

Caldwell RD, Potter TE. The Solubility of Inhaled Particles. Presented at the 14th AEC Bioassay and Analytical Chemistry Conference, Oct. 7-8, 1968;14:31.

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Keywords:

ICRP Task Group Lung Model

plutonium Pu-239 Pu-238

bioassay

excretion data

fecal sampling

urine sampling

in vivo counting

lung

lung burden

gastrointestinal tract

lognormal distribution

lung cancer

tumor

particle dissolution

particle size distribution

low-fired mixed plutonium oxide

high-fired mixed plutonium oxide

Pu metal

air sampling

PuO₂

THE SOLUBILITY OF INHALED PARTICLES

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(For Presentation Only)

SLIDE 1 THE 1966 ICRP PULMONARY RETENTION MODEL

In the proposed lung model, long term insoluble particles are supposed to clear principally by way of the G. I. Tract. This route depends upon some mechanism for getting deposited pulmonary particles to the ciliary escalator. Most investigators feel that mobile cells, called phagocytes, capture the particles and carry them to the ciliated parts of the lung.

A constant fraction of the lung burden is assumed removed per time interval. This should result in an exponential removal to the G. I. Tract and hence an exponential fecal excretion.

SLIDE 2 TROUBLE WITH THE RETENTION MODEL

When excretion data is compared with in-vivo counting (as shown by Saxby), we find the exponential model holds true for in-vivo counting, but not for fecal excretion. Instead after early clearance, fecal excretion generally follows the urine excretion. Both fall off faster than the directly measured lung burden.

Although NUMEC has not body counted exposed employees long enough to demonstrate this pattern as clearly as Saxby's example, we found fecal excretion lower than expected for several individuals with old lung burdens. Our in-vivo gamma counts show lung burdens several times higher than we would have estimated from an exponential fecal excretion.

On the other hand, we have always found good correlation with early fecal clearance and in-vivo counting of recently acquired lung burdens. Apparently, the ciliary - G. I. Tract route becomes less and less important with the age of the lung burden.

Another interesting aspect of Saxby's data is that excretion data does not account for all the radioactivity leaving old lung burdens. The amount excreted per unit time is less than that leaving the lung. This can only mean that translocation from the lung to other organs is taking place.

SLIDE 3 MERCER'S DISSOLUTION THEORY

Mercer recently proposed that long term insoluble particles only leave the lung by dissolving. He shows that the fraction remaining of an original lung burden is a function of a dimensionless parameter, β , and the geometric standard deviation, σ_g , of an assumed log-normal particle size distribution. β reduces to the product of the time, t , the solubility rate constant, k , and the ratio of s , the surface area, to the particle mass, m . This dependence on β is easy to understand, since the smaller s/m and k are, the longer t will be. Mercer found that M/M_0 could be satisfactorily approximated by the sum of two exponential terms for all values of σ_g greater than 1.65. He also shows excellent agreement of his theory with many animal experiments.

The dependence of M/M_0 upon σ_g may not be as readily apparent.

SLIDE 4 INCREASE OF MASS MEDIAN DIAMETER WITH σ_g

When particle size distributions, having the same count median diameter (i.e. half the number of particles are larger and half smaller than the CMD) but different σ_g 's are plotted on log probability paper, the reason why solubility is a function of σ_g becomes clearer. The higher the value of σ_g , the greater the percentage of larger particle sizes. When the mass median diameter (i.e. half of the mass is associated

with particles less than the MMD and half greater) is calculated for each σ_g , it is readily apparent what happens. The mass median diameter goes up sharply with increasing σ_g .

Particles, close in size to the MMD, contain most of the mass of typical distributions. Big particles dissolve slower than small particles. Thus, lung burdens of greater MMD have longer retention times. Steckel and West have demonstrated this for occupational exposures.

Bear in mind that Mercer's theory is for particles already deposited in the lung. The particle size dependent filtering action of the upper respiratory tract must be accounted for to apply the theory to airborne hazard analysis.

SLIDE 5 TUMOR RISK FROM SINGLE PARTICLES

Besides causing longer retention times, large particles subject surrounding lung cells to high doses. Dean and Langham extrapolated rat skin tumor experiments to calculate lung tumor risk from single PuO_2 particles. As the particle size increases, the dose is increased and the tumor induction probability goes up. A maximum probability occurs at a particle size, which varies with the specific activity of the particle. Beyond this size, the higher dose injures cells so that they no longer reproduce and the risk falls off. For a 500 day lung burden half life, the mean residence time for particles is 720 days. With these assumptions, Dean and Langham calculate some alarming tumor probabilities: 0.1 for a single 1 micron $^{238}\text{PuO}_2$ and 0.1 for a single 8 micron $^{239}\text{PuO}_2$ particle. It would then only take ten 8 micron particles of $^{239}\text{PuO}_2$ to guarantee a tumor.

SLIDE 6 PARTICLE SIZE DEPENDENCE OF PULMONARY DEPOSITION

It is often supposed that large particles are completely filtered out by the upper respiratory tract. However, the ICRP deposition model demonstrates a definite, if small, deposition all the way to 100 microns, MMD. Remember that for most distributions, the bulk of the mass is made up of particle sizes close to the MMD.

SLIDE 7 RELATIVE TUMOR RISK FOR EQUAL MASSES OF PuO₂

The Dean-Langham theory was not extrapolated to distributions of particles. We show here what happens to the tumor probability curve, when tumor risks are calculated for particle size distributions with varying MMD, but having equal masses. A 10 micron ²³⁹PuO₂ particle is equal to 0.37 nanocuries. This operation raises probabilities for smaller particles, since there are more of them.

We also show the effect of the URT filtering action for large particles when inhaled. We multiplied the top curve by the ICRP deposition curve. This also has the effect of raising tumor risks for small particles, because more of them reach the lung.

The actual tumor probabilities may not be realistic. Not enough experimental work on local tissue dose has been done. However, even taking size distributions and particle size dependent deposition into account, there is a band of particle sizes (0.8 to 8.0 microns for ²³⁹PuO₂) which appear to be most hazardous.

SLIDE 8 LOW FIRED MIXED OXIDE

Recalling Mercer's theory, in addition to the particle size effect, the surface area per gram also effects the half life. Bair has noted that high fired oxide exposures result in longer retention times than low fired oxides. The same effect of heat treatment was also noted by Steckel and West. Comparison of electron micrographs of low and high fired oxides reveals that heat treatment reduces the particle surface area. The low fired oxide has a small crystallite size (Most "particles" actually are agglomerates of crystallites) and a relatively high surface area per gram.

SLIDE 9 HIGH FIRED MIXED OXIDE

Crystallite size is much larger for high fired particles. Consequently, the surface area and solubility are both reduced. Ceramists tell us that crystallite perfection is an important parameter of solubility. Heat treatment also anneals the crystallite

getting rid of solubility-active lattice imperfections. An accompanying effect of this phenomenon is the reduction of the oxygen to metal ratio.

An interesting appearance of a second phase (ZrO_2) can be seen in this particular micrograph. This does not always occur, but is indicative of the changes heat treatment can bring about.

SLIDE 10 FIELD TEST FLOW CHART

Understanding that solubility of an aerosol directly effects the health hazard is not enough. What the Health Physicist needs is a technique for measuring aero-sol solubility. Assuming that aerosol characteristics are that of the material handled in an area is fraught with error. First, different kinds of materials may be handled in one working space, such as in an oxide conversion plant. Second, the aerosol particles may undergo physio-chemical changes when released from the parent material. (e.g. Pu metal particles may oxidize).

Aerosol particle sizing techniques are well-known. But field tests for solubility have not been published. Most solubility studies used lung equivalent fluids for long term studies. A quick test of air sample solubility appeared to be very useful to us. So, we developed a relative solubility test using 0.1N HNO_3 and 0.8u millipore filters. The test is not intended to duplicate solubility in the lung, but only allows us to differentiate between aerosols.

SLIDE 11 NUCLEAR FUEL AEROSOLS IDENTIFICATION

We tested aerosols in seven locations at NUMEC's Nuclear Fuel Plants. The samples were not truly representative of exposure, since they were obtained inside glove boxes or hoods. The idea was to obtain a sample representative of each chemical species.

A clear difference between aerosols resulted, especially between solution-generated

aerosols and oxides. But, we also got a reproduceable difference between low and high fired oxides.

We did not expect green Pu oxide scale (metal moss) to be so insoluble. But our experience duplicated that of our processing people, who say the stuff is very difficult for them to dissolve. The crystallite size may be large, since the oxide scale is formed slowly.

We held several air samples from the uranium locations for 48 hours. We wanted to see if any change would occur, if analysis was delayed. A detectable difference appeared for UNH. It seems the particles change characteristics with time.

SLIDE 12 SOLUBILITY OF PARTICLE SIZE FRACTIONS

To ascertain whether the solubility tests were merely an artifact caused by particle size differences, we collected several cascade impactor samples. After determining the stage activity, we subjected each size fraction to our field solubility test. The AMADs of the solution-generated aerosols were larger than for the oxides, so the solubility identification was not an artifact of particle size.

We may not know everything there is about particle size distribution, but not many of our measured distributions make single straight lines on a log probability graph. That is the reason for the uncertainty of the MMD for UNH and $\text{Pu}(\text{NO}_3)_4$.

Both oxides show poor solubility (Pu < U) for the larger sizes and increased solubility for the smallest fraction. The smaller the UNH particle, the less soluble it is. The Pu nitrate was relatively soluble except for the first stage, which was very insoluble. Possibly a different species represents the large particles and might explain the variability in our original field tests.

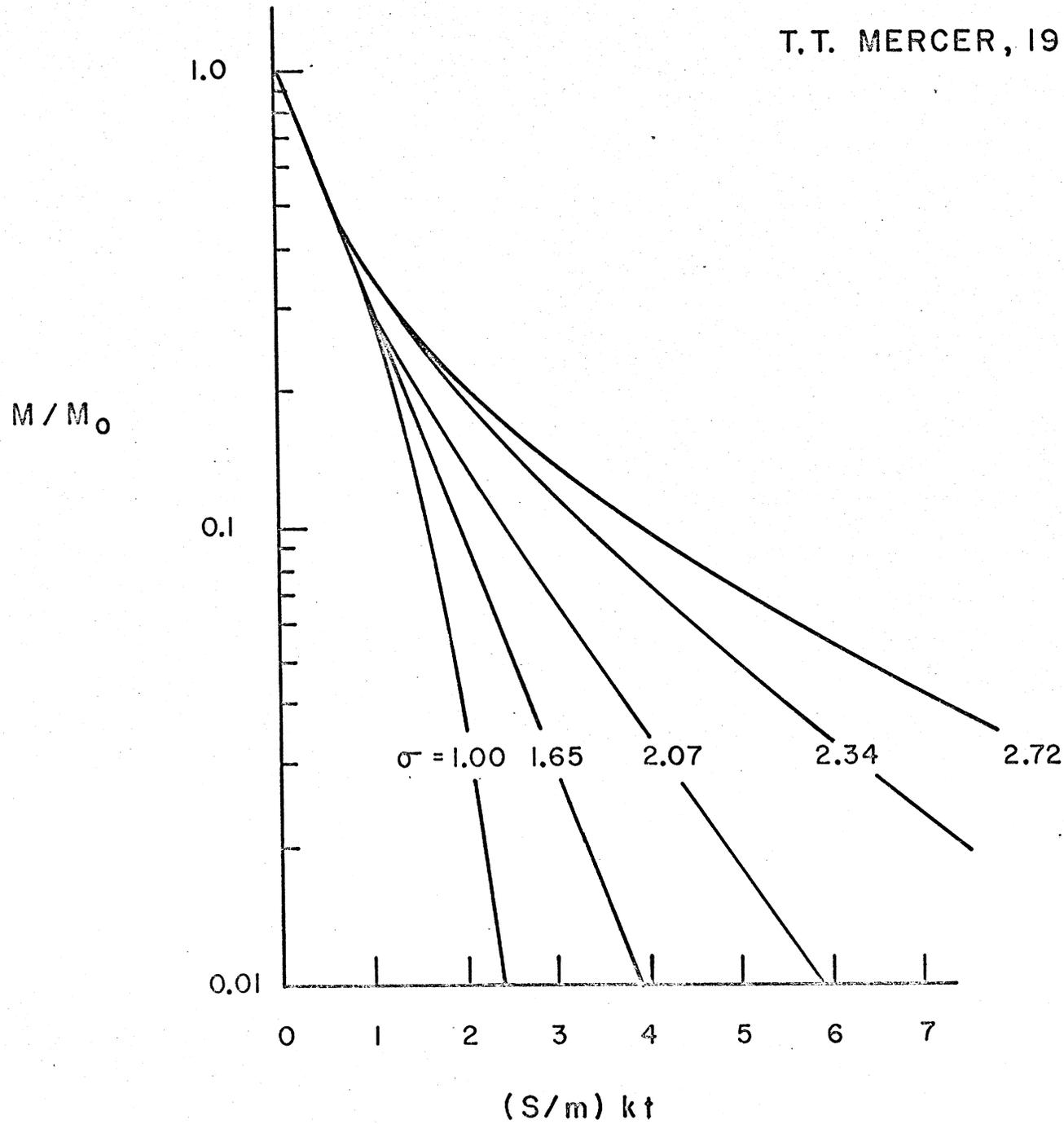
SUMMARY

Aerosol solubility and particle size both strongly effect lung retention time and thus are important parameters for hazard evaluation. We have developed a field test which permits us to identify different chemical species on air samples.

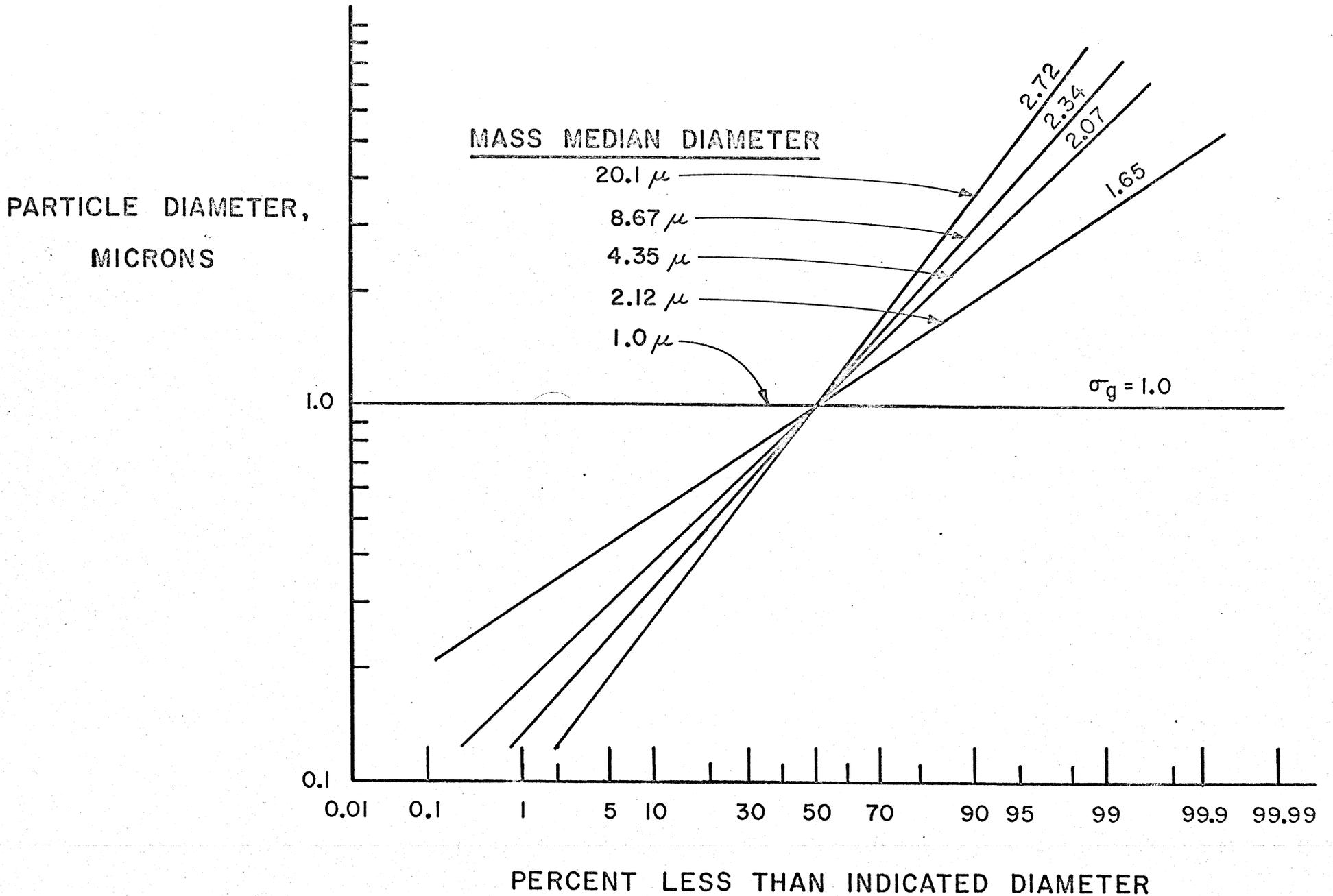
One last observation: maybe why phagocytes cannot move nuclear fuel particles out of the lung is because Pu or U oxide is as dense as lead. The phagocytes swallow the fuel particle and then they can't get the lead out.

LUNG BURDEN DISSOLUTION

T.T. MERCER, 1967

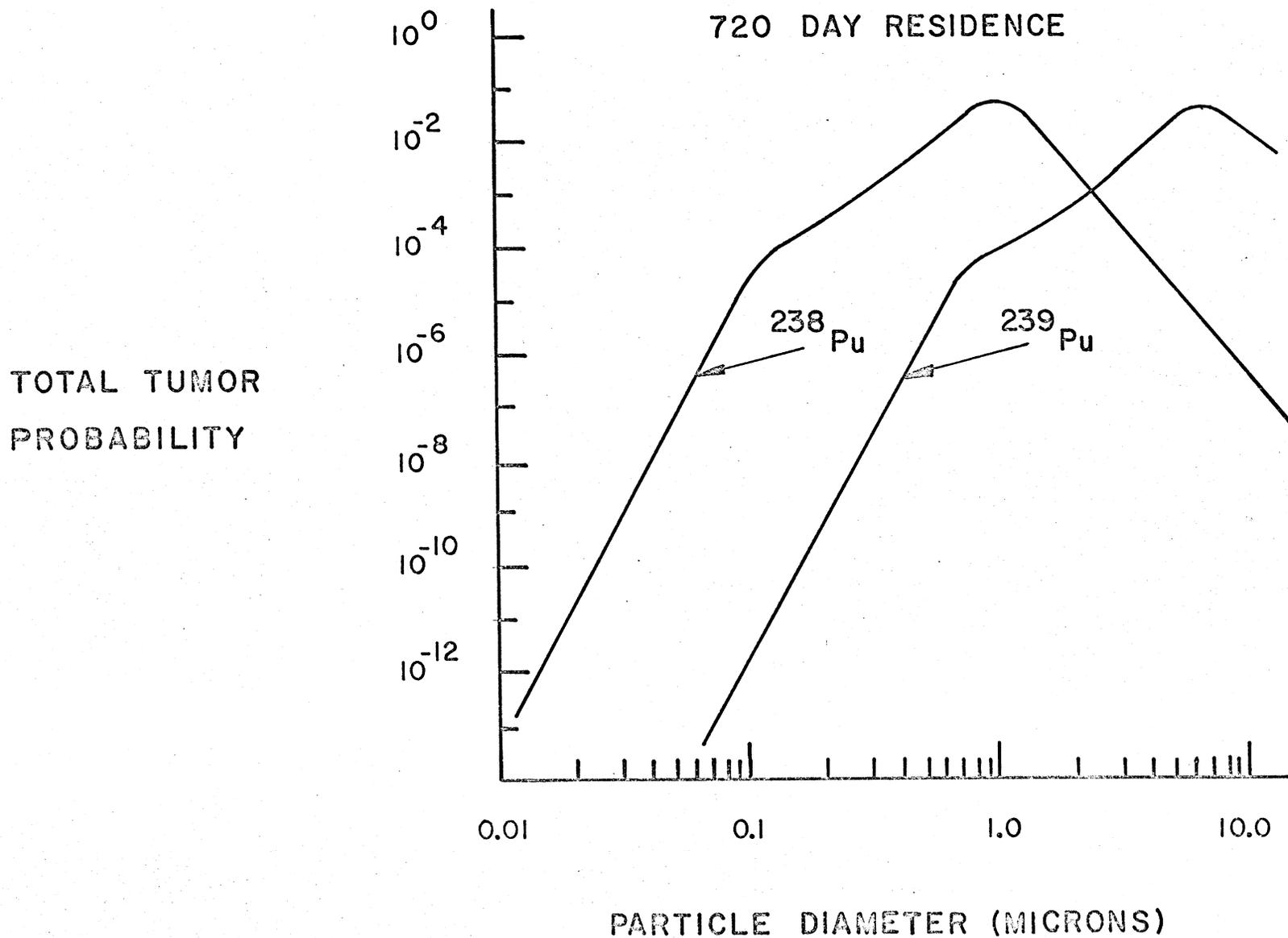


PARTICLE SIZE DISTRIBUTIONS
 AS A FUNCTION OF GEOMETRIC STANDARD DEVIATION, σ_g
 COUNT MEDIAN DIAMETER = 1.0μ



LUNG TUMOR RISK FROM A SINGLE PuO_2 PARTICLE

DEAN and LANGHAM, 1968

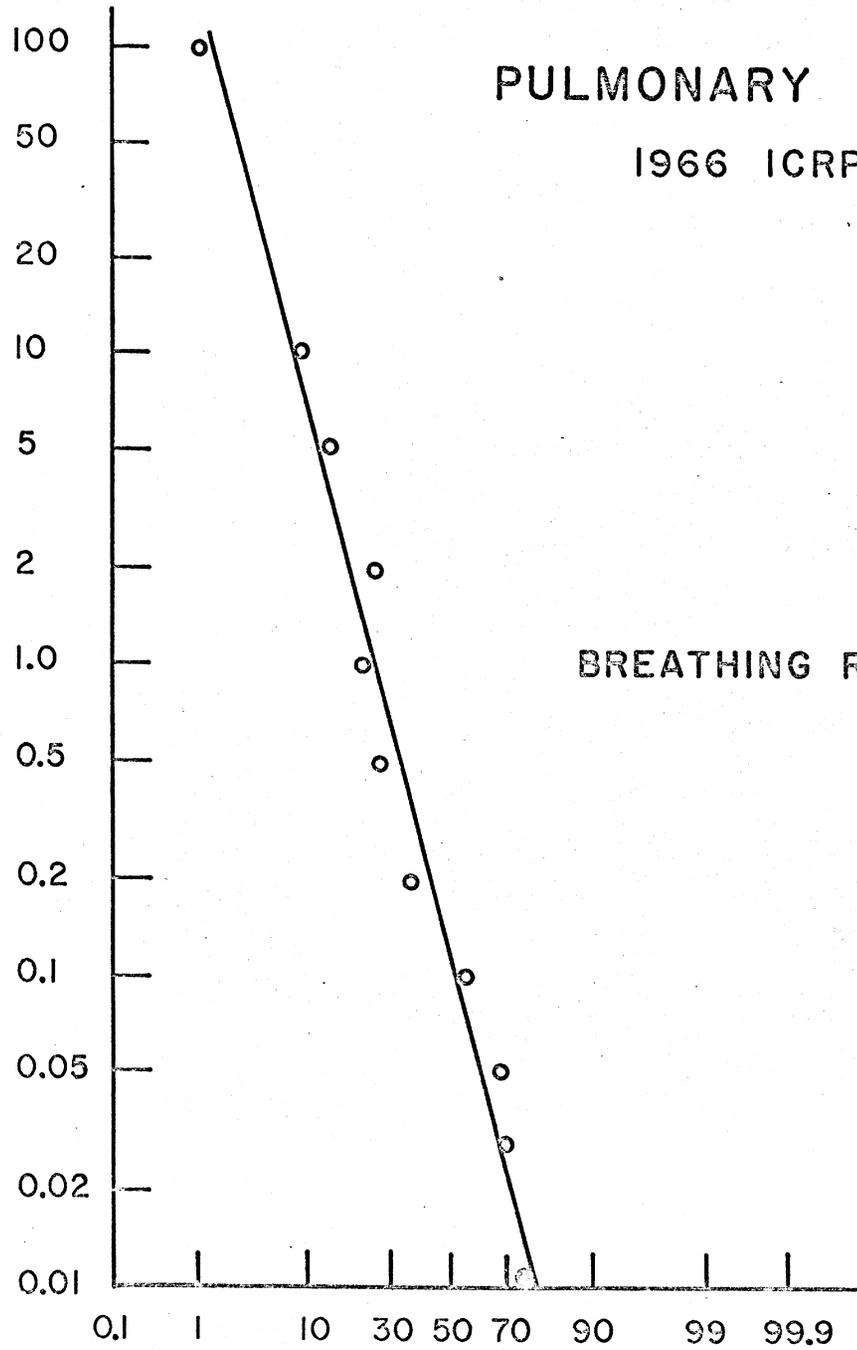


PULMONARY DEPOSITION

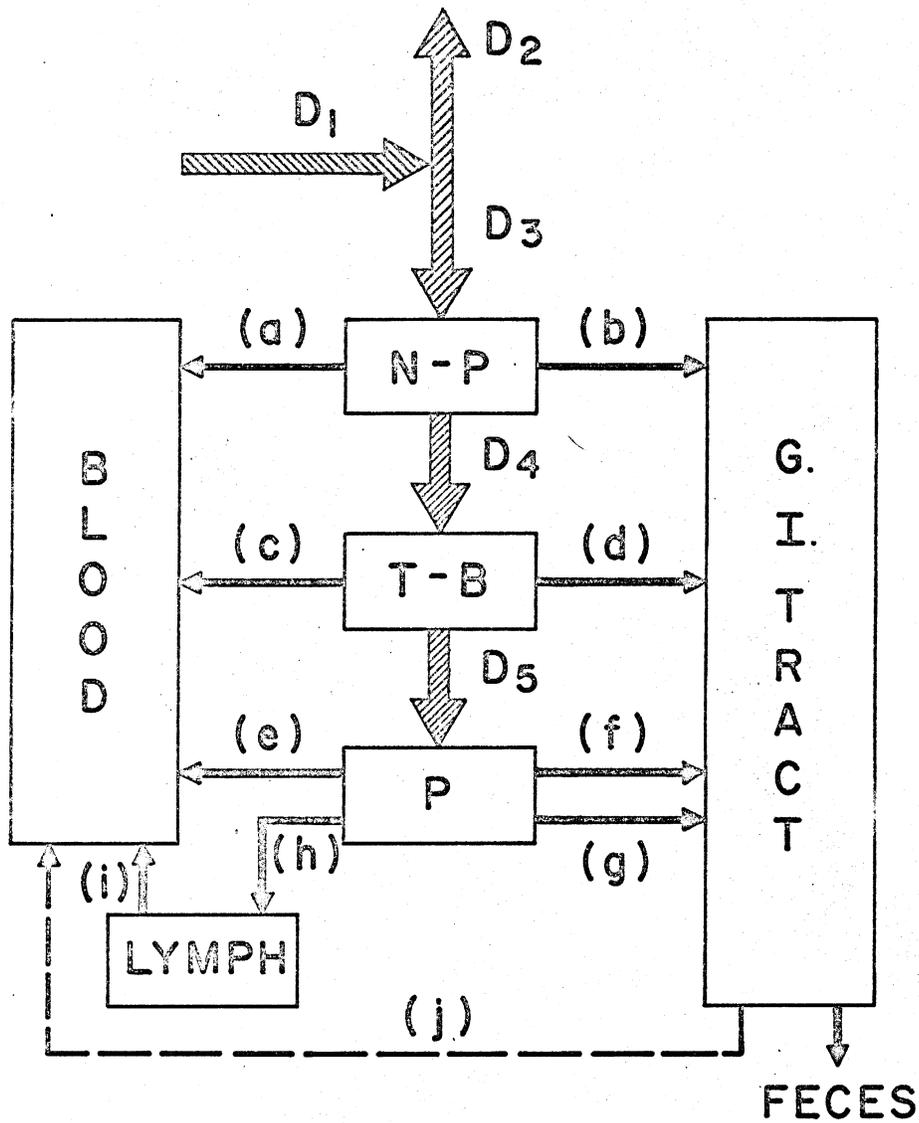
1966 ICRP MODEL

BREATHING RATE: 20 L/MIN.

MASS OR ACTIVITY
MEDIAN DIAMETER

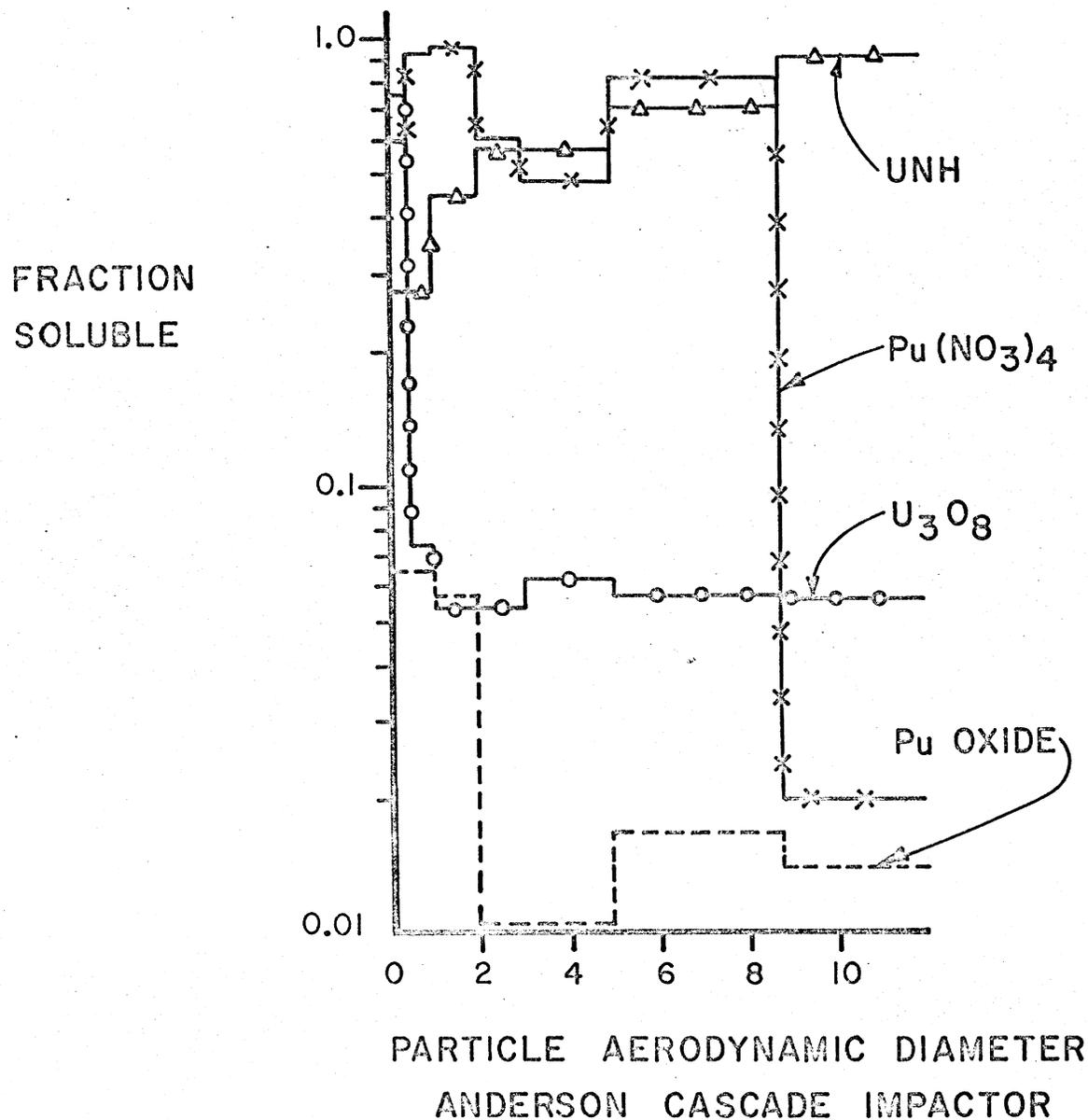


PERCENT DEPOSITION



ICRP DEPOSITION AND RETENTION MODEL

SOLUBILITY OF PARTICLE SIZE FRACTIONS
 NUCLEAR FUEL AEROSOLS
 NUMEC FIELD TEST



ACTIVITY MEDIAN AERODYNAMIC DIAMETER
UNH -> 8.4 μ
U ₃ O ₈ - 7.7 μ
Pu(NO ₃) ₄ -> 8.4 μ
Pu OXIDE - 4.5 μ

RELATIVE TUMOR RISK
FOR EQUAL MASSES
OF $^{239}\text{PuO}_2$

TUMOR
RISK

