Research Article

The Radiological Impact of ^{210}Pb and ^{210}Po Released from the Iron- and Steel-Making Plant ILVA in Taranto (Italy) on the Environment and the Public

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Lead-210 and ^{210}Po are naturally occurring radionuclides. Due to volatile characteristic of lead and polonium, environmental pollution of ^{210}Pb and ^{210}Po released from the coal power plant, steel-making industry and refractory material industry has been an exposure problem for the members of public. In this paper studies on the activity concentrations of ^{210}Po and ^{210}Pb in the raw materials, dust particles, surficial soils and atmospheric particulate samples collected in the area of the Iron- and Steel-Making Plant ILVA Taranto (Italy) were made. These data have been used to evaluate the source-term, distributions, inventories, mass balance, biological availability, ecological migration processes and public exposure risk of ^{210}Pb and ^{210}Po in the concerned environment.

1. Introduction

In general, the main source of ^{210}Pb and ^{210}Po in the environment is the exhalation of ^{222}Rn gas from the ground into the atmosphere. However, as a result of (i) the volatile characteristic of lead and polonium elements and (ii) the development of industries in recent decades, that is, mining, processing, and smelting of uranium, phosphate, lead and iron ore, burning of fossil fuels (coal), and burning leaded gasoline used for car engines during transportation, elevated activities of ^{210}Pb and ^{210}Po have been found in the atmosphere, hydrosphere, biosphere and surficial soil. A numerous of blood/food/dust/soil lead contamination cases have been reported [1–4]. Therefore, the artificial contamination of ^{210}Po , ^{210}Pb , and stable lead is more and more seriously affecting the public health via air inhalation and food ingestion.

Some years ago, Italian researchers started the contamination source-term survey on the natural occurring radionuclides, including uranium isotopes, thorium isotopes, radium isotopes, ^{210}Pb , and ^{210}Po [5]. The purpose of the project was mainly focused on studies of the contamination process, the exposure risk evaluation to the public, and the remedial measures for radiation protection from the radionuclides

released from the coal power plant, steel-making industry, and refractory material industry. The obtained results (Table 1) [5] showed that to lower the exposure risk to the public great attention should be given not only to the process of raw material supply and final product utility, but also to the process of by-product redistribution, high enriched waste management, and disposal of the natural occurring radionuclides in the studied industries, especially of the uranium isotopes, thorium isotopes, radium isotopes, ^{210}Pb and ^{210}Po , and so forth.

Following the warning by a press about the possible contamination of ^{210}Pb and ^{210}Po emitted from the chimney of the Iron- and Steel-Making Plant ILVA in Taranto (Italy) into the atmosphere, in October 2008, the Italian Ministry of Environment, Territory and Sea Protection asked the National Institute of Environmental Protection and Research (ISPRA) [6, 7] to prepare a detailed report to evaluate the radiological risk of the radioactive pollution arising from the plant itself. For this purpose, the ISPRA developed a program for the information acquisition, the contaminants and exposure pathway analyses, the realization of the contaminant monitoring, and the radiological impact evaluation. In the process of implementation of the program, the raw materials used in the plant, soil samples surrounding the plant, dust/ash

TABLE 1: The ^{238}U , ^{234}U , ^{235}U , ^{232}Th , ^{226}Ra , ^{228}Ra , ^{224}Ra , ^{210}Pb , and ^{210}Po concentrations (in Bq kg^{-1}) in samples collected from the coal power plant, steel-making industry, and refractory material industry in Italy in 2003 (the ^{232}Th concentrations were obtained from Zampieri et al., 2004 [5]).

Sample code	Sampling Site	Sample species	^{238}U	^{234}U	^{235}U	^{232}Th	^{226}Ra	^{228}Ra	^{224}Ra	^{210}Pb	^{210}Po
COD 1/A	Coal power plant GE	Bottom ash in winter 2001	92.0 ± 5.1	92.1 ± 5.1	4.49 ± 0.33	—	91.3 ± 4.7	74.9 ± 4.8	29.7 ± 3.4	43.6 ± 1.8	87.2 ± 4.9
COD 2/A	Coal power plant GE	Bottom ash in autumn 2000	48.2 ± 1.6	57.7 ± 1.8	2.58 ± 0.22	—	45.6 ± 2.4	29.6 ± 2.5	49.9 ± 4.7	25.7 ± 1.1	49.7 ± 3.0
COD 3/A	Coal power plant GE	Fly ash in autumn 2000	88.8 ± 4.8	92.4 ± 4.9	4.06 ± 0.64	—	73.7 ± 3.8	52.5 ± 3.8	85.8 ± 6.8	75.8 ± 3.2	74.6 ± 4.1
COD 4/A	Coal power plant Vado Ligure	Fly ash in spring 2000	154 ± 6	154 ± 6	7.83 ± 0.69	—	142 ± 7	89.1 ± 5.9	155 ± 11	143 ± 6	161 ± 9
COD 5/A	ILVA steel plant GE	Converter electrofilter dust	4.77 ± 0.24	4.77 ± 0.24	0.21 ± 0.03	—	8.08 ± 0.80	1.60 ± 0.83	2.08 ± 1.14	292 ± 13	444 ± 24
COD 6/A	ILVA steel plant GE	Blast furnace dust	10.0 ± 0.8	11.1 ± 0.9	0.40 ± 0.21	—	8.89 ± 0.87	5.45 ± 1.01	7.75 ± 1.79	43.4 ± 1.8	61.5 ± 5.0
COD 1	Refractory sepr	Zirconium sand	4215 ± 205	4175 ± 203	214 ± 17	517 ± 31	3878 ± 167	678 ± 32	1489 ± 93	2908 ± 119	3387 ± 187
COD 2	Refractory sepr	Melting furnace dust	327 ± 16	323 ± 16	13.2 ± 1.8	27.0 ± 1.6	92.9 ± 4.7	17.9 ± 1.8	46.3 ± 4.9	17076 ± 701	36953 ± 1818
COD 4	Refractory sepr	Product 1	1677 ± 78	1639 ± 77	72.4 ± 4.3	310 ± 19	1790 ± 78	397 ± 23	544 ± 38	882 ± 36	1004 ± 55
COD 5	Refractory sepr	Product 2	1629 ± 87	1655 ± 89	75.7 ± 5.1	262 ± 16	1617 ± 70	272 ± 21	405 ± 33	687 ± 28	668 ± 37
COD 6	Refractory sepr	Product 3	1776 ± 83	1753 ± 82	93.8 ± 6.5	250 ± 15	1596 ± 69	281 ± 20	423 ± 32	1198 ± 49	1015 ± 47
COD 7	Refractory sepr	Product 4	25.5 ± 1.4	24.7 ± 1.4	1.41 ± 0.15	2.00 ± 0.14	17.2 ± 1.2	6.01 ± 1.00	7.45 ± 1.67	17.6 ± 0.8	45.1 ± 2.4
COD 8	Refractory sepr	Grinding waste dust	1320 ± 57	1308 ± 56	59.1 ± 4.2	178 ± 11	1262 ± 54	203 ± 11	311 ± 26	992 ± 41	1134 ± 58
COD 9	Refractory sepr	Depuration sludge	1662 ± 76	1653 ± 75	74.3 ± 5.2	238 ± 14	1483 ± 63	235 ± 14	662 ± 52	1000 ± 41	1145 ± 61

particle deposited on the filtration systems, fly ash (dust particles) released from the chimney of the plant, and atmospheric particulate in air were collected. As part of the program, ^{210}Pb and ^{210}Po in the samples, which are volatile and very important radionuclides from radiation risk point of view, were preliminarily determined. Based on the relevant results obtained from these samples, ^{210}Po and ^{210}Pb inventory estimation and their radiological impact evaluation on members of public were made in the paper.

2. Materials and Methods

2.1. The Study Area and Sampling. Taranto city is located at a latitude (N) of $40^{\circ}28'00''$ and a longitude (E) of $17^{\circ}14'00''$ in Puglia region, the far south of Italy, with an area of 210 km^2 , 15 m above sea level, and population of 192 thousands. Meteorological data of the region collected in 30 years show that the mean annual precipitation, and minimum and maximum temperatures were 416.5 mm, $12.7 (6.0\text{--}20.9)^{\circ}\text{C}$, and $20.5 (12.2\text{--}29.9)^{\circ}\text{C}$, respectively.

The Iron- and Steel-making Plant ILVA in Taranto, just situated in the north of Taranto city and owned by the Riva Group, is the ILVA's core integrated iron- and steel-making plant and the largest plant in Europe. Production capacity is around 12 Mt of steel per year. The plant comprises five blast furnaces, AFO 1, 2, 3, 4, and 5. The iron mineral (85%), limestone (3–16%), coal (or coke: 3.5%), and lime (0.4–0.8%) are used as the principal raw materials in the plant. These materials are charged in batches into the blast furnaces at temperature of over 1000°C where the iron compounds in the ore give up excess oxygen and become liquid iron. At intervals of a few hours, the accumulated liquid iron is tapped from the blast furnace and either cast into pig iron or directed to other vessels for further steel-making operations.

Due to the fact that the raw materials always contain volatile materials, this process of iron- or steel-making has caused the environmental pollution of nitrogen oxides, sulphides, arsenide, carbon dioxide, organic compounds, heavy metals, naturally occurring radionuclides, and so forth in air, water, and soil or sediment. It was reported that in 2002 the ILVA in Taranto emitted 30.6% of total dioxin emission in Italy [8], and based on the data from National Emission Inventories and Sources (INES) the percentage of the dioxin emission from the ILVA in Taranto raised to 92% of the total dioxin emission in Italy in 2006. Mercury contamination from the ILVA in Taranto is another important environmental problem, and it was estimated that in 2005 the annual total mercury emission was over 2 t.

As far as naturally occurring radionuclides are concerned, attention should be given to uranium isotopes, thorium isotopes, radium isotopes and progenies (e.g., ^{210}Pb and ^{210}Po), and so forth. Especially, the relatively high contaminations of ^{210}Pb and ^{210}Po in air, water, and soil are often predicted to happen, due to the volatile behaviour of lead and polonium at the temperature $>1000^{\circ}\text{C}$. However, there is little information about the contamination level and exposure risk evaluation of ^{210}Pb and ^{210}Po released from the ILVA in Taranto.

For better understanding the source-term and inventory of ^{210}Pb and ^{210}Po in production process of the ILVA in Taranto, raw materials used in the plant, including lime, limestone, coal (coke), and iron minerals, were collected on May 5, 2009. For information about the redistribution and contamination level of ^{210}Pb and ^{210}Po in the environment, fly ash (dust particle) deposited in the filtration systems and dust particles emitted from the E312 chimney (latitude: $40^{\circ}28'00''\text{N}$, longitude: $17^{\circ}14'00''\text{E}$) were collected on May 5–7 2009. For the purpose of the exposure impact evaluation for the members of public, atmospheric particulate samples were collected by ISPRA in the sampling campaign of November 2008 and by ARPA Puglia in the sampling campaign May 2009. In the early days of November 2008 seven surficial soil samples were collected, in which five samples were taken in the areas around the ILVA in Taranto, one in Gioia del Colle about thirty miles northwest of Taranto and one in Castel Romano in Roma (CSM Roma). The sampling depth (0–5 cm) of all seven soil samples was the same. The sampling sites for air and soil samples were selected preferring areas of probable increased exposure on the basis of wind prevalence. Castel Romano in Roma being the Laboratory of the ISPRA was selected as a control site for both air and soil samples for comparison. The detailed information about the sampling site, strategy, date, and parameters for atmospheric particulate and soil was given in Tables 2, 3, and 4, respectively.

2.2. Apparatus and Reagents. Bismuth-210 for ^{210}Pb determination was measured by a 10-channel low-level β -counter (Berthold LB770, Germany). The instrument and reagent background of the counter for ^{210}Pb measurement is of $\leq 0.0053\text{ cps}$, and the counting efficiency was 48.2% that was calibrated with a PbSO_4 precipitate source obtained from a standard ^{210}Pb solution. Po-210 was determined by alpha spectrometry (Canberra, USA) with a counting efficiency of 31.2% and a background of $\leq 6 \times 10^{-6}\text{ cps}$ in the interested energy region.

A Perspex disk holder for polonium deposition was specially designed to fit 100–250 mL beakers [9]. Silver foil with a thickness of 0.15 mm was used for ^{210}Po spontaneous deposition, and it was cut into disks of 23 mm in diameter. Large volume ($67\text{ m}^3\text{ h}^{-1}$) air sampler was the Model Thermo G10557 equipped with analyzer of $\text{PM}_{2.5}$ or PM_{10} and with a glass-microfiber filter of dimensions of $20\text{ cm} \times 25\text{ cm}$ (Whatman GF/A cat. N. 1820-866).

Polonium-209 solution standard as a tracer for ^{210}Po determination by α -spectrometry and ^{210}Pb solution standard for β -instrument calibration, the reference material (IAEA-315) for quality control, and the BIO-RAD-AG 1-X4 resin (100–200 mesh) for lead separation were supplied by Amersham (UK), the IAEA, and the Bio-Rad Laboratories (Canada), respectively. TOPO (tri-octyl-phosphine oxide, 99%) used to isolate ^{210}Po , $\text{Pb}(\text{NO}_3)_2$ to prepare the carrier solution for lead separation, pure iron wire to prepare standard iron solution for chemical yield calculation and all other reagents were analytical grade.

TABLE 2: Characteristics of the sampling sites for atmospheric particulate (AP) collected in the sampling campaign of November 2008 (the mean (range) mass concentration of particle: 13.6 ± 8.4 (3.84–30.68) $\mu\text{g m}^{-3}$).

Sample code	Aerodynamic diameter, μm	Sampling site	Sampling date	Sampling volume, m^3	Mass concentration of particle, $\mu\text{g m}^{-3}$	Latitude (N)	Longitude (E)
AP 7	≤ 2.5	Via Machiavelli, Taranto	11-11-2008	1621.36	19.16	40°29'18.76"	17°13'33.47"
AP 8	≤ 2.5	Via Machiavelli, Taranto	12-11-2008	1632.73	13.91	40°29'18.76"	17°13'33.47"
AP 9	≤ 2.5	Via Machiavelli, Taranto	13-11-2008	1338.25	4.73	40°29'18.76"	17°13'33.47"
AP 10	≤ 2.5	Cisi, Taranto	14-11-2008	1565.65	5.87	40°29'39.74"	17°13'33.47"
AP 11	≤ 2.5	Cisi, Taranto	15-11-2008	216.03	3.84	40°29'39.74"	17°13'33.47"
AP 12	≤ 2.5	Cisi, Taranto	16-11-2008	1606.58	5.03	40°29'39.74"	17°13'33.47"
AP 1	≤ 10	Via Machiavelli, Taranto	11-11-2008	1609.99	30.68	40°29'18.76"	17°13'33.47"
AP 2	≤ 10	Via Machiavelli, Taranto	12-11-2008	1608.85	22.01	40°29'18.76"	17°13'33.47"
AP 3	≤ 10	Via Machiavelli, Taranto	13-11-2008	1331.43	9.82	40°29'18.76"	17°13'33.47"
AP 4	≤ 10	Cisi, Taranto	14-11-2008	1586.11	14.86	40°29'39.74"	17°13'33.47"
AP 5	≤ 10	Cisi, Taranto	15-11-2008	204.66	21.47	40°29'39.74"	17°13'33.47"
AP 6	≤ 10	Cisi, Taranto	16-11-2008	1590.66	11.85	40°29'39.74"	17°13'33.47"
AP 13	≤ 10	Castel Romano, Roma	19/29-11-2008	550.346	21.27	41°42'11.37"	12°26'52.77"

TABLE 3: Characteristics of the sampling sites for atmospheric particulate (AP) collected in the sampling campaign of May 2009.

Sample code	Aerodynamic diameter, μm	Sampling site	Sampling date	Sampling volume, m^3	Latitude (N)	Longitude (E)
AP14	≤ 2.5	Alto Adige, Taranto	4/5-5-2009	110.4	40°27'38.58"	17°15'49.04"
AP15	≤ 2.5	Alto Adige, Taranto	6/7-5-2009	110.4	40°27'38.58"	17°15'49.04"
AP16	≤ 2.5	Via Machiavelli, Taranto	4/5-5-2009	110.4	40°29'18.76"	17°13'33.15"
AP17	≤ 2.5	Via Machiavelli, Taranto	6/7-5-2009	110.4	40°29'18.76"	17°13'33.15"
AP18	≤ 10	Via delle Sorgenti, Statte	4/5-5-2009	110.4	40°33'44.95"	17°12'12.33"
AP19	≤ 10	Via delle Sorgenti, Statte	6/7-5-2009	110.4	40°33'44.95"	17°12'12.33"
AP20	≤ 10	Via delle Sorgenti, Statte	8/9-5-2009	110.4	40°33'44.95"	17°12'12.33"
AP21	≤ 10	Via delle Sorgenti, Statte	10/12-5-2009	165.6	40°33'44.95"	17°12'12.33"
AP22	≤ 10	San Vito, Taranto	4/5-5-2009	110.4	40°25'23.96"	17°13'31.66"
AP23	≤ 10	San Vito, Taranto	6/7-5-2009	110.4	40°25'23.96"	17°13'31.66"
AP24	≤ 10	San Vito, Taranto	8/9-5-2009	165.6	40°25'23.96"	17°13'31.66"

2.3. Microthene-TOPO Column Preparation. Six g of TOPO was dissolved by 100 mL of cyclohexane in a beaker, and 52 g of Microthene (polyethylene or polypropylene powder, 80–200 mesh) was added. The mixture was stirred for several minutes until it was homogeneous and was then evaporated to eliminate cyclohexane at 50°C. The porous powders thus obtained contain about 10.3% TOPO. A portion (2.5 g) of the Microthene-TOPO powder, slurred with 3 mL of concentrated HCl and some water, was transferred to a chromatographic column (10 mm internal diameter and 200 mm length). After conditioning with 20 mL of 1.5 M HCl, the column was ready for ^{210}Po separation.

2.4. Anion-Exchange Resin Column Preparation. The anion-exchange resin, BIO-RAD-AG 1-X4 (100–200 mesh), was sequentially treated with 6 M NaOH, 6 M HCl, and distilled water to remove any fine particles as well as other unexpected components. Twelve grams of the resin was then loaded in an

ion-exchange column (13 mm internal diameter and 200 mm length). Before use, the column was conditioned with 20 mL of 1.5 M HCl for lead separation and conditioned with 20 mL of 9.0 M HCl for Fe separation.

2.5. Methods

2.5.1. Determination of ^{210}Po and ^{210}Pb . Polonium-210 and ^{210}Pb in the normal soil samples were analyzed following the procedures reported in a literature [10]. The filtered air samples was composed of glass-fiber filter paper and dust/ash or the suspended particle from air. The analytical procedure for the air filter samples were as the same as that for normal soil samples, except for avoiding addition of 40% HF during leaching. Some samples, such as the raw materials, dust particles collected in the filtration systems, fly-ash, and so forth, showed a very high iron contents, and serious interferences for both ^{210}Pb and ^{210}Po determinations had

TABLE 4: Characteristics of the sampling sites for surficial soil (SS) samples (depth: 0–5 cm).

Sample code	Sampling site	Sampling date	Latitude (N)	Longitude (E)	Description of the sampling sites
SS1	Piazza del Gesù, Taranto	02-11-2008	40°29'39.74"	17°13'33.47"	1.6 km (S-SE) from the ILVA
SS2	Via Raimondello Orsini, Taranto	02-11-2008	40°29'46.98"	17°13'33.52"	1.3 km (S-SE) from the ILVA
SS3	Via Cannata, Taranto	02-11-2008	40°31'9.87"	17°16'15.18"	4.4 km (S-NE) from the ILVA (Paolo VI)
SS4	Via del Tratturello, Taranto	02-11-2008	40°29'39.74"	17°15'11.51"	3.0 km (NE) from the ILVA (Cisi)
SS5	Via delle Sorgenti, Statte	02-11-2008	40°33'45.00"	17°12'12.00"	6.5 km (N-NW) from the ILVA
SS6	Via Santerano, Gioia del Colle	02-11-2008	40°46'1.44"	16°52'25.57"	41 km (NW) from the ILVA (background)
SS7	Castel Romano, Roma	03-11-2008	41°42'12.15"	12°26'47.89"	Laboratory ISPRA, Roma (background)

occurred. Therefore, a modified procedure for determination of ^{210}Pb and ^{210}Po for such kinds of samples has been tested and recommended as given below.

Leaching of ^{210}Pb and ^{210}Po . Three g of sample together with 25 mg Pb^{2+} carrier, 0.025 Bq of ^{209}Po tracer, 10 mL of conc. HNO_3 , 10 mL of conc. HCl , and 10 mL of 40% HF were added to a 100 mL Teflon beaker. The beaker was heated at 250°C. Before drying, 10 mL of conc. HCl and 40% HF each were added, and the step was repeated. The solution was evaporated to incipient dryness and a 10 mL portion of 72% HClO_4 was added. The solution was evaporated to fuming to destroy the organic matter and to remove HF . The HClO_4 treatment was repeated until nearly the entire solid sample was decomposed. The residue was finally dissolved with 7 mL of conc. HCl and 40 mL of water. The obtained solution with an acidity of 1.5 M and a volume of 50 mL was filtered through a 0.1 μm Millipore filter paper.

^{210}Po Determination. Twenty percent of the leaching solution obtained from (title *Leaching of ^{210}Pb and ^{210}Po*) was put in a beaker. Then the solution was adjusted to pH 9–10 with conc. ammonia solution to coprecipitate ^{210}Po with iron (III) hydroxide. After centrifugation at 4000 rpm, the supernatant was discarded and the precipitate was dissolved with 3 mL of conc. HCl and 21 mL of water. The obtained solution was passed through a preconditioned TOPO column at a flow rate of 0.6–0.8 mL min^{-1} . After washing with 15 mL of 0.1 M HCl , polonium was eluted with 40 mL of 10 M HNO_3 at a flow rate of 0.3 mL min^{-1} . The eluant was evaporated to dryness and dissolved with 2 mL of conc. HCl and 10 mL of water. Five mL of 20% hydroxylamine hydrochloride and 5 mL of 25% sodium citrate solution were added to the obtained solution, which was then adjusted to pH 1.5 with 1:5 (v/v) ammonia. The solution was diluted to 40–50 mL, heated, and stirred on a hot-plate magnetic stirrer at 85–90°C. After disappearance of the yellow colour of Fe^{3+} (about 10 min), a Perspex holder with a silver disk was placed on the beaker and the silver disk was immersed into the solution. Any air bubbles trapped beneath the disk were removed by manipulation of the stirrer bar. The polonium deposition was continued for 4 h, and then the disk was removed, washed with distilled water and alcohol, dried, and assayed by α -spectrometry.

^{210}Pb Determination. Eighty percent of the leaching solution obtained from (title *Leaching of ^{210}Pb and ^{210}Po*) was evaporated to dryness. After cooling, 6 mL of conc. H_2SO_4 was carefully added to precipitate lead, calcium, and so forth as sulfate and heated until the white smoke appears. After cooling, about 40 mL of water was carefully added to dissolve the CaSO_4 and FeSO_4 by heating. After centrifugation at 4000 rpm, the supernatant was discarded and the PbSO_4 precipitate was dissolved with 15 mL of 6 M NH_4Ac by heating. Two mL of 0.5 M Na_2S was added, and in this case PbS was precipitated while the remaining Ca^{2+} and Mg^{2+} will remain in the solution. After centrifugation, the supernatant was discarded and the black precipitate was dissolved with 3 mL of concentrated HCl and 21 mL of distilled water by heating. The obtained solution was passed through a preconditioned anion-exchange resin column at room temperature and at a free flow rate. After washing with 40 mL of 1.5 M HCl , lead was eluted with 60 mL of distilled water at free flow rate, and the separation time of the pair $^{210}\text{Pb}/^{210}\text{Bi}$ was recorded. Two mL of conc. H_2SO_4 was added to the collected eluant, which was then evaporated until fuming to destroy the organic matters by oxidation with 1 mL of 30% H_2O_2 . Both the precipitate and the solution were centrifuged. The supernatant was discarded, and the precipitate was filtered on a weighed filter paper with a diameter of 24 mm (Whatman 42). The filter was dried at 110°C until constant weight (about 1 h) and weighed again to calculate the lead chemical yield.

Lead-210 was determined by measuring the ingrowth activity of its progeny ^{210}Bi ($T_{1/2}$: 120 h) by a low background β -counter some time after the separation (about one month of being suitable). The ^{210}Pb activity concentration ($C_{\text{Pb-210}}$) in soil sample (Bq kg^{-1}) or in air sample (Bq m^{-3}) was calculated according to the following equation:

$$C_{\text{Pb-210}} = \frac{A_{\text{Bi-210}}}{[(1 - e^{-\lambda_{\text{Bi-210}}t})\eta\gamma W]}, \quad (1)$$

where $A_{\text{Bi-210}}$ is the net count rate of ^{210}Bi (cps); λ_{Bi} , the ^{210}Bi decay constant (min^{-1}); t , is the ^{210}Bi ingrowth time after ^{210}Pb separation (min); η , is the detection efficiency for ^{210}Bi ;

y , is the chemical yield; W , is the sample weight (kg) for soil or the volume (m^3) for air.

Quality Control. Following approaches can be used to review the quality of a radioanalytical method: (1) to analyze the certified reference materials or similar matrices and to compare the obtained results with the recommended values, (2) to participate in the intercomparison activities between different international laboratories, and (3) to analyze the spiked samples.

For the purpose of quality control, the reference material IAEA-315 Marine Sediment supplied by the IAEA was used, in which the recommended value of ^{210}Pb was given. About 2 g of the reference material was analyzed following the recommended procedure of this paper. The precision was evaluated by the relative standard deviation obtained from a set of six analyses. The accuracy was assessed by the term of relative bias, which reflects the difference between the experimental mean and recommended value of ^{210}Pb activity concentration. Due to the presence of unsupported ^{210}Pb in the IAEA-315, the fraction of unsupported ^{210}Pb had to be corrected to the base date.

The obtained ^{210}Pb activity concentrations in the IAEA-315 were shown in Table 5. The mean ^{210}Pb concentration in the IAEA-315 was found to be $30.7 \pm 1.7 \text{ Bq kg}^{-1}$ (decay correction to the date of 1st January 1993). It was observed that the relative standard deviation is $\pm 5.5\%$ for ^{210}Pb . Since all being less than $\pm 10\%$ the precision for the analyses is well accepted as far as such a low activity is concerned. The relative bias obtained from the analyses was $+2.0\%$ for ^{210}Pb , showing that the mean activity concentrations of ^{210}Pb are in good agreement with the recommended value of 30.1 Bq kg^{-1} (the 95% confidence interval: $26.0\text{--}33.7 \text{ Bq kg}^{-1}$).

Due to its short half life, the reference materials for ^{210}Po are not available. The quality control for ^{210}Po analyses in this laboratory was carried out through participating in the intercomparison activities organized by the IAEA in March 29, 2007. The samples for intercomparison were a set of five water samples. The obtained activity concentrations of ^{210}Po were all in good agreement with the values given by the IAEA.

Detection Limits. Taking into account the 3σ of the blank count rates, the counting efficiencies of the instrument, the radiochemical yields, the ingrowth or decay factor (^{210}Pb : 100%) and the sample weight or volume, and the detection limit, or more precisely the minimum detectable activity (MDA) of the method for soil and air samples, are 0.25 Bq kg^{-1} and $1.7 \mu\text{Bq m}^{-3}$ for ^{210}Po and 0.73 Bq kg^{-1} and $1.7 \mu\text{Bq m}^{-3}$ for ^{210}Pb , respectively.

2.5.2. Determination of Lead in Dust Particle Samples Taken from Chimney. The procedure for stable lead separation and determination in the dust particle samples taken directly from chimney was the same as that for ^{210}Pb in soil samples, except for not adding lead carrier. The chemical yield for such dust particle sample determination was obtained through additional analyses of 16 soil samples without artificial lead contamination by addition of lead carrier. The lead

TABLE 5: Experimental values of ^{210}Pb activity concentrations (corrected to the date of 1st January 1993) in the IAEA-315 Marine Sediment*.

Sample no.	Sample weight, g	Pb yield, %	^{210}Pb , Bq kg^{-1}
IAEA-315-1	2.44352	91.7	32.7 ± 1.4
IAEA-315-2	2.47696	96.2	32.7 ± 1.4
IAEA-315-3	2.45552	88.6	29.4 ± 1.3
IAEA-315-4	2.53752	90.0	28.8 ± 1.3
IAEA-315-5	2.55880	93.4	30.9 ± 1.3
IAEA-315-6	2.52952	91.7	29.6 ± 1.3
Mean $\pm$ 1SD		91.9 ± 2.6	30.7 ± 1.7
Range	2.44352–2.47696	88.6–96.2	28.8–32.7

*The recommended value (95% confidence interval) of ^{210}Pb is 30.1 ($26.0\text{--}33.7$).

concentration was obtained from the PbSO_4 weight of the sample after weighing and chemical yield corrections. Taking into account the weighing deviation, chemical yield, and sample quantity, the estimated minimum detectable quantity for lead was 0.0032% .

2.5.3. Determination of Iron Concentration in Raw Materials

Determination of the Iron Chemical Yield. The standard iron solution ($40.0 \text{ mg Fe mL}^{-1}$) was prepared by dissolution of pure iron wire with 6 M HCl and some 30% H_2O_2 . One mL of the standard iron solution was put in a beaker, and the further treatment was done following the procedure title (*Iron Separation*). The iron chemical yield was calculated as the ratio of Fe weight in the residue over that in the standard solution taken. Four-time repeated analysis showed that the iron chemical yield of the procedure was $99.4 \pm 2.2\%$.

Leaching. Three g of raw material, 20 mL of conc. HCl, and 10 mL of 40% HF were added to an 100 mL Teflon beaker. The beaker was heated at 250°C . Before drying, 20 mL of conc. HCl and 10 mL of 40% HF each were added, and the step was repeated. The solution was evaporated to incipient dryness and a 10 mL portion of 72% HClO_4 was added. The solution was evaporated to fuming to destroy the organic matter and to remove HF. The HClO_4 treatment was repeated until nearly the entire solid sample was decomposed. The residue was finally dissolved with 15 mL of conc. HCl and 40 mL of water. The obtained solution was filtered through a $0.1 \mu\text{m}$ Millipore filter paper.

Iron Separation. A portion of the leaching solution was put in a beaker, and some conc. ammonia solution was added to precipitate iron as $\text{Fe}(\text{OH})_3$ at pH 9–10. After heating to flocculate the precipitate well, the sample was transferred to a plastic centrifugation tube and centrifuged at 4000 rpm for 5 min. The supernatant was discarded, and the iron precipitate was dissolved with 20–30 mL of conc. HCl. The obtained solution was passed through a preconditioned anion-exchange resin column at room temperature and at a free flow rate [11]. After washing with 5 mL of 9 M HCl, 30 mL of 6 M HCl + 1% H_2O_2 , and 30 mL of 4 M HCl, iron

TABLE 6: The ^{210}Po and ^{210}Pb concentrations in Bq kg^{-1} and iron concentrations in raw material samples collected from the ILVA Taranto on 5 May 2009.

Sample type	Sample weight, g	Po yield, %	^{210}Po	Pb yield, %	^{210}Pb	$^{210}\text{Po}/^{210}\text{Pb}$	Iron, %
Coke	5.27951	94.5	13.4 ± 1.3	99.1	16.4 ± 0.7	0.819	0.883 ± 0.005
Limestone	3.25234	99.5	11.7 ± 0.6	97.3	12.1 ± 0.6	0.965	0.0207 ± 0.0010
Lime	3.05868	94.7	12.2 ± 0.7	98.5	12.8 ± 0.6	0.950	0.178 ± 0.001
Iron mineral (MIN) 1	1.37134	84.8	13.3 ± 1.1	99.1	12.4 ± 0.8	1.072	65.5 ± 0.2
Iron mineral (MIN) 2	1.42930	81.6	17.9 ± 1.4	100	16.8 ± 0.9	1.067	58.6 ± 0.2
Iron mineral (MIN) 3	3.42674	73.4	3.46 ± 0.45	89.2	3.5 ± 0.3	0.991	66.6 ± 0.2
Homogenized sample	1.25528	89.5	26.6 ± 2.3	100	32.0 ± 1.5	0.833	58.1 ± 0.2
Mixture of agglomeration	1.17245	69.0	30.0 ± 2.5	100	27.7 ± 1.4	1.081	52.7 ± 0.1
Mean $\pm$ 1SD	—	85.9 ± 10.8	16.1 ± 8.6	97.9 ± 3.6	16.7 ± 9.1	0.972 ± 0.103	—
Range	1.17245–5.27951	69.0–99.3	3.46–30.0	89.2–100	3.49–32.0	0.819–1.08	0.0207–66.6

was eluted with 40 mL of 0.5 M HCl. Some conc. ammonia solution was added to the collected eluant to precipitate iron again as $\text{Fe}(\text{OH})_3$ at pH 9–10. The precipitate was filtered on an ashless filter paper (Whatman 42). The paper together with the iron precipitate was carbonised, transferred to a Muffle furnace, and burned into the iron chemical form of Fe_2O_3 at 700°C for 30 min. After cooling in a dryer, the Fe_2O_3 was weighed.

Calculation of the Iron Concentration. The Fe_2O_3 weight was corrected by the iron composition, the sample weight and chemical yield, and the iron concentration (%) in the sample was obtained. Taking into account the weighing deviation, chemical yield, and sample quantity, the estimated minimum detectable quantity for iron was 0.0030%.

3. Results and Discussion

Tables 5–11 show the activity concentrations of ^{210}Po and ^{210}Pb in the analyzed samples. The given uncertainties (1SD) were estimated taking into account the errors associated with the weighing samples, instrument calibration, yield calculation and the counting statistics of the sample and the blank sources, and so forth.

3.1. The ^{210}Po and ^{210}Pb Concentrations in Raw Material Samples. The raw material is the unique source of ^{210}Po and ^{210}Pb in the plant ILVA in Taranto. The contamination of ^{210}Po and ^{210}Pb in air is proportional to their volatile characteristics, to the activity concentrations of the ^{210}Po and ^{210}Pb in raw materials, and to the productivity of iron or steel and is inversely proportional to their adsorption characteristics on particle before filtration and to the filtration efficiency of the filtration system. The main raw materials used in the iron-/steel-making process are iron ore, and coke as a reductant and limestone and lime as flux to make a blast furnace more efficient. As shown in Table 6, the activity concentrations of ^{210}Po and ^{210}Pb in coke were low if compared with their concentrations in soil for Europe (a few Bq kg^{-1} up to hundreds of Bq kg^{-1}) [12]. It is not a surprise as the preparation process of coke could let the major part

of ^{210}Po and some parts of ^{210}Pb in coal volatilize. The data in Table 6 showed that some ^{210}Pb still remain in coke, and the secular equilibrium between ^{210}Pb and ^{210}Po has not been reached with a $^{210}\text{Po}/^{210}\text{Pb}$ ratio of only 0.819. The activity concentrations of ^{210}Po and ^{210}Pb in limestone, lime and iron minerals were also low, especially in MIN 3, but ^{210}Po and ^{210}Pb seem to be in secular equilibrium with $^{210}\text{Po}/^{210}\text{Pb}$ ratios of ~ 1 in these samples. However, The activity concentrations of ^{210}Po and ^{210}Pb in homogenized and mixture of agglomeration samples were similar and high if compared with those in the raw materials, and it was intimated that except for coke, limestone, lime, and iron minerals as raw materials there could be some other ingredients for the purpose of improving the characteristics of the iron/steel products, such as dolomite, quarts, phosphates, and so forth, which could contain much higher activities of ^{210}Po and ^{210}Pb .

3.2. The ^{210}Po and ^{210}Pb Inventory Estimation. The plant ILVA in Taranto comprises five blast furnaces. The iron minerals, limestone, lime, and coal (or coke) were used as the principal raw materials in the plant with a production capacity of around 12 Mt of steel per year. The obtained mean iron concentration in the mixture of agglomeration and homogenized samples was $55.4 \pm 3.8\%$. From Table 6, it was seen that the iron concentrations in coke, limestone, and lime were very low and their Fe contribution to the steel-making was negligible. Therefore, from the mean iron concentrations in the mixed raw materials, it was estimated that the total quantity of the raw materials consumed in the plant was 22 Mt y^{-1} . Based on the mean concentrations of ^{210}Po ($12.0 \pm 4.7 \text{ Bq kg}^{-1}$) and ^{210}Pb ($12.3 \pm 4.8 \text{ Bq kg}^{-1}$) in the raw materials and the total quantity of the raw materials consumed per year, the annual inventories of ^{210}Po and ^{210}Pb in the area of the ILVA in Taranto could be $2.64 \times 10^{11} \text{ Bq}$ and $2.71 \times 10^{11} \text{ Bq}$, respectively, if all of them were volatilized from the raw materials completely, of which some deposited in the first filtration system, some in the second filtration system, and some released from the E312 chimney and deposited in the nearby environment of the ILVA in Taranto.

TABLE 7: The ^{210}Po and ^{210}Pb concentrations in kBq kg^{-1} and lead contents in the dust particle samples collected from the first (DP1 and DP3) and second (DP2 and DP4) filtration systems of the ILVA Taranto on 16 March–5 May 2009.

Sample no.	Collection date	Sample weight, g	Po yield, %	^{210}Po	Pb yield, %	^{210}Pb	$^{210}\text{Po}/^{210}\text{Pb}$	Lead, %
DP1	16 March 2009	1.1318	73.3	10.9 ± 1.1	93.8	6.41 ± 0.26	1.70	0.470 ± 0.016
DP2	16 March 2009	1.1969	100	22.5 ± 1.0	93.8	13.5 ± 0.6	1.67	0.921 ± 0.032
DP3	5 May 2009	3.27400	100	$5.91 \pm 0.24^*$	99.4	5.44 ± 0.23	—	0.403 ± 0.002
DP4	5 May 2009	3.03932	100	$19.8 \pm 0.8^*$	99.5	18.8 ± 0.8	—	1.75 ± 0.01
Mean $\pm$ ISD	—	—	93.3 ± 13.4	—	96.6 ± 3.3	—	1.38 ± 0.36	—
Range	—	1.1318–3.2740	73.3–100	5.91–22.5	93.8–99.5	5.44–18.8	1.05–1.70	0.403–1.75

*The ^{210}Po concentrations in DP3 and DP4 sample is not representative due to the long storage time.

TABLE 8: The ^{210}Po and ^{210}Pb concentrations in kBq kg^{-1} and lead contents in the dust particle samples collected at the exit of E312 chimney of the ILVA Taranto on 5–7 May 2009.

Sample no.	Collection date	Sampling volume, m^3	Sample weight, g	Po yield, %	^{210}Po	Pb yield, %	^{210}Pb	$^{210}\text{Po}/^{210}\text{Pb}$	Lead, %
DP5	5 May 2009	0.949	0.00620	94.1	71.0 ± 3.2	93.8	28.1 ± 1.2	2.52	4.41 ± 0.22
DP6	6 May 2009	0.999	0.00424	95.9	85.6 ± 3.7	93.8	34.6 ± 1.4	2.48	5.24 ± 0.26
DP7	7 May 2009	0.978	0.00596	97.7	77.5 ± 3.6	93.8	34.5 ± 1.4	2.25	9.46 ± 0.47
Mean $\pm$ ISD	—	0.975 ± 0.025	0.00547 ± 0.00107	95.9 ± 1.8	78.0 ± 7.3	93.8	32.4 ± 3.7	2.42 ± 0.15	6.37 ± 2.71
Range	—	0.949–0.999	0.00424–0.00620	94.1–97.7	71.0–85.6	—	28.1–34.6	2.25–2.52	4.41–9.46

3.3. *The ^{210}Po , ^{210}Pb , and Lead Concentrations in the Dust Particles Collected from the Filtration Systems and the Exit of the E312 Chimney.* The raw material was charged into the blast furnaces at a temperature of over 1000°C where the iron compounds in it give up excess oxygen and become liquid iron, in the meantime where the volatile process of polonium (^{210}Po) and lead (^{210}Pb) compounds occurs due to their low melting point (mp: 254°C for polonium and 327°C for lead). The volatilized polonium and lead together with the flow air and the fly ash (dust particle) start to undergo a process of flowing-through/adsorption/filtration/deposition in the first (130 – 150°C) and second (120 – 140°C) filtration systems and releasing from the E312 chimney (100 – 130°C) and depositing in the environment of the ILVA in Taranto. At the beginning, the volatilized polonium and lead could be a state of smoke, which was then step-by-step attached on dust particles or atmospheric aerosols in the submicron size range at a suitable condition. It was reported that the average attachment times were from 40 s to 3 min at environmental temperature [13]. In Table 7, DP1 and DP3 were collected in March and May 2009 as dust particles at the first filtration system, and the contents were 5.91 – 10.9 kBq kg^{-1} of ^{210}Po , 5.44 – 6.41 kBq kg^{-1} of ^{210}Pb , and 0.403 – 0.470% of lead. DP2 and DP4 were collected at the second filtration system, and the contents were 19.8 – 22.5 kBq kg^{-1} of ^{210}Po , 13.5 – 18.8 kBq kg^{-1} of ^{210}Pb , and 0.921 – 1.75% of lead. The results showed that the concentrations of ^{210}Po , ^{210}Pb , and stable lead in the dust particles collected in the second filtration system were higher than those in the first one. This could be explained by the fact that McNeary and Baskaran [14] proposed the hypothesis that only a small portion of the aerosols scavenges effectively ^{210}Pb from the atmosphere and a major portion of the aerosols do not actively participate in the removal of these nuclides from

the air mass. The small portion of the aerosols should mean the portion of small particle size ($<1 \mu\text{m}$). Due to the fact that the particle sizes in the first filtration system are bigger than those in the second one, the adsorption rate is lowered.

DP5–DP7 in Table 8 as dust particle samples were collected in May 2009 from the exit of E312 chimney of the ILVA in Taranto using glass-fiber filter. The contents were 71.0 – 85.6 kBq kg^{-1} of ^{210}Po , 28.1 – 34.6 kBq kg^{-1} of ^{210}Pb , and 4.41 – 9.46% of lead. It was noticed that the specific concentrations of ^{210}Po , ^{210}Pb , and stable lead were the highest if compared with the relevant results obtained in the first and second filtration systems (Table 7). From the data in Tables 7 and 8, it can be seen that the activity concentrations of ^{210}Po in all the DP samples except for DP3 and DP4 were much higher than those of ^{210}Pb . This tendency could also be explained by the melting point difference between polonium and lead elements (mp: 254°C for polonium and 327°C for lead); therefore, ^{210}Po is more volatile than ^{210}Pb . The tendency in DP3 and DP4 should also be the same; unfortunately the two samples were analyzed with more than 2 years of delay after the sampling, so the activities of ^{210}Pb and ^{210}Po nearly reached the radioactive equilibrium due to the ^{210}Po decay.

The activity concentrations of ^{210}Po or ^{210}Pb as a function of the stable lead contents in all dust particle samples collected in the filtration systems and at the exit of the E312 chimney of the ILVA in Taranto were shown in Figure 1. The positive correlation illustrated that the ^{210}Po and ^{210}Pb activities in the raw materials used in the plant were proportional to their lead contents.

During the investigation, the plant was operated with only 30% capacity. Based on the working condition during 9–15 April 2008, the mean daily dust particle production was 5.00 t in the first filtration system and 1.71 t in the second one.

TABLE 9: The ^{210}Po and ^{210}Pb concentrations in Bq kg^{-1} in soil samples (depth: 0–5 cm) collected from the area of the ILVA Taranto (SS1–SS6) and from Castel Romano Roma (SS7) on 2 November 2008.

Sample no.	Sampling site	Sample weight, g	Po yield, %	^{210}Po	Pb yield, %	^{210}Pb	$^{210}\text{Po}/^{210}\text{Pb}$
SS1-1	Piazza del Gesù	3.2510	78.4	55.2 ± 3.1	91.5	52.5 ± 2.2	1.05
SS1-2	Piazza del Gesù	3.2125	93.9	49.3 ± 2.7	87.3	51.6 ± 2.2	0.955
SS2-1	Via Raimondello Orsini,	3.2095	87.0	64.7 ± 3.4	94.3	69.2 ± 2.9	0.935
SS2-2	Via Raimondello Orsini,	3.1815	101.8	60.1 ± 3.1	92.5	69.4 ± 2.9	0.866
SS3-1	Via Cannata	3.1419	90.8	86.3 ± 4.6	90.9	95.2 ± 3.9	0.906
SS3-2	Via Cannata	3.1775	97.4	85.0 ± 4.0	97.9	84.4 ± 3.5	1.01
SS4-1	Via del Tratturello	3.1980	79.2	92.3 ± 4.7	96.4	87.4 ± 3.6	1.06
SS4-2	Via del Tratturello	3.0564	96.9	88.9 ± 4.4	94.3	98.9 ± 4.1	0.899
SS5-1	Via delle Sorgenti, Statte	3.2292	93.4	140 ± 7	93.7	140 ± 6	0.998
SS5-2	Via delle Sorgenti, Statte	3.2858	100.5	130 ± 6	91.2	150 ± 6	0.863
SS6-1	Via Santerano, Gioia del Colle	3.2151	99.8	59.6 ± 3.4	88.2	66.5 ± 2.8	0.896
SS6-2	Via Santerano, Gioia del Colle	3.0566	86.6	65.5 ± 3.6	89.7	70.6 ± 2.9	0.927
Mean $\pm$ 1SD	—	—	92.1 ± 7.9	81.4 ± 28.9	92.3 ± 3.2	86.4 ± 31.4	0.947 ± 0.067
Range	—	3.0564–3.2858	78.4–101.8	49.3–140	87.3–97.9	51.6–150	0.863–1.06
SS7-1	Castel Romano, Roma	3.2114	87.4	52.8 ± 2.9	89.7	60.9 ± 2.5	0.866
SS7-2	Castel Romano, Roma	3.1919	83.2	52.5 ± 2.9	89.1	55.5 ± 2.3	0.946
Mean $\pm$ 1SD	—	—	85.3 ± 3.0	52.6 ± 0.2	89.4 ± 0.4	58.2 ± 3.9	0.906 ± 0.057
Range	—	3.1919–3.2114	83.2–87.4	52.5–52.8	89.1–89.7	55.5–60.9	0.866–0.946

Therefore, it was estimated that the mean annual deposited quantities of the dust particles were $1826 \pm 34 \text{ t y}^{-1}$ in the first filtration system and $626 \pm 7 \text{ t y}^{-1}$ in the second one. The diameter of the E312 chimney is 10.0 m, the air flow rate was 20.0 m s^{-1} and the mean content of the dust particle during sampling was 5.624 mg m^{-3} , so the estimated dust particle quantity released from the exit of the chimney was 279 t y^{-1} . According to the production quantities of the dust particles and their mean activity concentrations in the particles shown in Tables 7 and 8, the annual subinventories of ^{210}Po , ^{210}Pb , and stable lead could be $1.99 \times 10^{10} \text{ Bq}$, $1.08 \times 10^{10} \text{ Bq}$, and 7970 kg in the first filtration system, $1.41 \times 10^{10} \text{ Bq}$, $1.01 \times 10^{10} \text{ Bq}$, and 8360 kg in the second filtration system, and $2.18 \times 10^{10} \text{ Bq}$, $9.03 \times 10^9 \text{ Bq}$, and 17758 kg released from the exit of the chimney, respectively. The total inventories estimated in this way were $5.58 \times 10^{10} \text{ Bq y}^{-1}$ of ^{210}Po , $3.00 \times 10^{10} \text{ Bq y}^{-1}$ of ^{210}Pb , and 34089 kg of stable lead. From this data it was seen that 23.4–36.1% of ^{210}Po , ^{210}Pb and lead were deposited in the first filtration system, 24.5–33.8% in the second filtration system, and 30.1–52.9% emitted from the E312 chimney and deposited in the environment of the ILVA in Taranto. If the plant was in operation with full capacity, the total inventory could be $1.86 \times 10^{11} \text{ Bq y}^{-1}$ of ^{210}Po , $9.99 \times 10^{10} \text{ Bq y}^{-1}$ of ^{210}Pb , and $1.14 \times 10^5 \text{ kg}$ of Pb. These estimated inventories of ^{210}Po and ^{210}Pb were nearly in the same order of magnitude with that ($2.64 \times 10^{11} \text{ Bq}$ of ^{210}Po and $2.71 \times 10^{11} \text{ Bq}$ of ^{210}Pb) estimated from the raw materials in Section 3.2.

3.4. Polonium-210 and ^{210}Pb Concentrations in Surficial Soil Samples. (i) Scavenging in convective updraft, (ii) scavenging by large-scale precipitation, and (iii) dry deposition are

FIGURE 1: The activity concentrations of ^{210}Po or ^{210}Pb as a function of the Pb contents in the dust particle samples collected in the filter and chimney of the ILVA Taranto.

the three major mechanisms by which the scavenging of tropospheric ^{210}Pb and ^{210}Po takes place. Surficial (0–5 cm) soil sample is a good indicator of the ^{210}Pb and ^{210}Po deposition in air. As shown in Table 9, the activity concentrations in the collected surficial soil samples were in the range of $49.3\text{--}140 \text{ Bq kg}^{-1}$ for ^{210}Po and $51.6\text{--}150 \text{ Bq kg}^{-1}$ for ^{210}Pb . The reported values in soil by UNSCEAR [12] for Europe were from a few Bq kg^{-1} up to hundreds of Bq kg^{-1} . Was there any man-made contamination of ^{210}Po and ^{210}Pb in soil or not? It is really difficult to evaluate the obtained data only by comparison with the reported values with a wide variation for Europe. Therefore, it is very important to select a control

site, which is far from the sources of artificial contamination and geologically similar to the investigated sites in the ILVA in Taranto (SS1-SS6). The Laboratory of the ISPRA—Castel Romano Roma (SS7) was selected as an ideal site for control. The data in Table 9—showed that (i) the ^{210}Po and ^{210}Pb activity concentration in SS7 as a general background value were 52.6 ± 0.2 and $58.2 \pm 3.9 \text{ Bq kg}^{-1}$, respectively; (ii) except for SS1, the elevated ^{210}Po and ^{210}Pb activity concentrations in all other samples (SS2-SS6) at the ILVA in Taranto were observable and the mean concentrations of ^{210}Po and ^{210}Pb were 1.55 and 1.48 times as high as those at the control site; (iii) the highest values were found in SS5 with the mean ^{210}Po and ^{210}Pb concentrations of 135 ± 7 and $145 \pm 7 \text{ Bq kg}^{-1}$ which were about 2.57 and 2.49 times as high as those at the control site, respectively; (iv) it seems that the activity concentrations of ^{210}Po were a little bit less than those of ^{210}Pb , but they were nearly in equilibrium with a mean $^{210}\text{Po}/^{210}\text{Pb}$ ratio of 0.947 ± 0.067 .

The accumulation and mobility of ^{210}Po and ^{210}Pb in the terrestrial environment due to atmospheric fallout vary with the geographic locations and depend not only on physico-chemical properties of the radionuclides themselves but also on (i) climatic conditions, such as rainfall, temperature, humidity, wind direction and speed, and biological activity of microorganisms in soil and (ii) human activities, such as cultivation, irrigation, and fertilization [15, 16]. The deposition of ^{210}Po and ^{210}Pb from the smoke plume released from a chimney to surficial soil is a comprehensive process. In this study, the E312 chimney of the ILVA in Taranto was considered as the zero point (release point). After carefully fitting the data in Table 9 with the least square regression method, it was found that the relationships between the activity concentrations of ^{210}Po and ^{210}Pb in superficial soils and distance of the sampling sites from the release point can be well described by equations of polynomial (Figure 2). From Figure 2, it was observed that (i) the activity concentrations of ^{210}Po and ^{210}Pb in the soils are increasing with the increase of the distance within 20 km and (ii) the activity concentrations can be considered as background level beyond 40 km. Therefore, from Figure 2, it was predicted that the maximum activity concentrations of ^{210}Po and ^{210}Pb in the territory could occur at about 20 km from the release point. These data can also provide useful information for the reasonable site selection and distribution for the future radiological survey. The annual precipitation rate in Puglia region is much less than other regions of Italy; therefore, dry deposition could play an important role for the long distance transportation of ^{210}Po and ^{210}Pb released from the ILVA in Taranto.

3.5. Polonium-210 and ^{210}Pb Concentrations in Atmospheric Particulate. The atmospheric particulate samples in the first sampling campaign were taken with high volume samplers on 11 to 17 November 2008 at two sites located around the plant ILVA in Taranto, that is, Via Machiavelli and Cisi. At each site, two kinds of atmospheric particulate samples were collected, one with an atmospheric particulate mass concentration in

FIGURE 2: The activity concentration of ^{210}Po and ^{210}Pb as a function of the distance (D) between the ILVA Taranto and sampling site.

the fraction of an aerodynamic diameter $\leq 10 \mu\text{m}$ (PM_{10}), and another in the fraction of $\leq 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$).

As indicated in Table 10, the obtained activity concentrations of ^{210}Po in samples of $\text{PM}_{2.5}$ and PM_{10} were in the range of $45.8\text{--}214 \mu\text{Bq m}^{-3}$ and $43.1\text{--}226 \mu\text{Bq m}^{-3}$, that of ^{210}Pb in the range of $298\text{--}1054 \mu\text{Bq m}^{-3}$ and $331\text{--}1099 \mu\text{Bq m}^{-3}$, and the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios in the range of $0.133\text{--}0.231$ (mean: 0.172 ± 0.037) and $0.118\text{--}0.217$ (0.165 ± 0.040), respectively. The corresponding mean values of atmospheric particulate samples (PM_{10}) at the control site taken on 19–29 November 2008 were $48.5 \mu\text{Bq m}^{-3}$ for ^{210}Po , $399 \mu\text{Bq m}^{-3}$ for ^{210}Pb , and 0.122 for the $^{210}\text{Po}/^{210}\text{Pb}$ ratio.

At the first glance of the data in Table 10 and the weather record, the activity concentrations of ^{210}Po and ^{210}Pb were highly variable and similar to the mass concentration variation of the atmospheric particulate [7], in particular, depending on the variability of weather conditions encountered during the sampling period. In the raining days less particulates were collected, and, therefore, also lower activity concentrations of ^{210}Po and ^{210}Pb were detected.

In the UNSCEAR [12] reports that the reference concentrations in air are about $50 \mu\text{Bq m}^{-3}$ (ranged from 12 to $80 \mu\text{Bq m}^{-3}$) for ^{210}Po and $500 \mu\text{Bq m}^{-3}$ (ranged from 28 to $2250 \mu\text{Bq m}^{-3}$) for ^{210}Pb , respectively, and they are sites specific. It was reported that the yearly average concentrations of ^{210}Pb in surface air over Europe were about $200\text{--}700 \mu\text{Bq m}^{-3}$. Therefore, the obtained concentrations of ^{210}Po and ^{210}Pb in this study were in well agreement with the reported values.

Lead-210 and ^{210}Po in atmosphere come from several sources: (i) from volcanic dust [17, 18], (ii) from ^{222}Rn gas which is exhaled from the ground into the atmosphere [19, 20], (iii) from resuspended soil particles (see [21–24]), and (iv) from widespread dispersal of phosphate fertilizers, fossil fuel combustion, biomass burning, and industrial processes

TABLE 10: The ^{210}Po and ^{210}Pb activity concentrations in atmospheric particulate collected from the area of the ILVA Taranto (AP 1-12) on 11-17 November 2008 and from Castel Romano Roma (AP 13) on 19-29 November 2008.

Sample code	Sampling site	Aerodynamic diameter, μm	Po yield, %	^{210}Po , $\mu\text{Bq m}^{-3}$	^{210}Po , kBq kg^{-1}	Pb yield, %	^{210}Pb , $\mu\text{Bq m}^{-3}$	^{210}Pb , kBq kg^{-1}	$^{210}\text{Po}/^{210}\text{Pb}$
AP7	Via Machiavelli	≤ 2.5	92.5	130 ± 6	6.79 ± 0.29	84.5	975 ± 41	50.9 ± 2.2	0.133
AP8	Via Machiavelli	≤ 2.5	96.9	214 ± 9	15.4 ± 0.7	85.2	1054 ± 45	75.8 ± 3.2	0.203
AP9	Via Machiavelli	≤ 2.5	90.2	83.5 ± 4.2	17.7 ± 0.9	91.0	362 ± 15	76.4 ± 3.3	0.231
AP10	Cisi	≤ 2.5	99.8	75.0 ± 3.7	12.8 ± 0.6	90.0	464 ± 20	79.1 ± 3.4	0.162
AP11	Cisi	≤ 2.5	96.6	65.9 ± 6.8	17.2 ± 1.8	90.3	444 ± 20	116 ± 5	0.148
AP12	Cisi	≤ 2.5	96.7	45.8 ± 2.7	9.11 ± 0.5	82.8	298 ± 13	59.2 ± 2.5	0.154
AP1	Via Machiavelli	≤ 10	91.2	128 ± 7	4.18 ± 0.22	94.8	1083 ± 46	35.3 ± 1.5	0.118
AP2	Via Machiavelli	≤ 10	96.2	226 ± 11	10.3 ± 0.5	91.4	1099 ± 46	49.9 ± 2.1	0.206
AP3	Via Machiavelli	≤ 10	100.0	77.9 ± 3.8	7.93 ± 0.39	95.8	358 ± 15	36.5 ± 1.6	0.217
AP4	Cisi	≤ 10	93.0	81.5 ± 3.9	5.49 ± 0.26	91.4	484 ± 21	32.5 ± 1.4	0.169
AP5	Cisi	≤ 10	91.6	77.2 ± 7.5	3.60 ± 0.35	90.3	520 ± 23	24.2 ± 1.1	0.149
AP6	Cisi	≤ 10	100.1	43.1 ± 2.0	3.63 ± 0.17	87.2	331 ± 14	27.9 ± 1.2	0.130
Mean $\pm$ ISD		—	95.4 ± 3.6	104 ± 60	9.50 ± 5.18	89.6 ± 4.0	623 ± 325	55.3 ± 27.2	0.168 ± 0.037
Range		—	90.2–100.1	43.1–226	3.60–17.7	82.8–95.8	298–1099	24.2–116	0.118–0.231
AP 13	Castel Romano, Roma	≤ 10	96.7	48.5 ± 3.3	2.28 ± 0.15	84.2	399 ± 17	18.7 ± 0.8	0.122

including mining and smelting of uranium, phosphate, lead, and iron ore [20, 25]. The first three categories are natural sources, while the last is artificial one, that is, man-made contamination. The global mass balance calculation for the atmospheric ^{210}Pb or ^{210}Po was done by a number of researchers. Lambert et al. [26] estimated a global volcanic ^{210}Po output of $2.4 \times 10^{15} \text{ Bq y}^{-1}$, with a median $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio of 40, resulting in a ^{210}Pb contribution of $6.0 \times 10^{13} \text{ Bq y}^{-1}$. Wilkening et al. estimated the ^{210}Pb flux due to ^{222}Rn emanation from all the continental area of the world to be $3.50 \times 10^{16} \text{ Bq y}^{-1}$, and the corresponding value from surface water (ocean, rivers, and lakes) to be $9.5 \times 10^{14} \text{ Bq y}^{-1}$ [27, 28]. Robbins [29] estimated the ^{210}Pb global flux (assuming ^{210}Po and ^{210}Pb are in equilibrium) from resuspended dust to be $3.3 \times 10^{14} \text{ Bq y}^{-1}$, while the atmospheric inventory of ^{210}Pb or ^{210}Po from burning of coal and the widespread dispersal of phosphate fertilizers in land areas around the globe along with the gypsum byproducts of fertilizer production was estimated to be about $3.7\text{--}7.4 \times 10^{14} \text{ Bq y}^{-1}$. This investigation (Section 3.2) indicated that the emission from the ILVA in Taranto to the atmosphere is maximum $2.64 \times 10^{11} \text{ Bq y}^{-1}$ of ^{210}Po and $2.71 \times 10^{11} \text{ Bq y}^{-1}$ of ^{210}Pb .

It was said that the concentrations of ^{210}Po in air are extremely high for weeks and in some cases months before major volcanic eruption [21, 23]. Shortly after the eruption stops, over three orders of magnitude decrease in the activity of ^{210}Po in air even at >1300 km from the eruption site. Due to its volatility, ^{210}Po is highly enriched in volcanic gases with $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios up to 600 (57 to 614) in the Stromboli's plume which has been reported [30]. As far as the case of ILVA in Taranto is concerned, the affection from the source of volcanic eruption was not considered, as there was no any volcanic eruption in the investigation period in the studied region and nearby. In fact, the ^{222}Rn gas exhalation (the second category) is the most important source contribution of ^{210}Po and ^{210}Pb in the obtained samples, which can be confirmed by the similar concentrations of ^{210}Po and ^{210}Pb and the $^{210}\text{Po}/^{210}\text{Pb}$ ratios in some samples if compared to that at the control site. As shown in Table 10, the activity concentrations of ^{210}Po and ^{210}Pb found in the fraction of PM_{10} did not differ from the corresponding values found in the fractions of $\text{PM}_{2.5}$; thus, it was showed that almost all of ^{210}Po and ^{210}Pb were found only in the fraction of $\text{PM}_{2.5}$. This conclusion is consistent with that reported by Martell and Moore [31], who observed that 90% of ^{210}Pb ion aerosols are associated with particles $\leq 0.3 \mu\text{m}$.

Studies showed that, when inhaled, smaller particles can be more toxic than a comparable mass of larger particles of the same material, as the health effects are directly linked to their bigger particle surface area and higher solubility [32]. Due to the fact that (i) the sizes of the resuspended soil particle in most cases are $>2 \mu\text{m}$ and the $^{210}\text{Po}/^{210}\text{Pb}$ ratios are high and in the range of 0.77–1.0 and (ii) the $^{210}\text{Po}/^{210}\text{Pb}$ ratios in the collected samples are much less than 0.77–1.0, therefore, it was speculated that the ^{210}Po and ^{210}Pb contribution from the resuspended soil particles (the third category) in the collected samples (Table 10) were negligible.

According to the literature [33], the atmospheric residence times of the fine aerosols are 33–66 days, and the calculated $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios in air samples should be ≤ 0.092 . In fact, from the sampling to measuring, 10–20 days are needed in the study, and taking into account the ^{210}Po decay and ingrowth from ^{210}Pb in that period, the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio of 0.122 obtained at the control site is reasonable. If 0.122 was taken as the reference value, it was seen that the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios in Table 10 were only a little bit higher than the reference value in nearly all the atmosphere particulate samples collected from the ILVA in Taranto. The data indicated that there could exist a possibility of man-made dust contamination of ^{210}Po and ^{210}Pb in the area of ILVA in Taranto, and the contamination could be as a result of dust containing ^{210}Po and ^{210}Pb emitted from the plant chimney during the steel-making process, but the possibility seems not significant. The data also showed that ^{210}Po and ^{210}Pb activity concentrations at the nearer site to the chimney (Via Machiavelli) were higher than those at the farther site (Cisi), and the maximum contaminations of ^{210}Po and ^{210}Pb in the ILVA in Taranto samples were about 4.66 and 2.75 times as high as those at the control site, respectively. Moreover, correlation analysis (Figure 3.) showed that a positive correlation was found between the ^{210}Po and ^{210}Pb concentrations in atmospheric particulate collected at the site ILVA in Taranto, and their relation can be expressed as $[^{210}\text{Po}, \text{ in } \mu\text{Bq/m}^3] = 0.165[^{210}\text{Pb}, \text{ in } \mu\text{Bq/m}^3] + 1.72 \mu\text{Bq/m}^3$, ($R^2 = 0.793$, $n = 12$, $P < 0.01$).

As shown in Table 2, the mass concentration of particle in air samples collected in the ILVA in Taranto on 11–17 November 2008 ranged from 3.84 to $30.68 \mu\text{g m}^{-3}$ with a mean value of $13.6 \pm 8.4 \mu\text{g m}^{-3}$. The obtained value was much lower than the annual mean values of $42 \mu\text{g m}^{-3}$ at the site of Machiavelli and $34 \mu\text{g m}^{-3}$ in Cisi obtained by the Monitoring Center of Taranto. The lower value was attributed to a number of factors: (i) during the sampling period, the wind direction was changed frequently and it was not possible to relocate quickly the equipment for collecting particulates in a short time; therefore, the equipment was not in the well position of downwind of the plant emission; (ii) during the sampling period, namely, on days 12, 13, and 14 November 2008, the plant was operated with only one production line of two and partially reduced the emission of particles; (iii) during the sampling period, two or three days of raining occurred and the process could surely affect the scavenge of the particulates emitted from the chimney of the plant and reduce the particulate concentration in air. As shown in Figure 4, there exist positive correlations between the mass concentrations of particulate and ^{210}Po or ^{210}Pb concentrations in the air samples collected in the ILVA in Taranto on 11–17 November 2008. Therefore, based on the routine monitoring results of the mass concentrations of particulate in air in the area of the ILVA in Taranto, it is predicted that higher concentrations of ^{210}Po and ^{210}Pb derived from the industrial activity in the area in routine could be possible.

The mass specific activity concentrations of ^{210}Po and ^{210}Pb in the atmospheric particulate were also given in

FIGURE 3: Correlation between ^{210}Po and ^{210}Pb concentrations in atmospheric particulate collected at site ILVA Taranto.

FIGURE 4: The correlation between the mass concentration of particulate (PM) and ^{210}Po or ^{210}Pb concentrations in the air samples collected in ILVA Taranto in 11–17 November 2008.

Table 10. It is seen that the mass specific activity concentrations of ^{210}Po and ^{210}Pb in the samples collected in the area ILVA in Taranto were in the range of 3.60–17.7 (mean: 9.50 ± 5.18) and 24.2–116 (mean: 55.3 ± 27.2) kBq kg^{-1} , respectively. The corresponding values in the control site (Castel Romano, Roma) were 2.28 ± 0.15 and $18.7 \pm 0.8 \text{ kBq kg}^{-1}$, respectively. Compared with the mean activity concentrations in soil in the corresponding sampling sites, the ratios of the mass activity concentration in the atmospheric particulate and that in the top soil were 44.2–217 (mean: 117 ± 64) for ^{210}Po and 280–1340 (mean: 640 ± 315) for ^{210}Pb . It was observed that the mass specific activity concentrations of ^{210}Po and ^{210}Pb in the atmospheric particulate are one to three order of magnitude higher than that found in the top soil. Therefore, the efficiency of both ^{210}Po and ^{210}Pb entrainment into the atmospheric aerosols is very high, especially ^{210}Pb .

For further investigation, the second sampling campaign for atmospheric particulate samples at 4 sites in the ILVA in Taranto was carried out in 4–12 May 2009. As shown in

Table 11, the obtained activity concentrations of ^{210}Po in samples of $\text{PM}_{2.5}$ and PM_{10} were in the range of 154–564 $\mu\text{Bq m}^{-3}$, those of ^{210}Pb in the range of 618–1087 $\mu\text{Bq m}^{-3}$, and the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios in the range of 0.231–0.543 (mean: 0.343 ± 0.097). The maximum concentrations of ^{210}Po and ^{210}Pb were about 11.6 and 2.72 times as high as those at the control sites, respectively. It seems that a stable contamination factor for ^{210}Pb was found and that for ^{210}Po was much higher compared with the corresponding data in the first sampling campaign.

3.6. Estimation of the Committed Effective Dose for Members of the Public due to Inhalation of Air Contaminated with ^{210}Po and ^{210}Pb . The risk evaluation of ^{210}Po and ^{210}Pb for the general public could involve two aspects, that is, radiological and biological toxicities. In this paper only the radiological toxicity is evaluated. The possible radiation pathway could include the external exposure from cloud plume and ground and internal exposure due to inhalation of air and digestion of food and water contaminated with ^{210}Po and ^{210}Pb . Due to the fact that ^{210}Po is a pure α -emitter and ^{210}Pb is a weak β -emitter, internal exposure through inhalation of the contaminated air in this case could be a very important pathway for individual of the public. For estimation of the annual committed effective dose (E , Sv y^{-1}) for members of the public due to inhalation, the following equation was used:

$$E = \sum_j A_j R T C_{j,\text{inh}}, \quad (2)$$

where j is ^{210}Po or ^{210}Pb ; A_j is the mean or maximum activity concentration of ^{210}Po or ^{210}Pb in atmospheric particulate, Bq m^{-3} ; R is the mean respiration rate for individual of different age group [34], $\text{m}^3 \text{d}^{-1}$; T is the exposure time, 365 d y^{-1} ; $C_{j,\text{inh}}$, the age-dependent dose coefficient for inhalation of particulate aerosols containing ^{210}Po or ^{210}Pb . $C_{j,\text{inh}}$ also depends on the absorption rate of the materials inhaled. In general consideration, there could exist three types of ^{210}Po or ^{210}Pb materials in the taken atmospheric particulate samples, that is, F: fast absorption type, M: moderate type and S: slow type. In this study for sure to obtain the conservative dose results, the $C_{j,\text{inh}}$ value for the material of slow type was selected for the dose evaluation [35].

Based on the mean or maximum concentrations of ^{210}Po and ^{210}Pb in the samples collected on 19–29 November 2008 in Castel Romano Roma and on 11–17 November 2008 as well as in 4–12 May 2009 at sites of the ILVA in Taranto, the estimated annual committed effective doses for different age group of the public were given in Tables 12–14.

Only taking into account the adult population, from Table 12 it was seen that (i) the mean committed effective doses due to intake of ^{210}Po and ^{210}Pb from the atmospheric particulate at the control site (Castel Romano Roma) were 1.69 and 18.1 $\mu\text{Sv y}^{-1}$, respectively, and with a total dose of 19.8 $\mu\text{Sv y}^{-1}$ (ii) the dose from ^{210}Pb was 10.7 time as high as that from ^{210}Po , and (iii) the doses received for all age groups were nearly in the same level.

TABLE 11: The ^{210}Po and ^{210}Pb concentrations in $\mu\text{Bq m}^{-3}$ in atmospheric particulate (AP) collected from the area of the ILVA Taranto on 4–12 May 2009.

Sample code	Sampling site	Aerodynamic diameter, μm	Po yield, %	^{210}Po	Pb yield, %	^{210}Pb	$^{210}\text{Po}/^{210}\text{Pb}$
AP14	Alto Adige	≤ 2.5	100.3	246 ± 15	95.5	730 ± 32	0.337
AP15	Alto Adige	≤ 2.5	86.2	221 ± 13	100	655 ± 29	0.337
AP16	Via Machiavelli	≤ 2.5	100	217 ± 12	100	618 ± 27	0.352
AP17	Via Machiavelli	≤ 2.5	71.9	154 ± 11	100	662 ± 29	0.233
AP18	Via delle Sorgenti, Statte	≤ 10	88.5	193 ± 12	95.1	647 ± 28	0.299
AP19	Via delle Sorgenti, Statte	≤ 10	96.9	171 ± 13	91.0	739 ± 32	0.231
AP20	Via delle Sorgenti, Statte	≤ 10	90.8	265 ± 16	100.9	986 ± 42	0.268
AP21	Via delle Sorgenti, Statte	≤ 10	94.5	564 ± 28	100	1038 ± 44	0.543
AP22	San Vito	≤ 10	90.3	—	92.2	766 ± 33	—
AP23	San Vito	≤ 10	70.5	399 ± 23	76.7	1005 ± 43	0.396
AP24	San Vito	≤ 10	97.5	475 ± 23	100.4	1087 ± 45	0.437
Mean $\pm$ ISD		—	89.8 ± 10.3	291 ± 140	95.6 ± 7.2	812 ± 179	0.343 ± 0.097
Range		—	70.5–100.3	154–564	76.7–100.9	618–1087	0.231–0.543

As shown in Table 13, the mean committed effective doses for adult due to intake of ^{210}Po and ^{210}Pb from the atmospheric particulate at the sites of the ILVA in Taranto on 11–17 November 2008 were 3.62 and $28.3 \mu\text{Sv y}^{-1}$, respectively, and with a total dose of $31.9 \mu\text{Sv y}^{-1}$, and they were 1.6–2.1 times as high as that at the control site. Based on the maximum concentrations, the dose from ^{210}Po and ^{210}Pb could be 7.87 and $49.9 \mu\text{Sv y}^{-1}$, respectively, and with a total dose of $57.7 \mu\text{Sv y}^{-1}$. Therefore, about $12.1 \mu\text{Sv y}^{-1}$ at mean and $37.9 \mu\text{Sv y}^{-1}$ at maximum could be attributed to the ^{210}Po and ^{210}Pb emitted from the chimney of the ILVA in Taranto, of which about 84% were the contribution of ^{210}Pb . The estimated dose in this investigation campaign could be a underestimated value due to the particle scavenge effect of raining, the wind direction changing and the lower particle emitting rate during the plant production with only half-capacity.

Based on the results in the second investigation campaign (Table 14), the mean committed effective doses for adult due to intake of ^{210}Po and ^{210}Pb from the atmospheric particulate at the sites of the ILVA in Taranto in 4–12 May 2009 were 10.1 and $36.8 \mu\text{Sv y}^{-1}$, respectively, and with a total dose of $47.0 \mu\text{Sv y}^{-1}$, and they were 2.0–6.0 times as high as that at the control site. If based on the maximum concentrations, the doses from ^{210}Po and ^{210}Pb could be 19.7 and $49.3 \mu\text{Sv y}^{-1}$, respectively, and with a total dose of $69.0 \mu\text{Sv y}^{-1}$. Therefore, about $27.2 \mu\text{Sv y}^{-1}$ of the dose at mean and $49.2 \mu\text{Sv y}^{-1}$ at maximum could be attributed to the ^{210}Po and ^{210}Pb emitted from the chimney of the ILVA in Taranto, of which about 63.5–69.0% was the contribution of ^{210}Pb .

Due to the involvement of man-made release of ^{210}Po and ^{210}Pb , the operational process of the ILVA in Taranto could be considered as a planned and prolonged exposure situation. The radiological risk evaluation to members of public in Taranto was based on the guidance from ICRP and

the Italian law. The constrains and reference levels from ICRP to the public exposure from planned exposure situation is no more than 0.3 mSv y^{-1} from waste management operations [36] and is no more than 0.1 mSv y^{-1} from situation of prolonged exposure [37]. The guidance from ICRP is also the basic principle for the Italian law as well as its subsequent amendments [38]. The additional committed effective doses estimated to individuals of the public due to emission of ^{210}Po and ^{210}Pb from the plant of the ILVA in Taranto in the two times of investigation campaigns all were much lower than the established action level, even in the worst case ($49.2 \mu\text{Sv y}^{-1}$). Therefore, no specific interventions should be required, as far as only inhalation of ^{210}Po and ^{210}Pb was concerned.

It was reported that the average committed effective dose resulting from exposure to environmental background radiation in Italy, regardless of exposure to indoor radon, is greater than 1 mSv y^{-1} [39], and the global average human exposure from natural sources is 2.4 mSv y^{-1} [12, 40]. From radiation protection point of view, it is predicted that the exposure damage to individuals of public due to the inhalation of ^{210}Po and ^{210}Pb released from the ILVA in Taranto could not be observable and significant.

Being time-, labor-, and money-consuming, the survey at this stage was mainly focused on studies of the radiological impact of ^{210}Po and ^{210}Pb through inhalation on the adult members of the public. According to literatures [5], ^{210}Po and ^{210}Pb are the most important radionuclides released from the coal power plant, steel-making industry, and refractory material industry, and inhalation is a very important exposure pathway. However, the other possible radiological impacts of uranium, thorium, or radium isotopes, on workers and the public in the region of the ILVA in Taranto resulting from the treatment, storage, disposal, and reuse of the produced waste through ingestion of the contaminated food, vegetable, water,

TABLE 12: The committed effective dose for members of public estimated from the mean ^{210}Po and ^{210}Pb concentrations in atmospheric particulate collected from the Castel Romano Roma on 19–29 November 2008.

Age classes, y	Respiration rate, $\text{m}^3 \text{d}^{-1}$	Annual respiration volume, $\text{m}^3 \text{y}^{-1}$	^{210}Po dose coefficient, Sv Bq^{-1}	Mean ^{210}Po conc., Bq m^{-3}	Mean effective dose from ^{210}Po , Sv y^{-1}	^{210}Pb dose coefficient, Sv Bq^{-1}	Mean ^{210}Pb conc., Bq m^{-3}	Mean effective dose from ^{210}Pb , Sv y^{-1}	Total mean effective dose from ^{210}Po and ^{210}Pb , Sv y^{-1}
<1	2.86	1044	$1.80E-05$	$4.85E-05$	$9.11E-07$	$1.80E-05$	$3.99E-04$	$7.50E-06$	$8.41E-06$
1-2	5.16	1883	$1.40E-05$	$4.85E-05$	$1.28E-06$	$1.80E-05$	$3.99E-04$	$1.35E-05$	$1.48E-05$
2-7	8.72	3183	$8.60E-06$	$4.85E-05$	$1.33E-06$	$1.10E-05$	$3.99E-04$	$1.40E-05$	$1.53E-05$
7-12	15.3	5585	$5.90E-06$	$4.85E-05$	$1.60E-06$	$7.20E-06$	$3.99E-04$	$1.60E-05$	$1.76E-05$
12-17	20.1	7337	$5.10E-06$	$4.85E-05$	$1.81E-06$	$5.90E-06$	$3.99E-04$	$1.73E-05$	$1.91E-05$
Adult (>17)	22.2	8103	$4.30E-06$	$4.85E-05$	$1.69E-06$	$5.60E-06$	$3.99E-04$	$1.81E-05$	$1.98E-05$

TABLE 13: The committed effective dose for members of public estimated from the mean and maximum ^{210}Po and ^{210}Pb concentrations in atmospheric particulate collected from the area of the ILVA Taranto on 11-17 November 2008*.

Age classes, γ	Mean ^{210}Po conc., Bq m^{-3}	Maximum ^{210}Po conc., Bq m^{-3}	Mean effective dose from ^{210}Po , Sv y^{-1}	Maximum effective dose from ^{210}Po , Sv y^{-1}	Mean ^{210}Pb conc., Bq m^{-3}	Maximum ^{210}Pb conc., Bq m^{-3}	Mean effective dose from ^{210}Pb , Sv y^{-1}	Maximum effective dose from ^{210}Pb , Sv y^{-1}	Total mean effective dose from ^{210}Po and ^{210}Pb , Sv y^{-1}	Total maximum effective dose from ^{210}Po and ^{210}Pb , Sv y^{-1}
<1	1.04E-04	2.26E-04	1.95E-06	4.25E-06	6.23E-04	1.10E-03	1.17E-05	2.07E-05	1.37E-05	2.49E-05
1-2	1.04E-04	2.26E-04	2.74E-06	5.96E-06	6.23E-04	1.10E-03	2.11E-05	3.73E-05	2.39E-05	4.32E-05
2-7	1.04E-04	2.26E-04	2.85E-06	6.19E-06	6.23E-04	1.10E-03	2.18E-05	3.85E-05	2.47E-05	4.47E-05
7-12	1.04E-04	2.26E-04	3.43E-06	7.45E-06	6.23E-04	1.10E-03	2.50E-05	4.42E-05	2.85E-05	5.16E-05
12-17	1.04E-04	2.26E-04	3.89E-06	8.46E-06	6.23E-04	1.10E-03	2.70E-05	4.76E-05	3.09E-05	5.60E-05
Adult (>17)	1.04E-04	2.26E-04	3.62E-06	7.87E-06	6.23E-04	1.10E-03	2.83E-05	4.99E-05	3.19E-05	5.77E-05

*The respiration rate, annual respiration volume, and the ^{210}Po and ^{210}Pb dose coefficients used to calculate the committed effective dose for each age class are the same as those listed in Table 12.

TABLE 14: The committed effective dose for members of the public estimated from the mean and maximum ²¹⁰Po and ²¹⁰Pb concentrations in atmospheric particulate collected from the area of the ILVA Taranto on 4–12 May 2009*.

Age classes, y	Mean ²¹⁰ Po conc., Bq m ⁻³	Maximum ²¹⁰ Po conc., Bq m ⁻³	Mean effective dose from ²¹⁰ Po, Sv y ⁻¹	Maximum effective dose from ²¹⁰ Po, Sv y ⁻¹	Mean ²¹⁰ Pb conc., Bq m ⁻³	Maximum ²¹⁰ Pb conc., Bq m ⁻³	Mean effective dose from ²¹⁰ Pb, Sv y ⁻¹	Maximum effective dose from ²¹⁰ Pb, Sv y ⁻¹	Total mean effective dose from ²¹⁰ Po and ²¹⁰ Pb, Sv y ⁻¹	Total maximum effective dose from ²¹⁰ Po and ²¹⁰ Pb, Sv y ⁻¹
<1	2.91E-04	5.64E-04	5.47E-06	1.06E-05	8.12E-04	1.09E-03	1.53E-05	2.04E-05	2.07E-05	3.10E-05
1-2	2.91E-04	5.64E-04	7.67E-06	1.49E-05	8.12E-04	1.09E-03	2.75E-05	3.69E-05	3.52E-05	5.17E-05
2-7	2.91E-04	5.64E-04	7.97E-06	1.54E-05	8.12E-04	1.09E-03	2.84E-05	3.81E-05	3.64E-05	5.35E-05
7-12	2.91E-04	5.64E-04	9.59E-06	1.86E-05	8.12E-04	1.09E-03	3.26E-05	4.37E-05	4.22E-05	6.23E-05
12-17	2.91E-04	5.64E-04	1.09E-05	2.11E-05	8.12E-04	1.09E-03	3.51E-05	4.71E-05	4.60E-05	6.82E-05
Adult (>17)	2.91E-04	5.64E-04	1.01E-05	1.97E-05	8.12E-04	1.09E-03	3.68E-05	4.93E-05	4.70E-05	6.90E-05

*The respiration rate, annual respiration volume, and the ²¹⁰Po and ²¹⁰Pb dose coefficients used to calculate the committed effective dose for each age class are the same as those listed in Table 12.

and soil and inhalation of air all can be subjects of future researches.

In spite of the difficulties to statistically observe the bio-effect of radiation dose at very low exposure rate, additional concentrations of ^{210}Po and ^{210}Pb from the man-made release are usually considered toxic to public due to their radiation damage and chemical toxicity. One of the three well-known radiation protection principles (justification, optimization, and dose limitation) recommended by the ICRP for practices is that doses to individuals of public and to occupationally exposed workers should be kept as low as reasonably achievable (optimization principle). Thus, for realizing the optimization principle for the practice, or-so-called “planned and prolonged exposure situation,” the administrators of the plant ILVA in Taranto should keep in mind to better control their production procedures, for instance, (i) selecting the raw materials containing lower activity of radionuclides, (ii) keeping the filtration system always in good function, (iii) inventing and testing new technologies to improve the effect of dust particle removal (e.g., wet method), and (iv) well disposal of the wastes to prevent the second contamination of the environment, mankind, and so forth.

Moreover, the biological and chemical toxicities and epidemiological survey of lead in the area of the ILVA in Taranto should also be a subject of study as far as the public health is concerned because the release of 17.8 t y^{-1} or even more of lead smoke into the atmosphere, estimated in this work, is not a quantity negligible.

4. Conclusion

The radiological survey on the Iron- and Steel-making Plant ILVA in Taranto was mainly focused on contamination source-term investigation and exposure impact evaluation of the volatile radionuclides ^{210}Po and ^{210}Pb . The activity concentrations of ^{210}Po and ^{210}Pb in the raw materials, dust particles, surficial soils, and atmospheric particulate samples collected in the area of ILVA in Taranto were determined. The results showed that the activity concentrations in the raw materials were in the range of $3.46\text{--}17.9\text{ Bq kg}^{-1}$ of ^{210}Po and of $3.50\text{--}16.8\text{ Bq kg}^{-1}$ of ^{210}Pb , which were relatively low and could create a maximum annual inventories of $2.64 \times 10^{11}\text{ Bq}$ of ^{210}Po and $2.71 \times 10^{11}\text{ Bq}$ of ^{210}Pb if a total quantity of 22 Mt y^{-1} raw materials was consumed in the plant. The activity concentrations in dust particles emitted from the chimney of the ILVA in Taranto were in the range of $5.91\text{--}85.6\text{ kBq kg}^{-1}$ of ^{210}Po and of $5.44\text{--}34.6\text{ kBq kg}^{-1}$ of ^{210}Pb , releasing more ^{210}Po than ^{210}Pb . The activity concentrations in surficial soils (depth: $0\text{--}5\text{ cm}$) were in the range of $49.3\text{--}140\text{ Bq kg}^{-1}$ of ^{210}Po and of $51.6\text{--}150\text{ Bq kg}^{-1}$ of ^{210}Pb , being observable a variation of the activity concentrations with distance. The activity concentrations in atmospheric particulate were in the range of $43.1\text{--}564\text{ }\mu\text{Bq m}^{-3}$ of ^{210}Po and $618\text{--}1099\text{ }\mu\text{Bq m}^{-3}$ of ^{210}Pb , and it was observed that the mass specific activity concentrations of ^{210}Po and ^{210}Pb in the atmospheric particulate are one to three order of magnitude higher than that found in the top soil. In the worst case

detected, the mean committed effective doses for adult due to intake of ^{210}Po and ^{210}Pb from the inhaled atmospheric particulate at the sites of the ILVA in Taranto were about 10.1 and $36.8\text{ }\mu\text{Sv y}^{-1}$, respectively, and with a total dose of $47.0\text{ }\mu\text{Sv y}^{-1}$, and they were $2.0\text{--}6.0$ times as high as that at the control site. Based on the maximum concentrations, the doses from ^{210}Po and ^{210}Pb could be 19.7 and $49.3\text{ }\mu\text{Sv y}^{-1}$, respectively, and with a total dose of $69.0\text{ }\mu\text{Sv y}^{-1}$. After deduction of the background contribution, about $27.2\text{ }\mu\text{Sv y}^{-1}$ of the dose at mean and $49.2\text{ }\mu\text{Sv y}^{-1}$ at maximum could be attributed to the ^{210}Po and ^{210}Pb emitted from the E312 chimney of the ILVA in Taranto, of which about $63.5\text{--}69.0\%$ was the contribution of ^{210}Pb . The constrains and reference levels from ICRP to the public exposure from planned exposure situation is no more than 0.3 mSv y^{-1} from waste management operations and is no more than 0.1 mSv y^{-1} from situation of prolonged exposure. Therefore, no specific interventions should be required, as far as only inhalation of ^{210}Po and ^{210}Pb was concerned.

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