_i/A_0 versus storage time in 1.75-L PET bottles. Data are corrected for (a) decay and (b) decay and ^{226}Ra content in water (1.00 ± 0.09 Bq/L). Modified from Tuccimei et al. (2015).

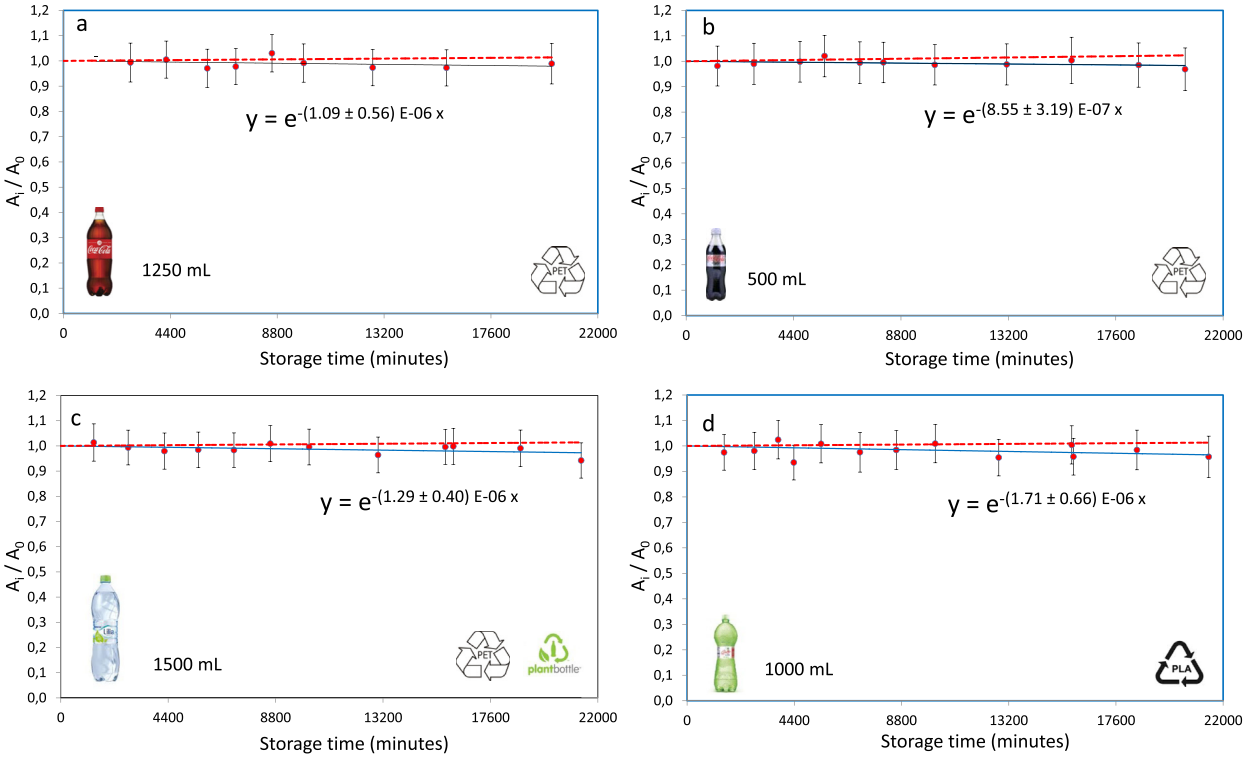

Fig. 5. Plot of A_i/A_0 versus storage time in different volumes of PET and PLA bottles. Data are corrected for decay and ^{226}Ra content in water (1.00 ± 0.09 Bq/L). Red dashed lines indicate the radon loss without correction for radium. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

included in the normal distribution (<32% beyond 1 σ and <5% beyond 2 σ), the test configuration is evaluated as efficient and the first datum for the interpolation is at 30 min. On the contrary, if deviations from the fit exceed normal distributions, the setup is not considered as completely efficient and the first datum for the interpolation is at 45 min. This may depend on the degassing efficiency if the aerator does not approach the bottom of the bottle and the equilibrium condition is reached later.

In the second step of evaluating radon data from classes with lower activities, if deviations of the 30 min datum for the class with the highest activity were included in the normal distribution, the data are checked singularly and the datum at 30 min is eliminated only in cases of large deviations. On the contrary, if deviations from the fit for the higher activity class were not included in the normal distribution, the 30-min datum is not considered and the interpolation started from the 45-min data point.

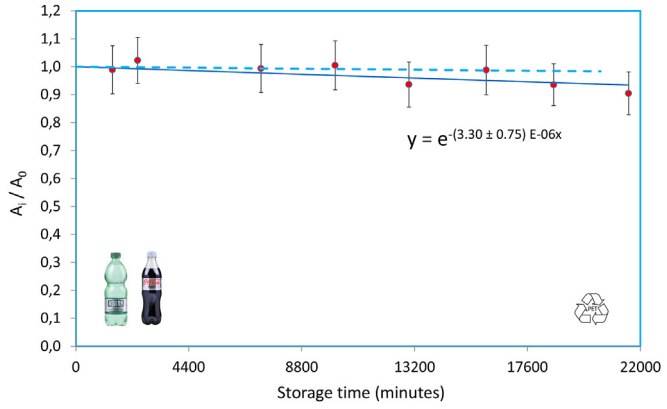

Fig. 6. Plot of A_i/A_0 versus storage time in 500-mL PET bottles with different thicknesses, but similar surface/volume ratios. Data are corrected for decay and ^{226}Ra content in water (1.00 ± 0.09 Bq/L). Data refer to 0.5-L Acqua di Nepi mineral water bottle, and blue dashed line indicates the radon loss in 0.5-L Coca-Cola bottles (Fig. 5). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

At ambient temperatures >23 °C, the correction due to radon loss in the head space was applied to consider the radon loss due to the thermal dilatancy of Ingeo™ PLA. When the cap is opened at the beginning of the measurement, the radon concentration in this volume (V_h) attains equilibrium with water and escapes from the system. This loss is expressed by V_h/α in Eq. (2). There was no evidence of this ratio becoming zero for petroleum- and bio-based PET. This may be due to the coefficient of thermal expansion of PET, which ranges from 20 to 80 × 10^{-6} °K^{-1} (Saleh and Lubineau, 2014), and is lower than that of PLA (80–90 × 10^{-6} °K^{-1}, Gao, 2012). In addition, PET becomes unstable at 72 °C, whereas PLA has a glass transition temperature of 55 °C.

No correction was applied for absolute humidity in the system for the temperature range of 15–23 °C, with a relative humidity inside the instrument of about 4–5%, because of no interference from the water molecules on the electrostatic collection of ^{218}Po. When the temperature inside RAD7 exceeds 23 °C and the relative humidity exceeds 5%, that is, 0.75 × 10^{-3} g of water in the RAD7 inner volume, a correction is applied to the radon concentration value at t = 30 min. This value is substituted in Eq. (1) for Ca $_{FIT30}$:

$$Ca_{FIT30\_corr} = Ca_{FIT30}(1.05 - (59 \times gH_2O_{RAD7})) \quad (3)$$

3. Results and discussion

3.1. Determination of dissolved ^{226}Ra

Total radon concentration in groundwater is the sum of ^{222}Rn in equilibrium with dissolved ^{226}Ra and excess radon from

Table 3
Radon loss during 15-day storage in PET and PLA bottles.

Bottle	Time (min)	A_1/A_0	$\sigma A_1/A_0$
1.75-L PET (Coca-Cola)	0	1.000	0.000
	1334	0.980	0.066
	2757	1.002	0.067
	4300	0.963	0.065
	5631	1.023	0.068
	7095	1.003	0.067
	8585	0.974	0.066
	9992	0.943	0.064
	10042	1.011	0.069
	10999	1.024	0.073
	12374	1.000	0.067
	12902	1.019	0.072
	13797	1.024	0.070
	15810	1.034	0.069
	18744	0.971	0.066
	19911	0.988	0.067
	21532	0.996	0.071
1.25-L PET (Coca-Cola)	0	1.000	0.000
	1335	1.017	0.077
	2758	0.993	0.073
	4231	1.005	0.076
	5921	0.971	0.071
	7096	0.977	0.074
	8583	1.030	0.076
	9890	0.991	0.072
	12732	0.973	0.072
	15769	0.972	0.071
	20102	0.989	0.075
0.5-L PET (Coca-Cola)	0	1.000	0.000
	1280	0.981	0.078
	2775	0.990	0.081
	4663	0.998	0.080
	5668	1.020	0.082
	7115	0.994	0.082
	8081	0.995	0.079
	10187	0.986	0.079
	13133	0.987	0.081
	15791	1.003	0.091
	18537	0.985	0.087
	20442	0.968	0.084
1.5-L PET (plantbottle™)	0	1.000	0.000
	1354	1.013	0.074
	2761	0.993	0.069
	4248	0.979	0.072
	5633	0.984	0.070
	7095	0.983	0.068
	8582	1.009	0.071
	10172	0.995	0.071
	12997	0.964	0.071
	15752	0.996	0.069
	16079	0.998	0.072
	18831	0.990	0.072
	21316	0.942	0.070
1.0-L PLA (Acqua S. Anna)	0	1.000	0.000
	1521	0.975	0.070
	2760	0.980	0.073
	3739	1.024	0.075
	4382	0.935	0.068
	5499	1.008	0.075
	7098	0.975	0.078
	8588	0.984	0.076
	10185	1.009	0.075
	12791	0.954	0.072
	15788	1.004	0.075
	15858	0.958	0.072
	18462	0.984	0.078
	21408	0.957	0.081
0.5-L PET (Acqua di Nepi)	0	1.000	0.000
	1426	0.989	0.086
	2406	1.022	0.083
	7215	0.994	0.086
	10104	1.005	0.088
	12979	0.936	0.081
	15994	0.988	0.088
	18621	0.935	0.075
	21541	0.904	0.076

water–rock interaction. This component may be interpreted as recoil flux from mineral surfaces (Krishnaswami et al., 1982) or as a combination of recoil and diffusion of radon from micro-fractures or aquifer solids surfaces (Rama and Moore, 1984; Davidson and Dickson, 1986; Andrews et al., 1989; Vinson et al., 2009). As described by Tuccimei et al. (2015), the effect of radium has to be considered when correcting radon data for radioactive decay, particularly for longer storage times. Moreover, if no correction is applied, radon concentration may apparently increase over time, in case of significant radium activity (Tuccimei et al., 2015).

The amount of dissolved radium in Valle della Caffarella spring was obtained by the interpolation of 33 radon data obtained by gamma spectrometry during a period of 2 months (Fig. 3), using an exponential function (Eq. (1)). The result of the interpolation is

$$y = 1.00 + 239.51\, e^{-1.26E-4x}$$

Consequently, 1.00 ± 0.09 Bq/L (y_0 in Eq. (1) and C_{Ra} in Eq. (2)) of radon has been subtracted in all radon measurements before applying the correction for radioactive decay (DF in Eq. (2)). The fitting also provides the initial excess radon (239.51 ± 0.84 Bq/L), which when summed to ^{222}Rn in equilibrium with dissolved radium agrees with the value of 236 ± 8 Bq/L, reported by Pizzino (2015). It is worth noting that the time constant (R_0 in Eq. (1)) corresponds to ^{222}Rn decay constant (expressed in minutes).

3.2. Radon loss during 15-day storage from PET and PLA plastics

A statistical test was conducted on the data reported in Table 2. As a result, the datum at 30 min was excluded from the data set used for the interpolation, only for 1.75-L PET bottles, when the air stone included in the standard soda bottle aerator kit was used.

A plot of radon loss (A_i/A_0) versus storage time in 1.75-L PET bottles (Fig. 4) was presented in Tuccimei et al. (2015). Data are corrected for decay (Fig. 4a) and decay and ^{226}Ra content in water (1.00 ± 0.09 Bq/L, Fig. 4b).

These data are compared and discussed against radon loss of groundwater from Valle della Caffarella spring stored in PET and PLA bottles for 15 days (Fig. 5). In all cases, ^{222}Rn activity concentration is corrected for radioactive decay and ^{226}Ra content (1.00 ± 0.09 Bq/L). Each graph also reports the exponential fitting of row data, not corrected for dissolved radium. If no correction for radium is applied, the exponential fitting of red dashed lines in Fig. 5 simulates a slight increase of radon concentration during storage in PET (petrol- and plant-based types) and PLA bottles, which is impossible. This confirms that a correction is needed, particularly for increasing storage time.

Corrected data demonstrate minor losses from all bottles, ranging from about 0.03% (1.75-L PET) to 0.25% (1-L PLA) per day, resulting in about 0.4 and 3.7% in 15 days. However, the relative differences are significant and depend on the surface/volume ratios of the bottles and the thickness of the plastic material. The discussion is reported in the following section.

Fig. 6 reports the radon loss from 0.5-L PET bottles used by Acqua di Nepi mineral water, with the volume and surface/volume ratio same as Coca-Cola, but different surface/weight ratio and hence plastic thickness. Radon loss is the highest among the measured values and approaches 0.35% per day, that is, 5.20% in 15 days. All radon data used in Figs. 4–6 are reported in Table 3.

The gas loss rates of bio-based PET and PLA bottles were about 0.19 and 0.25% per day, respectively, resulting in 2.8 and 3.7% after 15 days.

Table 4
Averages of volumes, surface/volume ratios, weights, surface/weight ratios, and radon loss rates of plastic bottles used for the experiments.

Bottle (L)	Water volume (L)	Surface (cm^2)	Surface/Volume (cm^{-1})	Weight (g)	Surface/Weight ($cm^2\ g^{-1}$)	Radon loss rate ($10^{-6}\ min^{-1}$)
1.75 – PET	1.804	913	0.506	36.1	25.3	−0.18 ± 0.52
1.5 – BIO PET	1.519	845	0.556	23.2	36.5	−1.29 ± 0.40
1.25 – PET	1.284	714	0.556	36.1	19.8	−1.09 ± 0.56
1 – PLA	1.114	663	0.596	24.6	27.0	−1.71 ± 0.66
0.5 – PET	0.524	412	0.786	17.4	23.7	−0.86 ± 0.32
0.5 – PET (Nepi)	0.516	402	0.779	12.4	32.3	−3.30 ± 0.75

3.3. Radon loss dependence on surface/volume ratios and thickness of plastic bottles

Table 4 presents the surface/volume and surface/weight ratios (roughly related to the plastic thickness) of PET (petrol- and bio-based types) and PLA bottles. Surface integral revolution (Eq. (4)) can be applied to the bottle profile, f(x), to calculate the surface area (S) of the bottles:

$$S = 2\pi \int_{x_1}^{x_2} f(x)\sqrt{1 + [f'(x)]^2}\, dx \quad (4)$$

Eq. (4) can be approximated using the geometry of the truncated cones (5) as

$$S = \pi \sum_i a_i (R_{2\,i} + R_{1\,i}) \quad (5)$$

where a is the apothem and R_2 and R_1 are the truncated cone radii.

The plot of radon loss rates versus surface/volume ratios (Fig. 7) shows the direct correlation between the two parameters, regardless of the plastic types, except for the 0.5-L PET (Coca-Cola) characterized by a surface/weight ratio significantly lower (i.e., larger thickness) than that of 0.5-L PET (Acqua di Nepi), having a similar surface/volume ratio. Consequently, with regard to radon loss rates, bottle thickness can partly balance higher surface/volume ratios.

4. Conclusions

The results suggest that PET, either petrol- or bio-based types, and PLA are much suitable for storing natural water for the assay of radon. Their performances are much better than those of other plastics investigated in the literature (LDPE and HDPE). If radon loss rates after 4 days of storing are compared with available data from literature, PET bottles loose from about 0.1 to 1.4% and PLA 1% against the rates of HDPE, from 15 to 22% (Saito, 1983; Leaney and Herczeg, 2006; De Simone et al., 2015) and LDPE 27% (Leaney and Herczeg, 2006).

Surface/volume ratios and thickness of different PET bottles were examined to verify their role on radon loss rates over a period of 15 days. The main factor affecting radon loss rate of a given material is its surface/volume ratio, because either diffusion or adsorption, indicated as possible involved processes in the literature (Saito, 1983; Arafa, 2002; Fernández et al., 2004; Ashry et al., 2011; De Simone et al., 2015), is surface dependent.

A higher bottle thickness reduces radon loss rates when considering bottles having similar surface/volume ratios as in the case of 0.5-L Coca-Cola and Acqua di Nepi mineral water PET bottles.

An accurate and precise determination of dissolved ^{226}Ra in water samples is necessary, particularly for measurements after at least a week from sampling.

Performances of Big Bottle RAD H2O device with the soda bottle aerator kit coupled to RAD7 radon monitor (Durridge Co., Inc.) were evaluated in terms of degassing efficiency, and the effects of temperature and grams of water in the RAD7 inner volume on the radon daughter electrostatic collection were investigated. Proper corrections were developed and applied.

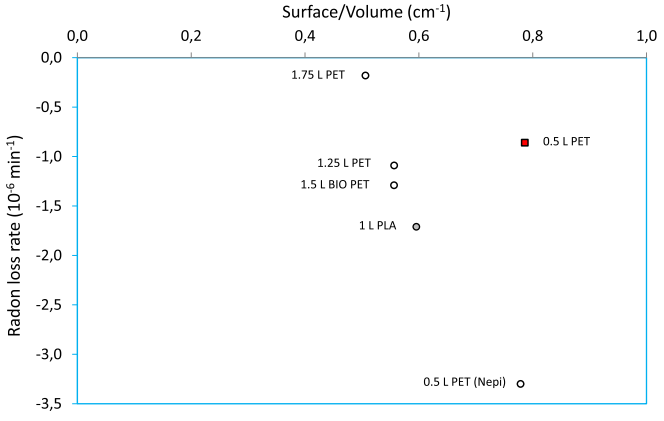

Fig. 7. Plot of radon loss rate versus surface/volume ratio of plastic bottles. 1-σ errors range from 0.3 × 10^{-6} to 0.7 × 10^{-6} min^{-1}.

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