Reprint from

"BEHAVIOUR OF TRITIUM IN THE ENVIRONMENT"

Internatl. Conf. Behaviour of Tritium in the Environment, San Francisco, CA, U.S.A., October 17 - 21, 1978. http://www-naweb.iaea.org/napc/ih/documents/IAEA%20Monographs/STI%20498%20Behaviour%20of%20Tritium%20in%20the%20Environment%201979.pdf

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INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1979

METHODOLOGY FOR HAZARD ASSESSMENT OF ENVIRONMENTAL TRITIUM

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Abstract

METHODOLOGY FOR HAZARD ASSESSMENT OF ENVIRONMENTAL TRITIUM.

The assumption is made that the ecological system immediately follows the variations of the tritium content in atmospheric water vapour. Tritium concentrations in the ecological subsystems (plant, animal and human) are calculated using balance equations for tritium intake and elimination. Due to consumption of non-contaminated food and water the tritium concentrations in animals and man are reduced by about the ratio of contaminated to non-contaminated food. The main exposure pathways are shown for several consumption habits. Since West Berlin imports a considerable amount of its food (more than 30%) from Niedersachsen (Lower Saxony), the proposed area for the West German nuclear fuel reprocessing plant, the exposure of the population of this city is used as an example.

1. INTRODUCTION

Tritium dose-rate calculations have to specify the concentration c_h (unit:nCi/ltr of body water, both as tissue water and non-aqueous tritium expressed as water) of tritium in human tissue. The dose rate H (mrem/a) is then related to c_h by the well-known relationship

$$H = 0.1\ Q\ c_h \tag{1}$$

Q is the quality factor for the beta radiation from tritium. For exposure to tritiated water vapour, Anspaugh and co-workers [1] have developed a dynamic model describing the tritium pathways among several compartments. Their model includes transfer rates and elimination rates (time constants).

In this study this dynamic method is reduced to a dynamic equilibrium approach which consequently does not need the time constants mentioned. It takes into account the mixing of contaminated and non-contaminated food (including water intake and water vapour absorption by the skin).

2. SYSTEM OF PATHWAYS

Figure 1 presents the well-known system of pathways of hydrogen. Each compartment may have to be divided into several subcompartments, i.e. may have several hydrogen pools with separate hydrogen pathways connected. The compartment system is assumed to be a linear time-invariant system, i.e.

(a) the superposition principle holds (e.g. doubling an input will double the pool content);

(b) the coefficients describing the amounts of input and output are independent of seasonal changes or weather influences.

The system has a tritium source in the air compartment. The tritium follows the non-radioactive hydrogen (protium) on all pathways into all compartments, and no process will enrich tritium with respect to protium. The behaviour of the tritium is then described on the basis of the following equations:

$$\begin{pmatrix} \dot{c}_p(t) \\ \dot{c}_a(t) \\ \dot{c}_h(t) \end{pmatrix} = \begin{pmatrix} -\lambda_p & 0 & 0 \\ \alpha_a/M_a & -\lambda_a & 0 \\ \alpha_h/M_h & \beta_h/M_h & -\lambda_h \end{pmatrix} \begin{pmatrix} c_p(t) \\ c_a(t) \\ c_h(t) \end{pmatrix} + \begin{pmatrix} R/M_p \\ (\beta_a + 2\theta_a f)/M_a \\ (\gamma_h + 2\theta_h f)/M_h \end{pmatrix} c_w(t) \tag{2}$$

$$d/dt\,\vec{c}(t) = \underline{A}\,\vec{c}(t) + \underline{B}\,\vec{c}_w(t)$$

where

c = concentration of tritium in the water of compartment i (nCi/ltr)

R = rate of rainwater consumption by plant (litre/a)

I = rate of consumption of irrigation water (litre/a)

M = mass of body water of animal (subscript a) or man (h) (litre)

f = absolute humidity of air = 5×10^{-3} litre/m^3

Greek letters mean intake rates (kg/a or kg/d) into the compartments denoted as subscripts of the letters. For further explanation see Fig.1. The elements λ_i in the diagonal of the matrix $\underline{A}$ give the time behaviour of the free system, i.e. the time constants of the system. Vector $\underline{B}$ specifies the coupling of the driving force $c_w(t)$ to the system of food pathways.

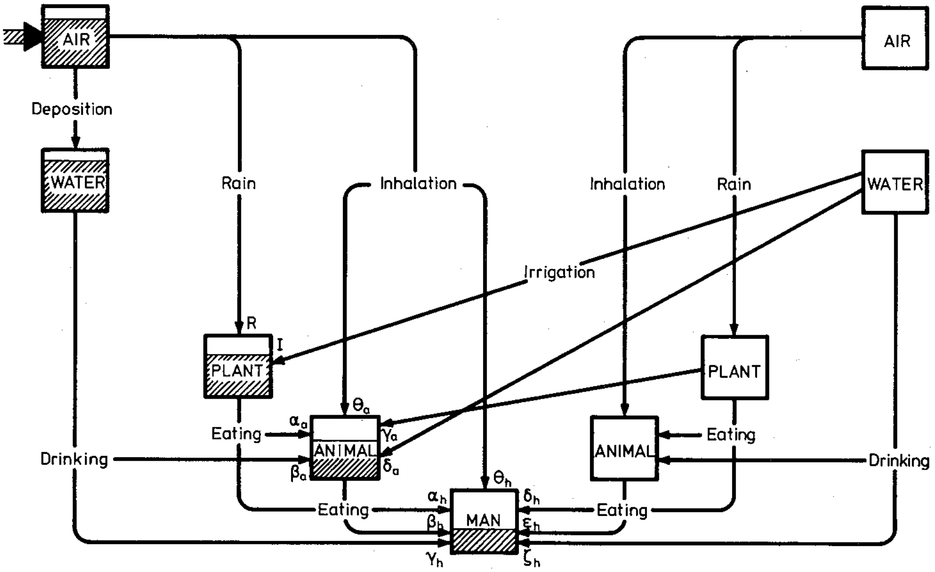

FIG.1. Compartment system describing the flow of hydrogen from the source (air compartment) to man.

Since tritium concentrations in air and rain vary through varying emissions and weather, it is important to know the response of the system $\underline{A}$ to periodic disturbances. The response of a single unconnected compartment i to a force $c_w(t) = \sin(\omega t)$ can be described by a transfer function g, which relates the concentration of the compartment i to the concentration in the environment:

$$c_i(t) = b_i /g_i(\omega)/\sin(\omega t - \phi) \tag{3}$$

$$/g_i(\omega)/ = 1/\sqrt{\lambda_i^2 + \omega^2} = \begin{cases} 1 \\ 0.7 \\ 0.1 \end{cases} \quad \text{for } \omega/\lambda_i = \begin{cases} 0 \\ 1 \\ 10 \end{cases}$$

$$\mathrm{tg}\phi = \omega/\lambda_i \tag{4}$$

One can see that if tritium concentrations in the air change slowly ($\omega < 10\lambda_i$), the compartment concentrations follow those changes closely. The amplitudes of the system remain of the same order of magnitude for $0 < \omega < 10\lambda_i$.

Anspaugh and co-workers [1, 2] report time constants $1/\lambda_p$ for the system: tritiated water vapour-plant-soil ranging from 1 hour for tritium in tissue water to several days for tissue-bound tritium, the tritium reaching the plants both from the air and the soil. The impulse responses measured for impulses of height c_w^0 and $\frac{1}{2}$ to 1 hour duration were

$$c_p(t = 0) \approx /g_p(\omega)/c_w^0 \tag{5}$$

for $\omega = 1 \ldots 2\ h^{-1}$, t = post-irradiation time and with

$$0.2 < /g_p(\omega)/ < 0.8 \tag{6}$$

which is not in contradiction with their time constants (see Ref.(3)).

Rosinski and Winter [3], and Lee and co-workers [4] recorded tritium concentrations in air, soil and in tissue water of several plants in the vicinity of the Karlsruhe Nuclear Research Center during seasons of late summer to winter. Although a thorough Fourier analysis of their measurements has not been performed, their data seem to point towards $/g_p(\omega)/$-values above 0.5 for very small ω, $1\ d < 1/\omega < 2\ m$.

3. STATE OF DYNAMIC EQUILIBRIUM

In this study we will take $/g_p(\omega)/ = 1$, which will be undoubtedly conservative depending upon how far the system is from stationariness, i.e. from the case $\omega \ll \lambda_i$ ('state of dynamic equilibrium').

There is another overestimation in this work, namely the omission of the subdivision of the plant and animal compartments. Although some transfer functions $g(\omega)$ of this system of subcompartments are small, their contribution to the food, however, is included with unreduced strength in our calculations, i.e. it is supposed that $g = 1$ for all subcompartments.

The concentrations c_i will be calculated for a stationary system. That means that the equation of state (2) will reduce to a balance equation of the type:

$$\text{rate of intake of tritium} = \text{rate of elimination of tritium} \tag{7}$$

For plants the following relation is used (M_p disappears for the stationary system):

$$R\ c_w = (R + I)\ c_p \tag{8}$$

c_p is smaller than c_w whenever a non-contaminated source of water is used, because the plant is not supposed to discern between tritium and protium.

TABLE I. POSSIBLE MAXIMUM CONTRIBUTIONS OF THE FOOD PATHWAYS TO THE DOSE RATE

Numbers do not add up to 1 because of rounding errors

Consumption habit of		Plant	Animal	Water	Inhalation and skin absorption
Child [5]	Maximum	0.37	0.25	0.34	0.05
	Average	0.27	0.28	0.35	0.09
Teenager [5]	Maximum	0.39	0.27	0.30	0.04
	Average	0.29	0.31	0.31	0.08
Adult [5]	Maximum	0.32	0.23	0.41	0.04
	Average	0.23	0.24	0.45	0.08
Anspaugh [1]		0.13	0.18	bal.	0.06
2.SSVO [6]		0.22	0.18	bal.	0.08

TABLE II. CONSUMPTION RATES FOR MAN (AFTER REFS [5, 7])

Meat and milk are calculated together; the values for West Berlin are averages over the whole population of 2 million inhabitants

	Maximum (average) consumption rates (kg/a)			
	$\alpha_h+\delta_h$	$\beta_h+\epsilon_h$	$\gamma_h+\zeta_h$	$2\theta_h f$
Child	550 (200)	370 (210)	510 (260)	70 (70
Teenager	670 (240)	470 (260)	510 (260)	70 (70)
Adult	580 (190)	420 (200)	730 (370)	70 (70)
West Berlin	(225)	(127)	(175)	70

TABLE III. CONSUMPTION RATES (kg/d) FOR ANIMALS (AFTER REF. [5])

Only beef/cattle considered

$\alpha_a + \gamma_a$	$\beta_a + \delta_a$	$2\theta_a f$
50	60	5

The balance equations for the other compartments will be obtained in the same way and may be interpreted similarly.

$$c_a/c_w = (\alpha_a(c_p/c_w) + \beta_a + 2\theta_a f)/S_a \tag{9}$$

$$c_h/c_w = (\alpha_h(c_p/c_w) + \beta_h(c_a/c_w) + \gamma_h + 2\theta_h f)/S_h \tag{10}$$

The total intake of food (including water) is called S_i here. It corresponds to R + I in the case of plants:

$$S_a = \alpha_a + \beta_a + 2\theta_a + \gamma_a + \delta_a \tag{11}$$

$$S_h = \alpha_h + \beta_h + \gamma_h + 2\theta_h + \delta_h + \epsilon_h + \zeta_h \tag{12}$$

Since c_h is related to the dose rate via Eq.(1), the dose rate consists of four contributions: each term in the sum (Eq.(10)) gives the contribution of one food pathway.

The amount of tritium entering via a pathway depends on the degree of contamination c_k of the species k of food considered, and on the consumption habits, α_i/S_i, β_i/S_i, γ_i/S_i, etc.

4. RESULTS

The worst case is the complete contamination of the food chain, i.e. when only contaminated food enters the chain ($c_i/c_w = 1$ for all compartments i). The four columns of Table I give the fraction of the dose rate for each of the four food pathways. Six different consumption habits are considered: consumption rates for children, teenagers and adults, values both for maximum and average intake used, as they are specified for 10CFR Part 50, App. I [5] (see Tables II and III). Since $c_i/c_w = 1$, the fractions in Table I are from left to right: α_h/S_h, β_h/S_h, γ_h/S_h, and $2\theta_h f/S_h$.

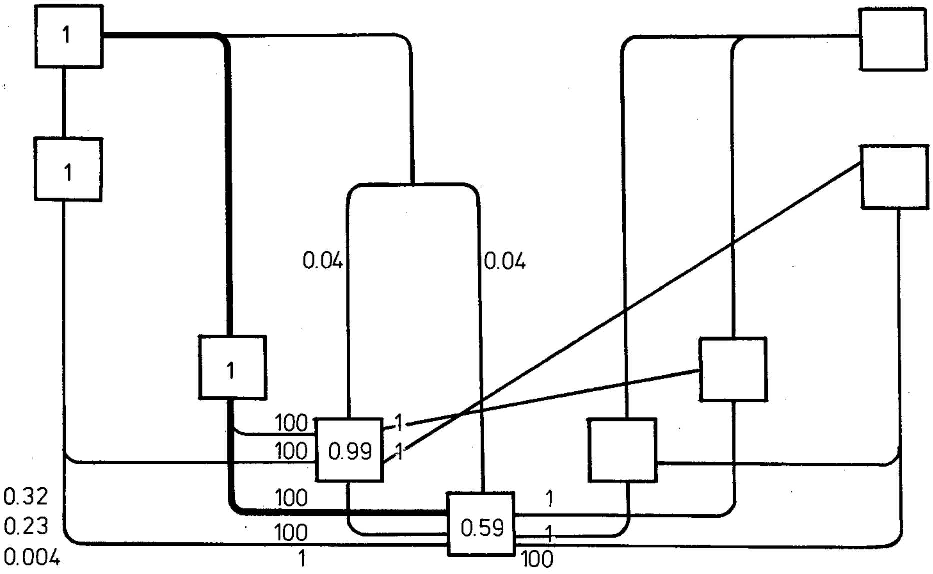

FIG.2. Adult's maximum consumption. Integers give the relative flow rates of contaminated and non-contaminated food. Fractions to the left are the first 3 summands of the sum (Eq.(10)). The summands for inhalation (0.04) are given on the inhalation pathways. The numbers in the compartments are c_a/c_w and c_h/c_w respectively.

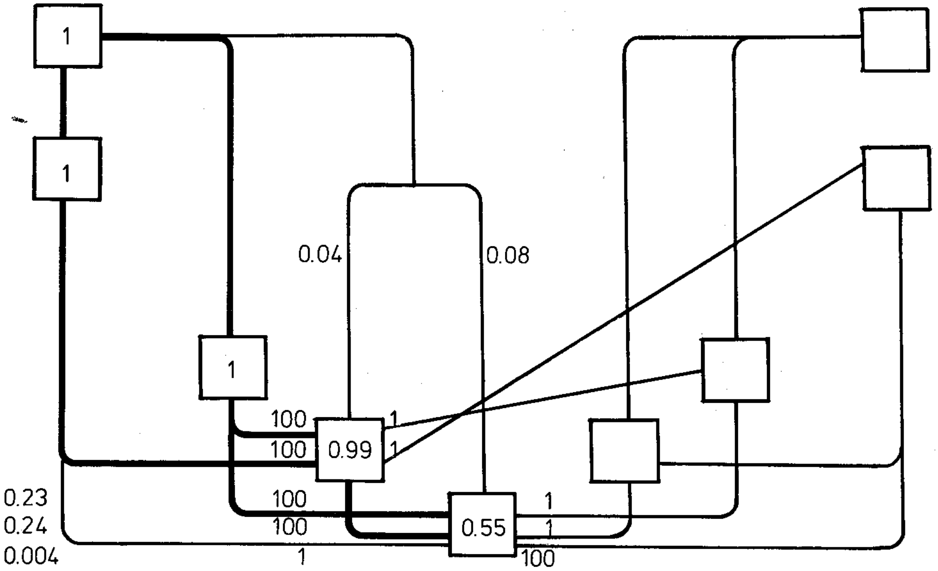

FIG.3. Adult's average consumption. Ratios of contaminated to non-contaminated food as given in Fig.2.

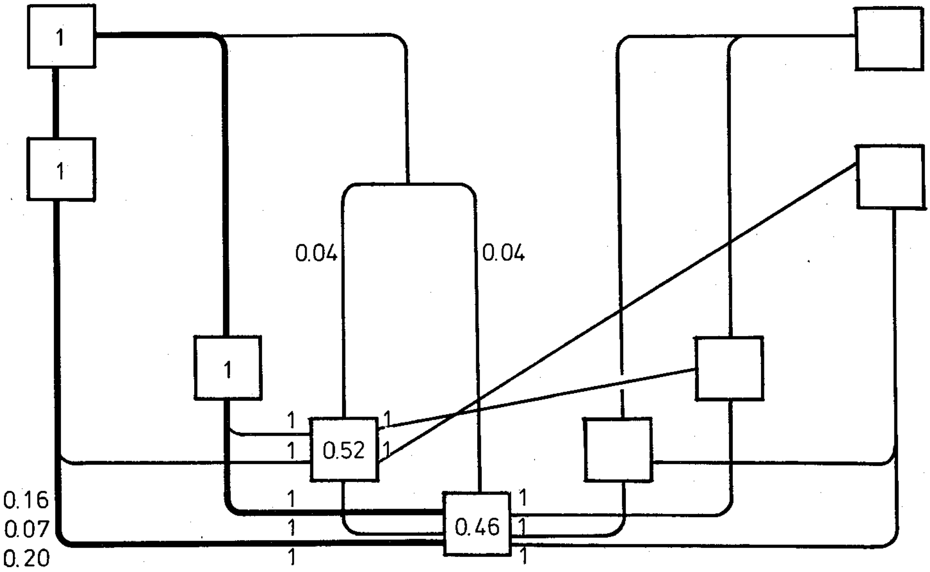

FIG.4. Adult's maximum consumption; 50% of the food is contaminated.

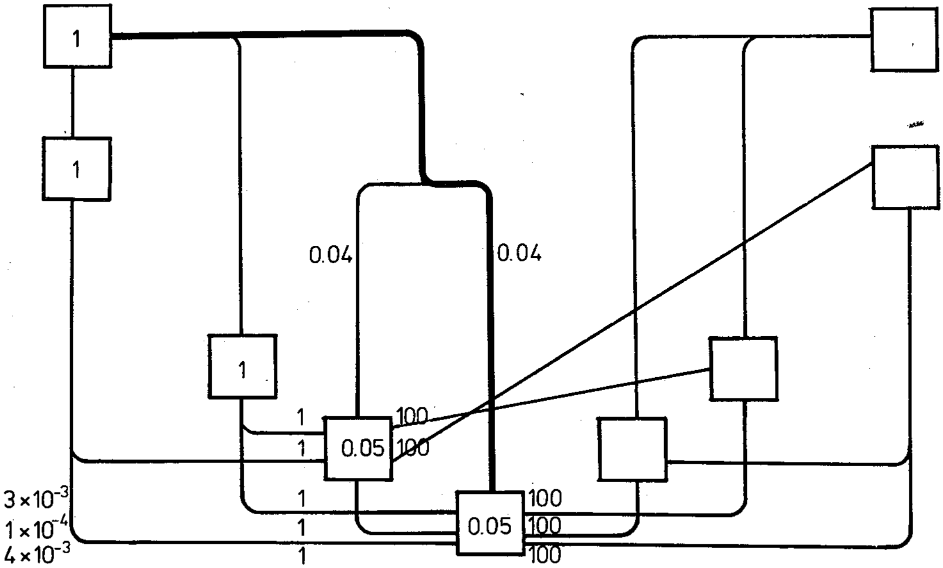

FIG.5. Adult's maximum consumption. Here inhalation contributes most of the contamination to man.

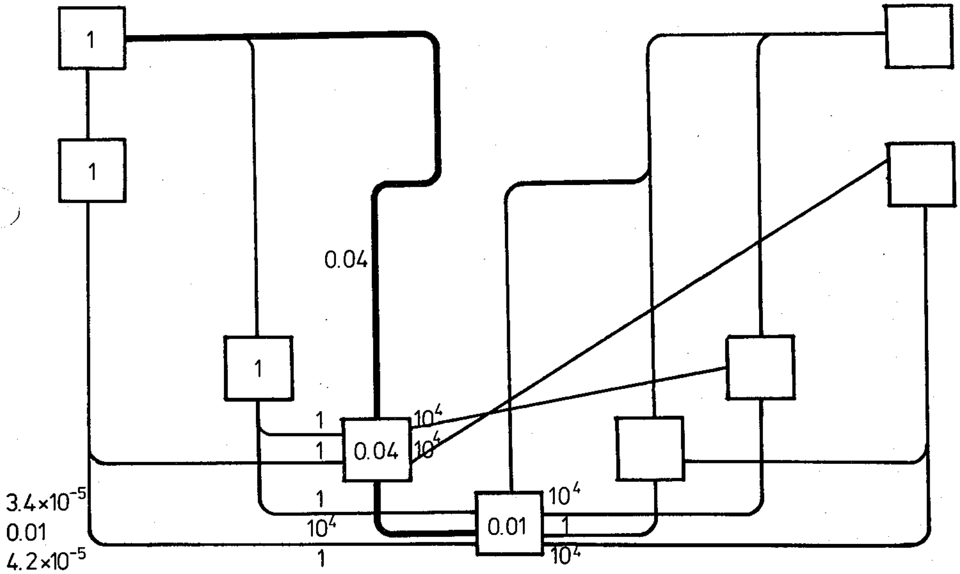

FIG.6. Adult's maximum consumption. Man lives outside the contaminated area (no inhalation of contaminated air by man), and only his milk and meat (beef) stems from the contaminated area. Cattle are fed non-contaminated food only, but breathe contaminated air.

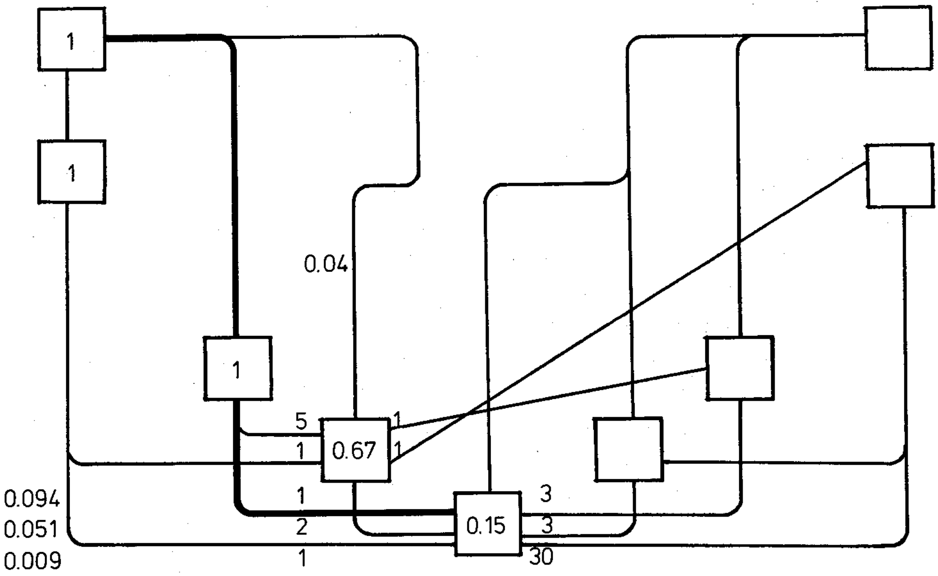

FIG.7. West Berlin's consumption habits. Transfer of tritium by food imported from Niedersachsen (Ns) to West Berlin causes the build-up of a tritium concentration equal to 15% of the average concentration in atmospheric water in Ns.

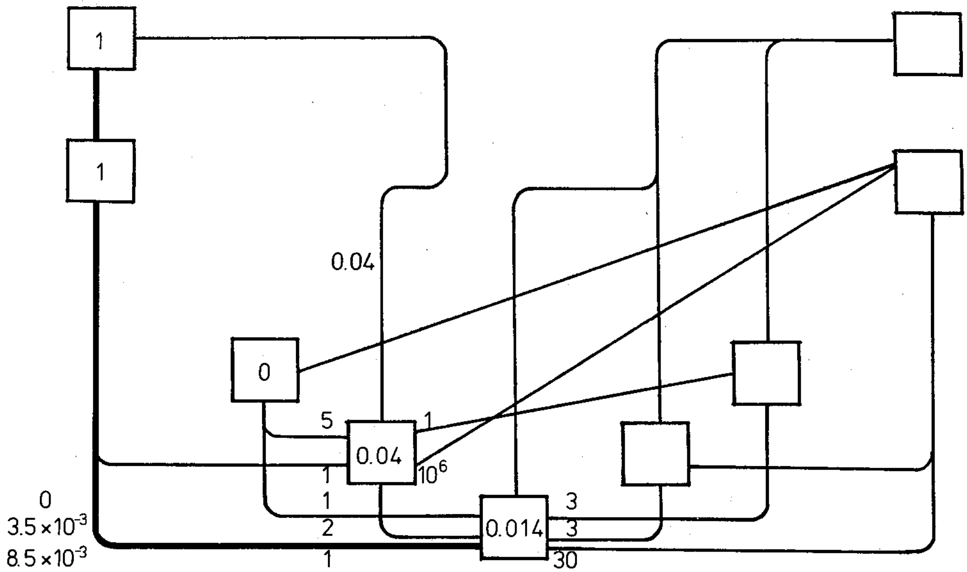

FIG.8. West Berlin's consumption habits. If irrigation water only is used in the contaminated area of Ns, the transfer of contamination to West Berlin is reduced by a factor of 10 compared with Fig.7, and is now dominated by the 10^4 m^3 of beer from Ns.

For comparison Table I also shows the values calculated on the basis of Anspaugh's data and those recommended by the Federal German Radiation Protection Commission. Both do not specify the pathway of drinking water. To be consistent with the assumptions underlying the data given in the first six lines, consumption of contaminated drinking water only is assumed here, and the fraction of the dose rate due to that pathway is taken to be 1-(fraction due to plant- and animal-product intake and inhalation).

The values in the line 'Anspaugh' were calculated using $S_h = \lambda_h M_h$.

In the last line of Table I the dose rates were first calculated according to the recommendations from Ref. [6]. Then the dose rate was converted to c_h with the help of Eq.(1) and using $c_i/c_w = 1$. Also c_a/c_w was taken to be unity even though the recommendations assign the values 0.75 X 55/100 (for milk) and 0.75 X 55/120 (for meat).

In the remaining figures various consumption situations of plants, animals and man are considered in more detail, giving more realistic values for c_i/c_w, which are denoted in the compartments i. The values for the total consumption rates (e.g. $\alpha_h + \delta_h$) again are taken from Ref.[5] and are listed in Tables II and III. They will be referred to as 'adult's maximum' or 'adult's average' in the figure captions.

The compartment system of Figs 2–8 is mainly the same as in Fig.1. The description of the pathways and the compartments could therefore be left out.

Again each food pathway (except inhalation) consists of two parts, one contaminated and one not. The integers L in the figures give the relative contribution of one part compared with the other. That means, for example (see Fig.2),

$$L_h^p = \alpha_h/\delta_h = 100/1 \quad (13)$$

and obviously

$$\alpha_h = (\alpha_h + \delta_h)/(1 + (1/L_h^p)) \quad (14)$$

A fraction denoted at the inhalation pathway and to the left of the plant-, animal- and water-pathway to man is the contribution of the pathway specified to the dose rate (for further explanation see, for example, caption of Fig.2).

The pathways contributing more than 50% of the dose rate are marked by heavy lines.

Figures 2 and 3 stand for consumption of contaminated food only, both by animals and man, except for drinking water for man, which is supposed to come mainly from fountains. The relative contributions of the plant- and animal-pathway have already been given in Table I. The slightly changed consumption habits of the average adult compared with the 'maximum adult' cause the slight change in the ratio c_h/c_w.

In Fig.2 the dose rate is 59% of the value it would be if only contaminated food were consumed, i.e. if the integers at all contaminated pathways were infinite; 32 of those 59% come via the vegetarian pathway.

In Fig.4 an equal mixture of contaminated and non-contaminated food is considered. The dose rate is reduced by about a factor of 2 in this case compared with the case of contaminated food only (Table I).

Figures 5 and 6 show that the inhalation begins to dominate the dose rate when the contaminated food fraction becomes less than 1%.

Figures 7 and 8 have been drawn to show the situation for West Berlin, which obtains about one third of its vegetarian and two thirds of its non-vegetarian food from the province of Niedersachsen (Lower Saxony), where a reprocessing plant for nuclear fuel is planned.

People in West Berlin breathe non-contaminated air. Because of the contaminated air breathed by cattle, a Berliner would (according to these calculations) obtain about 10% of the tritium concentration he would get if he inhaled the tritium while living in Niedersachsen, but without eating contaminated food (see Figs 5 and 8).

If the plants in Niedersachsen grow without irrigation because of a wet summer (Fig.7), a tritium concentration of 15% of the 'all contaminated food chain value' would be built up in the Berlin population.

During very dry summers the dose rate of the Berlin population is one tenth of the value of wet summers (Fig.8).

As with any model our model requires measured data throughout its various compartments to confirm its validity. Several assumptions have made this model simple. However, the final test is the confirmation in the environment.

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DISCUSSION

H.J. BURKI: It appears that this model applies primarily to a 'local' source of tritium contamination. It does not seem easily applicable to global contamination. Could you comment on this aspect? For example, could the model be applied to fusion reactor contamination?

A.A. MOGHISSI: As a co-author of the paper I should like to stress that the model does have global applicability because it is based on a specific activity concept and therefore does not require biological half-life data. It considers two compartments, one associated with the tritium source and the other with the environment. The entire system is subsequently homogeneously mixed.