

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 H₂O on radon daughter electrostatic collection was quantified.

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.

Keywords: Radon in water; Radon loss during storage; PET; PLA; surface/volume ratio; Big Bottle RAD H2O

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 CO₂ 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.

1 Soda drinks and mineral water plastic bottles provide a global and virtually unlimited supply of
2 water sample vials for the assay of radon in water, although glass is technically the best choice for
3 that. However, glass is fragile and its transport often leads to breakage. Thus, the need for
4 shatterproof bottles led us to test the performances of plastic bottles of different types, thicknesses,
5 and surface/volume ratios to store samples of water for a period of 2 weeks, before radon
6 measurement.
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11 Some investigations are reported in the literature for 1.3-L PET, 2.5-L HDPE (high-density
12 polyethylene), and 2-L LDPE (low-density polyethylene) bottles (Leaney and Herczeg, 2006).
13 Radon losses during a 12-day storage are lower in PET (about 7% after 4 days), and higher in
14 HDPE (about 15% after 4 days) and LDPE (27% after 4 days) bottles. In this study, radon losses are
15 reported after 4 days, for comparison with other shorter records. Saito (1983) showed that 1.1-L
16 HDPE bottles lose about 20% of initial radon after 4 days. The value is lower than that measured by
17 Leaney & Herczeg (2006), probably because of lower surface/volume ratio. De Simone et al. (2015)
18 tested 1-L HDPE bottles and found a radon loss of about 22% for a 4-day storage. This is the
19 highest loss among those quoted for HDPE, which could be attributed to a corresponding higher
20 surface/volume ratio. Finally, Tuccimei et al. (2015) tested 0.355- and 1.75-L PET bottles and
21 demonstrated a negligible decrease of radon concentration after 15 days of storage.
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32 These studies demonstrate that PET bottles show better performance than those of HDPE and LDPE
33 in storing water for the assay of radon, with the lowest loss. It is also evident that the lower the
34 surface/volume ratio of the bottle is, the better the performance, with other parameters being
35 unchanged. In this study, Coca-Cola PET bottles (1.75, 1.25, and 0.5 L) and two bio-based plastic
36 vials (1.5-L Acqua Lilia plantbottleTM and 1-L Acqua S. Anna Bio Bottle) were tested to evaluate
37 radon loss during storage. In order to investigate the way in which this parameter influences gas
38 loss, 0.5-L PET bottles (Acqua di Nepi mineral water) were also included in the second step of this
39 test, with approximately the same surface/volume ratios as 0.5-L Coca-Cola vials, but different
40 thicknesses of PET.
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52 **2. Materials and Methods**

53 *2.1. PET and PLA bottles*

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55 PET and PLA bottles are manufactured in two steps: (i) preforms, including the thread or the
56 mouthpiece for the cap of the finished bottle are produced by plastic injection into molds and (ii)
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1 the preforms are cast to their final shapes in a stretch blow molder. The weight of each bottle,
2 regardless of its volume, depends on the preform characteristics; weights ranging from 15 to 40 g
3 are commonly used. Hence, thickness will be affected, which needs to be investigated.
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8 2.2. Natural groundwater enriched with ^{222}Rn 9

10 Groundwater from a 5-L/min discharge spring in Valle della Caffarella area (Roma, Italy, Fig. 1)
11 was chosen for the experiments, because of its high radon content (236 ± 8 Bq/L, Pizzino, 2015)
12 and the location of the area being only few kilometers from Roma Tre University and Istituto
13 Nazionale di Geofisica e Vulcanologia (INGV) laboratories, where measurements were performed.
14 Groundwater belongs to “Complesso delle Vulcaniti Indifferenziate” hydrogeological unit (Capelli
15 et al., 2012), consisting of products from Colli Albani volcano (3 and 4 in Fig. 1b). Its composition
16 is Ca-HCO₃, with abundant potassium and sodium (Pizzino, 2015), typical of groundwater in high-
17 potassium volcanic areas of the Roman Comagmatic province (Conticelli et al., 1992). The salinity
18 of the source is about 740 mg/L (electrical conductivity at 25°C is 865 microSiemens/cm) and very
19 constant. The effect of salinity on radon solubility can be considered negligible in our experiments,
20 as reported in Leaney and Herczeg (2006), where much higher salinity solutions (NaCl = 80,
21 16,500, 35,000, and 53,000 mg L⁻¹) were tested.
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34 Groundwater was sampled nine times from January to June 2015, and radon activity concentration
35 was always measured using a RAD7 monitor with Big Bottle RAD H2O accessory and cross-
36 checked using activated charcoal collectors counted by gamma spectrometry (Galli et al., 1999).
37 This independent method shows radon activity concentration ranging from 236 to 240 Bq/L from
38 January to June 2015, also in agreement with data reported in Pizzino (2015). The variability of
39 ^{222}Rn concentration (238 ± 2 Bq/L) is lower than the average analytical uncertainty of gamma
40 spectrometry (238 ± 4 Bq/L). At least three different bottles of any investigated series were sampled
41 every time: 1.75-L PET (Coca-Cola), 1.25-L PET (Coca-Cola), 0.5-L PET Coca-Cola, 0.5-L PET
42 Acqua di Nepi, 1.5-L plantbottleTM (Acqua Lilia), and 1-L BioBottleTM (Acqua S. Anna), making
43 water overflow the bottle to replenish the volume at least thrice. The first bottle of all the six types
44 was measured in the following hours to have a zero time value (A_0), which could be used as a
45 reference for no radon loss during storage. The other bottles were measured in the following days,
46 following a scheduled program to complete the monitoring over 15 days of storage.
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2.3. RAD7 monitor with Big Bottle RAD H2O accessory

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3 The RAD7 monitor (DurrIDGE Co., Inc.) is equipped with an electrostatic PIPS collector (passivated
4 ion-implanted planar silicon detector) of alpha emitters and a spectrum analyzer, to select countings
5 of different radon daughters. Mode “Sniff” allows us to use only the short-lived ^{218}Po to detect
6 ^{222}Rn , which has the advantage of attaining equilibrium with the parent in just 15 min. Therefore, it
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8 is possible to set the cycle time at 15 min and repeat it for a minimum of seven times (and often up
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10 to 12 times). During the tests, the pump was on for the entire run to ensure equilibrium between
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12 dissolved and extracted radon. Air was extracted using a Teflon aerator, which consists of a single
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14 23-cm-long vinyl tubing with an air stone fixed at its lower end, and a cap at the upper end,
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16 delivering incoming air from RAD7 via a check valve to the bottle and retransmitting it through the
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18 bubble trap to the desiccant (drierite). Dried air is then conveyed to RAD7 in a closed-loop circuit.
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20 A data logger records the temperature at the bottle–elastic clinching strap interface during the
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22 measurement for calculating the radon solubility coefficient. The experimental apparatus is shown
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24 in [Figure 2](#). Typical analytical uncertainties for radon concentration values of 200 Bq/L are about 5
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26 % (i.e., 200 ± 10 Bq/L).
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2.4. Gamma-ray spectrometer

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33 Radon measurements can be made by γ rays emitted by ^{214}Pb and ^{214}Bi , radon short-lived daughters,
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35 using a γ spectrometer when the secular equilibrium is reached. The low-background spectrometer
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37 available at INGV laboratories, Rome, consists of a shield made of lead, either casting or pellets,
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39 surrounding a NaI(Tl) scintillator (3×3 in.), optically coupled to a photomultiplier. The pulse
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41 shaping is performed by a preamplifier and an amplifier, and the counting of peaks at 295, 352, and
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43 609 keV is done by a 4-k multichannel analyzer. The spectrometer response is verified daily by
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45 counting an activated charcoal canister containing a standard source of ^{226}Ra (376 ± 10 Bq).
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2.5. Radium calculation

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56 A Marinelli beaker (1.035 L) was filled with water from Valle della Caffarella spring, and analyzed
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58 33 times with a γ spectrometer over 2 months to evaluate the radium content. The radon
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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:

$$y = y_0 + A e^{R_0 x}, \quad (1)$$

where

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

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

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:

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

where

$$V_a = V_{\text{R7}} + V_d + V_t + V_b,$$

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 ($^{\circ}\text{C}$) (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 ($0.768\text{E}-03 \text{ m}^3$),

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V_d = equivalent desiccant column volume (0.673E-03 m³),

V_t = volume of tubing and aerator (0.053 E-03 m³),

V_b = volume of bubble trap (0.051E-03 m³),

V_a = total volume of air in the system (1.545E-03 m³),

$V_h/a(T)$ = 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} = ²²⁶Ra 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 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.

1 In the second step of evaluating radon data from classes with lower activities, if deviations of the 30
2 min datum for the class with the highest activity were included in the normal distribution, the data
3 are checked singularly and the datum at 30 min is eliminated only in cases of large deviations. On
4 the contrary, if deviations from the fit for the higher activity class were not included in the normal
5 distribution, the 30-min datum is not considered and the interpolation started from the 45-min data
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11 At ambient temperatures $>23^{\circ}\text{C}$, the correction due to radon loss in the head space was applied to
12 consider the radon loss due to the thermal dilatancy of IngeoTM PLA. When the cap is opened at the
13 beginning of the measurement, the radon concentration in this volume (V_h) attains equilibrium with
14 water and escapes from the system. This loss is expressed by V_h/α in Eq. 2. There was no evidence
15 of this ratio becoming zero for petroleum- and bio-based PET. This may be due to the coefficient of
16 thermal expansion of PET, which ranges from 20 to $80 \times 10^{-6} \text{ }^{\circ}\text{K}^{-1}$ (Saleh and Lubineau, 2014),
17 and is lower than that of PLA ($80\text{--}90 \times 10^{-6} \text{ }^{\circ}\text{K}^{-1}$, Gao, 2012). In addition, PET becomes unstable
18 at 72°C , whereas PLA has a glass transition temperature of 55°C .
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27 No correction was applied for absolute humidity in the system for the temperature range of 15–
28 23°C , with a relative humidity inside the instrument of about 4–5%, because of no interference from
29 the water molecules on the electrostatic collection of ^{218}Po . When the temperature inside RAD7
30 exceeds 23°C and the relative humidity exceeds 5%, that is, 0.75×10^{-3} g of water in the RAD7
31 inner volume, a correction is applied to the radon concentration value at $t = 30$ min. This value is
32 substituted in Eq. 1 for Ca_{FIT30} :
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$$39 \quad Ca_{FIT30_corr} = Ca_{FIT30} (1.05 - (59 \times gH_2O_{RAD7})). \quad (3)$$

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44 **3. Results and discussion**

45 *3.1. Determination of dissolved ^{226}Ra*

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50 Total radon concentration in groundwater is the sum of ^{222}Rn in equilibrium with dissolved ^{226}Ra
51 and excess radon from water–rock interaction. This component may be interpreted as recoil flux
52 from mineral surfaces (Krishnaswami et al., 1982) or as a combination of recoil and diffusion of
53 radon from microfractures or aquifer solids surfaces (Rama and Moore, 1984; Davidson and
54 Dickson, 1986; Andrews et al., 1989; Vinson et al., 2009). As described by Tuccimei et al. (2015),
55 the effect of radium has to be considered when correcting radon data for radioactive decay,
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1 particularly for longer storage times. Moreover, if no correction is applied, radon concentration may
2 apparently increase over time, in case of significant radium activity (Tuccimei et al., 2015).
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4 The amount of dissolved radium in Valle della Caffarella spring was obtained by the interpolation
5 of 33 radon data obtained by gamma spectrometry during a period of 2 months (Fig. 3), using an
6 exponential function (Eq. 1). The result of the interpolation is
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$$9 \quad y = 1.00 + 239.51 e^{-1.26E-4 x}$$

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12 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
13 radon measurements before applying the correction for radioactive decay (DF in Eq. 2). The fitting
14 also provides the initial excess radon (239.51 ± 0.84 Bq/L), which when summed to ^{222}Rn in
15 equilibrium with dissolved radium agrees with the value of 236 ± 8 Bq/L, reported by Pizzino
16 (2015). It is worth noting that the time constant (R_0 in Eq. 1) corresponds to ^{222}Rn decay constant
17 (expressed in minutes).
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28 3.2. Radon loss during 15-day storage from PET and PLA plastics

29 A statistical test was conducted on the data reported in Table 2. As a result, the datum at 30 min was
30 excluded from the data set used for the interpolation, only for 1.75-L PET bottles, when the air
31 stone included in the standard soda bottle aerator kit was used.
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37 A plot of radon loss (A_i/A_0) versus storage time in 1.75-L PET bottles (Fig. 4) was presented in
38 Tuccimei et al. (2015). Data are corrected for decay (Fig. 4a) and decay and ^{226}Ra content in water
39 (1.00 ± 0.09 Bq/L, Fig. 4b).
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43 These data are compared and discussed against radon loss of groundwater from Valle della
44 Caffarella spring stored in PET and PLA bottles for 15 days (Fig. 5). In all cases, ^{222}Rn activity
45 concentration is corrected for radioactive decay and ^{226}Ra content (1.00 ± 0.09 Bq/L). Each graph
46 also reports the exponential fitting of row data, not corrected for dissolved radium. If no correction
47 for radium is applied, the exponential fitting of red dashed lines in Figure 5 simulates a slight
48 increase of radon concentration during storage in PET (petrol- and plant-based types) and PLA
49 bottles, which is impossible. This confirms that a correction is needed, particularly for increasing
50 storage time.
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59 Corrected data demonstrate minor losses from all bottles, ranging from about 0.03% (1.75-L PET)
60 to 0.25% (1-L PLA) per day, resulting in about 0.4 and 3.7% in 15 days. However, the relative
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1 differences are significant and depend on the surface/volume ratios of the bottles and the thickness
2 of the plastic material. The discussion is reported in the following section.
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4 **Figure 6** reports the radon loss from 0.5-L PET bottles used by Acqua di Nepi mineral water, with
5 the volume and surface/volume ratio same as Coca-Cola, but different surface/weight ratio and
6 hence plastic thickness. Radon loss is the highest among the measured values and approaches
7 0.35% per day, that is, 5.20% in 15 days. All radon data used in **Figures 4, 5, and 6** are reported in
8 **Table 3**.
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14 The gas loss rates of bio-based PET and PLA bottles were about 0.19 and 0.25% per day,
15 respectively, resulting in 2.8 and 3.7% after 15 days.
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21 *3.3. Radon loss dependence on surface/volume ratios and thickness of plastic bottles*

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24 **Table 4** presents the surface/volume and surface/weight ratios (roughly related to the plastic
25 thickness) of PET (petrol- and bio-based types) and PLA bottles. Surface integral revolution (Eq. 4)
26 can be applied to the bottle profile, $f(x)$, to calculate the surface area (S) of the bottles:
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$$30 S = 2\pi \int_{x_1}^{x_2} f(x) \sqrt{1 + [f'(x)]^2} dx. \quad (4)$$

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

$$35 S = \pi \sum_i a_i (R_{2i} + R_{1i}), \quad (5)$$

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39 where a is the apothem and R_2 and R_1 are the truncated cone radii.
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42 The plot of radon loss rates versus surface/volume ratios (Fig. 7) shows the direct correlation
43 between the two parameters, regardless of the plastic types, except for the 0.5-L PET (Coca-Cola)
44 characterized by a surface/weight ratio significantly lower (i.e., larger thickness) than that of 0.5-L
45 PET (Acqua di Nepi), having a similar surface/volume ratio. Consequently, with regard to radon
46 loss rates, bottle thickness can partly balance higher surface/volume ratios.
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55 **4. Conclusions**

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57 The results suggest that PET, either petrol- or bio-based types, and PLA are much suitable for
58 storing natural water for the assay of radon. Their performances are much better than those of other
59 plastics investigated in the literature (LDPE and HDPE). If radon loss rates after 4 days of storing
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1 are compared with available data from literature, PET bottles loose from about 0.1 to 1.4% and
2 PLA 1% against the rates of HDPE, from 15 to 22% (Saito, 1983 ; Leaney and Herczeg, 2006; De
3 Simone et al., 2015) and LDPE 27 % (Leaney and Herczeg, 2006).
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6 Surface/volume ratios and thickness of different PET bottles were examined to verify their role on
7 radon loss rates over a period of 15 days. The main factor affecting radon loss rate of a given
8 material is its surface/volume ratio, because either diffusion or adsorption, indicated as possible
9 involved processes in the literature (Saito, 1983; Arafa, 2002; Fernandez et al., 2004; Ashry et al.,
10 2011; De Simone et al., 2015), is surface dependent.
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16 A higher bottle thickness reduces radon loss rates when considering bottles having similar
17 surface/volume ratios as in the case of 0.5-L Coca-Cola and Acqua di Nepi mineral water PET
18 bottles.
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22 An accurate and precise determination of dissolved ^{226}Ra in water samples is necessary, particularly
23 for measurements after at least a week from sampling.
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27 Performances of Big Bottle RAD H₂O device with the soda bottle aerator kit coupled to RAD7
28 radon monitor (DurrIDGE Co., Inc.) were evaluated in terms of degassing efficiency, and the effects
29 of temperature and grams of water in the RAD7 inner volume on the radon daughter electrostatic
30 collection were investigated. Proper corrections were developed and applied.
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38 **Figure captions**

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40 **Figure 1.** Valle della Caffarella spring (a) is placed in Roma (Italy). Its location is shown by the
41 closed circle in (b), where a simplified geological map of the city is reported. 1) Plio-Pleistocene
42 marine to transitional deposits; 2) Sabatini district volcanoes; 3) Colli Albani district ignimbrites; 4)
43 Colli Albani district lavas; 5) Alluvial sediments of Tiber River and its tributaries. Stars indicate
44 Roma Tre University (closed green) and INGV (open red) laboratories.
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51 **Figure 2.** Big Bottle RAD H2O configuration (a, modified from Big Bottle RAD H2O manual,
52 DurrIDGE Co., Inc., available at www.durrIDGE.com). 1) Plastic soda bottle; 2) Screw-on Teflon
53 aerator, with a single air stone; 3) Elastic clinching strap; 4) Temperature data logger; 5) Bubble
54 trap; 6) Laboratory dryer; 7) Clip; 8) Check valve; 9) Vinyl tubing; 10) RAD7 radon detector; 11)
55 Inlet filter. Plastic bottles used for the experiments (b). From left to right: 1.75-L PET Coca-Cola
56 bottle, 1.25-L PET Coca-Cola bottle, 0.5-L PET Coca-Cola Light bottle, 0.5-L PET Acqua di Nepi
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1 mineral water bottle, 1.5-L Coca-Cola plantbottleTM, commercialized in Italy by Acqua Lilia, 1-L
2 “BioBottle” made from IngeoTM PLA, used by Acqua S. Anna.
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4 **Figure 3.** Plot of radon measurements over 2 months for determination of dissolved ²²⁶Ra. Errors (1
5 σ) are enclosed in the symbols.
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9 **Figure 4.** Plot of A_i/A_0 versus storage time in 1.75-L PET bottles. Data are corrected for (a) decay
10 and (b) decay and ²²⁶Ra content in water (1.00 ± 0.09 Bq/L). Modified from (Tuccimei et al., 2015).
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13 **Figure 5.** Plot of A_i/A_0 versus storage time in different volumes of PET and PLA bottles. Data are
14 corrected for decay and ²²⁶Ra content in water (1.00 ± 0.09 Bq/L). Red dashed lines indicate the
15 radon loss without correction for radium.
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19 **Figure 6.** Plot of A_i/A_0 versus storage time in 500-mL PET bottles with different thicknesses, but
20 similar surface/volume ratios. Data are corrected for decay and ²²⁶Ra content in water (1.00 ± 0.09
21 Bq/L). Data refer to 0.5-L Acqua di Nepi mineral water bottle, and blue dashed line indicates the
22 radon loss in 0.5-L Coca-Cola bottles (Fig. 5).
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28 **Figure 7.** Plot of radon loss rate versus surface/volume ratio of plastic bottles. 1- σ errors range
29 from 0.3×10^{-6} to 0.7×10^{-6} min⁻¹.
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Table 1

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

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 1σ , within 2σ , or beyond 2σ). N denotes the number of measurements in the highest activity class for each bottle type.

Air stone included in the DurrIDGE soda bottle aerator kit

Bottle	N	> 50 Bq/L			< 50 Bq/L		
		<1 σ	1–2 σ	>2 σ	<1 σ	1–2 σ	>2 σ
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

Bottle	N	> 50 Bq/L			< 50 Bq/L		
		<1 σ	1–2 σ	>2 σ	<1 σ	1–2 σ	>2 σ
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	-

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

Table 4

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

Bottle	Water Volume	Surface	Surface/Volume	Weight	Surface/Weight	Radon Loss Rate
L	L	cm ²	cm ⁻¹	g	cm ² g ⁻¹	(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

Figure 1

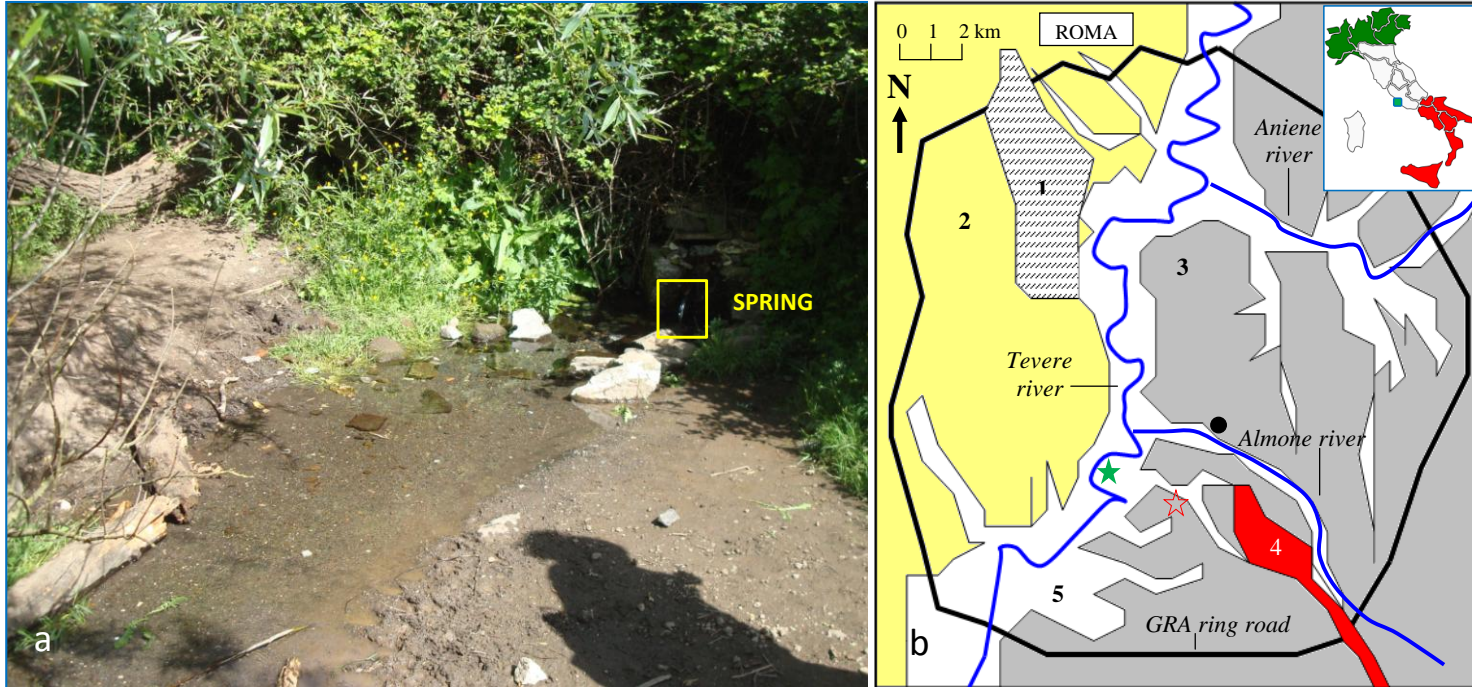
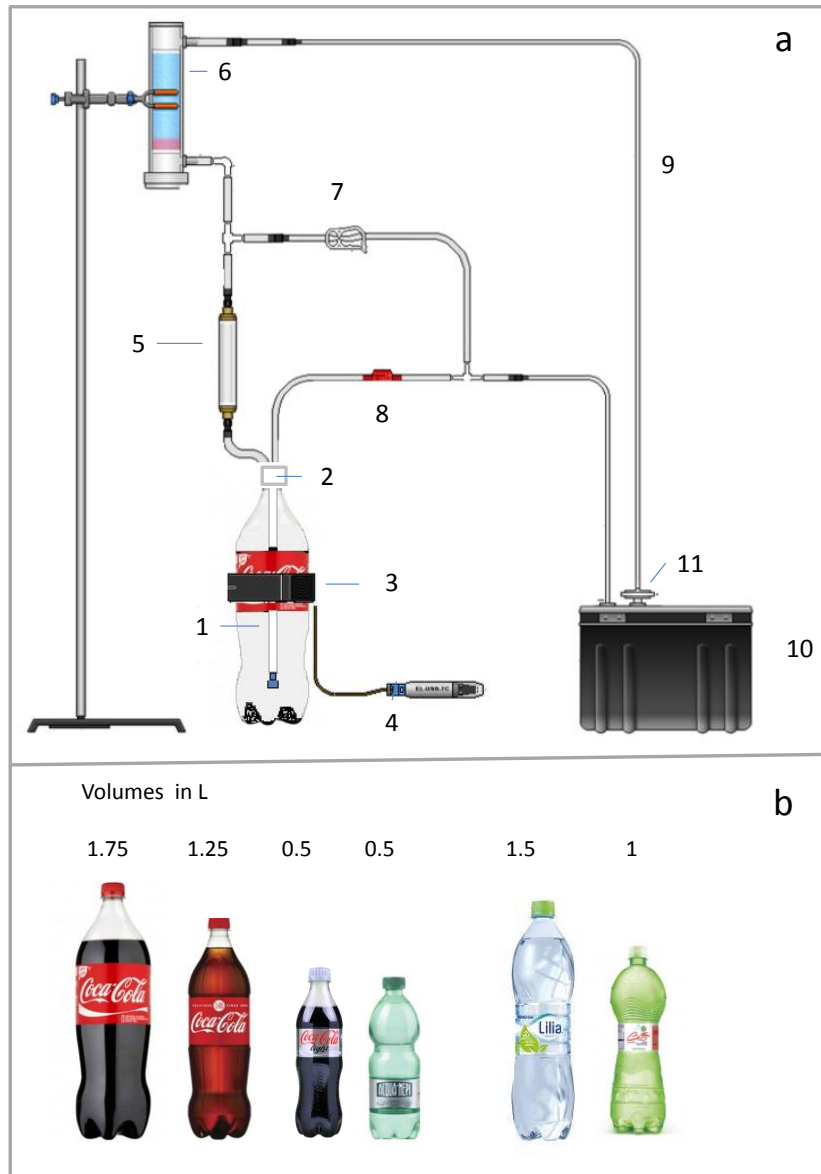


Figure 2



Volumes in L

1.75 1.25 0.5 0.5 1.5 1



b

Figure 3

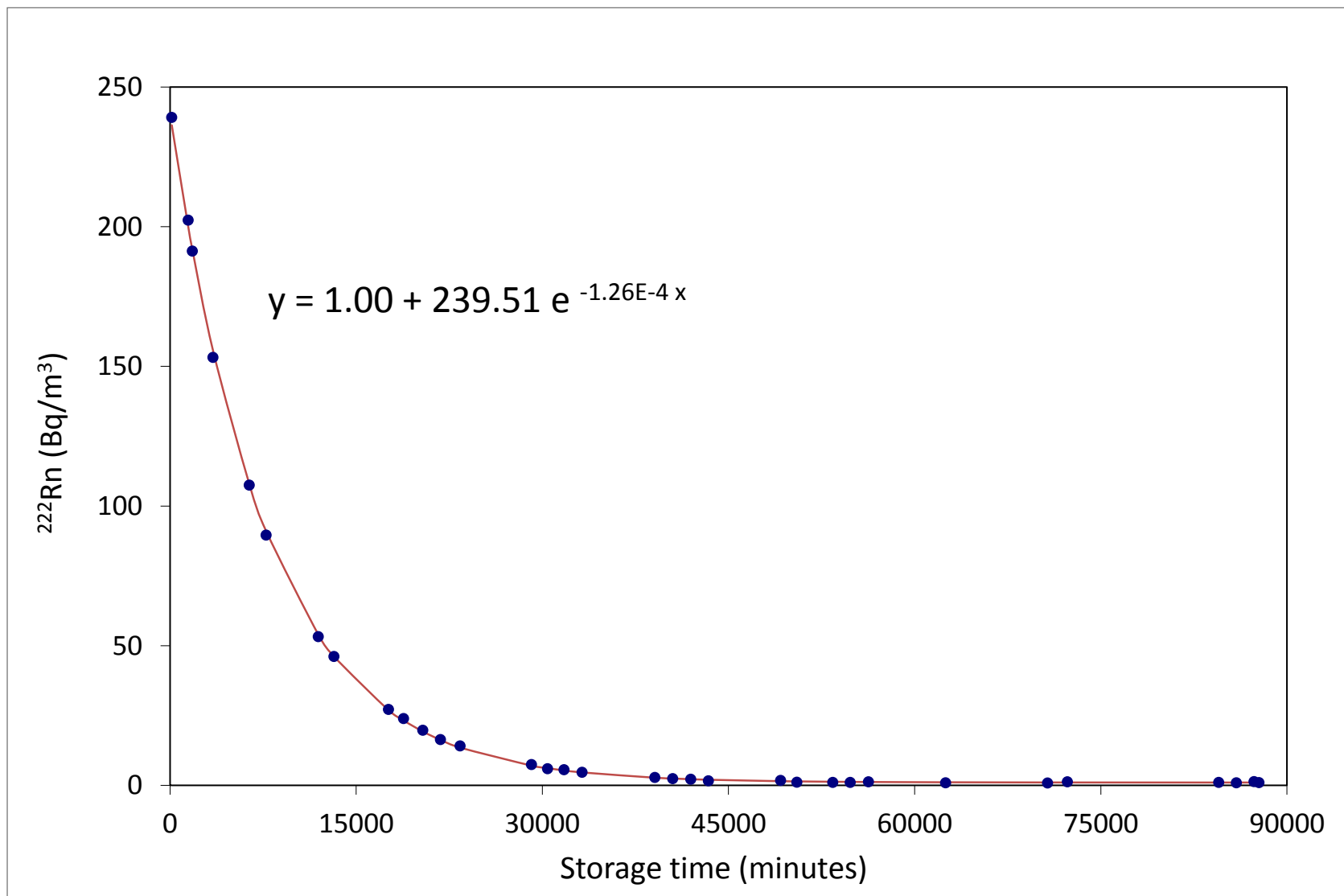


Figure 4

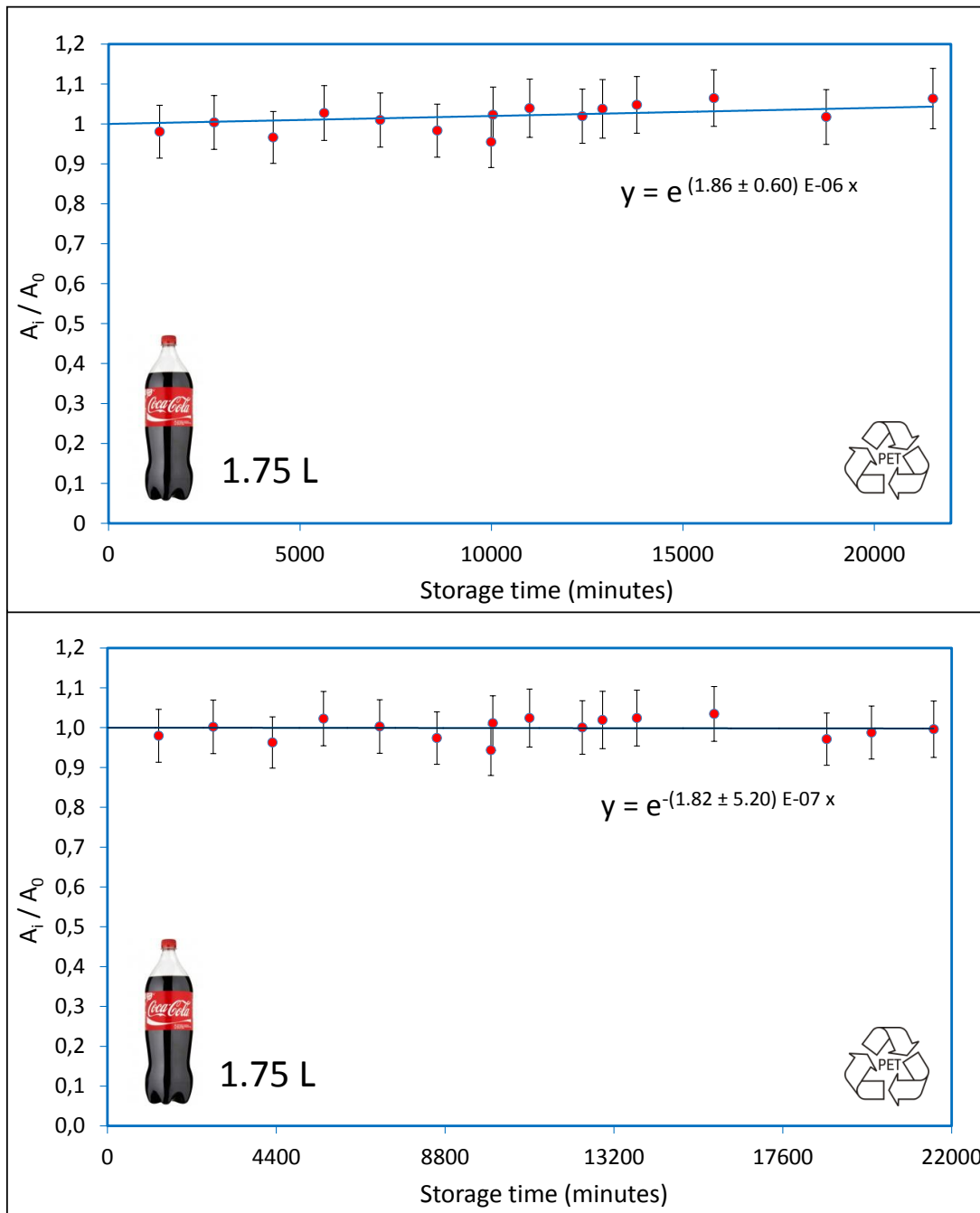


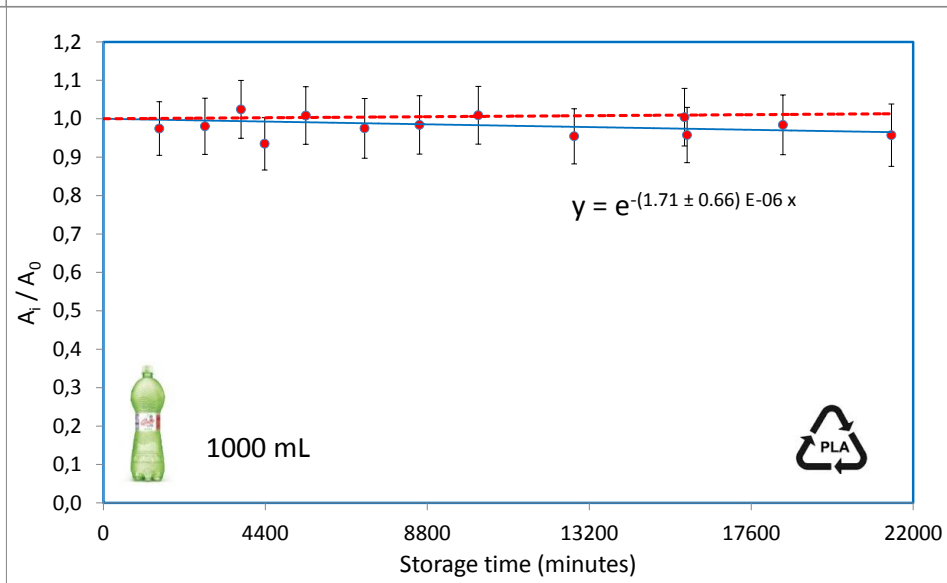
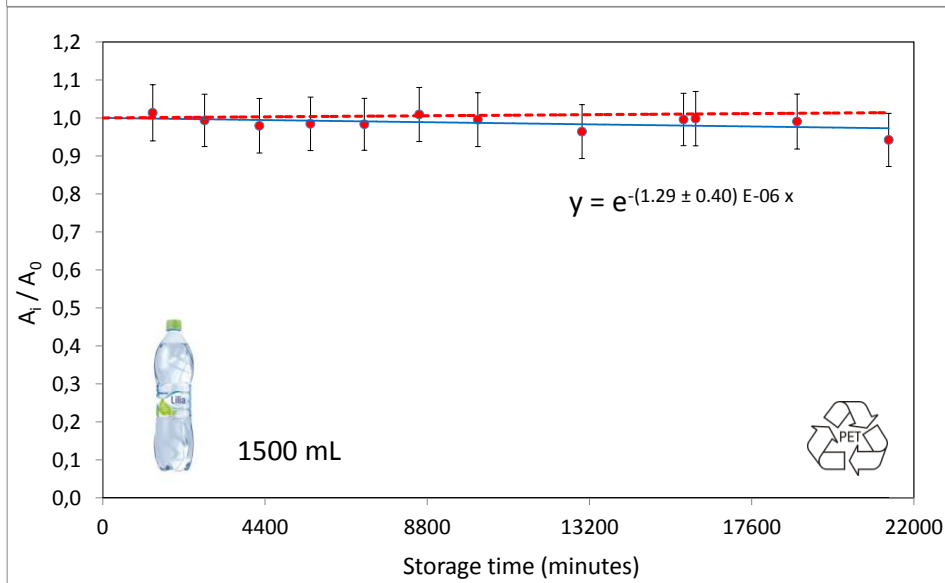
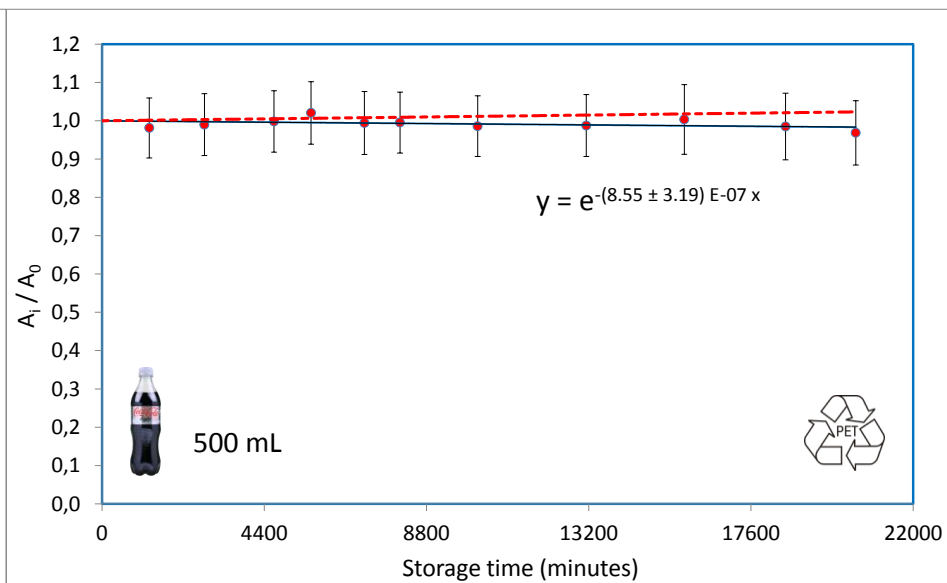
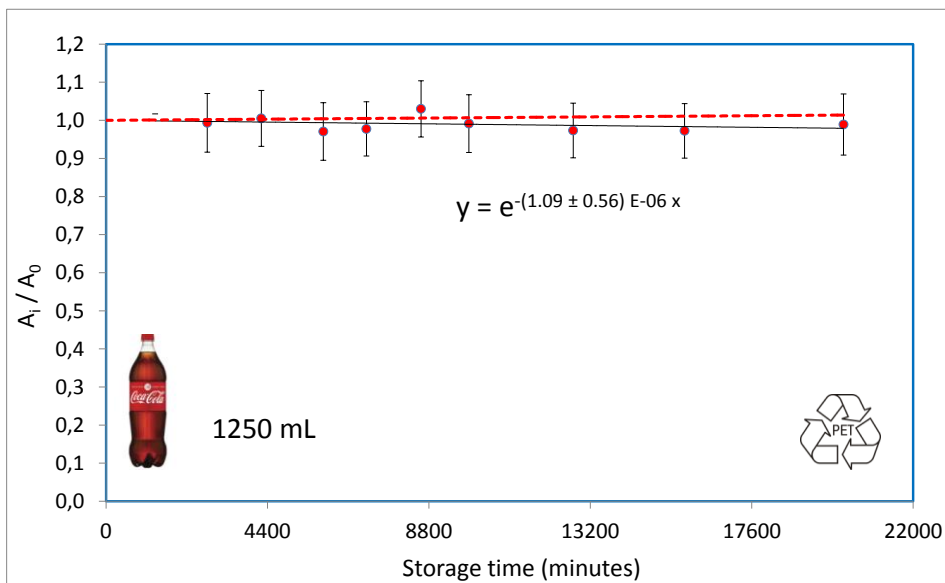
Figure 5

Figure 6

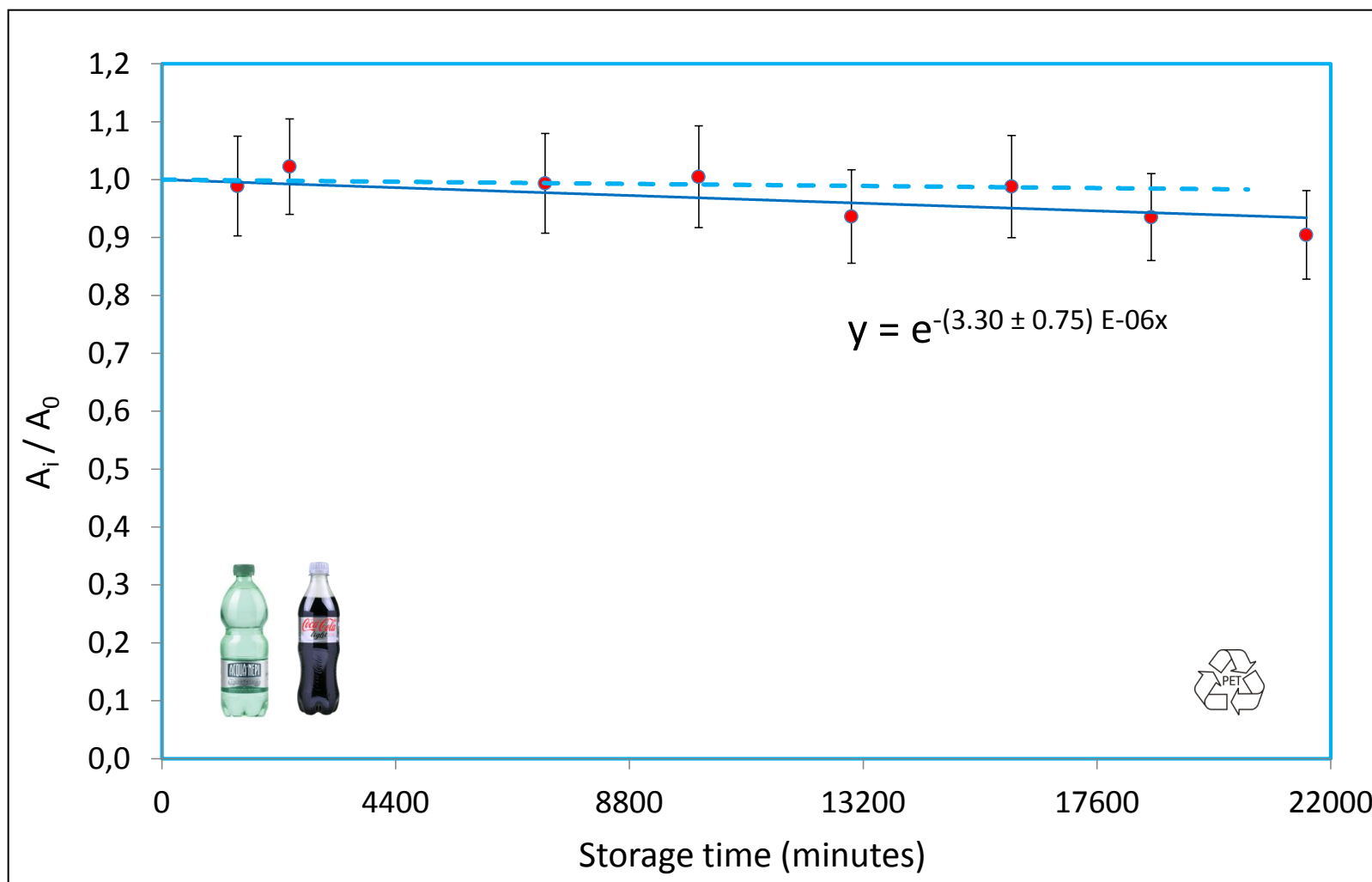


Figure 7

