PRELIMINARY IDENTIFICATION OF WORK ACTIVITIES INVOLVING NORM IN ITALY

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1 ABSTRACT

Implementation of EU Basic Safety Standards in Italian legislation (decree nº 241/2000) is introducing radiation exposure controls for a set of work activities involving NORM. A project aimed at estimating environmental impact for some activities with possible NORM risk is being carried out by the National Topic Centre on Physical Agents (CTN-AGF), coordination of Regional Agencies for the Environmental Protection (ARPA), which supports the National Agency for the Environmental Protection (ANPA) in the management of environmental information about physical pollutants (ionizing and non-ionizing radiations, noise). At this stage of the project, the following working categories have been selected: phosphate and fertilizers industry, integrated steelworks, processing of zircon sands, oil and gas extraction and oil refinery, uranium mines, coal-fired power plants. A questionnaire for collecting relevant information to assess environmental pressure, first step for radiological impact predictions, will be circulated amongst interested companies. In this paper, results of a first review on Italian potentially NORM involved industries are shown. Phosphogypsum is no more produced in the national phosphate industry, but past disposal areas do exist in the territory. Intense work activities are recorded for all italian sections of fertilizers production, zircon sands processing, integrated steelworking, hydrocarbon extraction and coal-fired energy production. Two closed uranium mines are also present.

2 INTRODUCTION

In Italy, the Basic Safety Standards (Directive 96/29/Euratom) for the health protection of the general public and workers against the dangers of ionizing radiation, have been implemented by means of a legislative decree (nº 241/2000) which provides the execution of controls of radiation exposure for a certain set of work activities involving NORM. These activities are:

- phosphate industry and warehouses for fertilizers wholesale trade;
- processing of ores in the extraction of tin, ferro-niobium from pyrochlore and aluminium from bauxite;
- processing of zircon sands and refractory materials;
- manufacture of rare earths;
- manufacture and use of thorium compounds;

- titanium dioxide pigment industry;
- oil extraction and refining industry and gas extraction industry.

This decree fixes an effective dose value not to be exceeded of 1 mSv/year for workers and of 0.3 mSv/year for general public.
Apart from that, a study project about NORM is being developed by the National Topic Centre on Physical Agents (CTN-AGF), a coordination of Regional Agencies for the Environmental Protection (ARPA), which supports the National Agency for the Environmental Protection (ANPA) in collecting environment quality information as far as physical pollutants (ionizing and non-ionizing radiations, noise) are concerned.
The NORM project, concerning above all the environmental aspect of the problem, aims at the estimation of radiological impact on the environment connected with some work activities dealing with NORM.
The methodology provides the creation of a data base of adequate information for characterising the environmental pressure of work activities, followed by impact assessments mainly based on provisional models.
The considered work activities have been chosen based on the importance of their predicted radiological impact on the environment (rather than on workplaces) and on economic significance and may differ from the decree nº 241/2000 list; they are indicated in the next paragraph.
In the first stage of the project some centralized data bases, such as Istat (National Institute of Statistics) ones and the Chamber of Commerce Industries Register, together with field reports and documents, have been consulted, representatives of sector associations or of the companies themselves have been contacted, the work processes have been investigated and work activity specific questionnaires have been prepared. In the next stage the schedules will be distributed to companies for the compilation and then collected through one or more representatives for each work activity, to set up the already mentioned data base.
The search through centralized data bases presents some problems because of Istat codes are often too general and the data sets of Industries Register are sometimes inexact.

3 INDUSTRIAL ACTIVITIES REVIEW

3.1 Phosphate and fertilizers industry

3.1.1 CURRENT PRODUCTIONS

Radiological significance for this activity is connected to phosphate ores and to products deriving from their use because of elevated activity concentration of natural uranium of phosphorites (calcium phosphates) (1). The wet process for the production of phosphoric acid, the alternative process that uses nitric acid to attack phosphorites and the thermal process to give elemental phosphorus P are particularly critical because of the formation of radioactive by-products (such as phosphogypsum). The superphosphate production, and the complex fertilizers production in general, have to be monitored. Another potential critical way is the use in agriculture of products with variable concentrations of U-238 and its decay products.

By consultation of Federchimica (chemical industries association) data base and through various contacts with Assofertilizzanti (main fertilizers companies association) it results that in Italy there is no phosphoric acid plant, no process using nitric acid with gypsum as a by-product, no thermal process to give elemental phosphorus in operation; several plants of this kind operated in past times. At present, one company treats the phosphorites with nitric acid without formation of any radioactive by-product and three factories produce superphosphates by balanced reaction between phosphorites and sulphuric acid. There are now 15-20 principal companies that produce complex fertilizers (and simple phosphate fertilizers) mainly by granulation, mixing and compacting; the number of industries rises to about a hundred including little productions or productions with a low title of phosphorus (such as organic fertilizers).
In 1998 the fertilizers total production was 2.421.578 t, made of superphosphates for 10% and of complex fertilizers for 38% (2).
The fertilizers industry is included in the Directive 96/61/CE (IPPC, Integrated Pollution and Prevention Control), that provides measures to reduce pollution of a list of work activities, considered as prioritary as far as general environment impact is concerned.
A recent survey has been carried out in a factory of complex fertilizers with measurements of activity concentrations (by means of gamma spectrometry) of decay products of U-238, Th-232 and K-40 in raw materials and products (3). Elevated K-40 activity concentrations are associated to the presence of potassium salts. The lack of equilibrium between U-238 and Ra-226 in H_3PO_4 and MAP, and in fertilizers that use such compounds as raw materials, is due to the fact that in phosphoric acid production radium coprecipitates with the gypsum, while uranium and thorium follow the phosphorus into the acid.

Table 1: Activity concentrations in the raw materials of fertilizers (Bq/kg).

Samples	K-40	Th-234	Bi-214
K_2SO_4	13000	< DL	< DL
KCl	15000	< DL	< DL
Phosphorites	31	1000	1200
H_3PO_4	19	1600	0.7
MAP	26	980	4.2

Table 2: Activity concentrations in the complex fertilizers (Bq/kg).

Samples	K-40	Th-234	Bi-214
Fert. 1	< DL	170	115
Fert. 2	2380	190	100
Fert. 3	3920	180	110
Fert. 4	2750	310	200
Fert. 5	< DL	200	110
Fert. 6	2790	230	130
Fert. 7	2200	180	110
Fert. 8	4810	360	150
Fert. 9	3830	290	100
Fert. 10	4400	515	62
Fert. 11	4400	280	55
Fert. 12	4100	560	58

3.1.2 PHOSPHOGYPSUM DISPOSALS

In past times several plants of a big public company (Enichem) produced phosphoric acid through the wet process, phosphogypsum being formed as a by-product. Three phosphogypsum disposal areas (Veneto, Sicilia, Sardegna) are known; one plant in Calabria has ceased the activity; there is uncertain information about one plant in Liguria.

While plants in Veneto and Sicilia produced fertilizers, those in Calabria and Sardegna operated in the field of detergents. Before the realization of phosphogypsum dumps, some plants used to discharge directly into the sea.

Since 1998 ANPA is surveying the phosphogypsum discharge site of Campalto (Venice lagoon), with measurements of Ra-226, Pb-210 and Po-210 activity concentrations in water, sediments and shellfishes around the site (4). The erosion of phosphogypsum, due to meteoric agents and tides, causes a higher level of Pb-210 and Po-210 in sediments near the dump, while it is not clear whether the level in shellfishes is correlated to the proximity to the dump or not. The estimated effective dose by ingestion of mussels (Po-210) is 50-250 μSv/year.

Table 3: Information on phosphogypsum disposal sites in Italy (local Enichem contacts).

Site	Volume (m^3)	Operation period of dump	Position	Reclamation	Notes
Veneto (Campalto-Venezia)	200.000-250.000	1965-80	Facing the lagoon	Started	Other three areas in the Venice Lagoon to investigate
Calabria (Crotone)	/	/	/	/	Plant operated in 1926-1986 (discharge to sea for long time)
Sardegna (Porto Torres)	800.000	1972-82	Old quarry 1 km from the sea	Concluded	
Sicilia (Gela)	6.000.000	1981-92	1 km from the sea	Programmed	

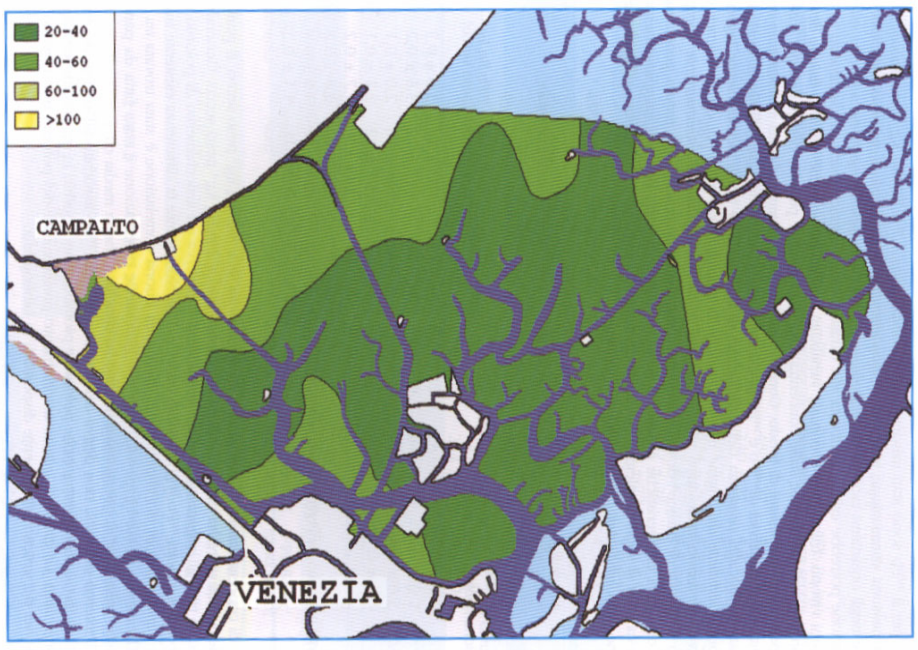

Fig. 1: Pb-210 activity concentration (Bq/kg, dry weight) in surface sediments at Campalto disposal site (4).

3.2 Steel production (integrated steelworks)

Iron ores have moderate contents of natural radionuclides; following high temperature treatments, emissions concentrate natural radionuclides, particularly Pb-210 and Po-210 (5). The processes of agglomeration of ore and smelting in the blast furnace are critical from this point of view. Most of the produced dust is captured by filters and then disposed in dumps, the rest is released to air. Similar problems are caused in coke distillation; furthermore, tar, the solid residue of distillate, is reported to concentrate Pb-210 and Po-210 (6).
Italy is the 10th steel producer in the world. In 2000, 40% of the overall production (26.7 million t) has been realized in four integrated steelworks, the rest in 38 electric-arc furnace steelworks.
All four integrated steelworks, located in Friuli Venezia-Giulia, Toscana, Liguria, Puglia and holded by two different private groups (Riva and Lucchini), own coke oven batteries, and two of them also agglomeration plants.
The four steelworks fall within the application field of the IPPC directive.

Table 4: Information on steelworks in Italy (Federacciai - main sector association - official reports and websites of companies).

Location	Yearly production (t of steel)	Blast furnaces (n°)	Agglomeration plants (n°)	Coke oven plants (n° batteries)
Genova (Liguria)	1.200.000	1	0	4
Trieste (Friuli V.Giulia)	800.000	2	1	2
Piombino (Toscana)	2.400.000	1	0	1
Taranto (Puglia)	6.300.000	5	2	9

3.3 Processing of Zircon sands

It is well known that zircon sands may present elevated natural radionuclides content (7). Critical work activities are the sand milling for production of zircon silicate flour, the manufacture of refractory materials, that can use zircon sands as raw materials, and the manufacture of ceramic and tiles, where zircon silicate flour is generally used in enamels or in the mixture itself of a particular tile named “white porcelain stoneware” (8). Potential environmental impact pathways are: air emissions, in particular Pb-210 and Po-210 occurring from smelting in the production of refractory materials, discharges of depuration waters and sludges, use of finished products in buildings.
Italy imports about 170.000 t per year of zircon sands, 70% of which is used in ceramic industry. There are about ten milling firms, located principally in Emilia-Romagna, Toscana and Liguria.
A centralized data-base has been used for the individuation of companies which manufacture refractory materials, a specific code for the extraction of this activity being available (9): 132 firms result, but their number should be sensibly

less (zircon sands are not used by each of them). With regard to the number of ceramic colourings and ceramic products manufacturers, the information available is that 20 and 78 firms join respective main sector associations: Ceramicolor (90% market representativity) and Federceramica (70% market representativity).
It is necessary to identify more precisely the industries which operate in the specific field of tiles and, probably, to select only firms with significant zircon sands use rate.
IPPC Directive includes the manufacture of ceramic products, provided that a certain production threshold is exceeded.
In Emilia-Romagna a relevant investigation has been carried out about the tiles working cycle. Activity measurements (by gamma spectrometry) of U-238, Th-232 decay products and of K-40 on an elevated number of raw materials, finished products and residues samples have been performed (8).

Table 5: U-238, Th-232 and K-40 activity concentrations (Bq/kg) measured on raw materials, residues and finished products in tiles working.

Samples	U-238	Th-232	K-40
Raw materials	26-58	38-73	422-1286
Zircon silicate (< 5 µm)	2334	880	/
Zircon silicate (< 45 µm)	2084	858	/
Sludges	68-354	30-119	266-427
White porcelain stoneware	118-247	40-89	528-1000
Red porcelain stoneware	42	42	625
Black porcelain stoneware	39	41	768
Other tiles	27-88	42-69	544-977

The majority of the finished products presents moderate values of concentrations, unlike white porcelain stoneware, that contains zircon flour into mixture. The activity values in sludges are not negligible on average and this suggests to keep monitored the water depuration cycle. Indoor use of the more common ceramic tiles seems not to significantly contribute to gamma irradiation or to radon gas immission; however attention should be paid to particular cases and to eventually high external beta irradiation (radionuclides from zircon sands are spread over the thin coat of enamel that covers tiles) (10).
With regard to the milling processes and to refractory materials manufacture, italian studies are documented only as far as workers exposure is concerned.

3.4 Oil and gas extraction /oil refineries

3.4.1 OIL AND GAS EXTRACTION

In many oil and gas extraction plants the build up of scales and sludges with high Ra-226/Pb-210 concentration in pipework, vessels and other components, constitutes a significant radiological hazard (11, 12). The main radiation protection problems are the management of this technologic waste, involving the removal and disposal of contaminated scales and sludges, the accidental dispersion of material that contains NORM, the release to the environment of connate water after separation (possibly). It must be emphasized that the attention paid to NORM management by the extraction industry is usually high.
ENI (AGIP division) and Edison GAS are the main companies that make oil and gas extraction activities in Italy. The prevailing activity is the gas extraction, with a production of 14.9×10^9 m^3 and 1.3×10^9 m^3 respectively from ENI and Edison GAS in 2000. 23% of the ENI overall hydrocarbons production in 2000 is made up of oil. In Figure 2, which shows the distribution of Italian ENI wells (approximately 7000 organized in 4 districts), the presence of off-shore wells and an important directrix (Appennini ridge) for on-shore wells can be noticed. Edison has 48 concessions and 40 explorative permissions. The 2000 companies reports and direct contacts with the companies have been used as information sources.
In 1992 an extensive investigation was carried out in ENI plants with measurements of gamma dose rate and of U-238, Th-232 and Ra-226 activity concentration in scales and connate water (12): hundreds of wells, tens of centrals, fields and platforms for both oil and gas were monitored in Italy and in Africa. Table 6 synthesizes data on activity concentration for samples from national plants: two high values of Ra-226 in oil wells scales (thousands of Bq/kg) and one high value of Ra-226 in a “mixed” well connate water (20 Bq/kg; Ra-226 in Italian drinking water ranges from 2×10^{-4} to 1.2 Bq/kg (13)) are evident.

Table 6: U-238, Th-232 and Ra-226 activity concentration in scales and waters of italian plants (Bq/kg).

Sample type	Site	Plant features	Extracted hydrocarbon	U-238	Th-232	Ra-226
Scales	Po valley	Extraction	Liquid	< 0.9	< 0.8	2890 ± 578
		Extraction	Liquid	< 0.9	< 0.8	1126 ± 225
		Extraction	Mixed	< 0.9	< 0.8	120 ± 24
		Collection	Gaseous	23.8 ± 4.3	18.9 ± 3.8	30 ± 6
		Collection	Gaseous	53.8 ± 10.8	< 0.8	< 2.7
	South Italy	Collection	Liquid	11.3 ± 2.3	< 0.8	110 ± 22
Waters	Po valley	Extraction	Mixed	$< 4.5\times10^{-3}$	$< 4.0\times10^{-3}$	$2.0\times10^{1}\pm4.0$
		Extraction	Liquid	$1.5\times10^{-2}\pm3.0\times10^{-3}$	$< 4.0\times10^{-3}$	$2.3\times10^{-1}\pm4.6\times10^{-2}$
	Adriatic Sea	Offshore platform	Gas	$7.3\times10^{-3}\pm1.5\times10^{-3}$	$< 4.0\times10^{-3}$	$6.0\times10^{-2}\pm1.2\times10^{-2}$

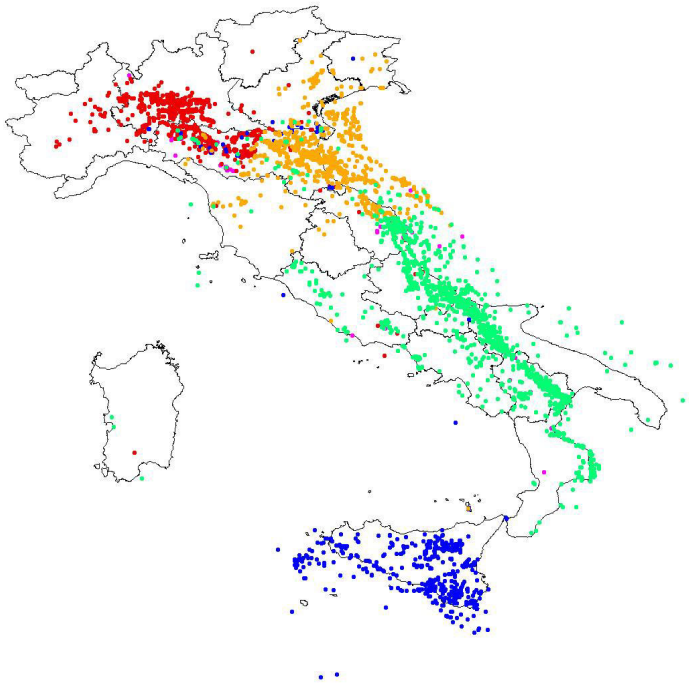

Fig. 2: Location of ENI (AGIP div.) Italian wells, organized in districts (red: Crema - orange: Ravenna - green: Ortona - blu: Gela).

3.4.2 OIL REFINERIES

Oil refineries are here accounted for because they fall within the application field of legislative decree n° 241/2000, but the working cycle has not been investigated as far as NORM are concerned. At present, there are 18 oil refineries in Italy (8 in the North, 6 in the Center and 4 in the South) according to the Ministry of Environment 2000 report (refineries are included in the legislation of industries at relevant accident risk thus being subjected to notification), but their number could be higher (source: 1999 Industries Register of Chamber of Commerce). Refineries are explicitly included in the Directive IPPC, too.

3.5 Uranium mines

At the end of Seventies two uranium mines, both located in Lombardia, started activity in Italy, for a research work; later on, they were closed because of declining of the national nuclear plan. The radiological impact on the environment should have been reduced by the technique used for this explorative phase, of digging “sterile” tunnels, parallel to uraniferous mineralization: therefore residues should have mainly a “sterile” origin (14).

3.6 Coal -fired power plants

Radiological significance of this activity arises from the ashes concentration of natural radionuclides originally present in the coal (1). The main environmental impact pathways are the release of fly ash to the atmosphere (critical is the ingestion of Pb-210 and Po-210 in foodstuffs grown in contaminated ground by nearby inhabitants), and the use of ash as cement additive in building industry (15); disposal of ash in landfill should also be considered.
At the moment, over 70% of electricity in Italy is provided by societies of ENEL Group, that produces almost entirely the electricity generated from coal. The Group has in all 13 coal-fired power plants: their activities in 1999 are shown in table 7.

Table 7: ENEL Group coal-fired power plants (1999).

Denomination	Location	Company	Gross production of electricity from coal (GWh)	Use of coal (t x 1000)
Vado Ligure	Liguria	Interpower	3712	1347
Genova	Liguria	Enel Produzione	2057	812
La Spezia	Liguria	Enel Produzione	2936	999
Fusina	Veneto	Enel Produzione	4078	1411
Porto Marghera	Veneto	Enel Produzione	781	326
Monfalcone	F.V.Giulia	Elettrogen	2115	714
Santa Barbara	Toscana	Enel Produzione	0	0
Bastardo	Umbria	Enel Produzione	996	376
Pietrafitta	Umbria	Enel Produzione	0	0
Brindisi	Puglia	Enel Produzione	0	0
Brindisi Sud	Puglia	Eurogen	5559	2005
Santa Gilla	Sardegna	Enel Produzione	0	0
Sulcis	Sardegna	Enel Produzione	1102	403
TOTAL			23340	8395

In 2000 the group's net production of electricity from coal was 23.3×10^6 kWh (about 13% of the total energy), and about 1.000.000 t of ash were produced,

over 96% of which is fly ash (nowadays many plants are provided with trituration sections that reduce the bottom ash component); the whole of the ash was used as additive in cement or road pavements. Used information sources are: 1999 and 2000 ENEL reports, direct contacts with the group.
The coal-fired power plants fall within the application field of the IPPC directive. Radiological assessments related to the coal-fired energy cycle have been performed several times in Italy (16, for instance). Recently, the Regional Agency for the Environmental Protection of Liguria (ARPA Liguria) has started up systematic monitoring about three local stations, measuring K-40, U-238 and Th-232 decay products (by means of gamma spectrometry) in various samples. Tables 8 and 9 present the mean values for coal and ash activity, resulted in the period 1998-2000 (17).
The mean values for coal are in accordance with literature references (1); there is an evident variability of radioactivity depending on the place of origin (in particular for U-238 and Th-232).
Also the mean values for ash are in accordance with the literature references (1, 15). No important differences among data from the three plants are evident. The predicted prevalence of radioactivity in fly ashes with respect to bottom ashes is confirmed. The ratio between the mean values of U-238 in ash and coal gives a coarse assessment of the coal ash content (14%), that it is compatible with literature references (15).

Table 8: Mean activity concentration (Bq/kg) for coal of various origin (in brackets the measured range) (17).

Place of origin	N° samples	Th-232	U-238	K-40
Usa	13	11.0 (5/13)	15.8 (7/21)	68.9 (48/103)
Colombia	30	3.3 (2/6)	5.8 (3/11)	36.2 (14/81)
South Africa	23	25.7 (15/34)	30.3 (14/42)	28.2 (17/70)
Indonesia	16	7.1 (4/18)	6.2 (3/17)	49.1 (10/76)
Poland	14	13.6 (9/18)	22.8 (14/31)	72.7 (37/94)
Venezuela	7	4.1 (3/5)	5.3 (4/6)	43.6 (3/58)
China	2	36.5 (36/37)	31.0 (31/31)	26.5 (23/3)
Russia	6	8.7 (7/11)	10.0 (8/12)	61.8 (42/93)
AVERAGE	111	13.7	15.9	48.4

Table 9: Mean Ash activity concentration in three ENEL coal-fired power stations (Bq/kg) (17).

Station	Type of ash	N° samples	Th-232	U-238(*)	K-40
Genova	Bottom	11	72	91	343
	Fly	11	92	118	452
Vado Ligure	Bottom	7	86	108	465
	Fly	10	96	137	478
La Spezia	Bottom	7	106	119	489
	Fly	7	104	123	445
AVERAGE	Bottom	25	88	106	432
	Fly	28	97	126	458

(*) It is assumed that U-238 equals the Ra-226 content.

4 CONCLUSIONS

In this paper the results of a first research on work activities characterized by a potential presence of NORM in Italy are presented; the research prepares the detailed data collection from the firms intended to provide information on these industries to adequately study their radiological pressure on the environment, for further impact predictions.

The NORM project is developed by the National Topic Centre on Physical Agents (CTN-AGF), coordination of Regional Environmental Agencies (ARPA), within the wider project of National Agency for the Environmental Protection (ANPA) for the set up of the National Environmental Informative System.

Even if the collected information is often indicative and typically describing only number and location of work activities, it is nevertheless possible to develop some considerations.

The national phosphate industry records today the absence of processes producing phosphoric acid; however, various phosphogypsum disposals sites occur throughout the country, although remediation measures have usually been adopted. Radiological hazards of fertilizers production has to be better investigated.

With regard to the processing of zircon sands, the most relevant fact consists maybe in the amount and variety of activity itself: investigations have to be conducted as far as emissions and discharges are concerned, but also with respect to the use of some finished products in building industry.

Impact assessments seem opportune, following fitting procedures (5,15), on the 4 integrated steelworks and the 13 coal-fired power plants; in fact these

industries cover respectively 40% of national steel production and 10% of national electricity production.
In Italy, oil and gas extraction is very intense (more than 7000 wells owned by the main national producer), however the attention paid to NORM problem by the operators is historically high.
Other work activities, outstanding as potentially hazardous in the scientific literature, have to be further considered.

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