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On the Contents of Helium and Other Constituents in the Natural Gases of Japan.

[Continued from the Report of this Institute No. 6 (1923).]

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and

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(with 3 plates).

Abstract.

An exhaustive examination is undertaken on the contents of helium in the natural gases of Japan, succeeding Mr. N. Yamada in his line of research.

Eighty four samples of natural gases in Taiwan, Hokkaidô, Honsyû, and Kyûsyû of Japan collected from various sources i. e. from petroleum oil reservoirs, coal mines, mineral springs and volcanoes are examined on their contents of helium and other constituents.

Some relations between the total composition and the helium content of a gas are discussed. Considerations are made on the geological features of gas reservoirs and also on the radioactivity of mineral gases.

The investigation show that the petroleum gases in Taiwan and Hokkaidô have little prospects as the source of helium. Considerable percentages of helium are found in some mineral gases, but unfortunately their evolution is at present not sufficient for any industrial purposes,

CORRECTIONS.

Page	Line	Misprint	Correction
348	26	the result throw	the result might throw
351	21–22	with result	with the result
353	28	considerably	considerable
356	7–8	between the the	between the

1. Introduction.

The inevitable danger which accompanies the use of hydrogen for inflating the airships and the balloons enlivens our serious interest for helium gas. And the helium-problem is of prominent importance in aerostation.

Two years ago Mr. N. Yamada[1] of this laboratory published his investigations on the contents of helium in all the principal petroleum gases and certain mineral gases in Honsyû and Kyûsyû of Japan. He finds that petroleum gases in those parts of Japan contain little or no helium, which, however, is contained in considerable quantities in some mineral-spring gases; and his conclusion is that it would subsequently be necessary to examine natural gases of Taiwan (Formosa), Hokkaidô and Karahuto (Saghalien), not only because that the quantities of their outflow are very abundant but also that the geological features of their reservoirs are different from those found in Honsyû and Kyusyû. He suggests that as volcanic gases of Japan are great in quantities, coming out of numerous sources, the investigation of them seems to be quite promising, though the sampling will be very difficult and need special attention and technique.

The authors of this papers succeeded Mr. Yamada in his line of research and undertook an exhaustive examination of helium contents not only in petroleum gases but also in gases of all other kinds, i.e. from mineral spring, coal mines and volcanoes in all the provinces of Japan. Further it was intended to extend the work to study their total composition, peculiarities of gas reservoirs and other factors, as the result throw some light on the problems regarding the fuel- and gas-industries of Japan.

Eighty four samples were collected. The analysis was carried out in the manner depicted in the next chapter. Of these twenty

(1) Report of this Institute No. 6. (1923):
Jour. Chem. Soc. Japan. **43**, 885 (1922): **44**, 1018 (1923)

TABLE I.

Sample No.	Locality: Prefecture	Locality: Prefectural District	Locality: Field	Source: Name	Source: Quantity of outflow m^3/day	Source: Geology	Source: Depth	Date Sampled	Composition of Gas (%): CO_2	SH_2	Heavy Hydro-carbons	O_2	CO	CH_4	C_2H_6	N_2 Inert Gases	He	Remark
1	Ôita	Hayami	Asahimura, Umidigoku	Natural outflow	520	Volcanic rock	—	Jan., 1924	90.68	trace	0	0.10	0	1.19	0	8.03	trace	Evolves from a fumarol.
2	do	do	Isigaki-mura, Hatimandigoku	do	320	Volcanic detritus	—	do	97.65	0.18	0	0.11	0	0	0	2.06	0	do
3	do	do	Okosi-mati, Sibaseki	do	1.4	Volcanic rock	—	do	26.10	0	0	0.56	0	0	0	73.34	0	With hot spring.
4	Hukuoka	Tukusi	Hutuka-iti-mati, Musasi	Hot spring well	0.4	Quarternary	60	do	1.11	0	0	0.21	0	0	0	98.68	0.087	do
5	do	Miike	Miike-mati, Kamoto	Gas well	0.6	Tertiary (coal bearing)	16	do	2.56	0	0	1.00	0	66.61	0	29.83	0	
6	Kumamoto	Hôtaku	Kozima-mati	Water well	1.4	Quarternery	64	do	11.01	2.09	0.26	0.31	0	83.47	0	2.86	0	With fresh water.
7	do	Kuma	Nakahara-mura, Hayasi	Hot spring well	1.5	do	124	do	2.51	0	0	0.12	0	0	0	97.37	0.023	With hot spring.
8	do	Ibusuki	Yamakawa-mura, Yamakawa	Natural outflow	0.7	Andesite	—	do	72.08	—	0	0.21	0	5.05	0	22.66	0	do
9	do	do	do , Unagiike-digoku	do	288	do	—	do	76.22	2.24	0	0.29	0	5.94	0	15.31	trace	Evolves from a solfatara.
10	do	Ibusuki	Ibusuki-mura, Surigahama	Natural outflow	3	Quarternary	—	do	15.64	trace	0	0.46	0	14.29	0	69.61	0.003	With hot sping.
11	do	Eira	Nisisoyama-mura, Myôken	Hot spring well	27	Tertiary	4	do	76.96	0	0	0.07	0	0.12	0	22.85	0	do
12	do	Hioki	Higasi-itiki-mura, Nagasato	Natural outflow	1.2	Volcanic ash	—	do	2.36	0	0	12.60	0	0	0	85.04	0.009	With fresh water.
13	Siduoka	Sida	Aosima-mura, Sida	Gas well	72	Tertiary	66	Dec., 1923	7.69	0	0.17	0.27	0	87.33	0	4.54	trace	With mineral spring.
14	do	do	Yaidu-mati	do	7	Quarternary	106	do	1.27	0	0	0.54	0	92.07	0	6.12	0.004	
15	do	do	Higasimasutu-mura	do	6	do	126	do	0.47	0	0.03	0.21	0	93.58	0	5.71	0.007	
16	do	Takata	Syûzenzi-mati	Hot spring well	0.05	Volcanic tuff	—	do	1.20	trace	0	0.22	0	0	0	98.58	0.037	With hot spring.
17	do	do	Nagaoka-mati	do	0.2	Tertiary	—	do	0	0	0.19	0	0	0.95	0	98.86	0.029	do
18	Kita-	Karahuto	Nutov	Natural outflow	480	Tertiary (coal bearing)	—	August, 1923	2.11	trace	0	0	0	96.21	0	1.68	0	With mineral spring & petroleum.
19	Yamanasi	Kitakoma	Masutomi, Yukubo	do	0.2	Granite	—	March, 1924	99.44	0	0	0	0	0	0	0.56	0	With mineral spring.
20	do	do	do , Tuganeyu-ura	do	0.2	do	—	do	99.32	0	0	0	0	0	0	0.68	0	do
21	do	do	do , Kuridaira (river-bed)	do	0.2	do	—	do	99.49	0	0	0	0	0	0	0.51	0	do
22	do	do	do , Kuridaira	do	0.02	do	—	do	98.50	0	0	0	0	0	0	1.50	0	do
23	do	do	do , Kamigawara	do	0.02	do	—	do	94.44	0	0	1.36	0	0	0	4.20	0	do
24	do	do	do , Obi	do	0.02	do	—	do	96.54	0	0	0.60	0	0	0	2.86	0	do
25	Gumma	Usui	Isobe-mati No. 1.	do	0.2	Tertiary	—	do	99.22	0	0	0	0	0	0	0.78	0	do
26	do	do	do No. 2.	do	0.2	do	—	do	97.16	0	0	0	0	0	0	2.84	0	do
27	do	Wagatuma	Kusatu, Yubatake-yu	do	156	Volcanic detritus	—	do	90.00	0.73	0	0.15	0	0	0	9.12	0	With hot spring.
28	do	do	do , Zizô-yu	do	30	do	—	do	89.09	0.50	0	0	0	0	0	10.41	0	do
29	do	do	do , Sirahata-yu	do	112	do	—	do	87.12	0.50	0	0.14	0	0	0	12.24	0	do
30	Nagano	Suwa	Kamisuwa, Nanatugama	do	54	Quarternary	—	do	2.85	—	0.07	0.76	0	87.91	0	8.41	trace	Evolves in the lake.
31	Hukuoka	Yama	Bantaisan, Nakano-yu	do	200	Volcanic detritus	—	May, 1924	83.39	2.10	0	0.39	0	13.90	0	0.22	0	With hot spring.
32	Hokkaidô	Isikari	Isikari-mati, Syunbetu	Oil well R_1 N.O.K.*	800	Tertiary Miocene (Oil bearing)	648	July, 1924	0	trace	2.95	0.08	0	37.96	58.48	0.53	0	
33	do	do	do	Oil well C_5 N.O.K.	1290	do	530	do	0.12	trace	3.34	0.19	0	34.14	58.47	3.74	0	
34	do	do	do	Oil well C_{25} N.O.K.	750	do	614	do	0.47	0	2.60	1.47	0	11.19	83.45	0.82	trace	
35	do	Sôya	Musuhoro	Natural outflow	4	Tertiary	—	do	1.63	0	0.22	0.11	0	93.04	0	5.00	0	
36	do	do	do	Oil well C_2 M.O.K.**	950	do	268	do	0.28	trace	0.48	0.35	0	97.39	0	1.50	0.003	
37	do	do	Wakkanai-mati, Tikappu	Oil well C_4 M.O.K.	440	Tertiary Miocene (coal bearing)	886	do	16.89	0.78	0.16	0.19	0	76.04	0	5.94	0	
38	do	Tesio	Horonobe-mura, Menasibetu	Natural outflow	7	do	—	do	1.51	0	0	0.24	0	94.34	0	3.91	0	
39	do	Sorati	Bibai-mati, Numagai	Coal prospecting well N.O.K.	50	Tertiary Eocene (coal bearing)	194	August, 1924	0.31	trace	0	0.25	0	99.29	0	0.15	0	With mineral spring; the well reaches no coal vein.
40	do	Yûbari	Yûbari-mati, Wakanabe No. 1.	do H. T. K. K.	20	do	263	do	0.27	trace	0	0.21	0	96.10	0	3.42	0.007	With hot spring; the well reaches a coal vein.
41	do	do	do , do No. 2.	do	50	do	157	do	0.12	0	0.06	0.38	0	92.86	0	6.58	0.004	The well reaches a coal vein.
42	do	do	do , Wakanabe, Sihorokabetu	Natural outflow	...	do	—	do	0.34	0	0.45	0.39	0	94.72	0	4.10	0	
43	do	Horobetu	Horobetu-mura, Noboribetu	do	—	Volcanic rock	—	do	80.73	5.87	0	0.19	0	0	0	3.54	0	Evolves from a fumarol. H_2=9.67 %.

* N. O. K. (Nippon Oil Kaisya) ** M. O. K. (Murai Oil Kaisya) ○ H. T. K. K. (Hokkaidô Tankô Kisen Kaisya)

TABLE 2.

Sample No.	Locality.			Source				Date Sampled.	Composition of Gas. (Per cent)								Remark.
	Prefecture	Prefectural District	Field	Name	Quantity of outflow m^3/day	Geology	Depth		CO_2 SH_2	Heavy Hydro-carbons	O_2	CO	CH_4	C_2H_6	N_2 Inert Gases	He	
44	(Syû) Taihoku	(Gôri) Kaisan	Dozyô-syô, Sekihekiryô	Natural outflow	1.2	Tertiary	—	Oct., 1924	16.29	0.01	0.15	0.01	82.66	0	0.88	0.007	
45	Tainan	Sinkwa	Nankwa-syô, Tikutôki	do	44	do	—	do	1.16	0.11	0.22	0.17	88.63	0	0.31	0.003	
46	Takao	Kisan	Kosen-syô, Kôsenpo	Disused well C_1	43	do	914.6	do	5.31	0.11	0.03	0.09	94.36	0	0.10	0.002	
47	do	do	Sanrin-syô, Zyuttyôri, Kasan	Natural outflow	9	do	—	do	0.67	0.40	0.12	0.15	92.37	3.27	3.02	0.002	
48	do	Takao	Ensô-syô, sensyûryô, Kunsuiko	do	414	do	—	do	1.04	0.26	0.11	0	95.80	1.25	1.54	0.005	
49	do	do	do , Sinsui	Disused well	12	do	—	do	1.56	0.08	0.07	0.13	97.86	0	0.30	0.003	
50	do	do	Nansi-syô, Konsuihei, Konsuisan	Natural outflow	22	do	—	do	2.52	0.17	0.18	0.13	95.87	0	1.13	0.004	
51	Tainan	Sinei	Sirakawa-syô, Kansirei	do	12	do	—	do	27.83	0.08	0.11	0.08	70.95	0	0.95	0.004	With hot spring.
52	do	do	do	do	10	do	—	do	40.01	0.33	0.20	0.25	57.24	0	1.97	0.003	do
53	do	do	do , Rokudyûkei	Disused well C_4	0.1	do	633.8	Nov., 1924	2.73	0.10	0.20	0	92.37	0	4.60	0.002	
54	do	do	do	do C_1	13	do	—	do	0.96	0.21	0	0.04	94.24	0	3.10	0.006	
55	do	do	do	Natural outflow	7	do	—	do	2.98	0.03	0	0	94.86	0	2.13	0.002	With hot spring.
56	do	do	do , Rokudyûkei, Kasan	do	45	do	—	do	1.74	0.42	0.17	0.04	93.05	2.70	1.88	0.002	
57	do	Kagi	Tyûho-syô, Tôsikyaku, Tikusigi	do	8	do	—	do	1.70	0.46	0.05	0.08	92.96	1.26	2.49	0.002	
58	do	do	do , Tyûron	do	362	do	—	do	89.96	0.03	0.05	0	3.64	0	6.32	0.003	
59	Sintiku	Byôritu	Kôkan-syô, Syukkôkô	Oil well $C_{22}C_{27}C_{13}$ $C_{18}C_{19}C_{29}C_{33}$ N.O.K.	7542	do	—	do	30.57	1.37	0.12	0.12	60.15	3.92	3.75	0.002	
60	do	do	do	Oil well C_{28} N.O.K.	297, 120	do	632.4	do	31.97	0.54	0.06	0.10	61.53	2.57	3.23	0.006	
61	do	do	Taiko-syô, Saidôhôkô	Natural outflow	116	do	—	do	9.18	0.87	0.14	0.09	87.02	0.92	1.78	0.006	
62	do	Tikunan	Zôkyô-syô, Sekikisi, Kinsui	Oil well R_1 N.O.K.	12	do	520.4	do	0.91	0.15	0.22	0.13	96.83	0	1.76	0.004	
63	do	do	do	Oil well R_5 N.O.K.	—	do	818.0	do	0.57	0.49	0.14	0.08	90.25	4.16	4.31	0.006	
64	Taihoku	Sitisei	Sirin-syô, Sôzan, Kôkeinai	Natural outflow	174	Volcanic detritus	—	do	95.13	0	0.15	0	1.98	0.14	2.60	0	Evolves from a solfatara.
65	do	do	do , Sitiko, Taiyûkô	do	—	do	—	do	96.99	0	0.23	0	0.12	0	2.66	0.004	do
66	do	do	Hokuto-syô, Hokuto, Iôdani	do	—	do	—	do	94.63	0.01	0.02	0	2.18	0	3.16	0.003	do
67	do	do	do , Digoku-dani	do	—	do	—	do	90.50	0.02	0	0.01	1.09	0	8.38	0.003	With hot spring.
68	do	Tansui	Sekimon-syô, Rôbai, Kyûkyûrin	do	36	Tertiary	—	do	89.42	0.05	0.15	0	7.23	0	3.15	0.002	
69	do	do	do	do	129	do	—	do	89.35	0.05	0.60	0	8.46	0	1.54	0.002	
70	do	do	do , Senzanko	do	116	do	—	do	88.53	0	0.28	0	8.19	0	2.95	0.002	
71	do	Kîrun	Kiusav-syô, Tyôtyûko, Sikôsihei	do	—	Volcanic detritus	—	do	97.10	0	0.13	0	0.44	0	2.33	0.002	Evolves from a solfatara.
72	do	do	Banri-syô, Simobanrikatô	do	—	Tertiary	—	do	88.04	0	0.12	0	4.85	0	6.99	0.004	With hot spring.
73	(Ken) Isikawa	Kasima	Hamura, Wakura	Hot spring well	1.0	do	3	April, 1924	0.13	0.21	trace	0	44.08	0	55.58	0.272	do
74	do	Enuma	Sakumi-mura, Katayamatu	do	1.2	do	—	do	0.14	0.02	0.11	0	4.90	0	94.83	0.308	do
75	Hukui	Sakai	Awara-mura, Hunatu No. 1.	do	0.8	Quarternary	60	do	0.52	0.09	trace	0	10.21	0	89.18	0.184	do
76	do	do	do No. 2.	do	0.4	do	63	do	0.24	0.08	0.01	0	9.07	0	90.50	0.186	do
77	Simane	Yama	Hukumitu-mura, Iti	Mineral spring well	7.2	Tertiary	—	do	99.03	—	—	—	—	—	—	0	With mineral spring.
78	do	do	Yunotu-mati	Hot spring well	43	do	—	do	84.58	0	0.45	0	0	0	4.97	0.007	With hot spring.
79	do	Annô	Sahimemura, Koyabara	do	29	Andesite	—	do	99.22	—	—	—	—	—	—	0	do
80	do	do	do , Ikeda	Natural outflow	2.1	Volcanic rock	—	do	98.35	—	—	—	—	—	—	0	With mineral spring.
81	do	Yatuka	Tamayu-mura, Tamadukusi	Hot spring well	2.4	Tertiary	—	do	1.18	0.08	0.12	0	0.71	0	97.91	0.088	With hot spring.
82	Tottori	Tôhaku	Misasa-mura, Misasa	do	0.01	Andesite	—	do	1.08	0	1.00	0	0.63	0	97.29	0.076	do
83	do	Tottori-si	Yosikata-mati	do	0.2	Quarternary	45	do	0.32	0	0.20	0	0.28	0	99.20	0.214	do
84	do	Mikata	Yumura-mura, Yumura	Natural outflow	1630	Volcanic rock	—	do	38.94	0	0.05	0	1.17	0	59.84	0.045	do

seven were found to be petroleum gases in Hokkaidô, Taiwan and Karahuto; others were mineral-spring gases, coal-mine gases and volcanic gases not only from the parts mentioned above but also Honsyû and Kyûsyû of Japan.

2. Sampling and Analysis.

The methods of sampling and analysis are practically identical with those used by Mr. Yamada.(1) It is an adaptation of the condensation method of Cady and Mc Farland(2) for the determination of helium (the apparatus is illustrated in Plate 23) and the Hempel's standard method for the analysis of common constituents.

In Hempel's standard method, the sum of carbon dioxide and hydrogen sulphide is determined by absorbing them in concentrated potassium hydrooxide solution. But, in some of our analysis the amounts of hydrogen sulphide have been separately determined in the following gravimetric way.

The desired quantity of a sample is passed slowly through a solution of lead acetate in order to precipitate sulphur completely as lead sulphide. This sulphide is converted into lead sulphate in usual manner and its quantity is determined by weighing.

The purity of helium is tested by observing its spectrum in the discharge tube attached to the condensation apparatus. The spectrum of the light emitted by the gas of each sample has been photographed with a small Hilger spectrograph. Three samples of spectrograms obtained in this way are illustrated in Plate 24. For the purpose of comparison, spectrograms of the discharge in helium, neon, argon, hydrogen and mercury vapour are also included in the plate.

3. Results of Analyses.

The results of analyses and informations concerning the sources

(1) loc. cit.
(2) Jour. Amer. Chem. Soc., **29** 1523, (1907).

from which the samples have been taken are shown in Tables 1 and 2. For the most part of geological informations the authors are indebted to Mr. K. Uwatoko. The places where the samples have been taken are marked on the accompanying map with black points and numerals corresponding to the "Sample Numbers." (Small circles on the map show Mr. Yamada's sampling places.)

The following table shows the nine samples which contain helium in the highest percentage, condensed from the Table I.

Table 3.

Sample Number	Source			Helium Content (%)
	Prefecture (Ken)	Prefectural District (Gôri)	Field	
74	Isikawa	Enuma	Sakumi-mura, Katayamatu	0.308
73	do	Kasima	Hamura, Wakura	0.272
83	Tottori	Tottori-si	Yosikata-mati	0.214
75	Hukui	Sakai	Awara-mura, Hunatu No. 1.	0.186
76	do	do	do , No. 2.	0.184
81	Tottori	Yatuka	Tamayu-mura, Tamadukuri	0.088
4	Hukuoka	Tukusi	Hutukaiti-mati, Musasi	0.087
82	Tottori	Tôhaku	Misasa-mura, Misasa	0.076
84	Hyôgo	Mikata	Yumura-mura, Yumura,	0.045

The sample (74) which is a hot spring gas in Tukimi-mura, Isikawa Prefecture is found to be richest in helium, and the sample (73) of the same kind in Hamura, Isikawa Prefecture takes the second place. Whether these helium-bearing gases can be regarded as the proper sources of helium or not depends chiefly upon the quantities of the evolution of these gases.

Unfortunately the gases above mentioned are so poor in their outflow that it would hardly be possible to be of practical use. Petroleum gases in Taiwan contain, with few exceptions, helium as shown in Table 2. Although its contents are very small, the quantities of outflow are enormous.

The constituent regarded as carbon dioxide and hydrogen sulphide are, as already mentioned, those which are absorbed in concentrated potassium hydroxide solution. In Table I the percentage of hydrogen sulphide determined separately are also given.

The part absorbed in ammonical cuprous chloride solution are usually regarded as carbon monoxide. However this constituent has been detected in none of the samples except those from Taiwan. Even of those samples from Taiwan, the contents of this constituent do not exceed 0·25 percent. It seems to have reason that Burrell and his collaborators[1] deny the presence of carbon monoxide in petroleum gases, ascribing the error on this point in previous works to the slight dissolution of higher paraffines in ammonical cuprous chloride solution. Burrell also denied the presence of olefine hydrocarbons in natural gases and ascribed the common point of error to the slight dissolution of higher paraffine hydrocarbons in fuming sulphuric acid. Nevertheless, in the results of the present authors the constituent absorbed in fuming sulphuric acid is expressed in the tables as heavy hydrocarbons. It is found to be contained in almost every petroleum gases in Taiwan and Hokkaidô. Also in mineral-spring gases it is contained in small quantities, even in cases where they evolve with hot spring and contain no higher members of paraffine hydrocarbons. This fact, together with result of Mr. Yamada who has found similar amounts of heavy hydrocarbons in the samples of the same sort as the authors' makes us belive that small quantities of heavy hydrocarbons are really present in such mineral-spring gases. It is difficult to believe that the contraction due to the absorption in fuming sulphuric acid in these cases is caused by the slight dissolution of higher paraffine hydrocarbons in this absorbent, while for petroleum gases which contain much larger amounts of higher paraffines, it will be possible to conceive that the error pointed out by Burrell may be made.

(1) G. A. Burrell and F. M. Seibert: Bull. 42, Bur. Mines, U. S. A., (1913), 116; Burrell and G. G. Oberfell, Tech. Paper 109, Bur. Mines, (1915), 11.

Worstal[1] asserted that methane was absorbed very slowly in fuming sulphuric acid at ordinary temperature, while on the other hand Burrell[2] remarked that methane was never absorbed in the absorbent in a few hour. In our experiments the time in which a sample is kept in contact with fuming sulphuric acid is no more than twenty minutes in all cases, and so there would be no error due to the dissolution of methane.

Oxygen is found in great majorities of samples and its content is less than 1 percent in every sample except No. (12), (23) and (34). These three gases have been presumably intermixed with air during sampling, owing to the insufficiency of outflow and inconvenient situations for sampling. As it is conceivable that a certain amount of air was entrapped previously in underground sediments or rocks at the time of geological changes, it may be natural that small quantities of oxygen have been found in most cases.

Hydrogen has been found in appreciable quantities in none of samples except No. (43), which belong to a kind of volcanic gas and contain 9·67 % hydrogen. Some hydrogen lines[3] have been, however, distinctly observed in some samples, when spectral observations have been carried out to test the purity of helium.

Helium has been determined by using from three to six litres of the gas samples. There are some samples which contain so small quantities of helium that its volume can not be directly measured, although its spectrum are clearly identified. For such cases, its content is less than 0·001 percent and denoted as "trace" in the tables.

4. Discussion of the Results.

An inspection on the results shows that natural gases in our country may be roughly classified into three types from their chemical

(1) Jour. Amer. Chem. Soc., **21**, 246.

(2) loc. cit.

(3) It is well known that one part of hydrogen in one hundred thousand parts of helium can be detected by the observation of discharge spectrum.

compositions. The first kind of them is composed chiefly of carbon dioxide, e.g. some volcanic and mineral-spring gases. The second is the gas which contains methane and other hydrocardon as the chief constituents and can be used as a gas fuel. Petroleum gases, coal-mine gases (Sample Nos. (5), (39), (40), (41)) and the gases which evolve from shallow geological horizon of the quarternary system are included in this type. The third consists mainly of nitrogen, some gases which issue with hot or cold mineral-springs belonging to this type.

There are, however, some gases of an intermediate nature ; samples Nos. (51), (52), (59) and (60) which issue in petroleum districts, contain methane and carbon dioxide both in great quantities : and the sample (73) which evolve with a hot spring has large contents of both methane and nitrogen.

In Japanese natural gases that have been ever examined untill to-day, helium is contained considerably only in nitrogenous gases. The gases which are chiefly composed of carbon dioxide do not generally contain helieum, except the gases of this type in Taiwan which contain very small amounts of it. Helium is also found in the gases of the methane type. And it will be noticed, as already remarked by Mr. Yamada that gases issuing from sources of quarternary system as Nos. (14), (15), (30) contain more helium than most of petroleum gases from the tertiary system.

Cady and Mac Farland[1] remarked that the percentage of helium in petrolum gases seemed to depend in a measure on the percentage of nitrogen. But Japanese petroleum gases thus far examined are too poor in helium to give any light on the relation between the helium and nitrogen content. In our case those gases which contain helium in considerably quantities are not petroleum gases but gases from mineral springs. However, the conclusion that gases rich in helium is nitrogenous, may hold also in this case.

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(1) loc. cit.

Mc Lennan and his collaborators[1] stated in the report of their investigation on the natural gases in Canada that it was impossible to establish a direct proportionality between the helium and emanation content of a gas. In the case of Japanese petrolum gases, it is impossible to study the relation between the helium content and the radioactivity, because all those examined previous to the report are too meagre in helium contents to establish a conclusion upon the evidence.

Of hot spring gases—those being Japanese gases containing helium in considerable quantities—the relation between helium and emanation contents is shown in the following table. (Table 4.)

Table 4.

Sample Number	Locality			Helium Content (%)	Emanation content per litre of gas (Maché unit)
	Prefecture (Ken)	Prefectural District (Gôri)	Field		
74	Isikawa	Enuma	Sakumi-mura, Katayamatu	0.308	24.18
73	do	Kasima	Ha-mura, Wakura	0.272	93.31
75	Hukui	Sakai	Awara-mura, Hunatu	0.186	39.43
81	Simane	Yatuka	Tamayu-mura, Tamadukuri	0.088	35.21
4	Hukuoka	Tukusi	Hutukaiti-mati, Musasi	0.087	32.54
82	Tottori	Tôhaku	Misasa-mura, Misasa	0.076	62.54
84	Hyôgo	Mikata	Yumura-mura, Yumura	0.045	25.98
16	Siduoka	Takata	Syûzenzi-mati	0.037	7.07
17	do	do	Nagaoka-mati	0.029	1.29
7	Kumamoto	Kuma	Nakahara-mura, Hayasi	0.023	5.64
78	Simane	Yama	Yunotu-mati	0.007	3.31
27	Gumma	Waga-tsuma	Kusatu, Yubatake-yu	0	0.88
28	do	do	do , Zizô-yu	0	0.65
29	do	do	do , Sirahato-yu	0	0.66

(1) J. G. M. Lennan: Report on some sources of Helium in British Empire.

In the table, gas samples are arranged in the order of the helium content. The figures showing the emanation contents are entirely due to Dr. Isidu's[1] measurement which was undertaken some years ago. Neverthless, it can be perceived that the percentage of helium seems to depend in a measure on the content of emanation of a gas, though there is no direct proportionality between them. The samples which contain no helium, for instance the last three in the table, possess in general merely feeble radioactivity. This aspect will stand in favour of the theory that ascribes the origin of helium in a natural gas to the disintegration of radioactive substances.

It is strange that the relation like above never holds in the case of cold spring gases—gases which issue with cold mineral-spring. For example samples Nos. (19) and (80) which evolve respectively in Masutomi, Yamanasi Prefecture and in Ikeda, Simane Prefecture contain no helium although considerable amounts of emanation have been found in them.

A wide variation in composition is observed of petroleum gases. While the gases belonging the same vein have nearly the same composition, those belonging to the different veins vary widely even when their situations are pretty near. It is conceivable that some relation may exist between the composition of these petroleum gases and the nature of petroleum oils accompanying them. Mr. Yamada pointed out already this relation for the petroleum gases in Honsyû.

Sample No.	Composition of Gas								Density of Oil (15° C)	Rates of Higher Hydrocarbons to Total Hydrocarbons
	CO_2 H_2S	Heavy Hydrocarbon	O_2	CO	CH_4	C_2H_9	N_2 Inert Gas	He		
37	17.67	0.16	0.19	0	76.04	0	5.94	0	0.927	0.002
36	0.28	0.48	0.35	0	97.39	0	1.50	0.003	0.870	0.005
32	0	2.95	0.08	0	37.96	58.48	0.53	0	0.797	0.618
34	0.47	2.60	1.47	0	11.19	83.45	0.82	trace	0.794	0.885
33	0.12	3.34	0.19	0	34.14	58.47	3.74	0	0.787	0.644

(1) R. Isidu: The Mineral Springs of Japan.

Here the composition of gas and the density of oil from the same field in Hokkaidô are tabulated. The density of oil was determined by Mr. K. Uwatoko who kindly furnished the results to the authors.

Mr. Yamada's conclusion on the relation between the nature of gases and accompanying oils of the Honsyû origin has been confirmed to hold true also for those of Hokkaidô.

There can be detected no definite quantitative relation between the the density of oils and the content of higher hydrocarbons, and the ratio of the amount of heavy hydrocarbons and ethane to the total amount of hydrocarbons does not always increase with the decrease of the density of oil. Qualitatively the content of higher members of hydrocarbons in a gas depend in a measure on the density of an oil from the same field. The higher the specific gravity of oil is, the poorer the content of higher hydrocarbons in gas—a relation which seems very natural.

The compositions of the petroleum gases in Japanese three islands —Honsyû, Hokkaidô and Taiwan are then compared in the following.

Carbon dioxide is generally contained about I percent, and its content rarely exceeds ten percent in the petroleum gases of Honsyû and Hokkaidô, while those of Kinsui district in Taiwan contain carbon dioxide of about thirty percent. The parafine hydrocarbons in the gases of Taiwan are chiefly composed of methane, and poor in higher paraffine hydrocarbons, the gases in Honsyû are richer in higher members of paraffine hydrocarbons. In Hokkaidô, the gases in Isikari oil-fields contain them in great quantities (60–80 percent), while those in Sôya oil-fields contain none. Helium is not found to be contained in most of the petroleum gases in Honsyû and Hokkaidô, while in Taiwan it is found to be distributed uniformly over all the oil-fields and the samples from various localities, very different in other respects, contains generally about 0.005 % helium. In Taiwan the gases which come out "naturally", as well as those from artificial wells contain helium in similar amounts, while it is found in other islands only in well gases. It is perhaps due to the large capacity of the source and outflow in Taiwan.

Samples Nos. (40) and (41) contain helium while No. (39) does not, notwithstanding the geological and other features of their resorvoirs are essentially similar. The sample (39) has been taken from a boring-well prospecting coal which could not reach any coal vein while wells giving samples (40) and (41) reach coal veins. From this fact it seems that helium frequently exist underground in the neighbour of a coal vein though in a small quantity.

The amount of helium in a petroleum gas seems to depend on the geological age of its resorvoir—a relation already manifested itself in the case of the natural gases in the United States of North America.[1] Oil resorvoirs of Mid Continent in that country, which are famous as excellent sources of helium, belong to the paleozoic system. On the other hand oil reservoirs of Hokkaidô and Taiwan belong to the tertiary system like that of Honsyû and do dot contain helium more than 0.007 percent.

In short, the petroleum gases in Taiwan and Hokkaidô have little prospects as the source of helium. Considerable percentage of helium are found, as already mentioned, in some spring gases, but unfortunately their evolution is at present not sufficient for any industrial purposes. Only few numbers of the volcanic gases in our country have been investigated, so that their future prospects can not yet be expected at present.

Summary.

(1) Twenty seven samples of the petroleum gases in Taiwan, Hokkaidô and Karahuto and fifty seven samples of the mineral spring, volcanic and coal-mine gases were collected and studied on their contents of helium and other constituents.

(1) G. S. Rogers: Helium-bearing Natural gases, U. S. G. Prof. Paper. 121 (1921).

(2) The helium content of some mineral spring gases is found to reach a few tenths of percent. Merely small quantities of helium are found in the gases from the resorvoirs in the petroleum fields of Taiwan which belong to the tertiary system.

(3) Natural gases are classified into three types respectively rich in carbon dioxide, in hydrocarbons, and in nitrogen. Considerable quantities of helium are found to be contained only in the gases of the last type.

(4) The percentage of helium in a "hot spring gas" seems to depend in a measure on the content of the emanation. But strangely this relation does not hold in the case of a gas which evolves with a cold mineral-spring.

(5) The compositions of the natural gases in Hokkaidô, Honsyû and Taiwan were briefly compared. Helium is not found to be contained practically in most of the petroleum gases in Honsyû and Hokkaidô, while in Taiwan it is found to be distributed uniformly in all the oil-fields and samples from very different localities contain generally about 0.005 % helium.

(6) The contents of higher hydrocarbons in the petroleum gases in Hokkaidô, as is to be expected, also have a certain relation with the density of accompanying petroleum oil. The higher the specific gravity of oil is, the poorer the content of higher hydrocarbons in gas.

In conclusion, the authors wish to express their gratefulness to Assistant Professor N. Yamada who kindly guided them in their work and also granted complete access to all of the facilities with which his valuable study had been carried out. Thanks are due to Prof. Katayama of the Faculty of Science for his valuable advices during the work and are also due to Mr. Uwatoko for his work in collecting gas samples and surveying the fields.

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抄　　録

本邦產天然瓦斯中の「ヘリウム」及其他の成分の含量に就て

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航空船,氣球等の塡充瓦斯として理想的不燃性瓦斯「ヘリウム」の本邦に於ける產出狀況如何に關して先に山田延男氏は東京帝國大學航空研究所に於て本州地方の代表的石油瓦斯並びに鑛泉瓦斯其他を分析し「ヘリウム」の含量を調査せり.

航空船塡充用として水素瓦斯を使用することに隨伴する危險の現下防止し難きことは吾人をして不燃性「ヘリウム」を渴望せしめる所以にして實に「ヘリウム」問題は國防上の意義深きものなりと信ぜらる.

この意味に於て著者等は山田氏の研究の後を繼ぎて本邦內の全般に亙りて單に石油瓦斯のみならず鑛泉瓦斯,溫泉瓦斯及び火山瓦斯等總て產出する所の天然瓦斯の代表的のものに就て「ヘリウム」の含量を決定せり.而して同時に此等の天然瓦斯中の諸成分及分布狀態を知ることは學術上にも興味あるのみならず將來燃料其他の化學工業の問題に關して參考となるものなれば併せて研究を行へり.

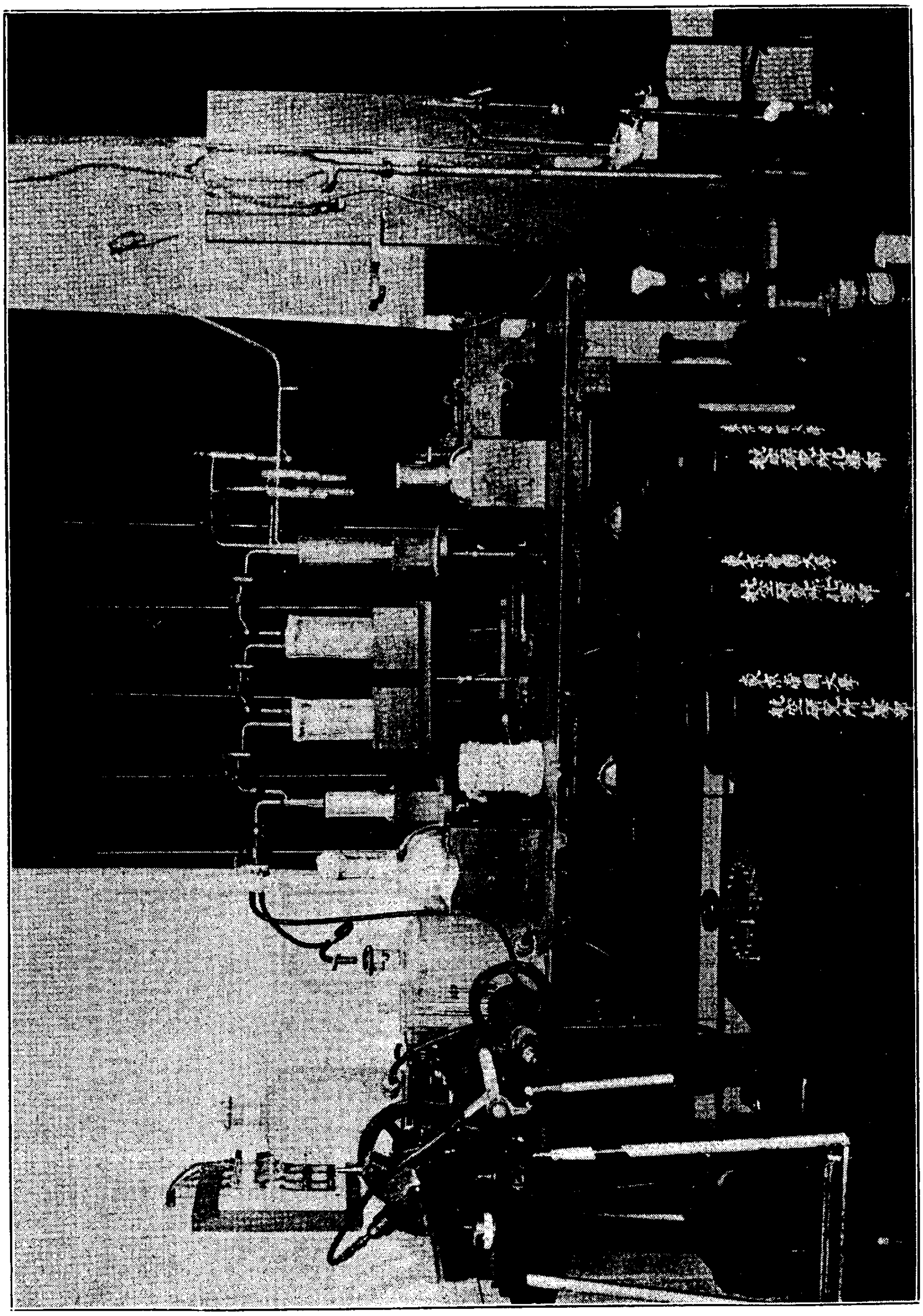
採集分析せし瓦斯は八十四種にして其の中二十六種は臺灣,北海道,樺太に產する石油瓦斯にして他は本州,九州,北海道及び臺灣の諸地方に於ける鑛泉,溫泉,石炭坑及び火山等より噴出する瓦斯なり. 臺灣に於ける石油瓦斯は其の噴出量甚大なる故多大の興味を以て分析せるも其の「ヘリウム」含量は何れの試料も 0.005％內外に止まれり. 「ヘリウム」の含量最大なりしは石川縣に於ける溫泉瓦斯にして江沼郡作見村に產するものは 0.30％, 鹿島郡端村に產するものは 0.272％の含量を示せり. 然れども其の噴出量は微少にして實用的價値に乏しきは遺憾なり. 北海道夕張に產するが如き石炭坑瓦斯にも小量ながら「ヘリウム」の含有せらるゝを見たり.

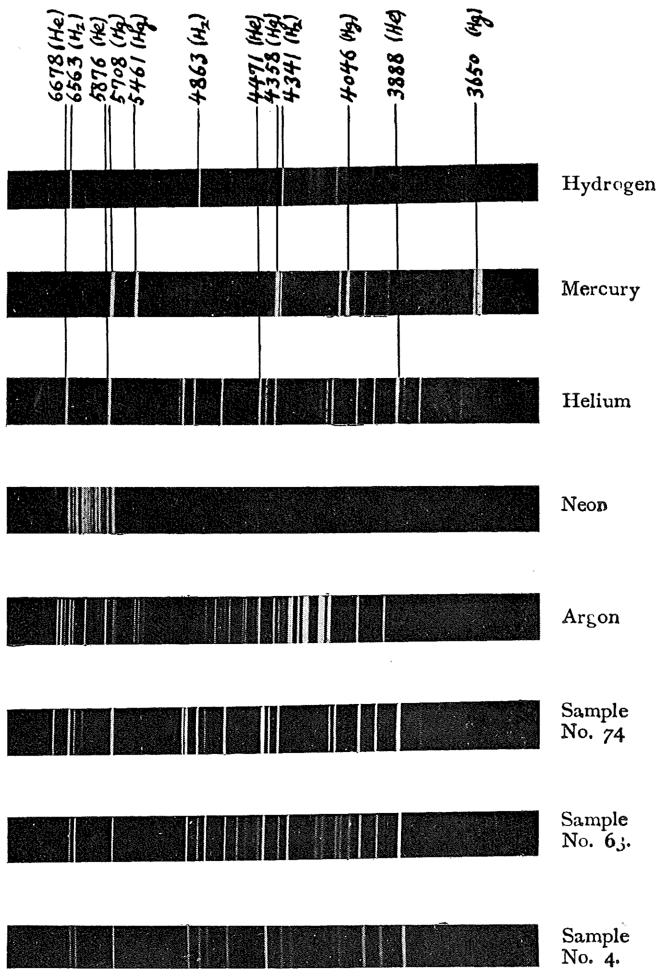
分析せし瓦斯八十四種を化學組成の上より分類し組成と「ヘリウム」の含量との關係を調査せり. 石油瓦斯に就ては組成と共成する石油の性質との關係を檢せり.

亞米利加合衆國に於ける例に鑑みるに石油瓦斯に於ては本邦の含油地層何れも新しき第三紀地層に屬する爲め之れに胚胎せらる瓦斯に「ヘリウム」の含量乏しきなりと思惟さる.

温泉瓦斯に於ては「ヘリウム」の含量大なる程其の瓦斯或は共生する温泉の放射能大にして其の間に何等かの關係の存するが如く見へたるは面白し鑛泉瓦斯に就ては全く斯る關係は成立せず.

火山瓦斯に就ては未だ調査乏しく其の前途を豫想するを得ず.

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Neon
Argon
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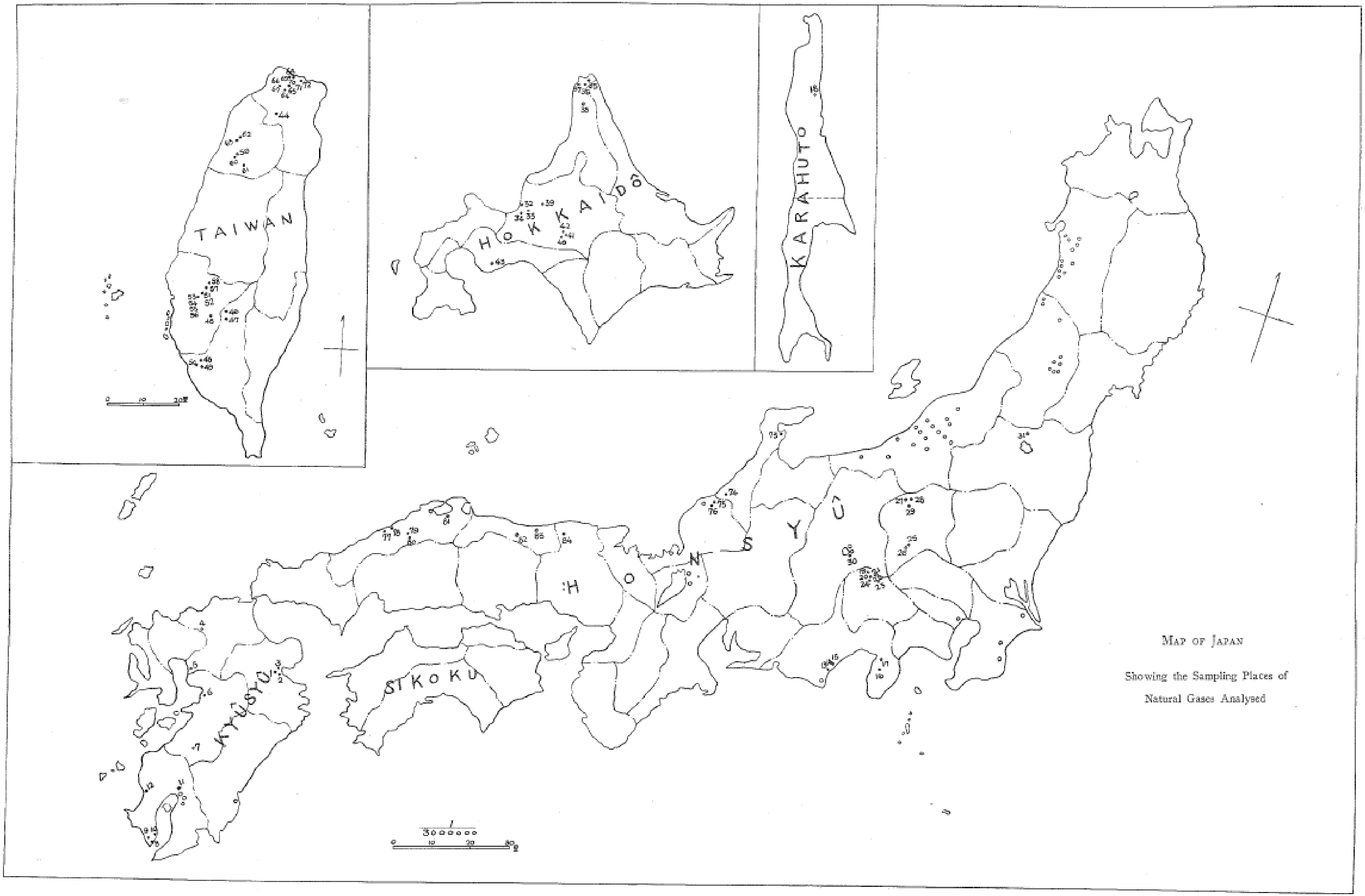
Sample No. 4.


TAIWAN
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SIKOKU
KYÛSYÛ
MAP OF JAPAN
Showing the Sampling Places of
Natural Gases Analysed