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Use of stable and radioactive traces for airmass characterisation and application to monsoon studies

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ABSTRACT. Several tracers, natural and man made, can be usefully employed for studying the source region and other characteristics of airmasses constituting the monsoon circulation. These include natural radioactive materials like radon, thoron and their daughter products, man-made radioisotopes like short-lived and long-lived fission products from nuclear explosions, trace elements present in the airmasses, synthetic compounds and devices like high pressure balloons, etc. These tracers have the advantage that integrated effects can be studied over long periods and distances and regions of airmass mixing can be clearly identified. Use has been made of some of these tracers (radon daughters) during Monex-1973 expedition over the Arabian Sea and extensive measurements have also been carried out on fission product levels at a number of countrywide stations for studying monsoon circulation. The results of these studies based on the measurements made at this laboratory and elsewhere have been given to illustrate the role of such tracers. The paper also describes some of the experiments proposed to be carried out during the forthcoming Monex projects, which are expected to give useful information about the monsoon.

1. Introduction

Tracers have the advantage that they can be followed for long distances along with the carriers without losing their identity. This enables the paths of the carrier to be inferred under dynamic situations, where the carrier identity is likely to become uncertain. Such tracers are extremely useful in following the movement and behaviour of the monsoon currents. The source region of the monsoon, its genesis, and areas of interaction with other airmasses are of interest in understanding its behaviour and predicting its course and intensity. The trajectories of the monsoon airmasses are also of interest to atomic energy as a system dispersing pollutants on a regional and interhemispheric scale. A brief account has been given of the tracers which have been used or which could be used to elucidate the specific characteristics of the summer monsoon currents. Some of these experiments will be carried out on a limited scale in the Monsoon-77 project and in a bigger way during Monex-79 project. The possibilities of others will be discussed.

2. Review of the tracers

A brief summary of the tracers relevant to the subject is given in Table 1. The desirable properties of such tracers are, the availability and mechanisms of production and introduction into the atmosphere, methods and possibilities of tracing its path, the detection and concentration estimation, the life time of the material, its capacity to follow faithfully the path of the carrier, etc. In addition, the tracers should be non-toxic and should have a low background, *i.e.*, its natural concentration should be sufficiently low to enable the levels to be assigned to the experiment. These properties and the use and future possibilities of several of the tracers will be briefly described below.

2.1. *Fission product particulate activity from nuclear tests*

These have the advantage that their source region and time of origin are known and detection fairly easy with the available instrumentation. Several laboratories round the globe document the

TABLE 1

Tracers and devices in monsoon studies

Device	Source	Detection method	Utility	Remarks
Short-lived fission products Ba^{140} and Zr^{95}	Nuclear tests	Nuclear counting and spectroscopy	Determining the source of the monsoon etc	Not an ideal conservative tracer due to washout of the particulates by rainfall (Rangarajan and Gopalakrishnan 1975-1976)
Rare gas fission products Xe^{133} and Kr^{85}	Nuclear tests	Cryogenic techniques	Determining the source of the monsoon etc	An ideal conservative tracer not subject to washout, etc—Detection difficult (Ehhalt *et al.* 1963; Kunz and Paperello 1976)
Radon and daughter activities	Continental land mass	Nuclear counting and cryogenic techniques	Airmass mixing and presence of continental air	Radon not subject to washout and supports daughter products. Hence an ideal tracer (Rangarajan *et al.* 1974 & 1976)
Sea salt/air dust	Sea and land	Chemical and gravimetric	Continental and maritime airmass distinction	Not a conservative tracer. Detection easy (Wilkness *et al.* 1974)
Devices and compounds	Man-made	Satellite tracking, mass spectrometer etc.	Airmass trajectory determination	Ideal tracer if done on a sufficient scale (Cowan *et al.* 1975; Sommeria and Cadet 1974)
Trace elements in aerosols	Continental land mass	Neutron activation	Airmass identification and source determination	Not a conservative tracer. Useful as low concentration of trace elements, can be detected (Internat. Symp., France 1973)

levels of fission products and these data are available for tracing the path of the debris although the monitoring stations may not be of sufficient numbers always. The short lived fission products Ba^{140} and Zr^{95} from the French tests of Polynesia (22°S) have been used in determining the source region of the monsoon (Rangarajan *et al.* 1975b, 1976). Fig. 1 gives the levels of Zr^{95} in surface air at Bombay, Thumba and other stations in India from several other locations round the globe following the French tests of 1966-1972, carried out in the summer monsoon period of the respective years. The figures clearly shows much higher activity on the west coast of India compared to other locations. The travel time as estimated from shortlived Ba^{140} is considerably less to the west coast, in comparison to other locations in the northern hemisphere — even the ones near the equatorial regions (Rangarajan *et al.* 1975 b). This argues strongly in favour of the southern hemispheric source for the monsoon which transfers radioactive debris across the equator in a massive way, presumably by the Somali jet stream. Estimation of the fraction of southern hemispheric air in the monsoon current is not easy from these data as particulate activity is subject to washout, but if the value just south of the equator is taken from the curve of Fig. 1 (*e.g.*, Guayaquil), the fraction reaching the western coast is certainly very high.

2.2. *Rare gas fission products*

The rare gases have the advantage that they are true conservative tracers since they are not subject to washout and chemical reactions. The fission products Xe^{133} and Kr^{85} have sufficiently high yields and half lives to be of use as a tracer. However, Kr^{85} has a long half-life of about ten

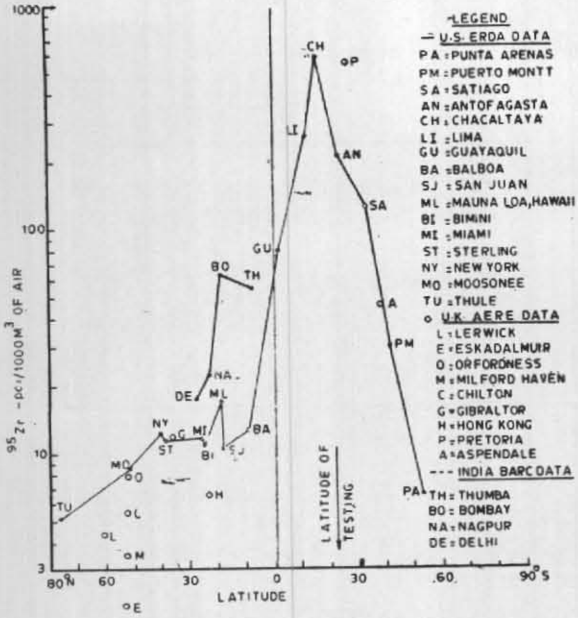

Fig. 1. Latitudinal variation of Z_r^{95} activity in surface air followin g the French tests of South Pacific (Average for the months of Aug 1966, Jul, Aug 1967, Aug, Sep 1968, Aug and Sep 1970)

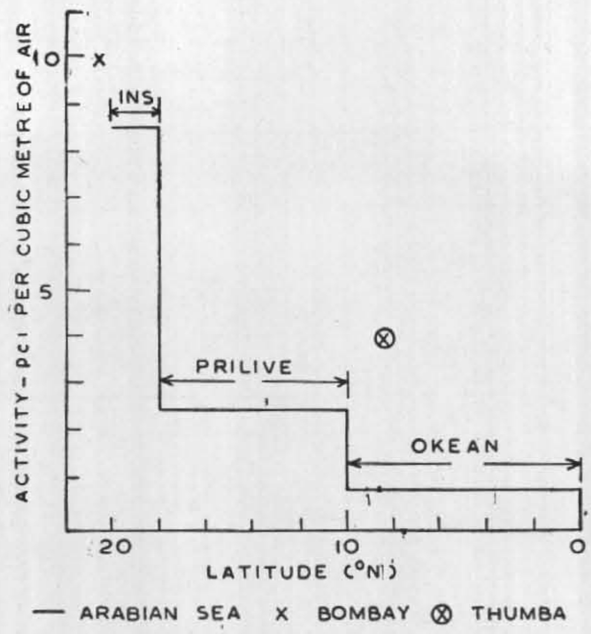

Fig. 2. Average radon daughters activities in the Arabian Sea as measured by Indian Navy and USSR ships during Monex-73 and at Bombay and Thumba

years and this makes it of limited applicability in view of its large accumulation in the atmosphere, from previous test (The same is the case with long-lived particulate fission products like Cs^{137}, Sr^{90} etc, although in this case the levels are reduced more rapidly due to ıain scavenging). Thus, Xe^{133} appears suitable with its half-life of about 5 days and fission yield of nearly 5 per cent (Ehhalt *et al.* 1963). It has already been detected from releases in nuclear reactors and tests (Ehhalt *et al.* 1963, Kunz *et al.* 1976). However their detection requires rather elaborate cryogenic techniques. Kr^{85} has been used in intermeridional diffusion studies (Pannetier 1970).

2.3. *Radon and daughters activities*

The activities of radon with a half-life of 3·8 days and its daughters of half-lives ranging from 3 to 30 minutes are ideal for detection of continental airmasses. The emanation of radon is mainly from the soil, the emanation over the ocean being negligibly small. The levels of radon in a true maritime air is less than about one pci/m^3 and any significant departure from the above value would indicate intrusion of continental air. Fig. 2 gives the radon daughter levels over the Arabian Sea at various latitudes from the data collected during Monex-73 (Rangarajan *et al.* 1976) from Indian Navy and USSR vessels ***Priliv*** and ***Okean.*** The latitudinal averaging is done in the figure as there is no significant difference along the latitudes except very near the coast (Rangarajan *et al.* 1974). The levels upto about 18°N indicate a general maritime origin of the airmass, there being some intrusion of continental air only above this latitude. This continential input is due to the presence of cyclones and depressions near the Gujarat coast during the period and the "seasonal low" over west Pakistan. This cyclonic circulation appears to be effective only upto about 20°N. The above findings of the presence of southerly maritime air in the main monsoon current is in line with the data from nuclear tests (Rangarajan *et al.* 1976).

The above conclusions will be confirmed with the more elaborate equipment in the monsoon-77 project.

In addition to radon levels, the ratio of radon to its long lived daughter ^{210}Pb is also an index of the continentality of the airmasses (Rangarajan *et al.* 1975b). This is because the ratio is small in maritime air, due to lack of input of short-lived radon.

TABLE 2*

Concentrations of some trace elements at Bombay during the monsoon months of 1972

(Nanograms/cubic metre of air)

Month	Na**	Se	Cr	Fe**	Co	Zn*	Sb	La	Ce
June	—	1·4	—	—	27·7	0·74	—	—	—
July	4·5	—	315	2·5	—	1·7	31·4	16·7	—
August	—	—	55·2	2·3	2·1	—	—	23·3	19·6
September	1·2	—	—	2·5	—	0·9	—	0·8	34·4

*From Kamath (1977) **Milligrams/cubic metre

The maritime values are around 100-200 while the continental values range from 5000-10,000 (Rangarajan *et al.* 1975b).

Thoron (half-life of one minute) and daughter Th-B (half-life 10·6 hr) are not of much use in view of their short half-life. Still it may be possible to detect Th-B if the continental component is significantly large. Even at 20°N no Th-B could be detected during the Monex-73 experiments. With more powerful blowers and sensitive counting equipment, Th-B can perhaps be followed for longer distances from the continents. In such cases it will be a sensitive indicator of the continental component.

2.4. *Sea salt and dust content*

The sea salt content and the dust content of the airmasses are of use in identifying airmasses from the continent in the marine atmosphere. It is known that continental dust can travel long distances into the sea in specific wind systems (Internat. Symp., France 1973). The ratios of sea salt to air dust is high in pure, maritime air, whereas in continental air it will be significantly lower (Wilkness *et al.* 1973). Hence, these, along with radon, will be useful in airmass identification. Measurements of the concentrations of air dust and sea salt are easy and no elaborate collection or measurement equipment is required.

2.5. *Aerosols*

The composition of the aerosols is an useful index of the source of the airmass carrying them (Internat. Symp., France 1973). The trace element content varies with the source region of the aerosols and this provides a method of identifying the source of the aerosols. The ratios of the concentration of various trace elements in the aerosols is an useful index for such studies. Detection is by neutron activation in reactors of the sample dust and counting is done in gamma spectrometers. Table 2 lists some of the trace elements detected in surface at Bombay (Kamath 1977) and their concentrations. Other trace elements (*e.g.*, Sr. V, Ba, Mn, Ni, Ga, Ca. Zn, Sn etc) can be included in the analysis by short term and long term irradiation and counting at various intervals. A single sample can be subject to multi-element analysis upto ppm levels. Thus this technique provides an useful tool in detecting the sources of the air present in the Indian Ocean.

2.6. *Devices and compounds*

The devices and compounds have the advantage that they can be introduced at times and places of interest and their trajectories followed. The devices include high pressure balloons moving at constant density levels and tracked by satellites (Sommeria and Cadet 1974). Such devices have been used by the above investigators in tracing the movement of airmassess across the equator. However these experiments are costly requiring elaborate launching and tracking devices and sufficient number should be launched to obtain representative tracks. Problems of maintaining the balloons at constant levels and for sufficiently long distances have to be solved.

In addition, special compounds such as Methane 21 (heavy methane) artificially produced by tagging with rare isotopes (*e.g.*, 13 CD_4) can be released into the atmosphere at selected points and their concentrations measured at required areas (Cowan *et al.* 1975). The instrumentation required for preparation of the compounds are complicated and measurements require sensitive mass spectrometers. The choice of the compound is critical as its natural background in the atmosphere should be very low. Subject to these criteria the compounds can serve a useful purpose.

3. Monsoon-77 and Monex-79 projects

The Monsoon-77 and the more elaborate Monex-79 projects provide an opportunity for carrying out several experiments with the devices listed above. Simultaneous use of some of the above tracers with relevant meteorological data will give a clear picture of the dynamics of the monsoon. It is, however, important to plan the experiment in a coordinated manner so that the experiments complement each other.

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COMMENTS

JAGDISH BAHADUR : Thank you, Mr. Chairman. I have some associated comments to offer. As the learned speaker has pointed out the difficulties of mass spectrometric analysis of stable isotopes of water molecule, I shall like to comment briefly on the use of these techniques for study of precipitation :

1. It has been recognised since early 50's that stable isotopes of water molecule can be potentially used for hydrometeorological investigations. Friedman in USA, Dansgaard in Denmark and Enkssen in Sweden have discussed the physical processes in details.

2. There exists a IAEA-WMO global network for environmental isotopic (^3H, ^2H and ^{18}O) concentrations in precipitation since 1961. Under this survey, more than 100 meteorological stations all over the world are participating in this programme. The main objective of this global endeavour is to provide basic information to all scientists who can use these data for solving their hydrological problems. Another scientific objective was provide information on ^3H input function at different locations and on some characteristics of moisture circulation patterns and mechanisms of the global and local water.

3. Data for stable isotopes from IAEA-Technical Reports was analysed for Delhi precipitation and the following aspects are noteworthy :

(*a*) Least square fits for $\delta^{18}O$ and δD show that the annual waters demonstrate the effect of evaporation, *i.e.*, the enrichment of oxygen isotope as compared to that of hydrogen in the precipitation. The effect is markedly low in monsoon precipitation where the rainfall follows the Graig line ($\delta D = 8\delta^{18}O + 10$).

(*b*) The δD—$\delta^{18}O$ diagram show a lot of scatter for winter precipitation and could be due to 'amount effect' as the raindrops during their descent have to traverse a longer path in dry atmospheric conditions. Part contribution to this scatter could be due to mixing of airmasses from different origins such as westerly disturbances NE and SE monsoons.

(*c*) Pre-monsoon showers show enrichment in δ-values and this could be due to presence of evaporated water from Arabian Sea.

(*d*) There exists an altitude effect for study of orographic phenomenon for the Himalayan Environment.

4. Sampling of snow and glacier melt waters during Jul-Oct have shown that the isotopic concentation indicates the result of predominant precipitation in higher altitudes of Himalayas during summer months. This may be due to moisture releases from retreating westerly disturbances.

The above data show that an inventory of stable isotopic concentration of water molecule in precipitation (both snow and rain) will be of immense help in sorting out some of the questions raised during this symposium, *e.g.*, origin of water vapour, amount of mixing of maritime and continental airmasses, evaporation effects from sea and inland stations and also resurrection of paleoclimatology from sampling of in cores in higher altitudes of glaciated land thereby helping us to know the water balance characteristics of these mountain catchments.