RADIOACTIVITY IN THE SCRAP RECYCLING PROCESS: RADIATION PROTECTION ASPECTS AND EXPERIMENTAL MONITORING PROBLEMS

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ABSTRACT

The steel scrap recycling by steel mill is increasing moved by profits and by pourpose of protection of environmental resources. Besides the use of radioactive sources in several fields (medical, industrial and in scientific reserches) on one hand, and the disposal of made radioactive materials from nuclear reactors on the other one, makes the likelihood no more negligible that some radionuclides could be found, accidentaly or fraudulently, in steel recycling scrap. Radiation protection problems for surveillance both employees in the production cycle and of people and environment in general arose.

First of all, we characterize different type of radioactive materials that can found in scraps, pointing out the potential hazards from exposure of workers and people and from environmental contamination, related to physical and chemical specifications of the involved radionuclides.

Some suitable monitoring equipements for scrap recycling facilities are discussed, related to the different step of production cycle (transport, storage, manipulation and melting).

At last experimental data, taken in some periods of the monitoring campaign made at the italian border on imported scraps, are presented.

INTRODUCTION

Possible Scrap Component	Possible Radioactive Material
Aircraft devices, luminous	H-3 Pm-147 Ra-226 Sr-90 Kr-85
Air ionizing devices	H-3 Po-21, Ra-226 Am-241
Automobile shift quadrants	H-3
Compass,marine:navigational equipement	H-3 Ra-226
Dewpoint gauges	Ra-226 Th
Electron & vacuum tubes	H-3 Co-60 Ni-63 Kr-85Cs-137 Pm-147 Ra-226 Th
Fire/smoke detectors	U Ra-226 Am-241
Gauges (thickness, calibration, porcess controll level measurement)	Am-241 Am-241/Be, Sr-90, Cs-137, Co-60 Ra-226 Kr-85 Ra-226/Be Ir-192
Ice detector	Sr-90
Industrial radiography sources	Co-60 Ir-192 Ra-226
Irradiators,self-contained	Co-60 Cs-137
Lightning rods	Ra-226 Th
Lock illuminators, automobile	H-3 Pm-147 C-14
Luminous signs	H-3 Pm-147 Kr-85 Ra-226
Medical devices (brachytherapy, teletherapy)	Am-241 Co-60 Cs-137 Ga-67 Ir-192 I-125 Ra-226 Sr-90
Radiation leak detectors	Kr-85
Refractory	Co-60
Shipping containers, shielded	Cs-137 Ra-226 Ir-192 Am-241
Spark gap irradiators (fuel oil burners)	Co-60
Static eliminators	Am-241 Po-210 Ra-226
Thermostat (dials, pointers)	H-3 Pm147
Well logging tools	Cs-137 Ra-226 Ra226/Be Am-241/Be
Timepieces	Th H-3 Pm-147 Ra-226

Table 1 : Scrap bearing sealed radioactive materials.

Although the use and disposal of the radioactive material is usually regulated in every industrialized country, the likelihood that radioactive sources and/or contaminated metals can enter the recycling process as scrap cannot be overlooked also if we exclude completely the fraudulent hypothesis.

A further reflection arises from tab.1, where some radioactive industrial devices or gauges are identified (a several thousand of licences/year in every industrialized contry) meanwhile the summary of known incidents occurred in the world with their cost of decontamination is presented in tab.2 [1] [2].

Year & location	Radionuclide	Activity (10exp10) Bq	Probable Origin	Decont. cost Milion ($)	Comments
1983 - New York	Co-60	93	ind. g-grap.	4	
1083 - Mexico	Co-60	1500	Ter.g-grap	?	.02-375 mR/h (°)
1984 - Alabama	Cs-137	37-190	Gauge ?	?	
1984 - Tawain	C0-60	37-74	Gauge	?	.08 mR/h (°)
1985 - Brazil	Co-60	?	Furn. wall	?	96 Bq/g (°)
1985 - California	Cs-137	5.6	Gauge	1.5	
1985 - Alabama	Cs-137	.037-.19	Gauge	0.6	18200 Bq/g in
1987 - Tennessee	Cs-137	.093	Gauge	0.21 (*)	soil contamination
1987 - Indiana (Al)	Ra-226	.074	Gauge	?	
1988 - Missouri (Cu)	Accel. Prod.	?	?	?	
1988 - California (Pb)	Cs-137	.074-.093	Gauge	?	
1989 - Louisiana	Cs-137	19	Gauge ?	0.05 (*)	
1989 - Italy	Cs-137	110	?	?	
1989 - Pennsylvania	Th	?	charge		
1989 - U.S.S.R. (Al)	?	?	?	?	
1990 - Italy	Cs-137	?	Gauge ?	?	
1990 - Ireland	Cs-137	?	Gauge ?	?	
1990 - Utah	Cs-137	?	Gauge	?	
1991 - Tennessee (Al)	Th	?	?	0.1	
1991 - India	Co-60	.74-19	?	?	
1992 - Kentucky	Cs-137	1.2	?	2 (*)	
1992 - Virginia (Al)	Ra-226	?	?		
1992 - Texas	Cs-137	.74	Gauge ?	app. 1(*)	
1992 - Russia (Cu)	Co-60	?	?	?	
1992 - Illinois	Cs-137	?	?	?	
1993 - New York	Cs-137	3.7	Gauge ?	app. 2(*)	
1993 - Kentucky	Cs-137	.74	Gauge ?	app. 2(*)	
1993 - Georgia (Zn)	U	?	U castings	?	
1993 - Kazakhstan	Co-60	<.074	?	?	

(*) Costs of disposal of flue dust not included
(°) Activity and/or exposition in final product

Table 2: Incidents.

IDENTIFICATION OF RADIOACTIVE SCRAP

Radioactive materials can enter the scrap recycling process in different ways:

inherently radioactive scrap, in which the radioactivity is an integral part of the metal and cannot be removed. This is the case of:

a) metals (Fe, Co, Mn, Ni) activated by nuclear reactors or accelerators;
b) industrial thoriated products made by adding the NORM (Naturally Occuring Radioactive Material) Thorium to usual components in order to improve mechanical, electrical and termical performances;
c) products coming from furnaces where a radioactive source was previously melted;

scrap bearing sealed radioactive materials, i.e. radioactive sources inside the industrial devices or gauges, or shielded/unshielded sealed sources among the pieces of scrap;

scrap contaminated with radioactive material, when the scrap becomes "dirty" with some radioactive material which is not chemically fixed to it, but can be dispersed by chemical and physical agents (oxidation, fuming, wind). This is the case of:

a) damaged unsealed sources come accidentally to scrap;
b) pipes and equipments coming from some industrial processes such as the extraction of gas,oil and minerals, which have deposited NORM radioactive material on internal walls.

WHICH ARE THE MOST PROBABLE RADIONUCLIDES IN THE SCRAP?

Tab. n°1 presents a list of several type of " gadget" which have a radioactive source inside, from more familiar therapeutic and medical devices or commercial applications in the form of smoke detector, till more precisely industrial applications like gauges to measure the thickness and/or the moisture in rolled products, the level or the specific gravity of liquids, or to weight accurately materials which are hot, abrasive, corrosive or difficult to weigh (on belt conveyor).This table individuates 20 differents radionuclides which it is possible to find in iron scrap in principle, but only few of them offer the combined suitableties of long half-life and large enough number of sources in use to make their entry in a steel - industry more probable.

Among them, the greatest potential hazard is represented by the following radionuclides: Co-60, Cs-137, Am-241, Ra-226, Sr-90.

Year & location	Radionuclide	Origin	Comments	
1984 - Pennsylvania	------	shield box of medic. diagn. device	symbol	°
1985 - Wahington NORM		scale ol well casing		
1985 - Pennsylvania	------	shield box	symbol	°
1985 - Pennsylvania	?	stainless steel tubes	.7mR/h at contact	
1985 - Florida	NORM	scrap steel from phospate plant	10 mR/h	" "
1986 - Texas	NORM	water softener housing in scrap	3mR/h	" "
1986 - Texas	NORM	scale in oil well casing	.045 mR/h	" "
1986 - Texas	?	pipe in scrap		
1986 - Florida	Sr-90	source found in empty rail car	800 mR/h	" "
1986 - Wisconsin	-------	indus. radiog.shield	symbol	°
1987 - Pennsylvania	Ra-226, Th	stain.s.pipes containing scale	1480 Bq/g - Ra 999 Bq/g - Th	
1987 - Pennsylvania	Ra-226, Th	stain.s.pipes from phospate p.p.	0.5 mR/h at contact	
1987 - Pennsylvania	NORM	stain.s.pipes containing scale	1 mR/h	" "

Table 3: Radioactive material and suspected objects findings.

TYPE OF SCRAPS

Name	Density (g/cm*3)	"probable "radioactive devices inside
Sheared 1	0.48--0.56	level gauges on tank wall, st.- eliminator
Sheared 2 (light conduit)	0.80--0.961	
Busheling (residue from punch)	0.48--1.20	
Bundles 1 (light scrap compressed)	0.64--1.12	
Bundles 2 (car and mix compressed)	1.12--1.44	less trascurable, because they catch-all
Turnings	0.80--1.28	
Plate and Structural (demolition)	0.96--1.12	indust.g-ray device,medical, NORM
Slitter	0.96--1.28	
Shredded (Frag)	1.04--1.2	level gauges on tank wall, st.- eliminator
Cast Borings (small chips)	1.44--2.24	
Cut Plate, Foundry and Ship Scrap	2.40--2.72	
Packed Bars	2.88--4.00	

Table 4: Typical scrap.

The grade of scrape used in steel mill depends on different factors:

a) size and type of furnaces;
b) kind and quantity of final product;
c) market-price.

All different metal scrap can be catalogued by size, component and by their origin, as listed in tab. 4 where, the name is associated with the density and industrial radioactive gadgets. This table points out that scrap's density has a quite large variability interval ((0.6 -- 4)g/cm*3), keeping almost always lower than the steel's half density (steel density = 7.84 g/cm*3).

RADIATION PROTECTION ASPECTS OF RADIOACTIVE MATERIAL SCRAP

The way a radioactive material enters a scrap streamline, together to radionuclide involved and its quantity, determines the degree of detectability, the potential hazard to employees and customers, and the likelihood of causing widespread contamination, with the final cost of the decontamination.

Once contaminated scrap is entered the consequences can be very different: environmental impact could extend not only to steel mill but also to outside areas, such as to cause internal and/or external exposition to workers and customers but also to population.

Obviously scrap-plant workers, handling metal scraps, are potentially at more risk to external exposition, because they could come to direct contact with g-radioactive gadgets; while internal exposition could result from rupture of sources's seal and from spreading of radioactive material in the workplace. In the last case a worker should unknowingly contaminate other areas either inside or outside the scrap facility (Mexican incident).

A sort of probability that a type of radioactive material, rather than another one, by chance enter the scrap, can be defined by examining the happened incidents, the type of radioactive scraps found in these past years, also considering the amount of radioactive devices into circulation, and the involved radionuclide. Furthermore, this "type of probability" can be also connected to the scrap type, grade, nature and origin.

In this view, it is more likely to find an industrial g-ray device, or scraps contaminated by NORM radioactive material in heavy scraps loads rather than a static-eliminator. This last one has the greatest chance to enter the cut plate scraps, either at the original condition, or pressed packages , sheared or shredded. From this point of view, the scraps coming from steel waste process (chips, punchings, turnings,plates)should offer less risks in principle.

If a radioactive material is melted, the nature of a specific radionuclide involved will determine the potential hazard and its consequences because it will react chemically with steel, other metals and impurities, and flux the same as its stable, non radioactive element.

Concerns about radionuclides listed above, and their likely pathways due to the melting are [1] [2]

Element	**pathway**	**trace**
Cobalt	Finished product (98.9%)	slag (1%), flue dust (.1%)
Cesium	flue dust (89.9%) + slag (10%)	finished product (.1%)
Americium, Radium, Thorium, Strontium	slag	flue dust

As regards material with uniformly distributed radioactivity is to be expected a minimal superficial contamination during the scrap handling phases, a not relevant external exposition for workers and a low radioactivity level on final products.

EXPERIMENTAL ASPECTS IN RADIOACTIVITY CONTROL IN SCRAPS

Before starting technical considerations, we would emphasize the importance of visual identification of suspect object, in scrap monitoring. Table 3 clearly shows the positive and encouraging results of this practice.

The main goal in scrap monitoring is maximize of the probability of g-emitting source detection, whichever position inside a load, activity, and shieled by means of lead or transportation box. In order to perform this, we have to identify the worst experimental conditions and the suitable instrumentation.

The first problem is that scrap itself is an "important" shield for gamma radiation, which energy is degradated due to the interations with the metal meets during its pathway.

The following table presents results on the energy degradation of a Cs-137, from tests with a gamma beam penetrating different steel, lead and scrap thickness (scrape density (.48-.8) g/cm*3, and thickness expressed in meter).

THICKNESS	none (primary)	.00625 steel	.0094 steel	.915 scrap	1.5-1.8 scrap totally degradate	.05 lead
Cs-137 PEAK	100%	75%	68%	11%	8%	

In this example the upper limit for scrap thikness allowing a significant chance of detection, is 1.2 m about, a value close to the halfwidth of a rail wagon or a TIR (2.7 m approximately).

Further a radioactive material still could be inside the housing shield. A bad enough situation (low radioactivity) can be simulated by a gauging device containing radioactive material covered by scrap; in fact a de facto standard exists for a such type of devices which limits the radiation emitted through the housing walls to 5 mR/h or less a distance of 0.3 m, from external surface, so that few centemetre ten thickness of scrap will reduce the exposition level to the background one [4].

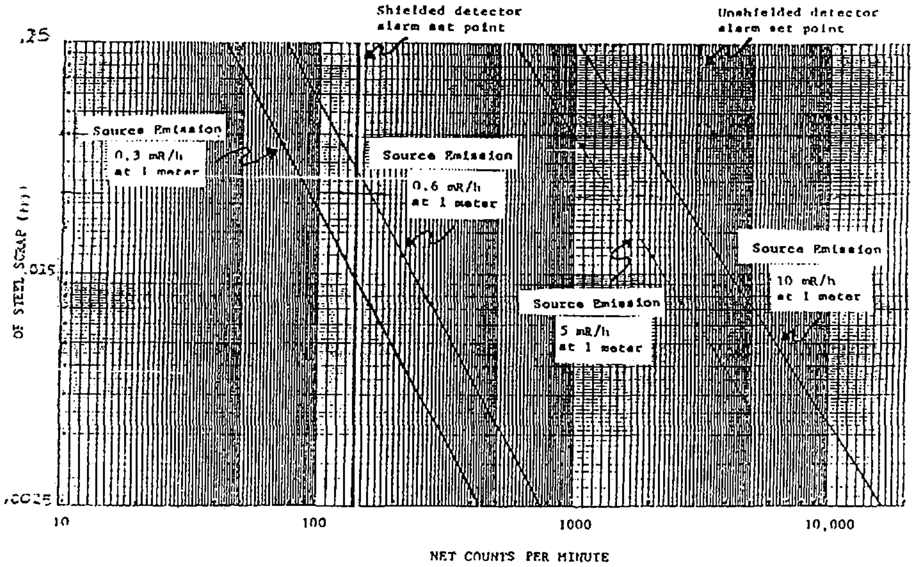


Fig. 1: Shielded/unshielded 2"×2" sodium iodide detector with ratemeter based circuit.

In fig. 1 data from a such type of simulation are presented; in this test has been used a shielded/unshielded 2x2 inches sodium iodide counting ratemeter, with an alarm threshold set at 2 times the backgrounnd one.

At last we have to note that positive factor is made instead by the presence of air inside the scrap, this allowing "radiation streamer " detection.

TECHNICAL FEATURES OF DETECTION EQUIPMENTS

The starting point in scrap monitoring is to have the chance to measure radioctivity level of the order of natural background, on a large amount of material (until 59 ton/railtruck).

An ideal system should have the following features:

1) high detection efficency;
2) large solid angle;
3) sensibility enabling to appreciate background variations (.01 mSv/h; .01 mGy/h);
4) the system would be shielded to consent the lowest alarm threshold. In such sense the results of the test in fig.1 are enlightening.
5) short response time (order of second)
6) continous monitoring of background: in fact it may change with atmospherical conditions, with presence of building material nearby and for the checked load itself shield effects.
7) alarm threshold easily programmable during background measurements.

8) low probability of false alarm, in order to not interfere with production line, and to avoid loss of trust from control personnel.

The individuation of the most available "scenarios" for the measurements of matter, is one of the most complex problems to define, involving not only technical aspects, but legal and economical too; in this context we mean as "scenario" both the site inside a factory (the weight and stocking places etc.), and the conditions in which the scrap is (inside TIR or rail-truck, loose, during the load-unload operations).

In order to simplify the analisys of the problem, let us identify two opposite measure scenarios, that is the control on the whole load and on loose material, examining of both advantages and disadvantages.

In the case of monitoring of a whole load still placed above the rail-truck, TIR, or container:

1 adv.) it is possible to prevent steel plant contamination;
2 adv.) there is no interference with factory producting cycle, because for example the measurement operation may take place at the moment of the entrance of material in the factory, in the weigh place area ;
3 adv.) one has memory of the origin place of the scrap;
4 adv.) it is possible to return scrap to the sender.

On the contrary:

1 dis.) there is maximun scrap shield, and minimum probability of detecting emerging radiation ;
2 dis.) it should require the use of a fixed detection system, computerized and expensive one, in order to maximize the detection of strongly shielded sources;

As what regards the loose material:

1 adv.) the shielding effect of the scrap is minimum;
2 adv.) it is possible to carry out at the same time the measurements and the visual recognition of suspect material;
3 dis.) portable instrumentation is available, with relatively low cost.

On the contrary:

1 dis.) the control is too much depending on human factor, both during measure execution, and in results interpretation. In this case it is of basic importance the technical training of assigned personnel about all the aspects of measure operations;
2 dis.) control interferes with the production cycle;
3 dis.) one loses memory of origin place of scrap;
4 dis.) it is unpracticable the return to the sender of possible contaninated material.

The opportunity of monitoring the loose scrap during some of the phases of a foundry or steel mill production cycle, is nowadays in Italy one of the most controversial matters , and one of the less accepted by manufacturers.

The place in which to carry out the control cannot be defined "a priori", as it strongly depends on the specific steel production reality, on the kind of scrap it employes, on the load/unload techiniques for scraps (crampon, electromagnet, etc.), on the kind of kiln supplying (belt-convoyer, hopper, etc.), and so on; it should be studied case by case.

A further step of the control in the factory, is the radioactivity monitoring at the last stage of the production chain, monitoring the finished product and flue dust, respectively by a gamma spectrometry measuresments on fusion tests, and the radioactivity measurements on the fume knocking down system.

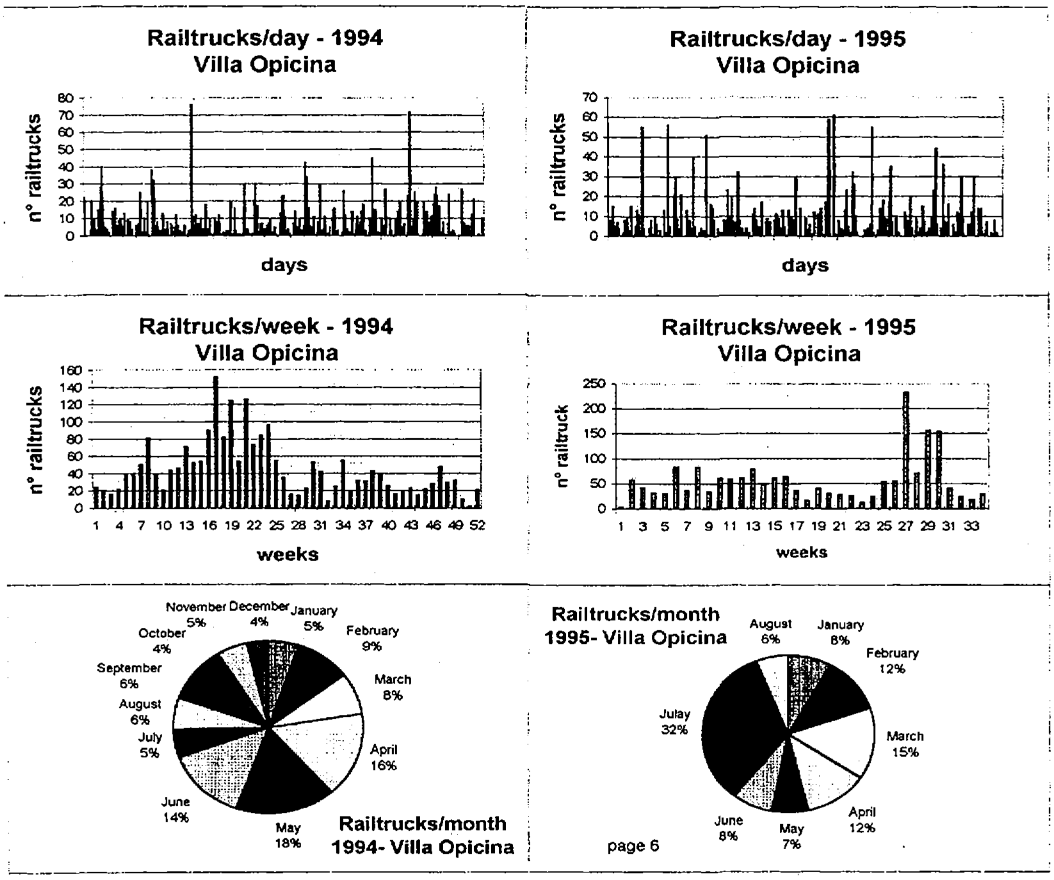
FIXED INSTRUMENTATION

Since 1989 the manifacturers of the field have been commercialized fixed systems "ad hoc", aimed to the radioactivity detection on scraps still placed on the transportation mean, (rail-trucks, TIR).

The most recent products use few plastc scintillators (2 - 4) of great dimensions (3700 - 7500 cm^3 approximatly), whose signals are managed and processed by a sophisticated software .The computer system is able to manage data while the load is moving, subtracting the background level, taking into account the shielding effect of the vehicle by measurements carried out before the load itself has passed through the detectors [3]. The software provides the calculus of the speed of the vehicle too, self-diagnosis tests, and the paper recording and monitor display of data relative to each scintillator, most of all in case of alarm.

ISPESL 'S CONTROL ACTIVITY ON BORDER

Since december 1993 ISPESL's radiation laboratory is engaged on radioactive monitoring of extra-Europe origin scrape; it has operated (and it is operating now) by Tarvisio (Udine) and Villa Opicina (Trieste) customs for what concerns rail-truck, Coccau (Udine), Fernetti (Trieste), Gorizia custom for TIR and trucks.



	Railt. Total Number			
1994			**1995**	
		Villa Opicina		
Accepted	2312		Accepted	1897
Returned	3		Returned	3
	0.13 %			0.16 %
		Tarvisio		
Accepted	21249			
Returned	12			
	0.06 %			

Fig. 2.

Fig.2 shows the number of for railtrucks/day, railtrucks/week during 1994 and 1995 in Villa Opicina, with the percentage of rejected ones. Some time, the big number of railtrucks/day,plus severe conditions for detection equipment and for workers (climatic conditions, rough ground, rail-truck moving, loud noise, large) make the control a hard work.

The protocol foresees a background measurement with an alarm threshold equal to background double value, checking both the sides of rail-truck with fixed or continuous measurements but in a time that sure to value the background level variation.

Experience made for so long a time, by monitoring so big a number of scrap loads, suggests us a number of "ergonomic features" which, together to the technical one's reported above, are usefull for a portable detector dedicated to radioactive monitoring of scraps, especially if it is used in a steel mill where more severe work conditions exist.

The "ergonomic features" are the following:

1) a large, well-lighted and visible display;
2) sound either for alarm, or to listen to counting rate, plus a headset to use in the case of loud noise;
3) possibility to fix the portable detector to the body with some special belt to have free hands;
4) possibility to select instrument functions simply and fast (threshold regulation, physical quantity selection, batteries substitution, etc..)by external switches, neither using screwdriwer nor removing covers;
5) availability of a 3-4 m long telescopic rod, solid and not heavy at the same time, whose parts can be attached simply and fast (for example a bajonet base);
6) a relatively not heavy detector to fix to the rod's extremity;
7) possibility to fix the rod to the body with a ad-hoc belt ;

A last a technical function seems to be very usefull ,that is the possibility to have background mean level on preselectable time intervals, (also short as few seconds),so to allow a simple and fast mapping of the measurement place.

CONSIDERATIONS ON BACKGROUND MEASURE RESULTS

In Villa Opicina custom place, the rail-trucks parking area presents an interesting tipology in which it is possible to identify several areas characterized by rather different values of background, due to the presence of building materials, penthouses and tuff, in some places and not in others.

By means of a sensible and fast counting ratemeter (as for example a 2x2 inches sodium iodide scintillation counter), it is possible to get a fast background map of these areas, and to select for each one the suitable alarm threshold. Right by means of such an instrument, we have measured sistematically different background mean values for each one of the defined areas, ranging from a minimum of 40 countings/s to a maximum of 120 countings/s, during a few minutes time interval.

With such a detector it is possible to check a high number of rail-truck, taking a reasonably brief time, however assuring a some meaning to the measurements.

As the last goal of scrape control is to discriminate the emission of the this material respect to the background, that is a performing relative measurement, we don't think important the instrument to necessarily provide the free air dose or equivalent dose rate, but it should be better to make reference to the primary quantity measured by itself, expressed as countings per time interval. The measurement of different radiation protection quantity would be left to further investigations, only in case of real alarm .

BIBLIOGRAPHY

1. "Radioactivity in the Scrap Recycling Process: Reccomended practice and Procedure", by Institute of Scrap Recycling Industries, Inc. ,January 12,1992.
2. " What to do when the alarm goes off:a suggested procedure for checking contamined metal products", by J.G.Yusko, (Pa. Department of Environmental Resources Bureau on Radiation Protection), and J.O.Lubenau (U.S.Nuclear Regulatory Commission State Programs).
3. "Materiale radioattivo nelle scorie metalliche", by J.G.Bellian, HARSHAW/BICRON, 6801 Cochran Road, Solon Ohio 44139,USA.
4. "Radioactivity material in steel scrap: its occurences, consequences and detection", by Lamastra, Health Physics Associates, inc., REV 03/89.