THE ITALIAN INVESTIGATION, RADIATION PROTECTION AND LEGISLATIVE APPROACH TO CONTAMINATED SCRAP METAL AND BUILDING MATERIAL RICH IN NATURAL RADIONUCLIDES

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1 ABSTRACT

A review has been carried out on two possible sources of exposure of workers and population to radiation in Italy, that is, radioactive contamination of scrap metal and building material rich in natural radionuclides.
In Italy a wide experience was gained in past years on possible radioactive contamination of scrap metal. This showed that such contamination is a problem that cannot be disregarded from a radiation protection point of view. The paper gives a picture on what has been done up to now in Italy on the issue and reviews the data on Naturally Occurring Radioactive Material (NORM) found in scrap metal and discusses the legislative frame.
Since the eighties building material rich in natural radioactivity has been largely studied in Italy and many research measurements were carried out. These materials were mainly of natural origin, however, over the years great attention has also been devoted by experts to the possible radiation protection consequences of the use of coal ash and zircon sand in them. The recent technical guide *Radiation Protection 112* (1) published by the European Union (EU) Commission for harmonising controls on the radioactivity of building materials within the EU gave the opportunity to classify these materials according to their radioactivity. The paper presents the main conclusions of this analysis and the Italian approach to the problem.

2 THE RECYCLING INDUSTRY

In the past ten years, in Italy, possible radioactive contamination of scrap metal received particular attention from the radiation protection community. As early as 1990 its awareness was originated by accidental meltings of ^{137}Cs sources in some foundries of the Lombardy Region, revealed only by an anomalous contamination of the Po river below the city of Milan. Italy is the first importer of scrap metal in the European Union (3.8 million tons each year) (2) and, particularly, the Brescia Province is the most important metallurgic hub in Italy: 7 million tons of scrap metal (steel, brass, copper, aluminium, etc.) are recycled every year in more than a hundred foundries and 13 steel mills (3). However, only in 1993, following reports by European countries that contaminated scrap materials were circulating in Europe, the Ministry of Health issued a directive on the necessity of radioactive controls of scrap metal shipment at Italian borders. Many national and regional institutions were charged of the radiometric control of imported material and at the same time melting companies were charged 1) to check on the presence of radiocontamination and/or radioactive sources at different steps of the production cycle and 2) to foresee in their contracts that, in case of radioactive contamination of shipment judged non-acceptable by the Italian national and/or local authorities, the importers should take the shipment back (2).

However, on the basis of one year of experience of controls at borders and in companies, it was concluded that external measurements of scrap metal consignments made at the border did not guarantee the absence of radioactivity. Therefore, in May 1994 a new directive of the Minister of Health required the companies to request the senders a certificate on the quality of scrap metal purchased and to carry out a radiometric control on the material during unloading. At the same time concerned authorities should continue normal surveillance activity.

Nevertheless, it became clear from the beginning that many certificates coming from non-EU member States were not reliable enough, hence border controls were continued up to the end of December 1995, when the publication of the Italian Radiation Protection Act (4) definitely entrusted (article 157) the monitoring of scrap metal to industrial and trade operators (excluding transporters), in order to detect possible misused sources. A decree to be issued by the Minister of Health, in agreement with the Ministers of Industry, Labour and Environment, is foreseen in that article to specify application conditions. Radiation protection experts from both national and regional institutions drafted and discussed a proposal for it. Their work was concluded more than three years ago. Nevertheless, this decree has still not been issued.

In the period 1990 - 1998 seven cases of radioactive contamination (mainly from ^{137}Cs, but from ^{60}Co and ^{241}Am, too) of scrap metal working environment were reported in the Brescia Province. They were of different degrees of severity, the worst one being that of 1997, when two different sources of ^{60}Co and ^{137}Cs were melted at a steel plant. Following this fact and still in the absence of the application decree, in June 1997 the Lombardy Region issued an urgent ordinance, implementing the duties concerning radiometric surveillance and detailing operative procedures. The regional ordinance was renewed a second time at its expiry date one year later (2). The ordinance is no longer in force as it cannot be deferred more than one time, but in the Brescia Province more than 150 local firms (steel mills, foundries and scrap recyclers) which established control procedures still carry them out, continuing to perform both the monitoring on incoming consignments and the visual inspection required by the regional ordinance (3). Many firms also perform checks at different steps of the melting process. Generally, the procedures utilised depend on dimensions and characteristics of the activity. However, a particular effort was made in teaching workers how to recognise labels, signs, and shapes of sources and in making them aware of the appearance of most commonly used sources and those out of use (smoke detectors, lightning rods, industrial source holders, etc.). Due to the absence of the foreseen decree the level of compliance differs in Lombardy and other Regions (3). However, it should be underlined that the attitude of industry towards controls has changed sensibly over the years, going from open opposition to the detailed application of the ordinance, to the advocation of national regulations (2). This is due to the awareness that accidental melting of sources is much more expensive, in terms of decontamination, shut-down of the plant, etc. than equipment and personnel for the controls. It is worth citing that, a recycler of furnace ash dust monitors possible radioactive contamination in all consignments and reports any abnormal case to the local authority (3).

A few years ago the large experience of local laboratories for environmental radioactivity and firms in monitoring scrap metal consignments was applied in drafting an experimental protocol which was recently issued (5) as a standard of the Ente Nazionale Italiano di Unificazione (UNI, Italian Organisation for Standardisation). This standard, devoted to detecting radionuclides in scrap consignments with gamma and X measurements, copes with both hand-held detectors and fixed large area systems. It details measurement procedures, criteria to judge when a consignment should be considered "radioactively anomalous" and supplies forms to be filled with measurement results. In appendix sensitivity of large area detectors is assessed for both point sources and extended sources and parameters which influence the measurements and false alarm are also discussed.

Even though two conferences on radioactive contamination of scrap metal were held in Italy (6, 7) and some other papers were published by Italian authors in international literature (see e.g. 3, 8), very scarce data is available about NORM finding in scrap metal in Italy. One paper (9), relevant to the period of control of the consignments at the borders, assessed in 54.2% the percentage distribution of natural radioisotopes in the consignments returned to the sender. Other authors (10) cited that during the effectiveness of the ordinance in the Lombardy Region out of 100 consignments found contaminated in the Brescia province, 50% contained ^{226}Ra and (20 - 25)% isotopes of natural origin (U and Th families). With regard to the Lombardy Region it is referred (11) that 17% of radioactivity found in scrap metal were actual sources, with or without their shielded container (mainly lightning conductors and smoke detectors), whereas 73% were various contaminated material (mainly luminescent quadrants and tubes from the oil extraction industry). In all these cases data is general and does not allow to distinguish NORM in scrap metal, originating from oil extraction or mines, from industrial sources or contamination (e.g ^{226}Ra in lightning conductors, smoke detectors, needles, luminescent quadrants, etc,). During local control activities ^{226}Ra concentration was detected to be up to 800 Bq g^{-1} in scales of valves, tubes, etc. and ^{228}Ra concentration one or two orders of magnitude lower (12). These figures are coherent with literature data.

In other countries, where this data is singled out, it can be noticed that in a few years the NORM percentage of radioactive material discovered in recycled metal increased from 37% to 62% (13, 14). Even if in the first paper only US cases are considered, whereas in the second one Canadian ones, too, the trend is clear. The authors of these two papers underline that if "NORM contaminated scrap was to be melted the finished steel products would not become significantly contaminated. NORM would end up with the slag, where the rest would go with the furnace dust" (13). However, the possible re-use of these by-products (e.g. in building materials) could be at risk. In conclusion, it can be agreed that "NORM contaminated scrap metal has been handled and consumed by the scrap metal industry for years but went unnoticed until the widespread use of radiation monitors that were intended to detect radioactive sources and devices" (14).

In Italy, from a legislative point of view the radiation protection problem of NORM in scrap metal is almost not dealt with. Without the applicative decree, the obligations of art. 157 are hardly applicable (the authors were informed of different opinion of courts on the territory) and anyway no mention to NORM in scrap metal is available in other regulations. Moreover, at EU level, in spite of various pushing initiatives of some countries, e.g. The Netherlands, Italy etc. (see 2), no directive or recommendation has ever been issued on either radioactive scrap metal in general, or on NORM in it. The Council Directive 96/29/EURATOM (15), the recommendation for its implementation (16) and other EU documents dealing with NORM (17, 18) do not mention the problem at all.

3 NATURAL RADIOACTIVITY IN BUILDING MATERIAL

In Italy, natural radioactivity in building material has been widely investigated since the eighties (see e.g. 19, 20, 21). Materials identified as containing high activity concentration were mainly of natural origin (tuff[1] and pozzolan[2]) but in the same years several research programmes were carried out on the use of fly ash and zircon sand in them.

As regards tuff and pozzolan many Italian papers are available in both national and international literature, studying the effect of their use as building material, but, as far as we know, no analysis has ever been made on work activity with these materials in quarries or industries.

Differently from tuff and pozzolan, the work activities with coal and zircon sand have been studied both in Italy and in other countries.

3.1 Coal combustion

In Italy coal burning is one of the main resources for energy production (about 10%) (22) and disposal of large amount of ash produced is made (40-60)% in the cement industry and (25-40)% in that of concrete (23).

In the past the possible health impact of coal combustion on the population was investigated by this Institute (24) analysing radioactivity content of coal and ash and possible atmospheric dispersion of radioactive effluents. A source-related control procedure before and during operation of the plant was suggested and applied, as a test, on a 72 MWe brown coal-fired power plant in Central Italy (25).

The use of 25% of coal ash in concrete has been assessed to increase up to 30% ^{226}Ra and ^{232}Th activity concentration and about 3% that of ^{40}K (23). This means that gamma dose rate indoors increases when ash is used in cement and concrete. However, in spite of the higher ^{226}Ra activity concentration, ^{222}Rn emanation coefficient is low (26). The total effect on radon exhalation is

[1] consolidated volcanic ash composed largely of fragments (less than 4 mm) produced directly by volcanic eruption.

[2] finely ground volcanic material used in cement because it hardens underwater.

controversial: in measurements made on cubes of fly ash concrete some authors found that the added fly ash causes a decrease in radon exhalation, on others there was no significant difference and in a few cases fly ash concrete showed to exhale somewhat more radon than ordinary concrete (27). Discrepancies are probably caused by differences in ^{226}Ra concentration, porosity, surface structure and fly ash content in concrete in the different countries. The added fly ash probably causes some change in the internal structure of the concrete, so that diffusion length and/or emanation coefficient are reduced, depending on the amount of ash and the structure of the ordinary concrete (27). Therefore, no unique conclusion can be drawn on the possible increased exposure indoors due to the use of ash. This is due, probably, to lack of systematic studies, as the authors already concluded in the past (24). In Italy a complete analysis would also require a comparison with cement or concrete added with pozzolan, due to its high activity concentration and large utilisation.
Coal burning is a typical NORM activity, but it was not considered among NORM activities in the study contract of the EU Commission (18) which deals with exposures in working environment. Neither was it considered by the recommendation deriving from this study (17), where a mention is only made citing that the handling and re-use of fly ash may also give rise to a radiological risk under some circumstances. It can be concluded that a study similar to (18) should be promoted to analyse population exposure from different work activities with NORM.

3.2 Zircon sand

Zircon sand is largely used in Italy. With this term two types of sandy minerals are identified: zircon, made of mainly zirconium silicate ($ZrSiO_4$) and baddeleyite, made of mainly zirconium oxide (ZrO_2), used as heat and corrosion resistant linings for furnaces and muffles. They generally have high activity concentration of U and Th families (28, 29). Main users of sands are foundries, ceramic and refractory material industries. Italian importation amounts to 60000-65000 ton y^{-1} and mostly is used in ceramic industry as opacifier in glaze of tiles (29). Italy is not a producer of this sand, but imports it mainly from Australia (58%), South Africa (24%), Ukraine (12%) and USA (6%) (30).
In Italy, the working environment in a factory producing refractory material with zircon sand was studied in the eighties (28). A low ^{222}Rn emanation from the sand, in both its original form and during its transformation, and a ^{210}Po enrichment in atmospheric dust were identified. The latter could be connected to the melting of material and volatilisation temperature of radionuclide in a way analogous to that which occurs in coal combustion. Effective doses to workers in the highest contaminated areas were assessed to be about 5 mSv y^{-1}, so that it was suggested to continue the analysis on the use of this sand (28). Recently another investigation was carried out in an industry for the production of sanitary equipment involving the use of enamel containing zircon (31). Industrial operation were mainly humid and with no foundry activity. ^{222}Rn concentration in air resulted to be low (20-50 Bq m^{-3}), no ^{210}Po activity concentration enrichment was found in the air and gamma dose rate was found significantly higher than background uniquely around zircon storage. These results confirm that the radiological situation is determined by the industrial process. Industrial activity

with zircon sand were analysed in the EU contract study (18) and are identified in the new Italian Radiation Protection Act (32) as " … work activities … within which the presence of natural radiation sources leads to a significant increase in the exposure of workers or of members of the public which cannot be disregarded from the radiation protection point of view" (15).
On the other hand the use of zircon sand as an opacifier in glaze of tiles seems to increase population exposure to radiation neither as a source of ^{222}Rn, due to its lower exhalation from glaze than from other building material, nor as a gamma source (33, 34), for the relatively low ^{226}Ra total activity in glazed tiles (34). However, the measurements performed by some Chinese researchers in some rooms with and without glazed tiles (33) showed that the average area density of total beta radionuclides in tile glaze was 12-13 times as much as that in ordinary building material. This causes an increase in the external radiation dose received by the public. Testicles are the human organ at the highest risk, particularly of boys who frequently play on ground. This possible beta dose is neglected in most national legislation on building material, and in the EU technical guide, too (1). Only the Austrian regulation (35), wishing to limit skin dose, due mainly to ^{238}U and its decay products, to less than 10 mSv y^{-1} (which means a skin dose rate lower than 3.4 μSv h^{-1}), requires tiles to have superficial activity lower than 1 Bq cm^{-2}. This value was obtained using a safety factor of about 0.5.

3.3 Italian building materials

In the last years the authors reviewed in detail national and international literature about radioactivity in Italian material, in order to build up a data base (36). This data base, recently up-dated (37), collects about 1000 data. It is not a representative picture of the national situation, but a conservative one, due to the fact that no national representative investigation has ever been made and measurements are generally made looking at possible high activity samples. Moreover, in order to assess the health effect of the use of these materials, percentage of utilisation should be known for the country. Data were compared with the EU technical guide (1) which establish an exemption dose level of 0.3 mSv y^{-1} and a dose criterion for controls of 1 mSv y^{-1}. Dose levels considered in the EU guide are the excess gamma doses to those received outdoors, due to the use of that material. The guide states that, within the EU, doses exceeding 1 mSv y^{-1} should be taken into account from a radiation protection point of view and higher doses should be accepted only in some very exceptional cases where materials are used locally. Controls can be based on a lower dose criterion, if it is judged that this is desirable and will not lead to impractical controls. An activity concentration index is suggested (1) for identifying materials which might be of concern, but any actual decision on restricting the use of the material should be based on a separate, more detailed and specific, dose assessment. This index should be lower than or equal to four different figures to satisfy the exemption dose level or the dose criteria for controls, depending on the use of the material in bulk amounts or superficially. A first identification of material which might be of concern was made by the authors, considering a dose of 1 mSv y^{-1} for materials used in bulk amount. Tuff,

basalt[3], pozzolan, syenite[3], granite[3], gneiss[4], lava and coal ash resulted to exceed the chosen dose level (see 36). This is an indication of the need to deepen the analysis on the use of these materials, and the possible consequent population exposure.
A comment can be made on lava: it is used locally both outdoors and indoors as a superficial layer and/or for decorative aim (e.g. statues, fireplaces, etc.). For this reason, its use should require on one hand, a more precise dose assessment, on the other hand a check to see if it satisfies the justification principle.
However, in spite of the fact that the EU Council Directive relating to construction products (38) was implemented in Italy about ten years ago, its requirements on possible radioactivity are not detailed and specific enough to limit the use of building materials rich in natural radioactivity.

4 CONCLUSIONS

Over the years, Italian experts have given great attention to the possible contamination of scrap metal and to its radiation protection aspects. Notwithstanding this a lack of political willingness should be underlined. A possible intervention of the EU Commission with a recommendation or some statements would probably have helped to conclude the legislative iter. The Italian experts, in fact, are convinced that the problem has no boundary limit, but should be dealt with at EU level, as it has been repeatedly maintained in EU formal and informal meetings (2). The possible NORM contamination of this scrap metal in our country has been neither studied enough nor dealt with from a legislative point of view and in the author's opinion this could represent a radiation protection problem particularly for by-products, that is, the possible use of ashes for building material or other products. Indeed, it is known that, during melting, natural radionuclide activity, particularly high in tubes and valves of some type of industries, are enriched in ash. The controls always required by this Institute at different stages of processing (external radiation measurements of the shipment, visual inspection and external radiation measurements of the unloaded scrap, gamma spectroscopy measurements of casting proof tests, radioactive control of flue dusts, etc.) (2) can be a safeguard even in case of NORM, for an improved detectability of possible contamination.
The control of trucks at the borders detected, among others, an unexpected case of radioactivity in consignments. Indeed, an increase of dose rate eight times the background value was detected near a truck carrying corundum[5] (39). From a subsequent spectroscopic analysis, activity concentration of U and Th families resulted to exceed the limit fixed by the Italian law for non-radioactive materials. This result suggested the relevant firm should monitor radon concentration in working environment. This fact shows that possibly new material, which can be considered NORM, can also be identified in industrial environment.

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[3] Basalt, syenite, granite are magmatic effusive and intrusive rocks
[4] Gneiss are rocks formed by regional metamorphism, from shale or granite.
[5] It is a hard mineral, crystallised in hexagonal system, used as emery in polishing and grinding.

As regards coal, the controversial data on ^{222}Rn exhalation from concrete cubes containing coal ash, together with the possibility of very high activity concentration in coal (see e.g. 40) should advise to check at least periodically qualities of ash used in the building industry. Indeed, ICRP recently reported, as an example of prolonged exposure situations, that in China the large production of coal ashes and their large use in building material production can expose the resident of building constructed with these materials to doses up to several mSv y^{-1} (41).
As regards building material rich in natural radioactivity, the recent issue of the EU recommendation should also promote in Italy the first provision to limit the use at least of the most active ones. However, it should be underlined that, in particular, tuff is the main building material found in old towns and villages, mainly in Central Italy, of significant historical and artistic value. Limiting its use, in restoration, for example, may thus create serious difficulties.

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