INIS-mf--13497

VARIATIONS OF CAESIUM ISOTOPE CONCENTRATIONS IN AIR AND FALLOUT AT DALAT, SOUTH VIETNAM, 1986-91

Pham Zuy Hien, Nguyen Thanh Binh, Truong Y,
Vuong Thu Bac, Nguyen Trong Ngo.

Dalat Nuclear Research Institute, Dalat, Vietnam[(1)]**.**

ABSTRACT

Monthly records of ^{137}Cs and ^{134}Cs concentrations in air and fallout at Dalat for the period 1986-91 are presented and discussed. The concentration variations exhibit distinct maxima during December- January, when dry fallout dominated. These peaks are explained by the intrusion of more radioactive cold air masses from temperate northern latitudes during the development of large-scale anticyclones frequently observed in the most active winter monsoon period. High dry fallout velocities (about 10 cm/s) determined from this data clearly demonstrate one of the most relevant characteristics of cold air masses : behind the cold front, vertical air motion is descending.

[(1)] **Work performed under IAEA Research Contract No. 4656/R1/RB**

INTRODUCTION

A preliminary radiation survey in Vietnam during 1982-85 revealed quite low environmental levels of ^{137}Cs, e.g. 0.2-0.3 μBq/m^{3} in aerosol, 0.1-0.2 Bq/m^{2}.month for fallout, 2 - 4 Bq/kg in surface soils, 0.1-0.2 Bq/kg in cereals. These levels are many times lower than those of most European and other higher latitude countries. Since 1986, monthly samples of air and fallout have been measured at Dalat (11^{o}57'N, 108^{o}26'E, 1500m asl). The results show a number of distinct maxima. The first, observed in May 1986, corresponds to the arrival of the radioactive cloud from the Chernobyl reactor accident: the monthly ^{137}Cs deposition increased by several orders of magnitude, and radionuclides such as ^{103}Ru, ^{106}Ru, ^{124}Sb, ^{131}I, ^{140}Ba and ^{134}Cs were detected for the first time. The levels of these Chernobyl products then decreased rapidly; only ^{137}Cs was detected after August. However, from November 1986, the Chernobyl activation products ^{134}Cs was again detected, accompanied by an increase of ^{137}Cs, reaching a second maximum in January 1987. Only the latter isotope was detected in subsequent years, with a regular maximum during December-February.

The peak levels of the two caesium isotopes were observed during periods of most active atmospheric circulation in the West Pacific tropical zone, under the influence of the quasi-permanent Asiatic high pressure centre. This suggests the role of the winter monsoon in transporting cold air with higher isotope concentrations from temperate northern latitudes into the tropical zone.

SAMPLING AND MEASUREMENT PROCEDURES

Particulate radioactivity was collected from about 100,000 m^3 of air on 0.48 m^2 chlorinated vinyl polychloride Petrianow filter FPP-15-1.7 using an air sampler 12-UC-34 with flow rate 760 m^3/hr. The intake was 1.5 m above ground. The filters were compressed into pellets of 36 mm diameter and 10 mm thickness for gamma spectrum measurement.

Wet and dry fallout were collected throughout the month in three stainless-steel trays, each of cross section 0.4 m^2. During dry periods, resuspension of dry fallout particles was prevented by adding distilled water to a depth of about 1 cm. Samples were passed through a filter to separate suspended and dissolved fractions. The filtrate was subsequently evaporated in vacuum. During the rainy season, when collected monthly rainwater reached some hundred litres, the filtrate was pre-concentrated by co-precipitation.

Gamma spectrum measurements were made using a low background system with relative efficiency 15%, peak-to-Compton ratio 41.5/1, and FHWM 1.92 keV for the 1332.5 keV ^{60}Co line . The integral background (100-2000 keV) was 1.7 cps. Detection limits for both caesium isotopes in fallout and air were respectively 0.01 Bq/m^2 and 0.02 μBq/m^3.

RESULTS AND DISCUSSION

Monthly variations of ^{137}Cs and ^{134}Cs levels are shown in Fig.1a,b covering the period 1986-91. Fig. 1c shows the monthly precipitation totals at Dalat meteorological station. The results for other Chernobyl products are not presented; these were detected only during May-August 1986. The $^{137}Cs/^{134}Cs$ ratio during May-June 1986 was 1.99 ± 0.14 for fallout and 2.2 ± 0.5 for aerosol, both in good agreement with other data (Aoyama, 1988; Chien Chung, 1989; Higuchi et al ., 1988; Mueck, 1988; Zhang Yongxing, 1988).

The intensities of the annual peaks are listed in Table 1. For comparison, Table 2 shows the ^{137}Cs concentration in air recorded in neighbouring countries. The levels at Dalat are very much lower. Needless to say,the Dalat data cannot be regarded as typical of the whole territory of Vietnam. Airborne dust was also collected during June 1986 at Hanoi (21°01'N, 105°48'E, 10 m asl) and Hochiminh City (10°47'N, 106°40'E, 10 m asl); the measured activity concentrations of Chernobyl-derived isotopes were 30-80 times greater in Hanoi and 2-3 times greater in Hochiminh City.

Such low activities of Chernobyl-derived radionuclides at Dalat can be explained by the high elevation. According to the observations of Higuchi et al. (1988), the height of the radioactive air passing over Japan was 1200-1800 m, whereas Dalat is located in the central highland of Indochina at altitude 1500m. The SW winds prevailing in South Indochina during May-July are considered to be also an important influence causing the low activity concentrations recorded at Dalat and HochiMinh City; at the same time, Hanoi was still influenced by air masses moving south from mid-latitudes to the West Pacific tropical zone (see e.g. the weather map shown in Chien Chung (1989)).

The intensity maximum observed in Dalat during December 1986 - February 1987 appeared surprisingly high when compared to the Chernobyl peak observed 8 months earlier (Table 1). Both ^{137}Cs and ^{134}Cs were observed. The activity ratio, corrected to April 1986, was 2.0 ± 0.2 for fallout and 2.2 ± 0.7 for air, which indicates the predominant Chernobyl origin. In subsequent years, only ^{137}Cs was observed and the winter maxima decreased quite rapidly.

To identify the origin of the observed winter peaks, some possible mechanisms can be assessed. The first is resuspension of soil particles. The role of this process can be evaluated on the basis of the $^{137}Cs/^{134}Cs$ activity ratios measured in surface soil and in air and fallout (Aoyama, 1988). The mean ^{137}Cs activity in the surface soil at Dalat in 1987 was 3 Bq/kg. ^{134}Cs has not been detected in soil in Vietnam. Measurements of the soil samples collected in an area with ultrabasic rocks having very low uranium concentration (less than 10^{-2} ppm) yield an upper limit 0.05 Bq/kg for ^{134}Cs concentration in surface soil, corresponding to a $^{137}Cs/^{134}Cs$ activity ratio greater than 60. This is in contrast with the occurrence of ^{134}Cs in both the air and fallout in winter 1986-87 with a $^{137}Cs/^{134}Cs$ ratio typical of Chernobyl-derived caesium isotopes.

The 500kW Dalat nuclear research reactor can be considered as a possible source of caesium isotopes. But regular measurements of radionuclides in water and ion exchange resins of the reactor primary cooling system have not detected any fuel element failure which could cause discharge of fission products to the atmosphere.

Thus there are believed to be no local sources of the observed peaks. There seem to be two possible non-local origins, viz. increased stratospheric-tropospheric exchange (stratospheric fallout), or intrusion of radioactive tropospheric air from temperate latitudes into the tropical zone.

Injection of radioactivity from stratosphere into the northern troposphere during spring-early summer has been a regular feature of the stratospheric fallout from nuclear tests, with variations depending on the latitude of the collecting station (Ehhalt & Haumacher, 1970). Aoyama (1988) observed a spring maximum of Chernobyl isotopes in April 1987 in fallout at Tsukuba, Japan. Its intensities was almost three orders of magnitude lower than the initial tropospheric Chernobyl peak, suggesting that the stratospheric inventory was only about 0.5% of the total release of caesium isotopes. These features are considerably different from the maxima observed in this study.

The coincidence of the peaks observed at Dalat with the most active periods of the winter tropical monsoon suggests that cold continental polar (cPk) air masses move equatorwards from temperate latitudes to supply with higher caesium isotope concentrations. Such a circulation of cPk air from the NE often reaches very low latitudes during December-January, when the intertropical convergence zone is aligned in the Southern Hemisphere up to 10°S. cPk air moves from a source region over the Asian interior (the quasi-permanent Asiatic high-pressure centre). It is well known that the latitudinal distribution of stratospheric fallout from nuclear weapons tests shows a maximum around this latitude (40-50°N). Concerning the Chernobyl debris, after the initial rapid fallout in 1986 , maximum concentrations of caesium isotopes should also have been located around this latitude belt; this assumption is based on the known intensified flow of radioactivity from stratosphere-troposphere exchange during the northern spring, when the accident occurred, and the influence of the westerly zonal circulation in the upper atmosphere. Fallout from the upper atmosphere is considered to be the main process leading to the presence of fission products in the lower troposphere after the decline of the initial phase of the Chernobyl-derived fallout.

Measurements from the post-Chernobyl period are so scarce that it is not possible to establish a latitudinal distribution of ^{137}Cs concentrations in air for the Far East region. However a few data shown in Table 2 for winter 1986-87 appear to confirm a considerable difference between concentrations at Dalat and those at higher latitude locations of the region (Mishra, 1990; Radioactivity Survey Data in Japan, 1991). Thus, the increase of 0.3 $\mu Bq/m^3$ at Dalat during this period (Fig.1) is ascribed to intrusion of the colder, more radioactive air from higher latitudes.

The role of cold air masses in transporting more radioactive air from temperate latitudes to the tropical zone was well demonstrated by Dmitrieva et al. (1970) in a study of atmospheric β-activity of air during an Atlantic, Indian and Pacific Ocean cruise. Sharp increases in near-surface air were observed when the ship crossed the cold front from the warm air side, and always just behind the cold front in the ridge of the anticyclone. These authors suggested that more radioactive air was supplied firstly from the stratosphere at temperate and subtropical latitudes by a process called "stratospheric break", and was then brought towards the equator during the development of the anticyclone.

The monthly sampling procedures adopted in this study could not reveal the fluctuations of air radioactivity in connection with the passage of any cold front. Moreover, such cold fronts are usually weakly defined at low latitudes (south of 15^{o} in southern Indochina). But, as shown below, a relevant characteristics of the cold air behind the cold front, its descending vertical motion (Godske et al.,1957), can be identified from the obtained above data. The dry fallout velocity can be approximated by dividing the fallout concentrations ($Bq/m^2.s$) by the air concentrations (Bq/m^3). The resulting velocities are in the range 5-15 cm/s; these values are much higher than those measured in May-June 1986 in Europe (Monte, 1990; Mueck, 1988), North America (Smith & Ellis, 1990) and China (Zhang Yongxing,1988) (about 0.1 cm/s) and indicate descending motion of cold air during December-January at Dalat. This result agrees fairly well with values up to 7.2 cm/s determined during the dry season at Bombay, India (Rangarajan et al., 1985), suggesting, probably, similar downwards movement of air at both locations.

CONCLUSIONS

Winter peaks of ^{137}Cs in air and fallout were observed at Dalat in five successive years from 1986. The presence of ^{134}Cs in the first of these peaks (1986-87) showed that some Chernobyl debris was present. The regular occurrence of these annual peaks at the season of the most active winter monsoon suggests that they are due to intrusion of more radioactive air from higher latitudes. The ^{137}Cs concentrations yield quite high estimates of dry fallout velocity ascribed to the descent of air behind the cold front in the ridges of the associated anticyclones.

REFERENCES

Aoyama, M. (1988). Evidence of stratospheric fallout of caesium isotopes from Chernobyl accident. *Geophys. Res. Lett.*, 15, 327-30.

Chien Chung (1989). Environmental radioactivity and dose evaluation in Taiwan after the Chernobyl accident. *Health Phys.*, 56, 465-71.

Dmitrieva, G. V., Krasnopevtsev, Ju. V. & Malakhov, S. G. (1970). Some peculiarities of fission product concentration distributions over Oceans in the tropical zone and their connection with atmospheric processes. *J. Geophys. Res.*, 75, 3675-85.

Ehhalt, D. H. & Haumacher, G. (1970). The seasonal variation in the concentration of ^{90}Sr in rain and its dependence on latitude. *J. Geophys. Res.*, 75, 3027-31.

Godske, C. L., Bergeron, T., Bjerknes, J. & Bundgaard, R. C. (1957). Dynamic meteorology and weather forecasting. *Joint Publication of AMS, Boston and Carnegie Institution of Washington, USA.*

Higuchi, H., Fukatsu, H., Hashimoto, T., Nonaka, N., Yoshimizu, K., Omine, M., Takano, N. & Abe, T. (1988). Radioactivity in surface air and precipitation in Japan after the Chernobyl accident. *J. Environ. Radioactivity*, 6, 131-44.

Mishra, U. C. (1990). Comparison of radionuclides levels from the Chernobyl reactor accident and from global fallout. *J. Radioannal. and Nucl. Chem.*, 138, 119-25.

Monte, L. (1990). Evaluation of the environmental transfer parameters for ^{131}I and ^{137}Cs using the contamination produced by the Chernobyl accident at a site in Central Italy. *J. Environ. Radioactivity*, 12, 13-22.

Mueck, K. (1988). Variations in activity concentration and radionuclide ratio in air after the Chernobyl accident and its relevance to inhalation dose estimates. *Radiation Protection Dosimetry*, 22, 219-29.

Radioactivity Survey Data in Japan, Part 1, *Environmental Materials*, 94, Oct 1991.

Rangarajan, C., Eapen, C. D. & Gopalakrishnan, SMT. S. (1985). Measured values of the dry deposition velocities of atmospheric aerosols carrying natural and fallout radionuclides using artificial collectors. *Reprint, Bhabha Atomic Research Center, Bombay, India.*

Smith, J. N. & Ellis, K. M. (1990). Time dependent transport of Chernobyl radioactivity between atmospheric and lichen phases in Eastern Canada. *J. Environ. Radioactivity*, 11,151-68.

Zhang Yongxing (1988). Evaluation of release amount from Chernobyl accident and of resulting radiological consequence in China. *Reprint, Institute of Atomic Energy, Beijing (in Chinese).*

TABLE 1

Caesium isotopes concentrations peaks observed in air and fallout at Dalat during 1986-91

Period	*Maximum concentration in air ($\mu Bq/m^3$)*		*Integrated deposition (Bq/m^2)*	
	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs
May-July 1986	1.30±0.13	0.59±0.09	3.19±0.13	1.60±0.05
Nov 1986 -Feb 87	0.82±0.12	0.32±0.06	4.24±0.18	1.64±0.16
Dec 1987	0.80±0.12	*)	0.30±0.08	*)
Dec 1988 -Jan 89	0.47±0.09	*)	0.12±0.03	*)
Jan-Mar 1990	0.5 ± 0.1	*)	0.08±0.01	*)
Dec 1990	0.46±0.09	*)	0.11±0.02	*)

*) Value below detection limit

TABLE 2

^{137}Cs concentration in air (mBq/m^3) measured in neighbouring countries

Country, location	*May-June 1986*	*Nov 1986- Feb 87*	*Reference*
China, Beijing (40°N, 116°E)	7.4		Zhang, 1988
Japan, Chiba (36°N, 140°E)	16.4	0.001 - 0.01	Higuchi, 1988 RSDJ, 1991
Taiwan, Hsinchu (25°N, 121°E)	2.5		Chien, 1989
India, Bombay (19°N, 73°E)	1.6	0.03	Mishra, 1990
Vietnam, Dalat (11°N, 108°E)	0.0013	0.0005-0.0008	

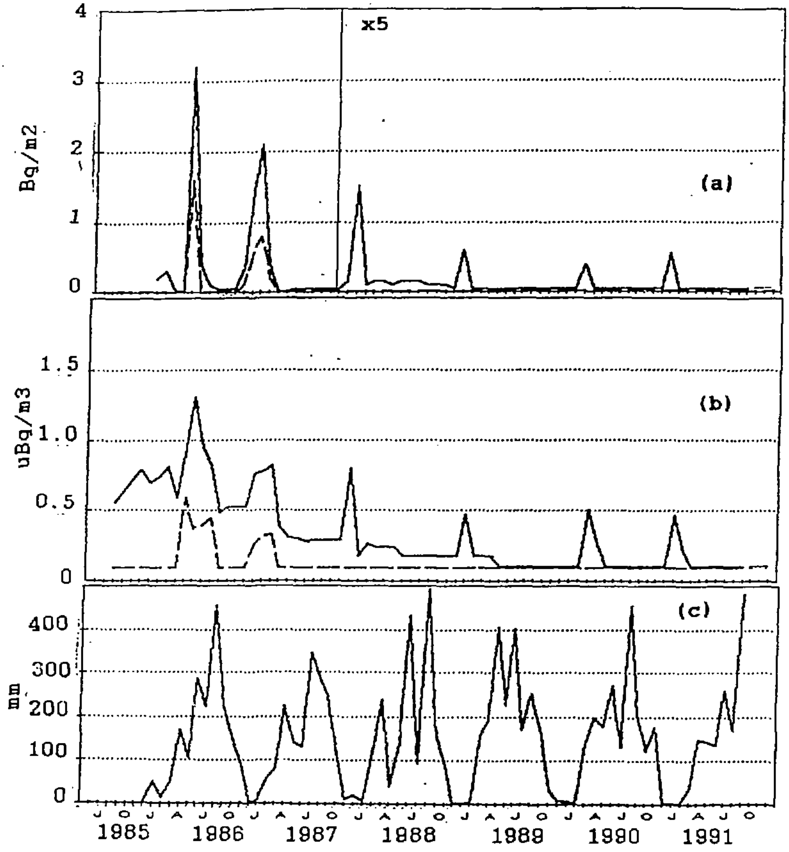

Fig. 1

Monthly variations of caesium isotopes activities in fallout and airborne at Dalat during 1985-1991 period.

a. Cs-137 (———), Cs-134 (---) in deposition

b. Cs-137 (———), Cs-134 (---) in airborne

c. Monthly precipitation