SHORTER CONTRIBUTION

Measurement of radioactive xenon in the atmosphere

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In the course of a previous investigation (D. EHHALT *et al.*, 1963) we tried to measure the Xe-133 (5.27 days half-life, 347 keV β-emitter) released to the atmosphere by bomb tests during 1961. At that time we could only give an upper limit for this activity. The raw samples at our disposal consist of krypton, xenon, argon and oxygen. The considerable activity due to Kr-85 ($\approx$500 dpm Kr-85/millimole Kr in 1965) makes it difficult to assess small amounts of Xe-133 in this gas mixture. A further complication arises because practically all the radon contained in the atmospheric air (in the order of 10^5 dpm Rn/millimole Xe) is usually found in this air fraction. The similar half-lifes of Xe-133 and radon make it virtually impossible to assess even considerable amounts of Xe-133 (see figure 4 of ref. 1).

We have now developed a gas chromatographic method (J. SCHÖLCH & W. STICH, 1965) for the separation of xenon from krypton and radon. (Oxygen and argon are separated from the mixture by fractional distillation (D. EHHALT *et al.*, 1963)). An activated charcoal column (120 cm length, 0.6 cm diameter, working temperature -80°C) is suitable for the isolation of about 3 millimoles of xenon containing only a small amount of krypton (0.2–0.5 %), and undetectable amounts of radon. The radon content can be checked very sensitively by α-counting. The residual krypton content of the xenon is checked by mass spectrometry.

The assay of the Xe-133 β-radiation is made in low level proportional counters normally used for C-14 dating in our laboratory. These counters have large volumes (several litres, counting background less than 10 cpm), and are brought to working pressure by addition of inactive Co_2. The xenon thus comprises only about 1 % of the counter filling. This means that the detection limit (3σ) of 2 dmp/millimole Xe or 10^{-2} dmp/m^3 of air (STP) reached in this way could easily be lowered considerably by using a specially made small counter with lower background. Another improvement easy to achieve would be to to use a larger separation column. However, as will be seen below, the sensitivity of the present method is fully adequate for a survey of Xe-133 in European air. If one assumes that the fission products are equally distributed over the entire atmosphere, then the detection limit mentioned above (10^{-2} dpm Xe-133/m^3 of air (STP)) corresponds to the fission yield from 2 gm of U-235 (0.04 Kilotons TNT equivalent) (see table 1, ref. 1). Such equal distribution will of course never occur during the short half-life. Therefore in practice much smaller amounts can be detected if the source is not too far away from the sampling site. Immediately after the fission process the activity due to Xe-133 is by about four orders of magnitude higher and thus much easier to detect than the Kr-85, since the half-life of Xe-133 is much shorter and the fission yield higher. Moreover there is a Kr-85 "background" in the atmosphere which corresponds to about 100 tons of fissioned U-235 and makes freshly produced Kr-85 difficult to detect, (D. EHHALT *et al.*, 1964).

Fig. 1 shows the Xe-133 measurements obtained to the time of writing. The concentrations are corrected to the date of sampling. It is evident that there is a considerable "noise level" even if we exclude the concentrations of June, 1965, which are probably due to the Chinese bomb test of May 14, 1965. If we deduce from the data given in Fig. 1 an average atmospheric concentration at the sampling site

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of about 70 dpm/millimole Xe, an average daily addition of about 10 dpm Xe-133 would be necessary to compensate the radioactive decay and to keep the average concentration steady. This daily addition corresponds to the fission release of about 10 gm U-235/day if the fission products are equally distributed over the entire atmosphere. This is not of course the case, and the necessary amount of Xe-133 immediately released from freshly fissioned material will depend strongly on the distances between the source or sources and the sampling site, but will probably be less than the quoted 10 gm U-235 per day, if the sources lie roughly at the same geographical latitude as the sampling site. If we exclude the possibility that the sources are by chance very near to the sampling site in Western Germany, but assume that they lie in approximately the same latitudinal belt, we shall come to a reasonable estimate if we assume that the xenon released at ground level will be distributed on the average over only 1/10 of the entire atmosphere. This means that about 1/10 of the 10 gm U-235 per day calculated above would be sufficient to account for the average concentration of Xe-133 found at the sampling site. It is interesting to compare this amount of Xe-133 released to the atmosphere with the amount of Kr-85 which is responsible for the steadily rising concentration of that isotope. From our measurements of the Kr-85 activity (D. EHHALT *et al.*, 1963 and 1964) we find a yearly increase of about 50 dpm/millimole Kr corresponding to an amount of fissioned U-235 of 10 ton/year or 200 kg/week. This is a factor about 3×10^4 higher than we calculated from the Xe-133 activity. This means

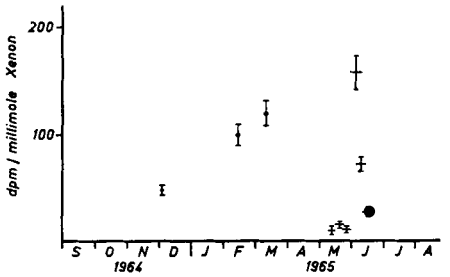

FIG. 1. Concentration of atmospheric Xe-133 from Decembre 1964 to June 1965. The results of the measurements after the Chinese bomb test of May 14, 1965, are also shown in fig. 2.

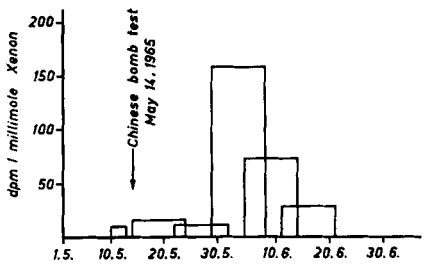

FIG. 2. Concentration of Xe-133 in the atmosphere during and after the Chinese bomb test of May 14, 1965. In order to have an uninterrupted sequence of measurements, the sampling periods were extended to 10 days each.

that the prompt release of fission-produced noble gases is on the average less than about 3×10^{-5} of the total amount eventually released to the atmosphere. The bulk of these fission produced gases must be more than 2 months old before its release to the atmosphere, otherwise more Xe-133 would be found.

After the Chinese bomb test of May 14, 1965 a series of measurements of Xe-133 was made. In order to have an uninterrupted sequence of activity measurements the time of sampling was extended to 10 days each. The results for this period alone are shown in fig. 2. At the time of the test the Xe-133 level was quite low. Three weeks later a marked rise of the activity was found which might be attributed to the Chinese test. However, as can be seen from fig. 1, peaks of activity with comparable height were found in the time before this test. Therefore we can use the observed activities only to deduce an upper limit for the release of fissioned material to the troposphere due to the Chinese bomb test. Extrapolation back to the date of the explosion gives an activity of 2300 dpm/millimole xenon. If we assume the xenon to be equally distributed over the Northern Hemisphere at the time of sampling, this corresponds to an amount of fissioned U-235 of 1.2 kg or 25 kilotons TNT equivalent. Since the decrease of the Xe-133 activity can exclusively be attributed to radioactive decay, (see fig. 2), it is evident that the Xe-133 was already distributed homogeneously over a very large area, and the 25 kilotons calculated therefore should not be a serious overestimate.

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REFERENCES

D. EHHALT, K. O. MÜNNICH, W. ROETHER, J. SCHÖLCH and W. STICH, Artificially produced radioactive noble gases in the atmosphere. *J. Geophys. Res.*, **68**, 3817 (1963).

D. EHHALT, K. O. MÜNNICH, W. ROETHER, J. SCHÖLCH and W. STICH, Krypton 85 in the atmosphere. Paper A, Conf. 28/P/541, presented at the Third International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964.

J. SCHÖLCH and W. STICH, Measurement of radioactive xenon in the atmosphere. Paper submitted to the conference on "Low level radioactive measurements — limitations and new techniques", Imperial College, London, 5–6 July, 1965.