Contamination levels observed on the Belgian territory subsequent to the Chernobyl accident.

COTTENS E. Radiation Protection Service - Public Health Department - BRUSSELS.

The author wishes to emphasize that the summary of data presented here, concerning the influence of the Chernobyl emissions on the Belgian territory results from the joint effort done by the IHE at Brussels, the SCK/CEN at Mol and the IRE at Fleurus. These institutions provided the authorities with the necessary daily updated figures for judging the radiological situation on the territory. Faced with this urgent need for monitoring data in different pathways, the staff and technicians of those institutions realized a remarkable performance during the month of May and they do have the greatest merits in achieving this review.

1. Introduction

Contaminated air masses reached the Belgian territory from the South during the night of the first to the second of May. At this stage however the origin of this contamination was already identified through earlier observations over the Scandinavian area and the subsequent message about the reactor accident at the Chernobyl site. Later on radioactive clouds were also detected over het central part of Europe, demonstrating the persistent nature of the emissions from the damaged reactor. Consequently the influence on the Belgian territory was not unexpected. The authorities called on the SCK/CEN at Mol, and the IRE at Fleurus to assist the IHE at Brussels in collecting the necessary data for judging the radiological situation in our country. The KMI/IRM at Brussels was involved for the follow-up of meteorological conditions and analysis of the trajectories of contaminated air masses.

Early detection possibilities for the arrival of contaminated air were provided by the continuous environmental monitoring apparatus for ambient γ- dose rate or for β activity of airborne dust, available ot nuclear institutions and nuclear power plants.

On detection of enhanced air radioactivity, the sampling period of routine air dust samplers was significantly shortened to allow for the hour to hour renewal of data for gross B activity as a general indication of the evolution of the air contamination. γ- spectrometric analysis of those filters provided the necessary data for the estimation of the dose equivalent due to inhalation.

Ground deposition data at the location of the participating institutions were obtained by daily analysis of the radioactivity contents of a water container collecting both dust and rainwater.
Field gamma spectrometry was used later on at a number of other locations, to estimate the integrated ground deposition of radioactivity and its distribution over the country.

As the grazing season was just started or was about to be started in the following days for regions of higher altitude (Ardennes), the food chain grass-milk was intensively surveyed in first instance in respect of iodine-131 contamination.
Extensive grass sample measurements were carried out during the first week of May, until further significant deposition ceased. Milk sampling covered the most important dairies of the country, while some individual farms and even individual cows were followed continuously over the whole period of significant milk contamination.

High priority was also given to the follow up of contamination on leafy vegetables by γ-spectrometry.
The radioactivity in surface waters, especially those used for drinking water preparation, and in finished drinking water was monitored at high frequency for about two weeks mostly by gross B measurements.

γ-spectrometry on meat samples, essentially to determine the long lived isotopes of Cesium, started after the critical period for iodine contamination during the second half of May. The build up of cesium in meat is indeed a much slower process.

Besides the fission products analysed by γ-spectrometry, some measurements were done for Strontium - 89 and 90 in various samples. As the gross alpha-activity on some dust samples revealed also the presence of supplementary α - activity, some of them were further analysed by α- spectrometry.

2. Results

2.1. Air

The evolution of gross B-air activity at ground level calculated from activity measurements on air dust filters is shown on fig. 1. During the night from the first to the second of may, the air activity starts rising sharply.

The mean concentration over a one hour sampling period reaches a maximum value of about 60 Bq/m³ against noon on the second of May shifted by some hours between the different sampling places. Concentrations remained at a level of about 10-20 Bq/m³ on the third of May and decreased towards values below 1 Bq/m³ in the evening.

From the seventh of May concentrations are about 10 m Bq/m³ or below, to be compared with normal values before the accident of about 1 m Bq/m³ for the same time delay between sampling and measurement. Observed concentrations over the country were very similar, except for a monitoring station near the coast where the mean concentration on the second of May amounted only to about half of the values measured elsewhere.

γ-spectrometry measurements on exposed dust filters revealed ^{132}Te as the dominant radionuclide in particulate form.

Table 1 gives the isotopic composition of particulate air activity at its maximum value at Brussels, Mol and Fleurus. Similar fission product concentrations were also observed at Ghent (1). Fig. 2 shows the particulate ^{131}I concentration during the first week of May. The concentration of the other isotopes follows a quite similar pattern.

Non-particulate ^{131}I was sampled in a charcoal cartridge in series with a glass fiber filter on different occasions during the second and third of May. A ratio of 2 was found between the non-particulate ^{131}I fraction and the particulate one. Maximum total ^{131}I concentration on the second of May can consequently be estimated as about 25 to 30 Bq/m³.

Alpha spectrometry of filters exposed at maximum air activity revealed concentrations of ^{239}Pu + ^{240}Pu in the range of 25-46 μ Bq/m³ and of 80 μ Bq/m³ for ^{242}Cm.

For ^{90}Sr a maximum air concentration of 35 mBq/m³ was found.

2.2. Ground deposition

Integrated fall out captured in a water container at Mol and Fleurus is given in Table 2 for the most important radionuclides. Large differences in rainfall quantities on the third and fourth of May (from nearly zero for the western part of the country up to a local maximum of 36 mm in the province of Luxembourg) caused considerable differences in deposited activity over the country. At Mol, about 60 % of the deposited activity in Table 2 was due to the rainfall during the night of the 3rd to the 4th of May. The concentration of ^{131}I in rain water amounted to 1.4 kBq/l. ^{132}Te was significantly less abundant with respect to the composition of air activity. Cesium isotopes and ^{103}Ru became relatively more important in the rainfall from the 6th of May.

In situ γ spectrometry on various locations over the country allowed to measure deposited activity on soil surface directly. From this measurements the distribution of deposited activity over the country could be mapped as on fig. 3. The distribution of deposited activity correlates well with the rainfall distribution from the third to the sixth of May. Mean deposition is estimated at 4 kBq/m² and 1,5 kBq/m² for ^{131}I and ^{134}Cs + ^{137}Cs respectively. Global deposition of those radionuclides on the Belgium territory amounts to 120 TBq and 45 TBq respectively.

2.3. Ambient γ-dose rate

As shown on fig. 4, the ambient dose rate level measured at I m above the ground is increasing at passage of the contaminated cloud. However due to deposited activity, the dose rate doesn't fall off again to its normal value. On the following days, sharp increases of the dose rate level correspond to the deposition of fission products with rainfall.
With the decay of short lived isotopes such as the ^{132}Te-^{132}I couple, ^{131}I, the ^{140}Ba - ^{140}La couple and ^{103}Ru the dose rate decreases again until finally the long lived isotopes ^{134}Cs and ^{137}Cs determine the residual supplementary γ- dose rate level.

Fig. 5 shows the variation of ambient γ-dose rate at Mol, Fleurus, Tihange and near Chooz. Maximum increase of ambient γ- dose rate levels was observed on the fifth of May and ranges from about 40 nSv/h up to about 120 nSv/h in high deposition areas.

2.4. Food chain

2.4.1. Milk

This pathway was intensively monitored by measurements on grass as well as on milk samples.
Intensive sampling of grass all over the country was done by regional officials from the Ministry of Agriculture until the tenth of May. After that date the evolution of grass contamination was further followed on a limited number of sampling places near the measuring laboratories.
Fig. 6 shows the evolution of the minimum, maximum and median values of the daily observed distributions of ^{131}I on grass. In the Western part of the country with the lowest integrated ground deposition (see fig. 3) maximum ^{131}I contamination on grass was in the range 500 - 1.000 Bq/m². For the average contaminated areas values are in the range of 500 - 2.000 Bq/m², while for the highest deposition area values in the range 1.000-4.000 Bq/m² were observed.

Some characteristics of the observed distributions for grass contamination with Cesium isotopes in each province are given in table 3.

Milk monitoring was carried out on three distinct levels :

- individual cows
- individual farms
- blended milk samples at delivery at the dairies.

Fig. 7 shows the day by day evolution of milk contamination by ^{131}I for two individual cows, one of them being permanently on the meadow, the other one only by daytime.

The typical evolution of ^{131}I concentration in milk from an individual farm can be observed on Fig. 8. Maximum ^{131}I concentrations for this type of monitoring were in the range 100 - 660 Bq/l.

Fig. 9 and 10 show the evolution of mixed milk contamination at delivery at the dairy for respectively a dairy collecting in the province of Luxemburg and West/East Flanders. Nearly the whole country was covered by this sampling as all important dairies were included. The maximum value for ^{131}I concentration found for this type of controls amounts to 225 Bq/l at the fifth of May. A striking feature is that this maximum value is found for a dairy near the border between West and East Flanders situated within the low ground deposition area. The spread of ^{131}I contamination in milk proved to be less than the spread in grass contamination and there is also little correlation with the deposition zones of Fig. 3.

The fact that the beginning of the grazing season in the southern part of the country is generally shifted backwards with respect to the other areas, possibly influenced milk contamination during the first week of May in those areas.

Better correlation is observed with Cesium concentration in milk. Table 4 demonstrates that ^{137}Cs concentration in the province of Luxemburg and the individuel cows at Mol remained at a level of about 20 Bq/l (about 50-55 % or 10 Bq/l for ^{134}Cs) near the end of May, which is about a factor two higher compared to the other regions of the country.

2.4.2. Vegetables

Leafy vegetables exposed to the air activity and/or contaminated rain water form a direct pathway for human ingestion. The most important leafy vegetables on the market at the beginning of May were : spinach, lettuce, cornsalad, leek, watercress, selery and endive.

Due to its great capture surface and open structure spinach proved to be the most sensible species. Fig. 11 and 12 give maximum, minimum and mean observed concentrations of ^{131}I and ^{137}Cs (^{134}Cs is again about 50 % of ^{137}Cs) on spinach.

On cornsalad maximum 131 I concentration was about 400 Bq/kg (Fig. 13). For lettuce this was 300 Bq/kg (Fig. 14). For the other mentioned vegetables maximum concentrations were in the range 100-200 Bq/kg.

While Cesium contamination on spinach decreased rather rapidly from a maximum value of 200 Bq/kg towards values below 30 Bq/kg in the second half of May, this was not so with most of the other vegetables. However the latter showed only concentration levels of some tens of Bq/kg.

2.4.3. Water

Surface waters were monitored by gross B laboratory measurements and some automatic measuring stations permanently installed along the Meuse and Sambre measuring ^{131}I concentration.

With those permanent stations ^{131}I concentrations up to 25 Bq/l were measured. Gross B activities from 1 Bq/l up to about 60 Bq/l were observed on the fifth of May depending largely on the dilution capacity of the watercourse. These values decreased rapidly below a few Bq/kg by the fifteenth of May.

Intensive monitoring of surface waters and water basins used for the production of drinking water was performed until the middle of May. The maximum gross B activity observed was about 4 Bq/l at the ninth of May. By the fifteenth of May most of the results were less than 1 Bq/l.

Finished drinking water was also intensively sampled. Fig. 15 shows the evolution of drinking water activity during May. Maximum activity found was 2 Bq/l at the eight of May. Drinking water treatment proved to reduce gross B activity by about a factor 2. The removal efficiendy of individual γ-emitting nuclides such as ^{131}I, ^{103}Pu, ^{134}Cs and ^{137}Cs was investigated by a drinking water society and ranges from 30 to 70 % (2).

2.4.4. Meat

Meat sampling was started only during the second half of May, as the nuclides of interest here are the long lived Cesium isotopes.

Ranges and median values of the results are presented in table 5 for the period May - August 1986.
For cattle 80 % of samples showed concentrations below the detection limits of a few Bq/kg. Horse samples proved to be more generally contaminated but at relatively low concentrations. As expected sheep and lamb in particular show the highest concentrations.
As observed concentrations have a large spread, it is impossible to recognize a clear trend in the evolution of contamination over the period june - august.

References

(1) J. Uyttenhove - Internal report of the Nuclear Physics Laboratory. State University of Ghent.

(2) J.W. Maschelein et al.
Report of the Laboratories of the Brussels Waterboard.

IHE : Daily communications of the IHE.

SCK/CEN : Accident of Chernobyl - Report of the measurements (86-675) - Working document.

IRE : Accident of Chernobyl - Report of the measurements - Working document.

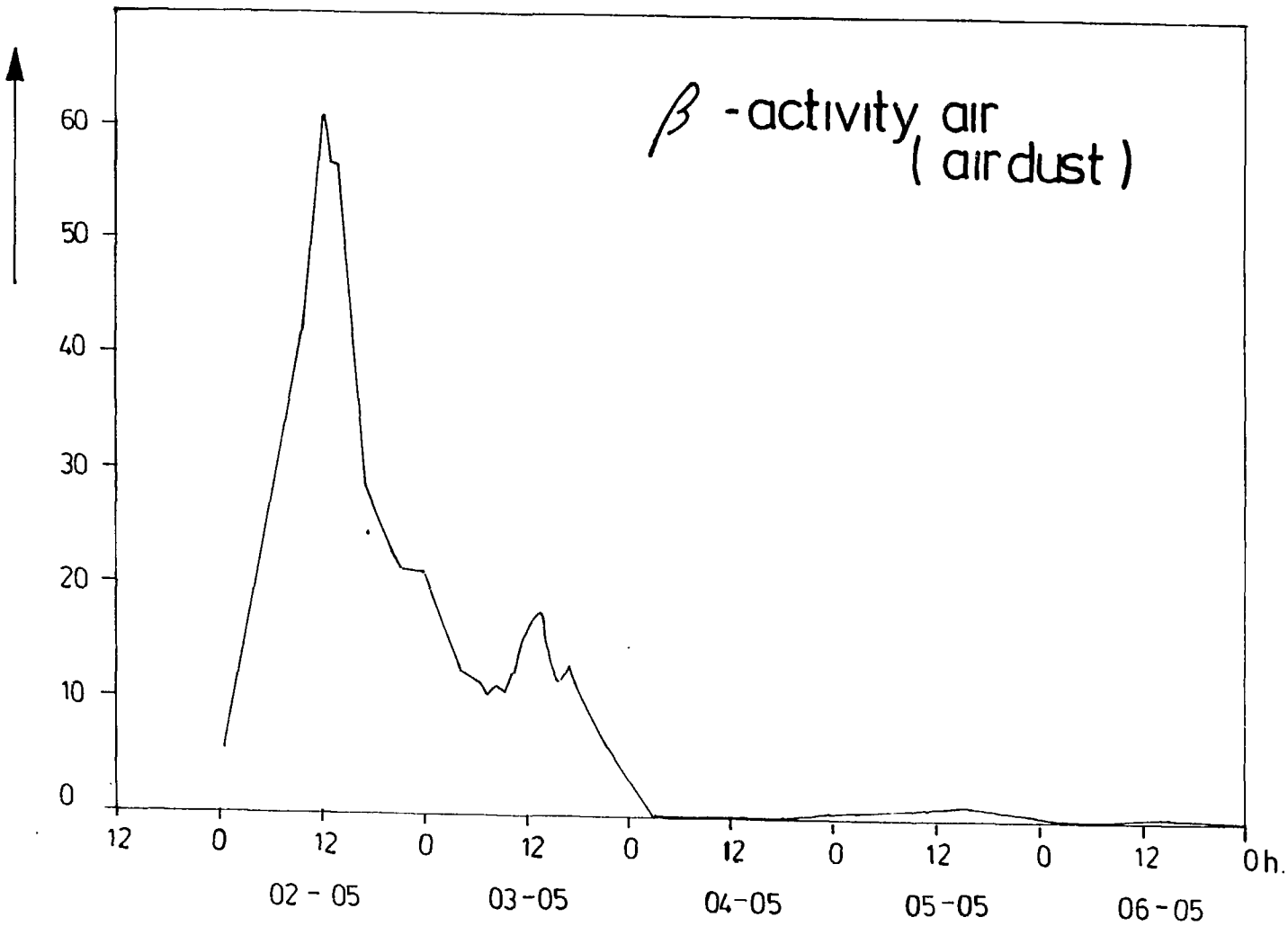

β -activity air
(air dust)
60
50
40
30
20
10
0
12
0
12
0
12
0
12
0
12
0
12
0 h.
02 - 05
03-05
04-05
05-05
06-05

FIG. 1

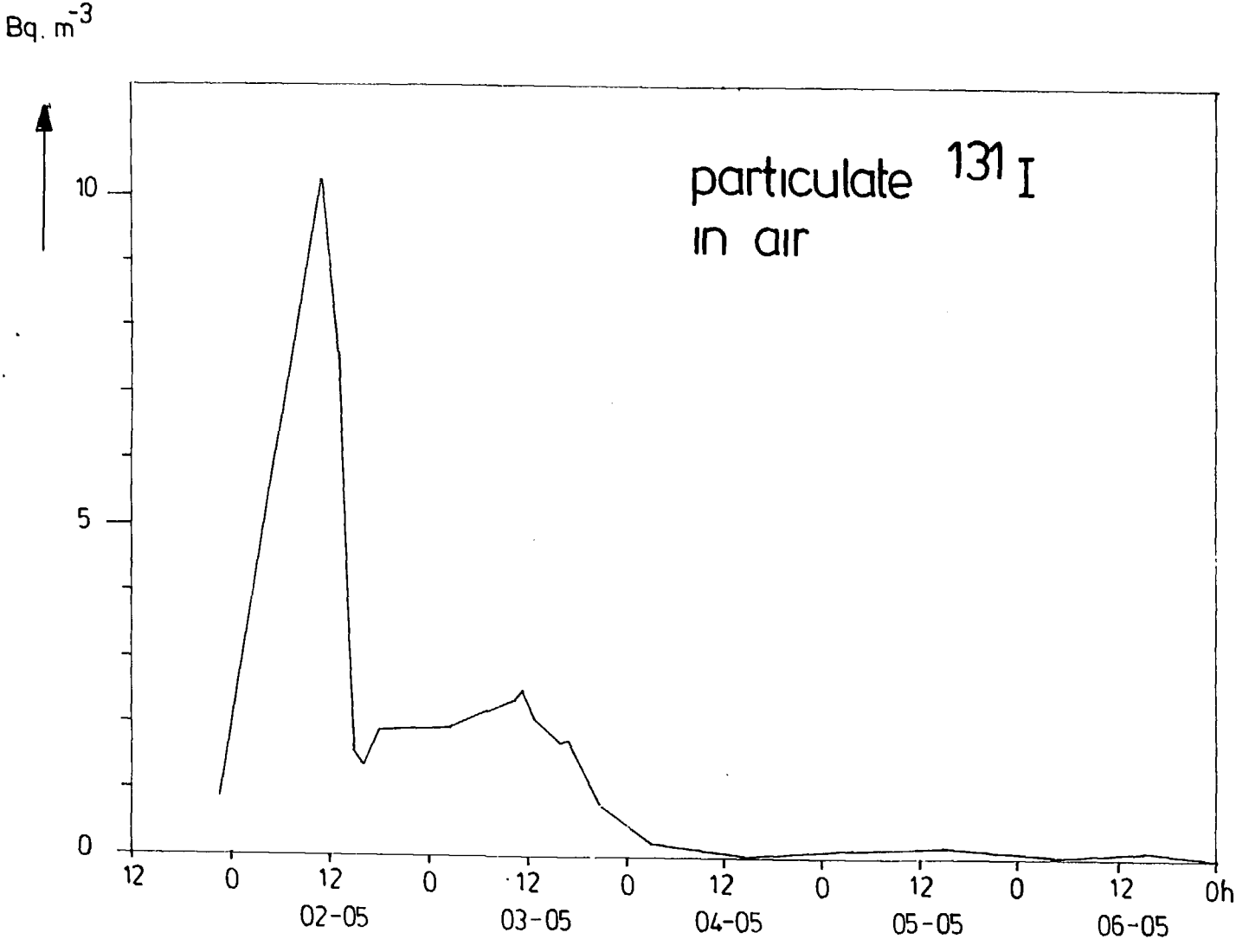

Bq. m-3
10
5
0
particulate 131 I
in air
12
0
12
0
12
0
12
0
12
0
12
0h
02-05
03-05
04-05
05-05
06-05

FIG. 2

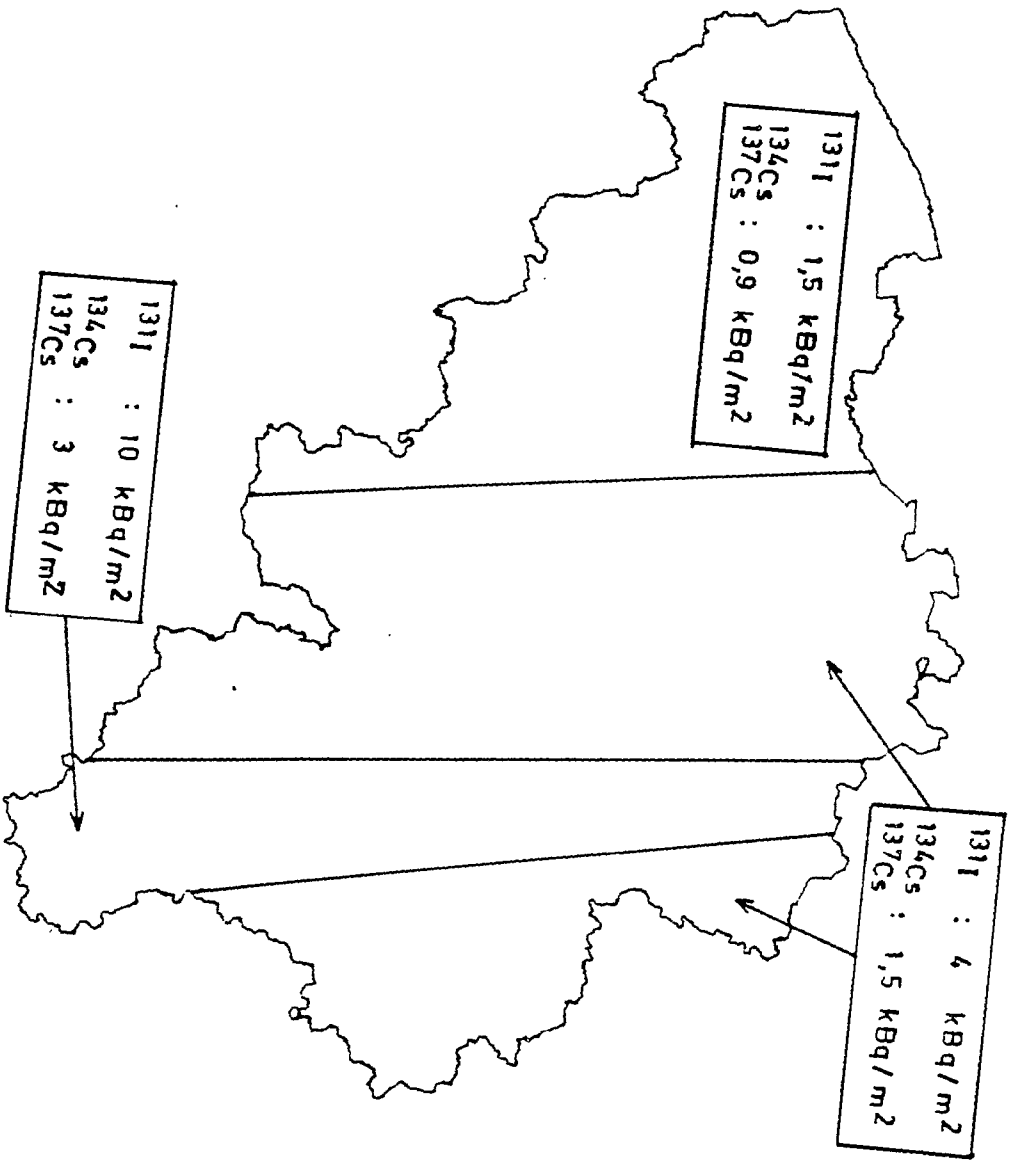


FIG. 3

INTEGRATED DEPOSITION MAP

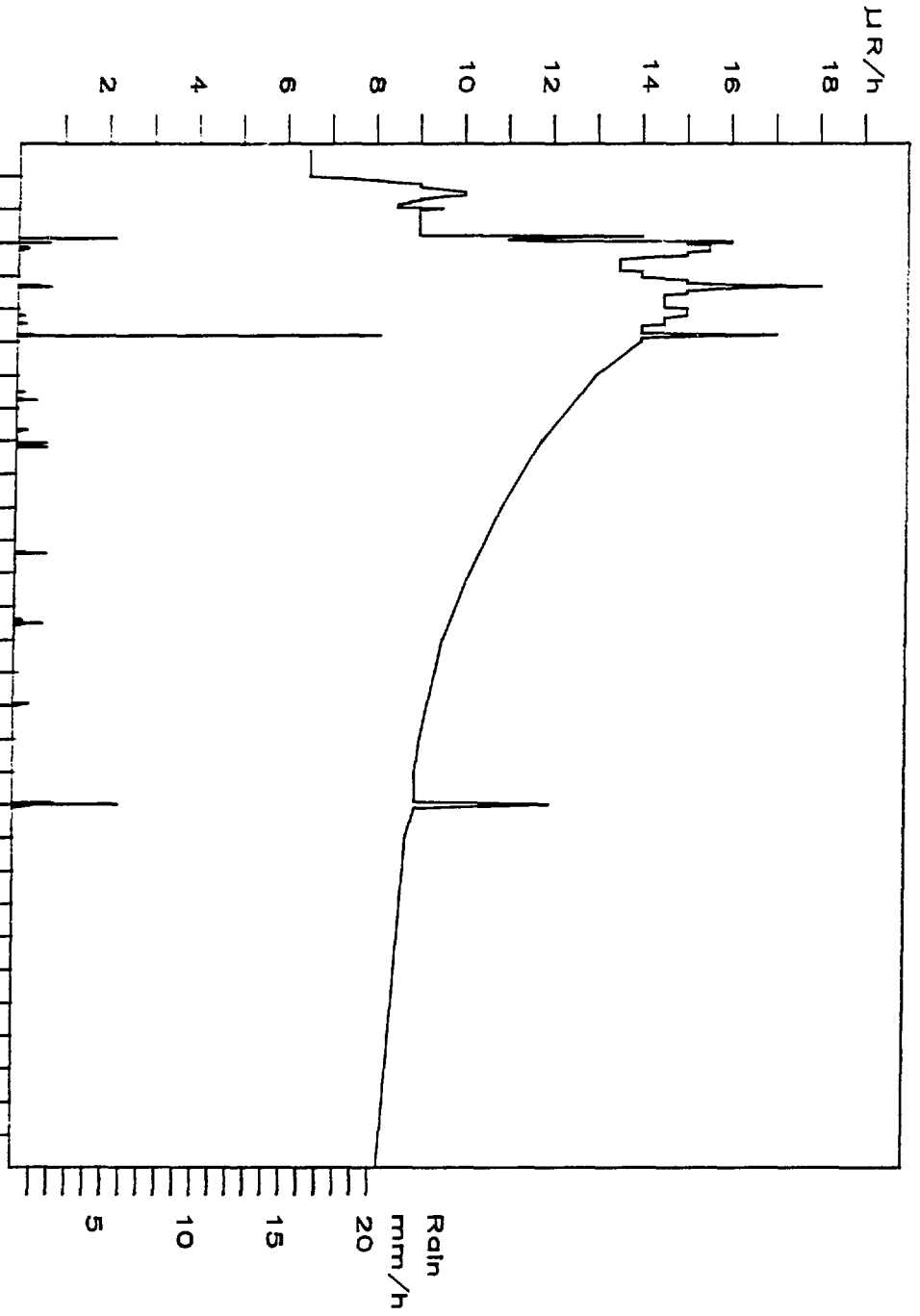
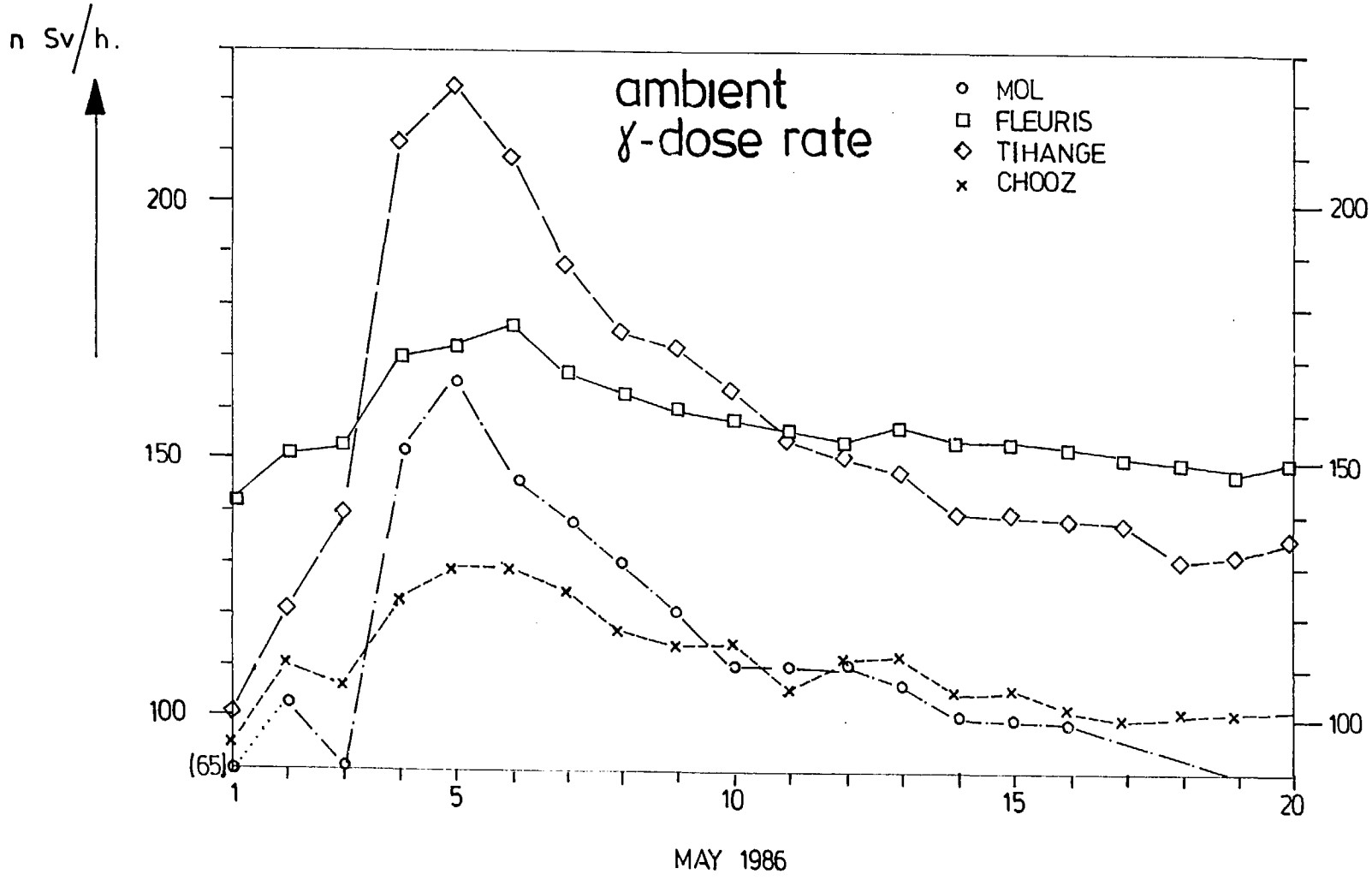


FIG. 4


n Sv/h.
ambient
γ-dose rate
MOL
FLEURIS
TIHANGE
CHOOZ
200
150
100
(65)
1
5
10
15
20
MAY 1986

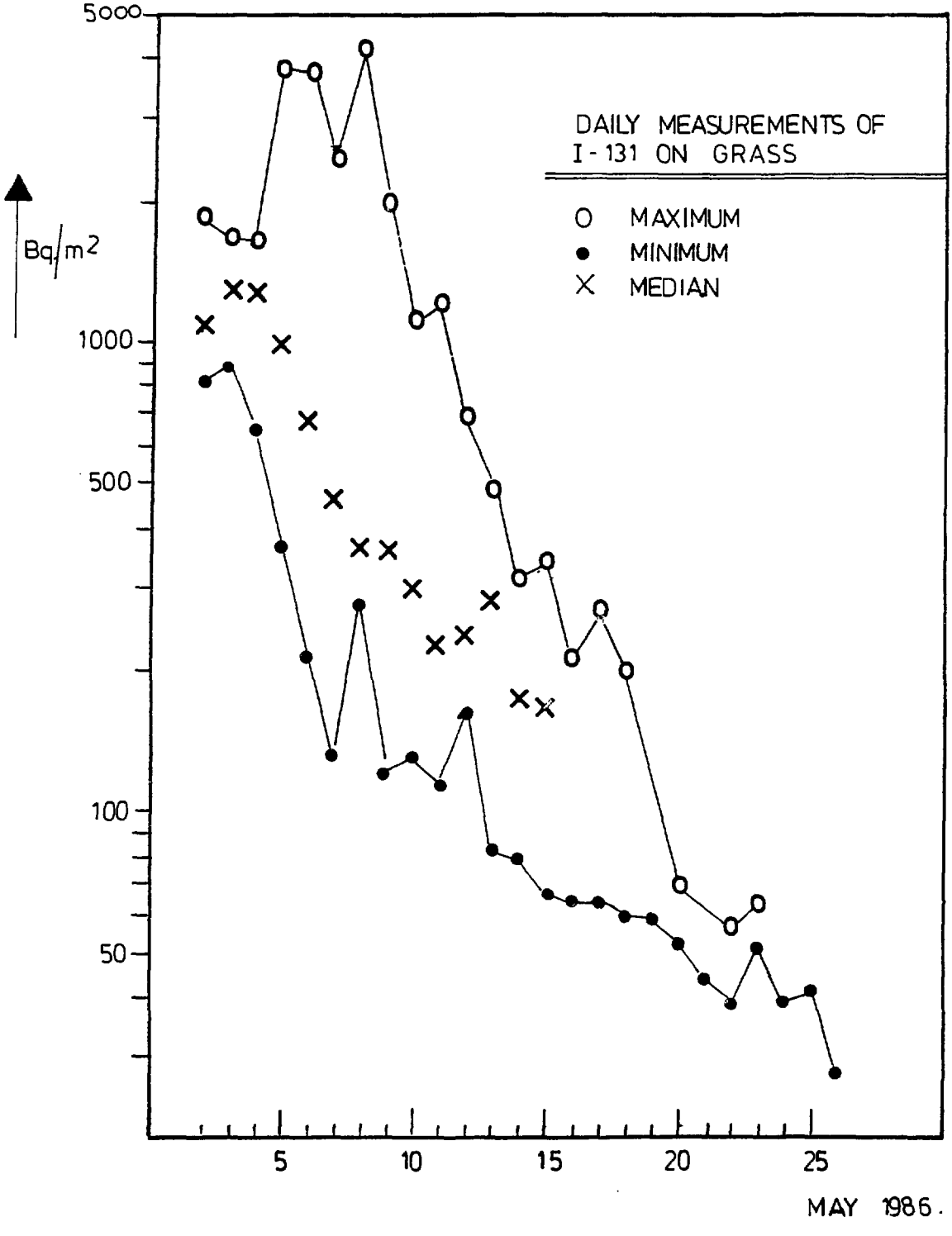

DAILY MEASUREMENTS OF
I - 131 ON GRASS
O MAXIMUM
● MINIMUM
X MEDIAN
Bq/m2
5000
1000
500
100
50
5
10
15
20
25
MAY 1986.

FIG. 6

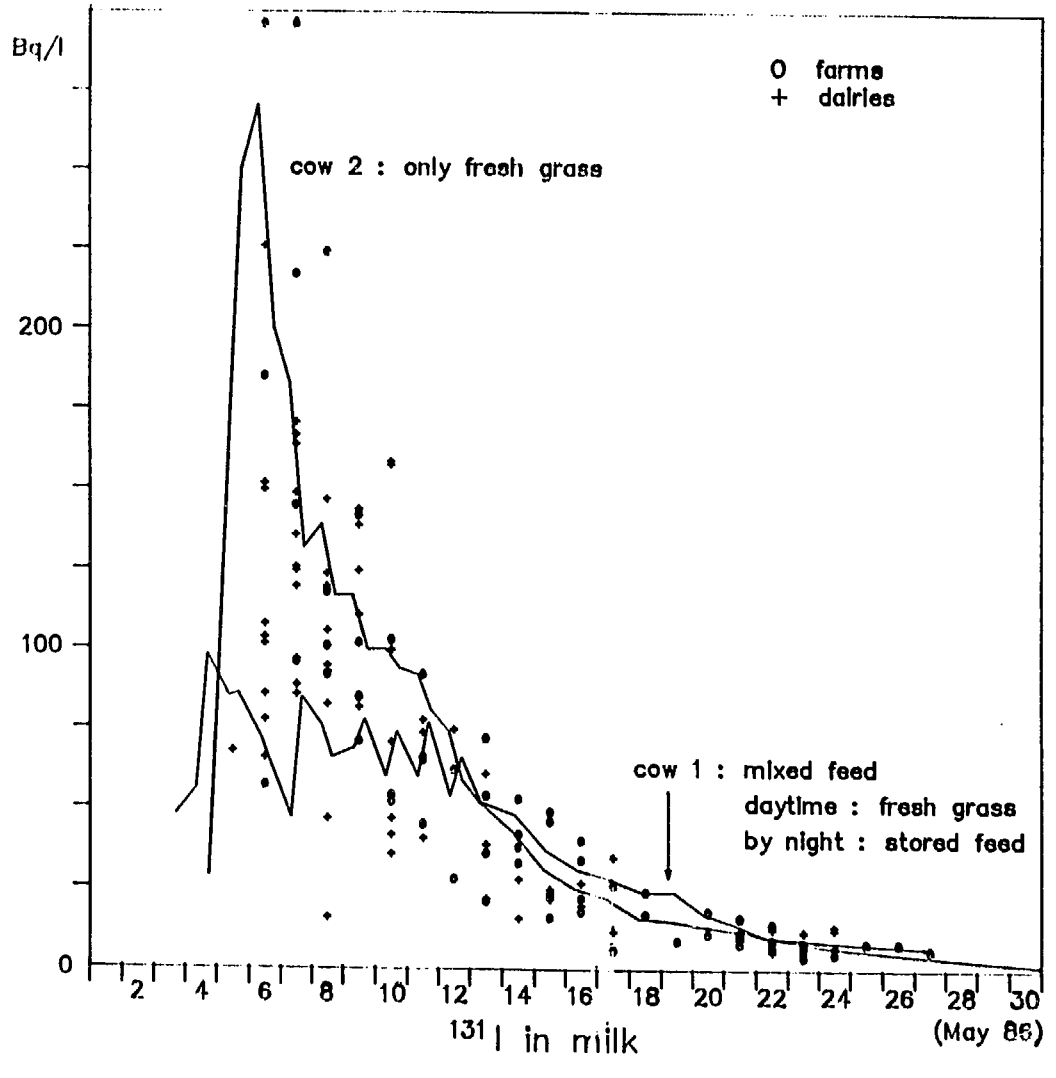

Bq/l
200
100
0
o farms
+ dairies
cow 2 : only fresh grass
cow 1 : mixed feed
daytime : fresh grass
by night : stored feed
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
(May 86)
131I in milk

FIG. 7

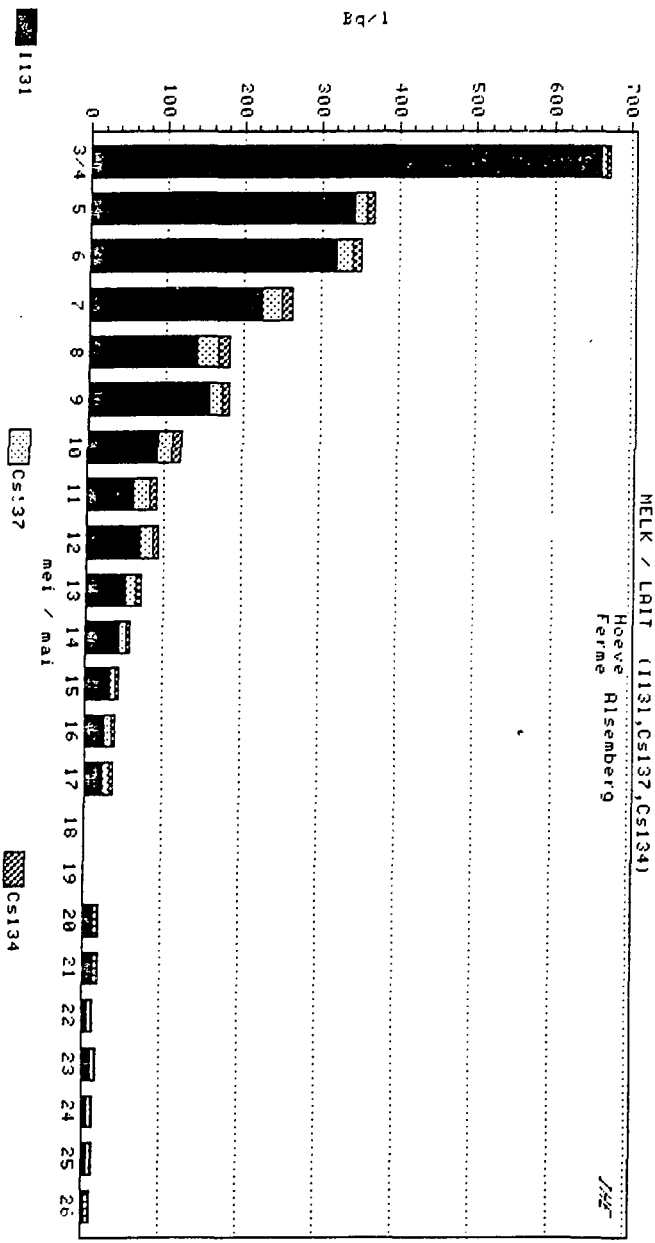

Farm Milk
MELK / LAIT (I131,Cs137,Cs134)
Hoeve
Ferme Alsemberg
Bq/l
700
600
500
400
300
200
100
0
3-4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
mei / mai
I131
Cs137
Cs134

FIG. 8

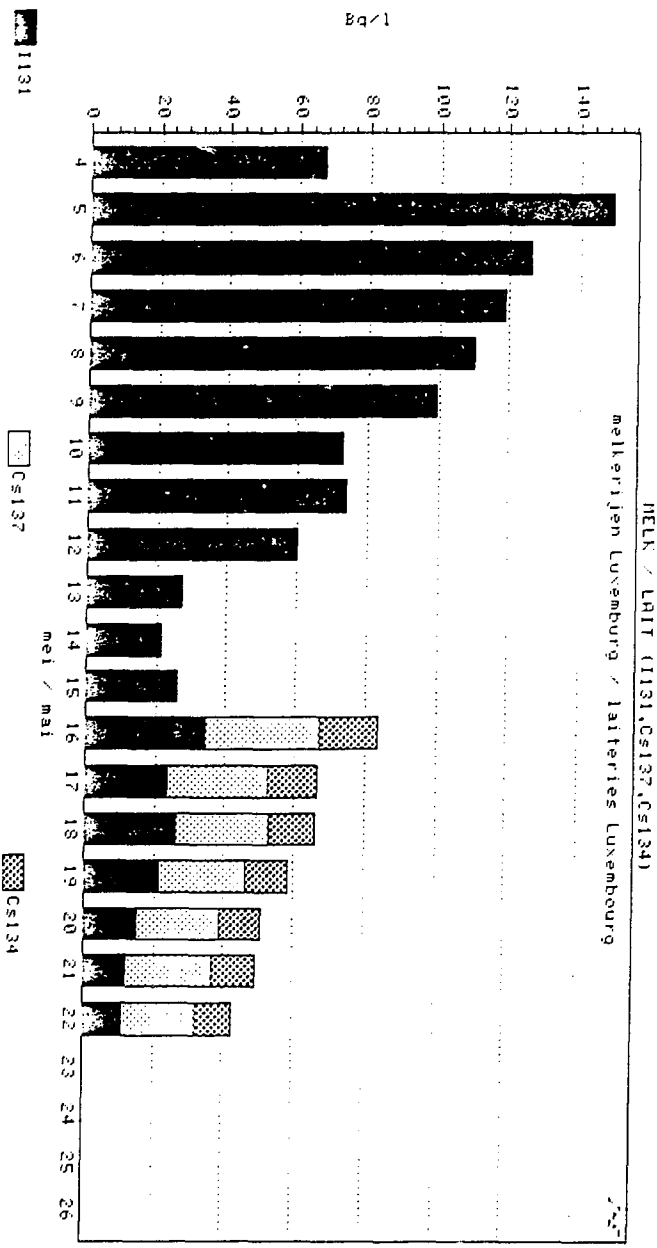

Dairy Milk
MELK / LAIT (I131,Cs137,Cs134)
melkerijen Luxembourg / laiteries Luxembourg
Bq/l
0
20
40
60
80
100
120
140
I131
Cs137
Cs134
mei / mai
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26

FIG. 9

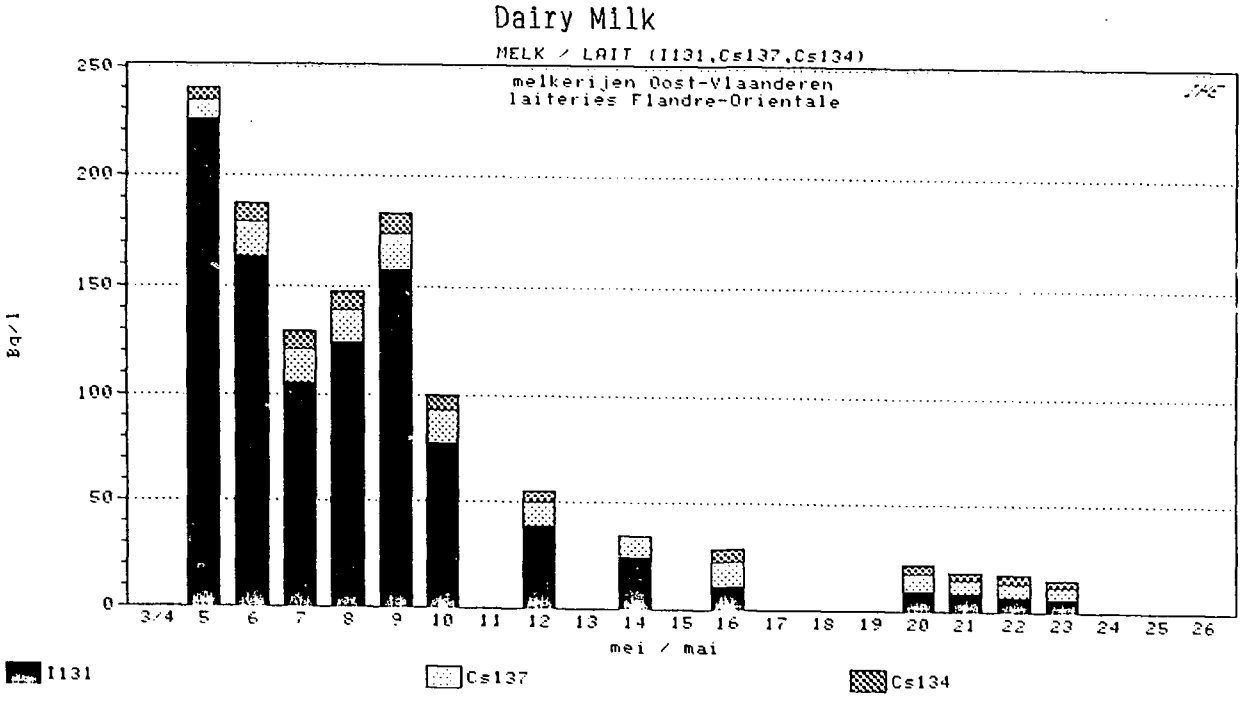

Dairy Milk
MELK / LAIT (I131,Cs137,Cs134)
melkerijen Oost-Vlaanderen
laiteries Flandre-Orientale
Bq/l
250
200
150
100
50
0
3/4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26
mei / mai
I131
Cs137
Cs134

FIG. 10

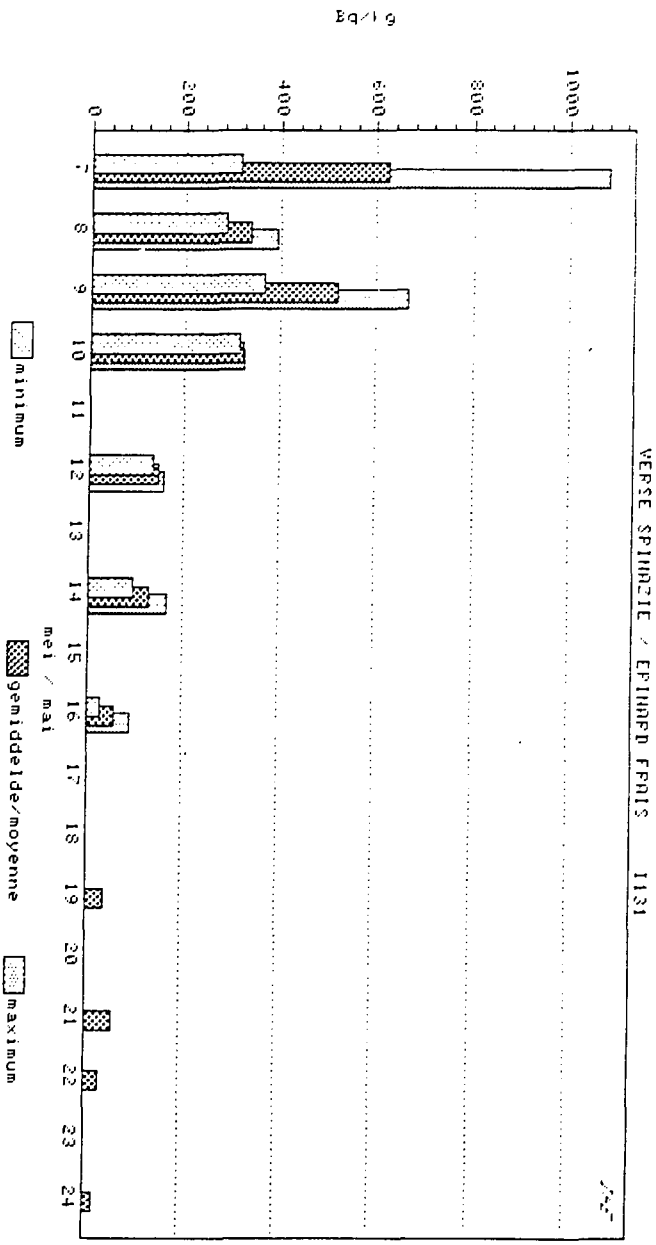

Spinach
VERSE SPINAZIE / EPINARD FRAIS 1131
Bq/kg
0
200
400
600
800
1000
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
mei / mai
minimum
gemiddelde/moyenne
maximum

FIG. 11

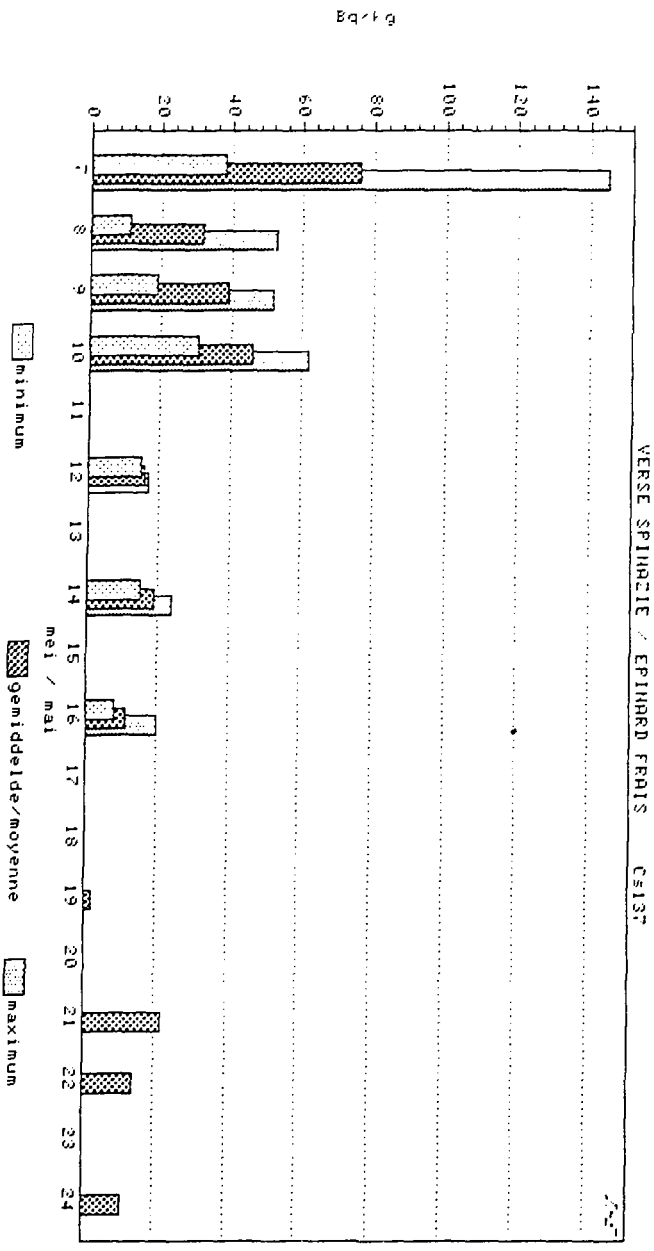

Spinach
Bq/kg
0
20
40
60
80
100
120
140
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
mei / mai
minimum
gemiddelde/moyenne
maximum

FIG. 12

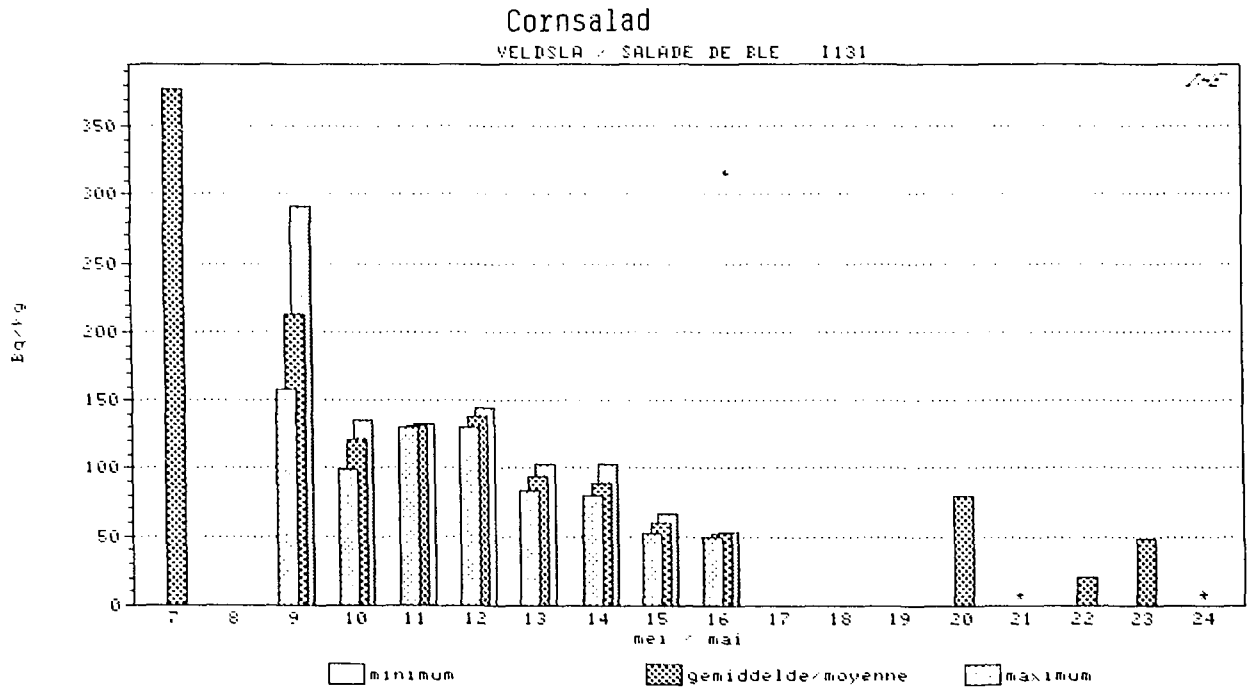

Cornsalad
VELDSLA / SALADE DE BLE 1131
Eq/ha
350
300
250
200
150
100
50
0
7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24
mei / mai
minimum
gemiddelde/moyenne
maximum

FIG. 13

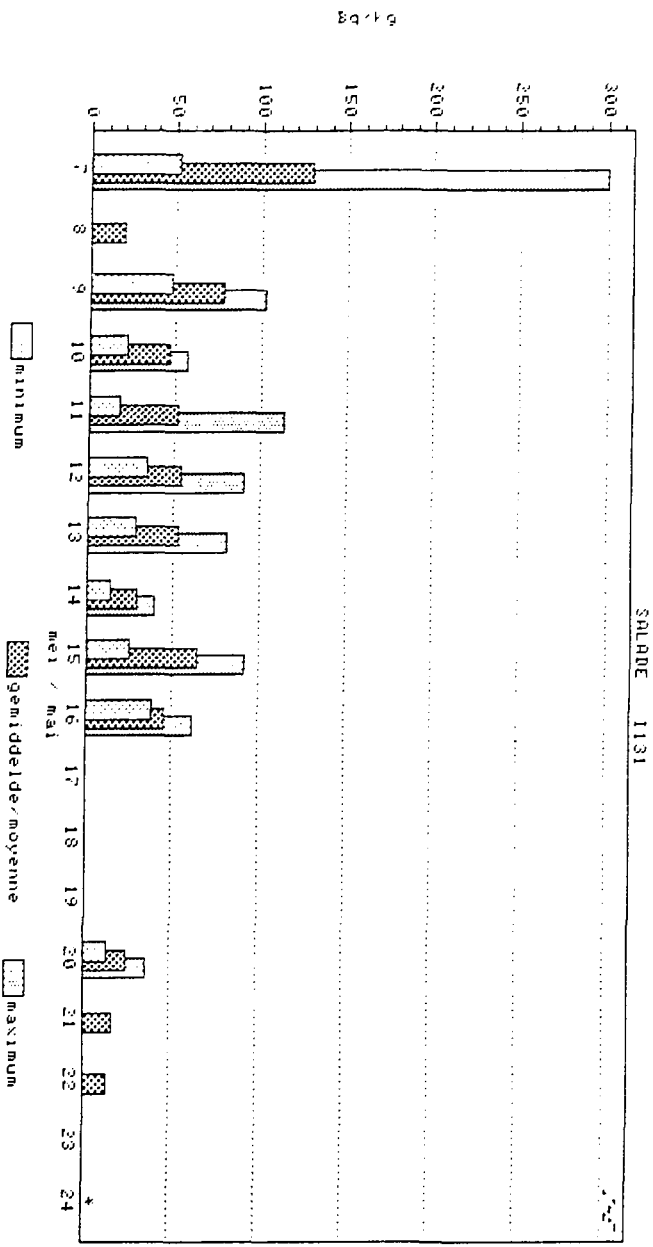

Lettuce
SALADE 1131
minimum
gemiddelde/moyenne
maximum
mei / mai
0
50
100
150
200
250
300
1
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
*

FIG. 14

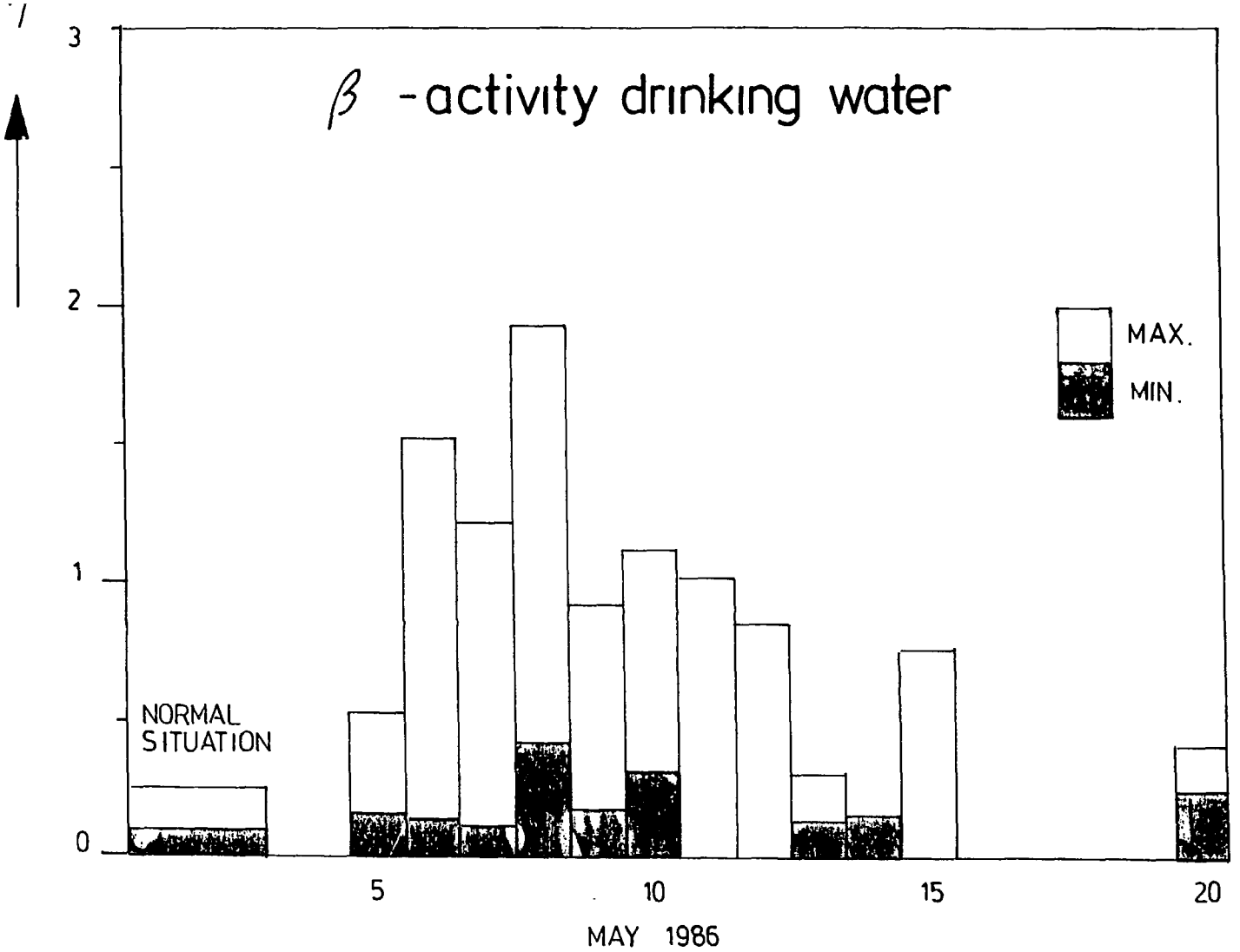

β -activity drinking water
3
2
1
0
MAX.
MIN.
NORMAL
SITUATION
5
10
15
20
MAY 1986

FIG. 15

Table 1

Isotopic composition of airborne activity
as measured on paper filter
(May, 2nd - Bq/m³)

	MOL (9h-15h)*	BRUSSELS (10h30-11h30)*	FLEURUS (8h30-9h30)*
I-131 (particulate)	7.8	10.2	6.8
Cs-137	3.6	6.0	3.3
Cs-134	1.9	2.4	2.0
Cs-136	0.6		0.6
Mo-99	1.5		
Te-132	17		9.6
Ru-103	3.7		3.7
Ru-106	1.9		
Ba-140	1.5		
^{90}Sr	$3.5\ 10^{-2}$		
$^{239}Pu + {}^{240}Pu$	$2.5\ 10^{-5}$		$4.6\ 10^{-5}$
^{242}Cm	$8.0\ 10^{-5}$		

* sampling time at maximum observed air concentrations.

Table 2

Integrated fall out captured in a water container (k Bq/m²)

	Mol	Fleurus
I-131	13	6
Te-132	4.9	3
Cs-137	2.1	0.5
Cs-134	1.0	0.4
Ru-103	3.5	0.9

Table 3

Cesium isotopes on grass * (Bq/m²)

(May 4th - 10th)

Province	Cs-137		Cs-134	
	max - min	Median	max - min	Median
Antwerpen	85 - 191	130	53 - 105	70
Limburg	55 - 145	110		
West-Vlaanderen	32 - 94	60	20 - 55	30
Oost-Vlaanderen	25 - 320	110	12 - 172	65
Brabant	48 - 196	100	22 - 123	60
Hainaut	17 - 245	30	10 - 80	15
Namur	35 - 306	130	23 - 219	60
Liège	33 - 239	110	33 - 205	65
Luxembourg	92 - 1.422	360	55 - 859	170

* Daily measurements of samples from the same meadows near Mol, Brussels and Fleurus are not included in the table.

Table 4

^{137}Cs concentration in milk (Bq/l)

DATE (may)	05/08	09/13	14/18	19/23
Dairies				
- West/Oost-Vlaanderen	14	15	11	7
- Antwerpen	13	13	11	11
- Luxembourg			30	21
Farms				
- Fleurus	16	17	10	7
- Alsemberg	22	17	9	5
- Bellem	24	14	15	5
Individual cows				
Mol 1			23	23
2			28	20

Table 5

Cesium isotopes in Belgian meat production (Bq/kg)
(May - August 1986)

		Cs - 134		Cs-137	
		maximum	median	maximum	median
Cattle (*)	kidney	22	D.L.	55	D.L.
	liver	15	D.L.	28	D.L.
	meat	21	D.L.	48	D.L.
Horse	kidney	41	11	82	24
	liver	27	8	56	10
	meat	23	7	42	14
Sheep/	kidney	75	10	197	19
Lamb	liver	37	D.L.	80	13
	meat	164	12	392	25

(*) 80 % of analysed samples show concentrations below the detection limit (D.L.) of a few Bq/kg.