

# NATURAL RADIOACTIVITY IN BOTTLED MINERAL WATERS: A SURVEY IN NORTHERN ITALY

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A natural radioactivity monitoring on 21 best sold bottled mineral waters produced in Lombardia (Northern Italy) has been performed. Gross alpha and beta activities were first evaluated; subsequently  $^{226}\text{Ra}$  and uranium were measured, respectively by emanometric technique and ICP-MS. In order to assess with higher accuracy the dose contribution due to uranium isotopes in most active waters,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  were determined by alpha spectrometry on electrodeposited samples.  $^{234}\text{U}/^{238}\text{U}$  ratios were found to be higher than 1 in most cases. A correlation with chemical parameters of waters has also been attempted. Dose calculation for different classes of age was finally performed.

## **Introduction**

Recent national and international regulations on water intended for human consumption expressly exclude bottled mineral waters.<sup>1,2,3</sup> Bottled mineral waters have always been regarded as a voluptuary good, something

between a soft drink and a dietary or medical aid with a limited importance in human diet.

In the last years, anyway, consumption of bottled mineral water has widely increased in all western countries. For example, in the period 1990-2000,<sup>4</sup> mineral water consumption rose of 50 % in Germany and of 130 % in U.S.A.

Italy is the main mineral water producer in the world: in 2000, 9,500 million liters have been bottled (more than 1/3 of the whole European production), 700 million of which have been exported. Between 1995 and 2000 national production rose of 55 % and export of 120 %. Italy is also the main consumer of bottled mineral water. The national market absorbs more than 90% of the whole production. At present, about 9,000 million liters are yearly sold in Italy for a 3,000 million euro value. The average individual consumption is 160 l/year.

More than 50 % of bottled water is sold in northern Italy. Lombardia district is the main consumer (2 million l/year) and here individual consumption reaches 190 l/year.<sup>4</sup> Moreover Lombardia produces 36 brands of mineral water over the 257 Italian brands.

The increasing use of bottled mineral water makes it obvious to consider it an important element of the human diet, with special regard to children in lactation age: in this case mineral water is extensively used for powder milk preparation. As a consequence, Italian national regulation on tap and bottled water got closer in recent years.

All these facts, added to the lack of radioactivity data on mineral water, pushed us to carry on a monitoring campaign in Lombardia district. We have taken into consideration the 21 most widely sold bottled mineral brands produced in Lombardia, namely: San Pellegrino, Pracastello, Bracca, Boario, Gaverina, San Carlo Spinone, Orobica, Primula, Tavina, Limpia, Fonte Laura, Gajum, Pineta, Chiarella, San Francesco, Sant'Antonio, Frisia, Levissima, Daggio, Stella Alpina, Bernina. Gross alpha and beta,  $^{238}\text{U}$  and  $^{226}\text{Ra}$  concentrations were determined on every sample. In 15 selected cases the measurement of all uranium isotopes ( $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$ ) has been performed.

### **Experimental**

All reagents were analytical grade.  $^{90}\text{Sr}$ ,  $^{226}\text{Ra}$ ,  $^{241}\text{Am}$  and  $^{232}\text{U}$  CEA-DAMRI liquid standard solutions have been used. ICP-MS uranium standard (1000 ppm) and anionic resin Dowex 1X8 (50-100 mesh) were purchased from Aldrich and Chelex 100 resin (100-200 mesh) from Biorad.

Gross alpha activity was measured by a zinc sulphide scintillation counter (ASPN SE 105); gross beta activity was measured by a plastic, anticoincidence scintillation detector (ASPN CRS/1 PH). Emanometric measurement were performed by a Pylon WG-1001 vacuum water degassing system.

ICP-MS analysis were accomplished by a Finnigan Mat ITS 40 instrument.

Uranium spectrometric measurements were performed by a 7937/A Silena alpha spectrometer equipped with a BU-021 Perkin Elmer PIPS detector.

Water samples were obtained through normal distribution channels. When possible, still waters, stored in plastic bottles, were chosen.

#### *Gross alpha and beta activity*

EPA 00-01 method was used for alpha and beta activity measurements.<sup>5</sup> A proper amount of sample was first acidified by HNO<sub>3</sub> and the volume was evaporated to a small volume on a hot plate. The solution was transferred on a tared 50 mm diameter steel planchet and dried under a heat lamp. The residue was then ground by a glass rod and evenly dispersed. The planchet was then heated in a muffle furnace at 350°C for one hour, cooled in a dessiccator and weighed. The final weight ranged between 80 and 120 mg ( $5 \pm 1$  mg/cm<sup>2</sup>).

The calibration was performed by a calcium sulphate nitric solution traced by <sup>241</sup>Am or <sup>90</sup>Sr/<sup>90</sup>Y standards: six standard planchets were prepared for alpha measurements and six for beta ones, following the above described procedure and with residue weight ranging from 50 to 150 mg. This allowed to calculate the efficiency for different layer thickness of actual samples.

Samples were measured for 60.000 seconds for both alpha and beta activity. Measurements were performed immediately after the preparation of the sample to minimize the influence of <sup>226</sup>Ra daughters ingrowth.

### *<sup>238</sup>U determination*

<sup>238</sup>U concentration was measured by ICP mass spectrometry directly on the sample without any further pre-treatment. The lower limit of detection was 0.2 µg/l corresponding to 2.5 mBq/l activity of <sup>238</sup>U.

### *Uranium isotopes determination*

<sup>238</sup>U, <sup>235</sup>U and <sup>234</sup>U concentrations were determined by alpha spectrometry on electrodeposited samples after pre-concentration and chemical separation.

Water samples were first selectively preconcentrated by ionic exchange on Chelex 100 chelating resin.<sup>6,7</sup> A 20 ml volume of Chelex resin was transferred in a glass chromatographic column, washed with 2 M nitric acid and conditioned by elution of 1 M acetic buffer. A 2 liters water sample was acidified with 100 ml of concentrated nitric acid, traced by <sup>232</sup>U standard solution and stirred for one hour. 200 ml of 1 M acetic buffer and NH<sub>4</sub>OH up to pH 5.3 were then added.

The solution was slowly eluted on the resin (5 ml/min c.a.). After a 50 ml water washing, uranium was recovered by elution with 4 M HCl (100 ml). The volume of hydrochloric solution was reduced to 40 ml by evaporation on a hot plate.

Uranium was then purified by ionic chromatography on Dowex 1X8 anionic resin.<sup>8</sup> The formerly obtained hydrochloric solution was slowly eluted (1 ml/min) in a small column containing 10 ml of resin, previously washed with 8 M HCl. The column was then rinsed with a solution of 57 ml HCl 8M and 6 ml of HI 57% (phosphate free, unstabilized). After a further washing with 40 ml of HCl 8 M, the uranium was eluted with 100 ml of HCl 0.1 M. The solution containing uranium was evaporated to dryness. Few drops of nitric acid were added before the end of the evaporation to eliminate completely iodine. The procedure was repeated till the end of red-brown fumes evolution.

Several authors described electrodeposition methods.<sup>9-11</sup> In this work, Italian Standard Method UNI 9778 was adopted.<sup>12</sup> The residue was dissolved with 1 ml of concentrated H<sub>2</sub>SO<sub>4</sub> and 1 ml of H<sub>2</sub>O<sub>2</sub> (120 vol.) by gentle heating. 10 ml of water, 1 ml of H<sub>2</sub>O<sub>2</sub> and some drops of methylred were then added. The pH was raised by 1 M NH<sub>4</sub>OH till a salmon-pink solution was obtained. The uranium was then electrodeposited on a stainless steel planchet (3 hrs, 5 mA, 5.7 V). The planchet was counted for 4000 minutes on an alpha spectrometer. Chemical yields, determined by <sup>232</sup>U internal tracer, ranged from 50 to 80%, the minimum detectable activity (MDA) was around 0.1 mBq/l.

#### *Radium determination*

<sup>226</sup>Ra was determined by emanation procedure followed by scintillation cell counting. 1-5 liters samples were acidified with nitric acid

and slowly evaporated on hot plate up to 500 ml (pH 2 c.a.). The solution was transferred in a glass bubbler (diam. 45 mm, h. 400 mm) with a teflon stopper equipped with two stopcocks. Standard stoppers for HPLC solvents degassing fit the purpose. After 20 days ingrowth period, the emanation procedure described by EML manual (Ra-02-RC) has been followed.

Calibration was accomplished with 500 ml radium standard solutions brought to conditions similar to the real samples by addition of  $\text{CaCl}_2$  and  $\text{HNO}_3$  in proper amounts. The MDA was 2 mBq/l for 5 liter samples.

#### *Chemical parameters*

Chemical and physical parameters of waters have not been re-determined. Most recent data as specified from producers have been used.

## Results

In Tab.1 chemical and physical data of waters are listed. Aquifer lithology (igneous, calcareous, sedimentary rocks), source temperature, dissolved CO<sub>2</sub>, pH, conductivity, dry residue and main dissolved ions concentrations are reported. Waters are ordered by decreasing dry residue. First 5 waters belong to the medium-mineral class (residue > 500 mg/l), the last one to the low-mineral class (residue < 50 mg), all the others to oligo-mineral class. It should be remarked that all the 5 medium mineral waters are produced in calcareous aquifers.

### Tab. 1

In Tab. 2 radioactivity data are reported. Gross alpha and beta activities were determined by total counts on the dried residue, <sup>238</sup>U by ICP mass spectrometry and <sup>226</sup>Ra by emanometry. Since ICP-MS analysis provides mass concentration results, <sup>238</sup>U activity concentrations were obtained multiplying mg/l uranium concentrations by <sup>238</sup>U specific activity (12.4 mBq/μg).

### Tab. 2

Gross alpha values range from < 3 to 550 mBq/l; gross beta values range from 27 to 1108 mBq/l (Fig. 1). Recommended WHO guideline activity concentrations for drinking water are exceeded in 4 cases for gross alpha activity (WHO recommended concentration < 0.1 Bq/l) and in 1 case for gross beta activity (WHO recommended concentration < 1 Bq/l).

**Fig. 1**

Higher alpha and beta activity values are observed in the most mineralised waters. A rough correlation can be found between gross alpha activity and dry residue ( $R^2= 0.76$ ) (Fig. 2). The correlation, however, appears to be driven by the most active samples; the  $R^2$  value drops to 0.16 if medium mineral waters (samples 1-5) are not considered (Fig. 3). The same considerations apply to beta activity values.

**Fig. 2**

**Fig. 3**

$^{238}\text{U}$  concentrations range from  $< 2.5$  to 122 mBq/l. Eighteen samples over twenty-one exhibit  $^{238}\text{U}$  values equal or lower than 50 mBq/l and  $^{226}\text{Ra}$  values lower than 10 mBq/l (Fig. 4).  $^{226}\text{Ra}$  concentrations are usually lower than  $^{238}\text{U}$  ones, except in the three most mineralised waters. Radium and uranium concentrations seem to be poorly correlated each other and to dry residue, especially if medium mineral waters are excluded.

**Fig. 4**

In selected cases, uranium isotopes concentration has been determined by alpha spectrometry on electrodeposited samples after uranium chemical separation. Results are reported in table 3.

Measured  $^{238}\text{U}$  concentrations generally agree with ICP-MS results. It can be noticed that  $^{234}\text{U}/^{238}\text{U}$  ratio is equal (within experimental uncertainty) or higher than one. Values up to 1.6 have been observed. The lack of uranium isotopic equilibrium in waters is a well-known phenomenon and has been ascribed to erosion mechanism of rocks.<sup>13</sup> These results show that

calculation of total uranium from  $^{238}\text{U}$  concentration under equilibrium hypothesis can lead to a significant underestimation.

### **Tab. 3**

#### **Doses**

As stated before, national and international regulations on drinking water expressly exclude bottled mineral waters, for which no reference level of committed effective dose is recommended. Nevertheless, in this work dose calculation was performed using drinking water intake values usually suggested by different international organisations, including WHO. Moreover, the dose reference level of 0.1 mSv/y suggested by WHO Guidelines and EC Directive 98/83 has been used for comparison purposes, though the assumed intake hypothesis may lead to overestimate the actual doses.

The total equivalent dose due to  $^{226}\text{Ra}$  and uranium isotopes was calculated for babies in lactation age (< 1 year), for children (2-7 years) and for adults (> 17 years). Activity-dose conversion factors reported by EC Directive 96/29 have been used. A water consumption of 250, 350 and 730 l/year has been supposed respectively for babies, children and adults. For radionuclide concentrations below the detection limit, the MDA was considered. When possible, dose was calculated considering measured activities of all uranium isotopes, otherwise the hypothesis of secular equilibrium between  $^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{235}\text{U}$  was assumed, even if it can lead to underestimate actual dose, as stated before.

For children and adults class ages, calculated doses are quite similar and range from 1 to 50  $\mu\text{Sv/y}$ . For babies class age, doses are higher and reach 0.26  $\text{mSv/y}$ ; the 0.1  $\text{mSv/year}$  reference level recommended in EC Directive 98/83 is exceeded in two cases. Highest doses are observed in medium-mineral water group (samples 1-5).

In Tab. 4 percentages of dose due to  $^{226}\text{Ra}$  are reported for samples with at least one radionuclide concentration higher than MDA. Because of its high radiotoxicity,  $^{226}\text{Ra}$  contribution to dose is large especially for the first age class. In children and adults age classes its contribution is generally lower than uranium one, except for most medium-mineral waters (samples 1-5).

### **Conclusions**

All most popular bottled mineral water produced in Lombardia have been examined from the radiometric point of view.

Gross alpha and beta, uranium and  $^{226}\text{Ra}$  analysis allow to get a fairly complete information on radioactive characteristics of most samples. Furthermore, gross alpha and beta measurements and uranium mass spectrometry analysis can be performed in a short time and give approximate but reliable information.

In most samples uranium isotopes represent the major radioactivity source in water; however, since the isotopic equilibrium is not always attained, all isotopes, with special regard to  $^{238}\text{U}$  and  $^{234}\text{U}$ , should be

individually measured. The determination method chosen, based on preconcentration by chelating resins, radiochemical purification, electrodeposition followed by alpha spectrometry measurement, gave good results. High spectral resolutions and, subsequently, precise  $^{234}\text{U}/^{238}\text{U}$  ratios were obtained. Most samples exhibited isotopic ratios higher than one and up to 1.6.

$^{226}\text{Ra}$  is usually present in lower amounts than uranium. Sample preconcentration combined to emanometric technique allowed to reach a MDA of 2 mBq/l, low enough for most waters.

No clear correlation exists between chemical-physical parameters and radioactivity, except for most mineralised waters. Over a wide range of dry residue values (up to 500 mg/l) all radiometric parameters appear to be poorly correlated, though the most radioactive samples belong to the medium-mineral water class (residue > 500 mg/l) and are produced in calcareous aquifers.

The first screening on Lombardia bottled waters showed that reference levels of gross alpha and beta activities given by international regulations are exceeded in few cases. Moreover, the reference level of committed effective dose for drinking water<sup>1,2,3</sup> is exceeded in only two cases and for the lowest age class (< 1 year) alone.

$^{226}\text{Ra}$  gives a relevant contribution to the equivalent dose, especially when babies age class is concerned. It's worth mentioning that the real value of the total dose should include the dose due to all isotopes, with special

regard to  $^{228}\text{Ra}$  and  $^{224}\text{Ra}$ , which show an even higher radiotoxicity. This is the aim of our job at present.

Very few radiometric data on Italian bottled water are available besides these ones. Recently 43 mineral water brands produced in Spain, Portugal and France have been analyzed.<sup>14</sup> It was not possible a direct comparison with our results due to differences in analysis sensitivities and the lack of chemical parameters. Nevertheless it should be remarked that at least one third of them exceeded 0.1 Bq/l gross alpha activity. Most waters of this group exhibited relatively high  $^{226}\text{Ra}$  concentrations and, as a consequence, equivalent doses were often higher than 0.1 mSv/year even for oligo-mineral waters. This behaviour stresses once more that no prediction on the radiometric activity of waters can be done on the basis of their chemical-physical parameters.

International regulations expressly exclude bottled mineral waters. Furthermore, derived limits for gross alpha and beta activities, intended to assure compliance to the reference dose limit of 0.1 mSv/y, were derived using activity-dose conversion factors proposed for adults. Nevertheless, mineral water is extensively used for powder milk preparation; activity-dose conversion factors for babies are the highest. As a consequence, bottled waters may greatly contribute to committed dose for the lowest classes of age. Monitoring of bottled waters radioactivity should therefore be strongly

recommended; special regulations should be enforced to protect the most exposed class of age.

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## Captions

Fig. 1 Frequency distribution of gross alpha and beta activities

Fig. 2 Correlation between the dry residue and gross alpha activity (all samples). Error bars are quoted for  $1\sigma$

Fig. 3 Correlation between the dry residue and gross alpha activity (excluding medium-mineral waters). Error bars are quoted for  $1\sigma$

Fig. 4 Frequency distribution of  $^{238}\text{U}$  and  $^{226}\text{Ra}$  activities

Table 1: Chemical-physical parameters of waters

Sample	Litol.	Temp. °C	CO <sub>2</sub> mg/l	pH	Conduct. μS/cm	Residue mg/l	Na mg/l	K mg/l	Mg mg/l	Ca mg/l	Cl mg/l	HCO <sub>3</sub> mg/l	SO <sub>4</sub> mg/l	SiO <sub>2</sub> mg/l
1	Cal	25.9	5.3	7.7	1306	1109	43.6	2.7	56	208	74	220	549	9
2	Cal	20.2	7.6	7.1	1191	892	25.1	2.3	49	164	45	244	395	6
3	Cal	7.8	4.1	7,4	881	682	19.4	1.7	42	134	31	259	286	7
4	Cal	14.8	12.5	7,2	756	597	6.4	2	41	124	6	305	235	8
5	Cal	13.2	2.6	7.8	703	521	16.3	0.9	32	112	9	339	153	12
6	Cal	11.6	6.2	7.7	630	406	7.2	0.7	31	93	3	351	74	9
7	Sed	7	9.7	7.5	599	385	17.1	1.4	22	90	15	323	55	11
8	Cal	8	6.7	7.6	512	375	10.8	0.7	30	80	3	317	85	10
9	Ign	13.5	6.2	7.3	564	354	8.9	2.1	26	88	8	360	27	14
10	Cal	12.9	1.4	7.9	344	212	0.5	0.1	23	46	1	229	108	1
11	Ign	11.5	4.7	7.8	339	211	1.7	0.7	22	46	1	240	10	5
12	Ign	8.7	0.8	7.7	288	205	1	0.4	9	60	2	197	15	3
13	Cal	8	1.5	8.1	276	196	0.4	0.2	10	58	1	208	7	4
14	Cal	10.5	6.6	7.7	265	187	0.9	0.2	22	39	1	211	10	4
15	Sed	14.1	3.5	7.8	208	135	3.8	0.7	5	35	1	134	1	14
16	Sed	14.1	2.1	8.0	210	133	4.9	0.7	6	32	1	135	2	15
17	Ign	10.2	7.3	7.4	111	82	2.7	2.9	2	17	2	49	14	10
18	Ign	5.8	0.8	7.8	108	74	1.7	1.8	2	20	1	56	14	6
19	Ign	14	1.5	7.3	87	52	1.9	0.7	3	9	1	41	5	7
20	Ign	10.7	0.7	7.7	85	50	1.1	0.8	3	10	1	40	6	6
21	Ign	6.2	7.5	7.5	67	37	0.8	0.8	1	8	0	20	7	5

Cal = calcareous rocks Sed = sedimentary rocks Ign = igneous rocks

Table 2: Water radioactivity data

Sample	Gross alpha		Gross beta		<sup>238</sup> U		<sup>226</sup> Ra	
	mBq/l	+/-	mBq/l	+/-	mBq/l	+/-	mBq/l	+/-
1	500	55	440	75	122	12	200	18
2	550	54	1108	99	89	9	70	9
3	315	38	178	45	102	10	140	15
4	103	22	110	31	50	5	7.6	0.8
5	54	14	144	29	3.6	0.4	9	1.4
6	61	12	94	24	9.1	0.9	< 2	
7	35	11	94	26	25	3	3,3	0.7
8	52	11	147	25	9.7	1.0	2	0.3
9	40	10	170	24	22	2	< 2	
10	17	3	47	13	17	2	< 2	
11	62	7	122	13	63	6	< 2	
12	< 10		109	15	4.6	0.5	< 2	
13	28	5	43	12	12.5	1.3	4.5	0.8
14	49	6	83	12	38	4	6.5	1.2
15	< 10		31	9	7.3	0.7	< 2	
16	14	4	52	9	6.6	0.7	< 2	
17	55	5	160	13	34	3	7,5	1.2
18	64	7	124	10	62	6	< 2	
19	< 3		27	4	< 2.5		< 2	
20	15	2	44	4	< 2.5		7	1
21	16	2	32	3	< 2.5		< 2	

Table 3: Uranium isotopes concentrations by alpha semiconductor spectrometry

Sample	<sup>238</sup> U		<sup>234</sup> U		<sup>235</sup> U		Total U		<sup>234</sup> U/ <sup>238</sup> U ratio	
	mBq/l	+/-	mBq/l	+/-	mBq/l	+/-	mBq/l	+/-		+/-
1	111.6	7.5	118.9	7.9	8.0	1.1	238.5	10.9	1.07	0.10
2	73.3	4.4	85.1	5.1	< 4		158.3	6.8	1.16	0.10
3	97.0	6.2	119.5	7.5	< 4		216.5	9.7	1.23	0.11
4	47.6	4.6	41.5	4.4	2.09	0.70	91.2	6.4	0.87	0.13
5	3.1	0.3	5.0	0.4	0.24	0.08	8.3	0.5	1.63	0.20
6	8.5	0.5	12.8	0.8	0.43	0.07	21.7	0.9	1.51	0.13
7	21.3	1.4	28.3	1.8	< 0,6		49.6	2.3	1.33	0.12
9	21.1	1.4	20.9	1.4	0.97	0.17	43.0	2.0	0.99	0.09
11	56.0	3.8	59.0	4.0	2.00	0.40	117.0	5.5	1.05	0.10
12	3.9	0.4	5.6	0.5	0.26	0.10	9.8	0.6	1.44	0.19
15	7.2	0.8	8.5	0.9	< 0.4		15.7	1.2	1.18	0.18
16	5.6	0.4	7.5	0.5	0.33	0.07	13.4	0.6	1.34	0.12
17	20.3	1.3	20.6	1.3	0.98	0.17	41.9	1.9	1.02	0.09
18	52.2	3.2	56.0	3.4	2.50	0.34	110.7	4.7	1.07	0.09
21	3.3	0.3	3.1	0.2	< 0.2		6.5	0.4	0.94	0.10



Fig. 1: R. Rusconi

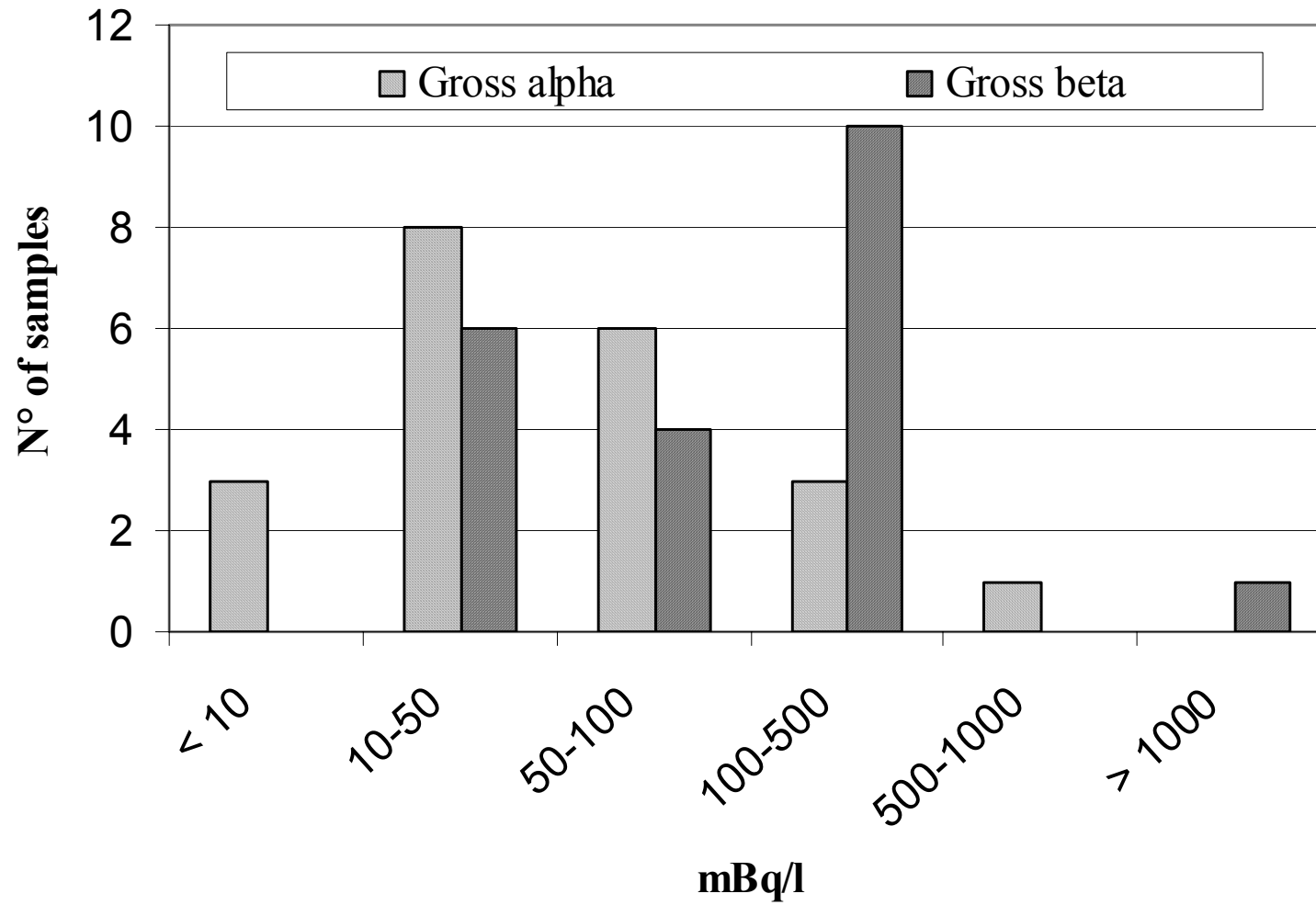


Fig. 2 R. Rusconi

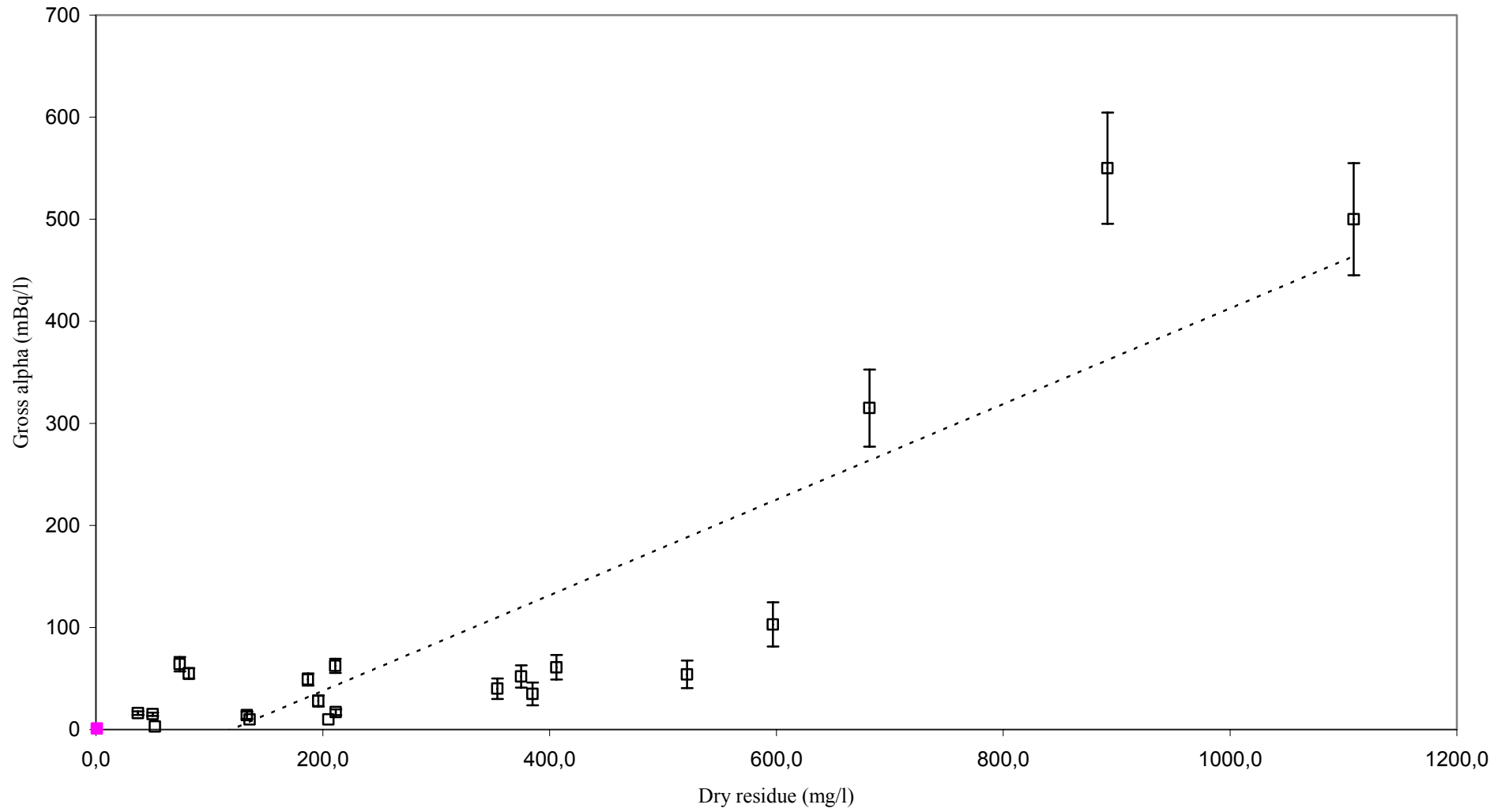


Fig. 3 R. Rusconi

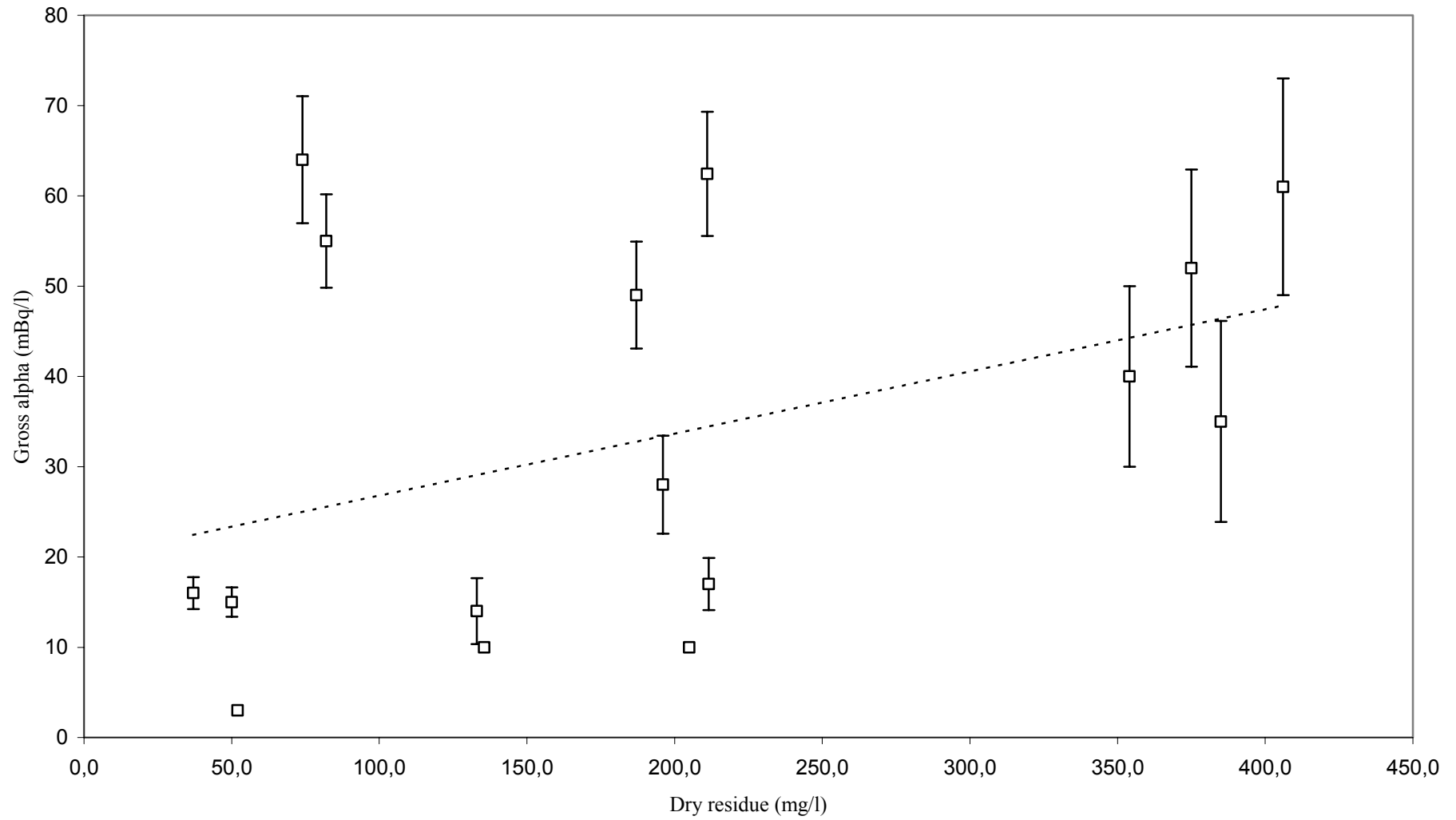


Fig. 4: R. Rusconi

