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LONG RANGE TRANSPORT OF CAESIUM ISOTOPES FROM TEMPERATE LATITUDES TO THE EQUATORIAL ZONE DURING THE WINTER MONSOON PERIOD

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ABSTRACT

An air radioactivity monitoring study carried out in Dalat, Vietnam since 1986 has revealed distinct peaks of caesium isotope concentrations in air and fallout during December-January, when the monthly average air temperature was lowest and dry fallout dominated. These peaks provide evidence of 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 velocity (about 10 cm/s) determined from the measured concentrations, clearly demonstrates one of the most relevant features of cold air masses: behind the cold front, vertical air motion is descending. The role of other processes,such as injection of radioactive air from stratosphere and local resuspension of soil dust, has been shown to be insignificant. The interpretation of the experimental results was based on the analysis of environmental-meteorological factors as well as the behaviour of other naturally-occurring radionuclides.

1. INTRODUCTION

A radioactivity survey conducted in Vietnam during the 1980's showed quite low levels of ^{137}Cs in air, fallout and other environmental objects. The typical values were many times lower than those of most European and higher latitude countries. This is in agreement with the latitudinal distribution of the global radioactive fallout from nuclear weapon tests. Other fission and activation products were not detected, except during the period from May to July 1986, immediately following the Chernobyl accident. Under such conditions, an air radioactivity monitoring study is still of importance in providing a basis for assessing suspected accidental releases of radioactivity into the atmosphere. Furthermore, such a monitoring study could provide information on seasonal variations of air radioactivity and their correlation with the seasonal changes of weather and meteorological conditions as well as long-range air circulation involving high activity air masses.

Sampling and measurement of radionuclide concentrations with respect to various aspects of the environment have been routinely conducted in Dalat ($11^0$57'N, $108^0$26'E, 1500m asl) since 1986. The study has revealed distinct maxima of airborne and deposited caesium isotopes.

During the period from May through July 1986, a variety of fission and activation products was detected in both the airborne filter and fallout in Dalat, about 10,000 km away from the damaged Chernobyl reactor. Chernobyl accident debris was observed also in Hanoi ($21^0$01'N, $105^0$48'E, 10m asl) and Hochiminh City ($10^0$47'N, $106^0$40'E, 10m asl) during task force campaigns. It has been shown in our previous paper [1] that during that period these radioactive species followed different trajectories to Vietnam, resulting in quite different levels of activity concentrations in air and fallout at the three above cited locations.

Besides the Chernobyl peaks, the monitoring study has revealed regular peaks of ^{137}Cs for both the air and fallout during December and January, when the monthly average temperature was lowest and dry fallout dominated. For activation product of Chernobyl origin ^{134}Cs, such a peak was detected in both the air and fallout only during the period from December 1986 to February 1987.

The peak levels of caesium isotopes were observed during the period of most active atmospheric circulations in the West Pacific tropical zone, under the influence of the quasi-permanent Asiatic high pressure centre. This indicates the role of winter monsoons in transporting cold air with higher isotope concentrations from temperate northern latitudes into the tropical zone. A preliminary explanation of this fact was reported in our previous paper [1]. In this paper the evidence of this process will be shown by involving information on meteorological and weather factors and on the behaviour of other environmental radionuclides. Some typical features of cold air masses transported by winter monsoons to the equatorial zone will also be revealed from the analysis of the experimental data.

2. EXPERIMENTAL AND RESULTS

As the concentrations of caesium isotopes in air were very low, a large volume of air (about $100{,}000 m^3$) was collected every month, followed by radioactivity measurement of the filter on a low background gamma spectrometry system. An air sampler with flow rate $760 m^3$/hr was used in the experiments. Air dust was collected on a

$0.48m^2$ Petrianow filter FPP-15-1.7. The uncertainty of the flow rate given above is estimated to be not less than 15-20%. However, this has caused only a little concern, as such an uncertainty affected the experimental activity concentrations uniformly for all aerosol samples and all radionuclides. Wet and dry fallout were collected throughout the month in three stainless-steel trays, each of cross section $0.4m^2$. Gamma spectra were measured using a low background system with high purity Ge-detector of 15% efficiency and 1.9 keV FWHM for the 1332.5 keV ^{60}Co line. The measuring time was often not less than 20 hrs yielding detection limits for both the caesium isotopes in the air and fallout not exceeding 0.02 Bq/m^3 and 0.01 Bq/m^2 respectively. Concentrations of naturally-occurring radionuclides, such as ^{7}Be, ^{40}K, ^{232}Th etc, were also quantitatively determined through their gamma lines.

Monthly variations of ^{137}Cs and ^{134}Cs activity concentrations are shown in Fig. 1a,b covering the period 1986-91. Fig. 1c shows the monthly precipitation totals recorded at the Dalat meteorological station. The caesium isotope maximum observed in Dalat during December 1986 -February 1987 appeared surprisingly high compared to the Chernobyl peak observed 8 months earlier [1]. Both the caesium *isotopes were detected in that winter. The activity ratio* indicates their predominant Chernobyl origin. In subsequent years the ^{137}Cs peak decreased quite rapidly, while the ^{134}Cs level was apparently lower than its detection limit.

3. ASSESSMENT OF SOME POSSIBLE MECHANISMS

Seasonal variations of airborne and deposited radionuclides were observed and discussed by many researchers. Owing to the short residence time of these species in the lower troposphere (about 1 month), any seasonal variations of their activities should reflect the seasonal change of their inputs into the atmosphere. To identify the origin of the observed winter peaks some mechanisms potentially responsible for that seasonal change will be assessed.

(a) Injection of radioactive air from the stratosphere by the stratosphere-troposphere exchange (STE) process.

For cosmogenic and nuclear weapons produced radionuclides with rather large inventory in the stratosphere, the so-called spring maximum has been well documented, especially at monitoring stations of temperate northern latitudes [2,3]. Such a maximum has been explained by the injection of radioactivity from the stratosphere due to the STE process, being intensified during spring - early summer near the polar front and subtropical jet stream of the Northern Hemisphere. In this work, peaks were observed during December and January, about 4-5 months earlier, when the STE rate was still at its annual minimum level. Moreover, the caesium isotope concentrations in air and fallout decreased very rapidly to their former levels within 1-2 months, while the rate of STE varied rather slowly and quasi-sinusoidally with time [2].

Aoyama [3] observed a STE spring maximum of Chernobyl-derived caesium isotopes in fallout in April 1987 at Tsukuba, Japan. Its intensity 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. In this work, caesium isotope peak detected in the 1986-87 winter was also predominantly of Chernobyl origin. However, it was as pronounced as the Chernobyl peak observed 8 months earlier [1]. This again indicates that STE was not responsible for the peaks observed in this work. It could be said that no spring maximum induced by STE process was detected in our monitoring study. This is true for ^{7}Be as well. Although this radionuclide was detected with much higher accuracy owing to its relatively high activity level in air (about 1 mBq/m^3), the temporal variation of ^{7}Be is so irregular that no seasonal change trend could be unambiguously identified. It is likely that STE play an insignificant role at low latitudes.

(b) Local resuspension of soil dust

Intensified local resuspension of soil dust in dry season may induce the increase of airborne and deposited caesium isotopes. As shown by data in Fig. 2c, covering the period from April 1986 to March 1988, the dust concentration in air exhibits maximum in February, slightly later (about 1-2 months) than the maximum of caesium isotopes. The role of the local resuspension of soil dust can be assessed by utilizing data on other nuclides, and comparing the activity ratios of nuclides in surface soil with those in airborne and deposited dust. The $^{137}Cs/^{134}Cs$ and $^{137}Cs/^{232}Th$ activity ratios would serve as criteria for that comparison.

The mean ^{137}Cs activity in the surface soil at Dalat in 1987 was (3.0$\pm$0.4) Bq/kg, while ^{134}Cs has not been detected in soil in Vietnam. Measurements of soil samples collected after the Chernobyl accident in an area with weathered ultrabasic rocks having very low uranium and thorium contents (less than 10^{-1} ppm) yielded an upper limit of 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 to the occurrence of ^{134}Cs in both the air and fallout in winter 1986-87 with a $^{137}Cs/^{134}Cs$ ratio of about 2.8, typical of Chernobyl-derived caesium isotopes.

The activity ratio $^{137}Cs/^{232}Th$ could not support as well the local resuspension mechanism. Thorium in atmospheric dust has been considered as being among crustally-derived elements, such as Al, Si, Sc,... contained in aluminosilicate dust particles (see, e.g.[4]). In our experiment, maximum of ^{232}Th activity concentrations in air and fallout occurred in February, at the same time as the maximum of dust concentration was observed. As an illustration, Fig. 2d shows the monthly ^{232}Th deposition rate for the period September 1986 - March 1988. It is evident that ^{232}Th can be regarded as a marker of resuspended soil particles. The average activity concentration of ^{232}Th in surface soil in Dalat is (90 ± 4) Bq/kg, yielding a ^{137}Cs/^{232}Th activity ratio of (0.033 ± 0.006). The corresponding values for the deposited dust ranged from 5.0 to 10.6 in December and January (when caesium isotope maximum occurred), and decreased to 0.08-0.6 in February and March (when the dust concentration in air was maximum). Very high enrichment of ^{137}Cs in dry deposition during December-January period again indicates a very insignificant contribution of resuspended soil dust in the observed peaks of caesium isotopes.

The temporal behaviour of ^{40}K has showed poorer correlation with that of the dust concentration in air, as was the case of ^{232}Th. Probably, there were another sources of airborne ^{40}K, e.g. products emitted into the atmosphere from forest fires, which affect a large forest area in the Central Plateau of Vietnam during the dry season. The assessment of such a potential source of ^{40}K needs further studies.

4. THE ROLE OF WINTER MONSOON IN TRANSPORTING HIGH ACTIVITY AIR FROM TEMPERATE NORTHERN LATITUDES

The peaks of caesium isotopes were observed in December and January, when the monthly average temperature was lowest and dry fallout dominated (Fig. 2a,b). This suggests that cold continental polar (cPk) air masses move equatorwards during the most active period of winter tropical monsoons, to supply higher caesium isotope concentrations. Such a circulation of cPk air from the NE often reaches very low latitudes during December and January, when the intertropical convergence zone is aligned in the Southern Hemisphere up to 10^{o}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^{o}N). Concerning the Chernobyl debris, a modelling prediction [5] calculated for the beginning of May 1986, showed a marked radioactive plume dispersed around this latitude band over the Asian continent with elevated activity concentration over the region of the Asiatic high- pressure centre. As a consequence, the soil in that region could be enriched with caesium isotopes and the resuspension of soil particles could, to a much extent, maintain a high radioactivity level of the surface air there. Furthermore, in the upper atmosphere, Chernobyl isotopes should also have maximum concentration 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 stratosphere was another source contributing to the high level of caesium isotope concentrations in the lower troposphere of that region. Measurements from the post-Chernobyl period are so scarce that it is impossible to establish a latitudinal distribution of ^{137}Cs concentration in air for the Far East Asia. However, a few data available for the years 1986-87 [1] appear to confirm a considerable difference between concentrations at Dalat and those at higher latitude locations of the region. Thus, the increase of 0.3 Bq/m^3 at Dalat in December 1986 (Fig.1) can be attributed to the 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 has been well demonstrated by Dmitrieva et al. [6] in a study of atmospheric -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. 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, i.e. its descending vertical motion [7] can be identified from the above obtained 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 for airborne caesium isotopes are in the range of 5-15 cm/s; these values are much higher than those measured in the following months after the Chernobyl accident in Europe, North America and China [1] (about 0.1 cm/s), and indicate descending motion of cold air during December and January at Dalat. Meanwhile, quite different values of dry fallout velocities, ranging in the interval of 0.4-1 cm/s, were obtained for the two naturally occurring isotopes ^{232}Th and ^{40}K during the periods of caesium isotope maximum. This again indicates different sources of these radioactive species. While particles bearing naturally-occurring radionuclides are being continuously generated in the surface air from the soil, air masses with elevated caesium isotope concentration intrude from subtropical latitudes and descend to the surface layer only several times a month in December and January during the propagation and development of anticyclones.

5. *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 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 the intrusion of more radioactive air from higher latitudes. The ^{137}Cs concentrations yielded quite high estimates of dry fallout velocity ascribed to the descent of air behind the cold front in the ridges of the associated anticyclones.

The obtained results clearly demonstrate that air pollutants from mid-latitude Asian industrialized regions would be transported far into the equatorial zone in the period of active winter monsoons. Furthermore, in the weather conditions affected by winter monsoons, any air pollution caused by local industries would be horizontally, rather than vertically, dispersed owing to the descending motion of dry air behind the cold front.

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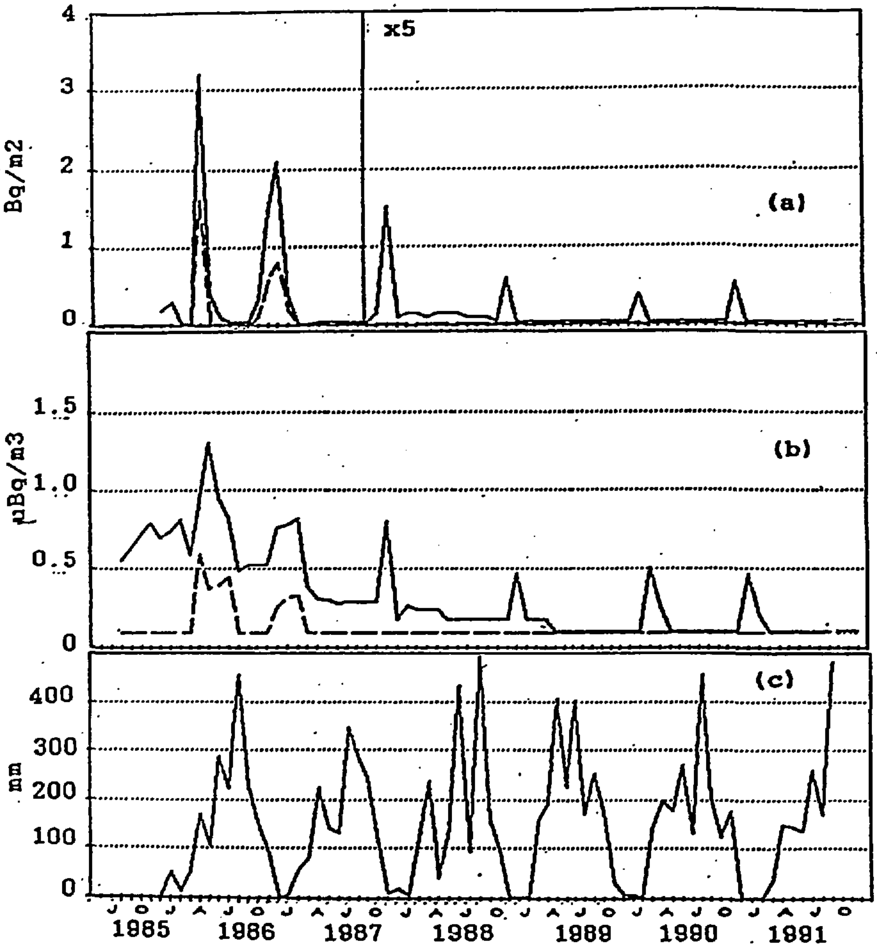
CAPTIONS TO FIGURES (IAEA-SM-329/13)

Fig. 1. Monthly variations of caesium isotope concentrations in fallout and air in Dalat, 1986-91.

(a) ^{137}Cs (———), ^{134}Cs (-----) in fallout
(b) ^{137}Cs (———), ^{134}Cs (-----) in air
(c) Precipitation

Fig. 2. Monthly variation of precipitation (a), average air temperature (b), dust concentration in surface air (c) and deposition rate of ^{232}Th (d). The arrows indicate the caesium isotope maximum.



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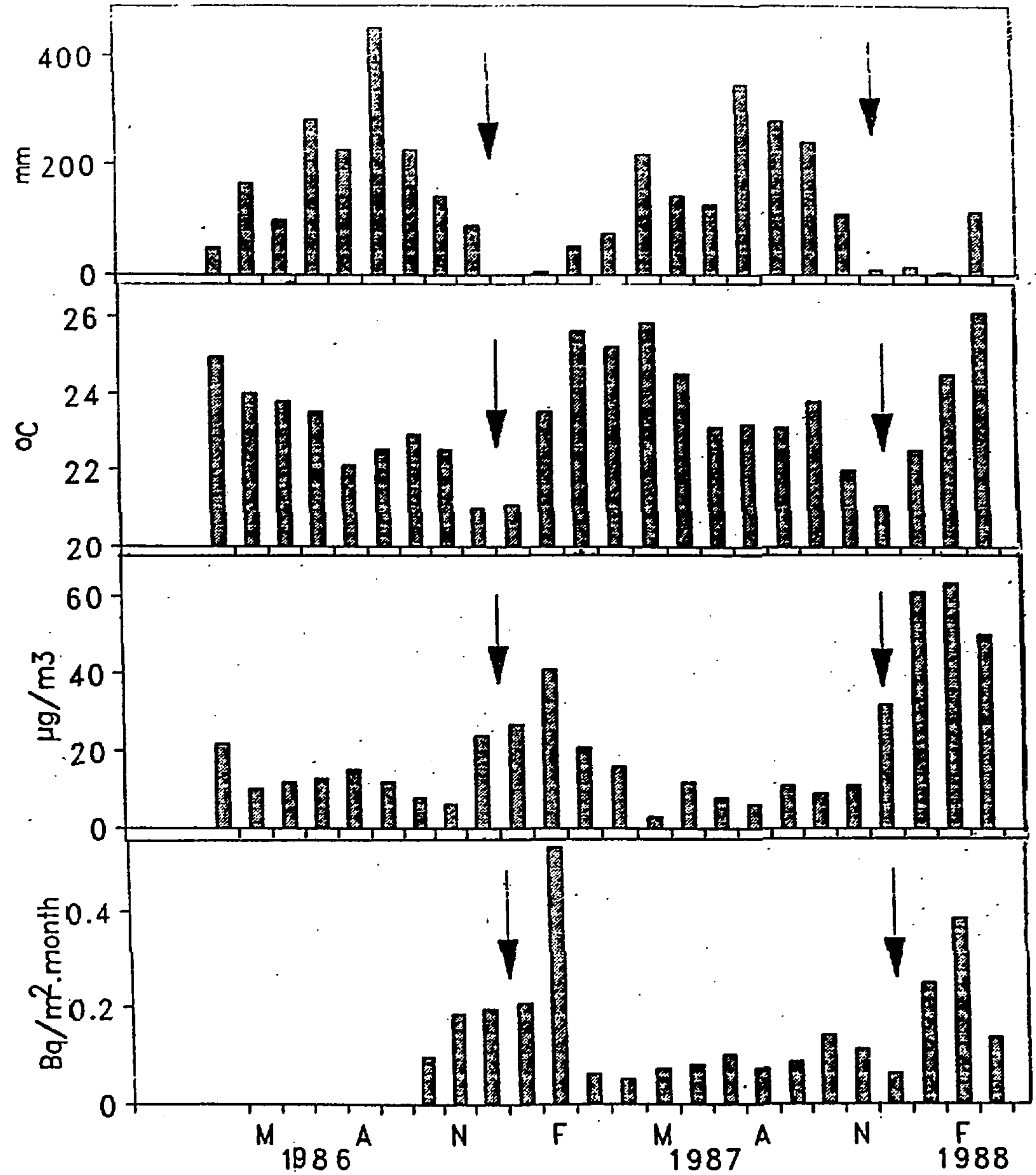

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