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JACKSON LABORATORY
RESEARCH AND DEVELOPMENT DIVISION TECHNICAL REPORT
CHEMICALS AND PIGMENTS DEPARTMENT
E. I. DU PONT DE NEMOURS & COMPANY

KINETICS OF SORPTION AND ELIMINATION OF FLUORINATED SURFACTANTS IN BLOOD

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Type of technical work: [redacted] RD
Period covered: [redacted] August 1984 to February 1986
Personnel: [redacted] Ward Gibson and Paul A. Saincon

ABSTRACT

Blood samples of a Haskell Laboratory inhalation subchronic study on [redacted] have been analyzed for organic fluorine and inorganic fluoride. The kinetics of sorption and elimination of [redacted] in blood have been determined. The concentration of [redacted] related organic fluorine in blood was found to increase approximately with the square root of the increasing [redacted] concentration in the air inhaled. The elimination of [redacted] from blood appears to obey first-order kinetics, but the rate coefficient decreases with the square root of the increasing postexposure time.

The elimination rate coefficients of [redacted] and perfluorooctanoic acid in blood do not differ greatly. However, [redacted] is less volatile than perfluorooctanoic acid and the concentrations of [redacted] in air and consequently in blood are therefore likely to be lower than those of perfluorooctanoic acid.

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OBJECTIVES

To determine fluorine in rat blood samples, submitted by [redacted] at Haskell Laboratory, of the inhalation subchronic study on [redacted] to estimate the [redacted] levels in blood following exposure and the rate of elimination from blood.

BACKGROUND

The observation by Taves (1,2) that human blood serum contains both inorganic fluoride and organic fluorine have increased the interest in the retention of organic fluorochemicals in blood. In a study on the exposure of industrial workers to fluorochemicals (3), elevated fluorine concentrations, ranging from 1 to 71 ppm, were found in the blood of workers handling the ammonium salt of perfluorooctanoic acid. The long retention time of perfluorooctanoic acid in blood has lead to efforts to replace perfluorooctanoic acid salts by less volatile fluorinated surfactants. [redacted] an ammonium salt of a sulfonic acid derived from Telomer B, was designed to replace ammonium perfluorooctanoate in Teflon® polymerization processes.

CONCLUSIONS

1. The amount of organic fluorine in blood increases with the square root of increasing [redacted] concentration in the air inhaled.

2. The elimination of [redacted] from blood approximates first-order kinetics. However, the rate coefficient is not a constant, but decreases with the square root of the increasing postexposure time.

3. The elimination rate coefficients of [redacted] and that of perfluorooctanoic acid do not differ markedly. However, [redacted] is less volatile and its concentrations in air and consequently in blood are likely to be lower than those of perfluorooctanoic acid.

4. The inorganic fluoride levels in blood increased slightly, if significantly, after exposure to [redacted] in air but decreased to normal values in a relatively short time.

PATENT SITUATION

Patent Protection

A patent application [redacted] has been filed.

Proposed Filing Action

Patent action, related to this study, is not planned.

PUBLICATION STATUS

The kinetic interpretation of sorption and desorption of fluorosurfactants in blood is novel and probably of general validity. Publication will be considered.

The method for the determination of inorganic fluoride in blood has been published (E. Kissa, Clin. Chem. 33, 253 (1987)).

FUTURE WORK

The [redacted] inhalation study has been completed and further work is not planned.

SAFETY

No unusual considerations.

ENVIRONMENTAL CONSIDERATIONS

No unusual environmental considerations.

TABLE OF CONTENTS

DISCUSSION

1. Sorption of [redacted] in Blood.

Haskell Laboratory conducted an inhalation subchronic study on [redacted] using rats as the test animals. Blood samples were collected after the tenth exposure, after two weeks, six weeks, and twelve weeks of recovery. We analyzed the blood specimen for total fluorine by the oxyhydrogen torch method [6] and for inorganic fluoride by our analyte addition method [4]. The concentration of organic fluorine in blood was calculated as the difference between the total fluorine concentration and the inorganic fluoride concentration in blood.

The concentration of organic fluorine in blood, c_b, was found to increase exponentially with the increasing concentration of [redacted] in air, c_a, expressed as

$$c_b = k\, c_a^{\,n} \qquad (1)$$

A plot of c_b versus c_a on logarithmic scales yields straight lines with the slope n (Fig. 1). The plot shows that the values of the coefficient, k, decreased with the increasing recovery time (Table I). The value of the exponent n is approximately 0.5, suggesting that c_b may be a function of the square root of c_a (Table II). A plot of the organic fluorine concentration in blood versus the square root of [redacted] concentration in air yields indeed a straight line, the slope of which decreases with the increasing recovery time. The lines do not pass through the origin, however, but intercept the x-axis at distances which decrease with the increasing recovery time (Fig. 2). The nonlinearity of this relationship at very low [redacted] concentrations requires more data for a definite interpretation.

2. Elimination of [redacted] from Blood.

The presence of organic fluorine in blood has been attributed to fluorinated carboxylic acids which are volatile and absorbed in the body mainly by inhalation [3,6]. Because of concern about their long residence time in blood, perfluorooctanoic acid, used as the dispersant in the Teflon® process, was replaced by [redacted] a fluorinated surfactant of low volatility.

The subchronic inhalation study on [redacted] showed that [redacted] like perfluooctanoic acid, also has a long residence time in blood. Twelve weeks after exposure to a low concentration (7.6 mg/m3) of [redacted] in air the organofluorine concentration in blood returned to a normal level. However, rats exposed to higher [redacted] concentrations in air had a significantly elevated organofluorine concentration in their blood still after twelve weeks (Table I).

In order to compare the rate at which [redacted] is eliminated from blood to the elimination rate of perfluorooctanoic acid, the data have to be fitted to a kinetic expression which permits the calculation of rate constants or rate coefficients. This is, however, not straightforward, because the elimination of fluorinated surfactants from blood does not obey simple kinetics.

It is usually assumed that the elimination of fluorocompounds from blood obeys, like a mass transport process, first-order kinetics. The half-life of the fluorocompound in blood has been used to express the elimination rate. However, the calculation of the half-life assumes uncomplicated first-order kinetics and this assumption is not always valid.

I have reported [6] that the decrease of organic fluorine in blood during the postexposure period of a perfluorooctanoic acid feeding study approximates first-order kinetics:

$$\log c_b/c_o = k_f t \qquad (2)$$

where c_b is the concentration of the fluorinated surfactant in blood at time t, c_o the initial concentration of the fluorinated surfactant in blood, and k_f the rate coefficient of the elimination of the fluorinated surfactant from blood. If the fluorinated surfactant is not metabolized, which is a reasonable asssumption, then the organic fluorine concentrations in blood can represent the concentrations of fluorinated surfactants.

A plot of log c_b versus time (Fig. 3) exhibits considerable curvature and consequently a significant departure from uncomplicated first-order kinetics. The elimination rate of perfluorooctanoic acid from blood is more rapid in the beginning than during the latter part of the recovery period. This suggests that the desorption rate of perfluorooctanoic acid molecules in blood may vary. Perfluorooctanoic acid is known to adsorb on protein in blood [7,8]. It is possible that the unadsorbed and the less strongly held perfluorooctanoic acid molecules are eliminated at a faster rate, leaving the more strongly held perfluorooctanoic acid molecules back. It is also possible that the strength of the adsorptive bond of perfluorooctanoic acid molecules varies, depending on the adsorption site in blood.

A first-order plot of [redacted] elimination from blood also shows curvature (Fig. 4). This deviation from uncomplicated first-order kinetics can be explained by an adsorptive site dependent variation of adsorptive bonding, similar to that of the perfluorooctanoic acid. The heterogeneous composition of [redacted] may also complicate its elimination kinetics. [redacted] consists of molecules (telomers) of different chain length which may affect the desorption and the diffusion rate of [redacted] molecules.

The average rate coefficient k_t for the elimination of a fluorinated surfactant from blood during a time interval Δt is given by the general kinetic expression

$$\Delta c/\Delta t = k_t \, c_t^{\,z} \qquad (3)$$

where Δc is the average organofluorine concentration decrease and c_t the average organofluorine concentration during the postexposure time interval Δt.

It follows that

$$\log (\Delta c/\Delta t) = \log k_t + z \log c_t \qquad (4)$$

The organofluorine concentrations in blood are plotted according to equation (4) in Fig. 5 for [redacted] and in Fig. 6 for perfluorooctanoic acid. The plot yields lines with the slope of about one, indicating that the elimination process is kinetically of first-order. However, the values of k_t are not constant, as expected for an uncomplicated first-order process, but decrease with the increasing recovery time (Table III). Consequently,

$$\log (c_b/c_o) = k_t \, t \qquad (5)$$

where k_t is a time dependent rate coefficient at time t.

The logarithm of organofluorine concentration in blood plotted against the square root of time yields straight lines (Fig. 7). Hence

$$\log (c_b/c_o) = k_r \, t^{0.5} \qquad (6)$$

The question remains, why does the organofluorine concentration in blood fit the equation (5) and also equation (6). Since the left sides of the equations are identical, the right sides have to be equal

$$k_t \, t = k_r \, t^{0.5} \qquad (7)$$

It follows that

$$k_t = k_r \, / \, t^{0.5} \qquad (8)$$

and the equation (5) becomes equation (7):

$$\log (c_b/c_o) = k_r \, t \, / \, t^{0.5} = k_r \, t^{0.5} \qquad (9)$$

Although the time intervals, at which the [redacted] concentration in blood was determined, were not selected for determining the time dependence of the elimination rate coefficient, it is apparent that the values of k_t vary approximately with the square root of time (Table IV).

In summary, the elimination of fluorinated surfactants from blood is a first-order kinetic process having a time dependent rate coefficient. A logarithmic plot , according to equation (4) yields the average rate coefficient, k_t, for the time interval Δt.

A comparison of the [redacted] and perfluorooctanoic acid concentrations in blood suggests that the rate coefficient values for anionic fluorinated surfactants may be similar. However, [redacted] is much less volatile than perfluorooctanoic acid and poses therefore a lesser environmental problem.

It should be remembered that the disappearance of organofluorine from blood does not necessarily indicate that the fluorinated surfactant has been completely eliminated from the body. Organs, especially liver and spleen, may also harbor organic fluorine.

3. Inorganic Fluoride in Blood.

The inorganic fluoride in the blood increased slightly, but perhaps significantly, as a result of exposure to [redacted] (Table V). The average inorganic fluoride value increased from 0.017 ppm in control samples to 0.051 ppm after exposure to 480 mg/m3 [redacted] in air. If this increase was significant, the inorganic fluoride concentration in blood returned to normal values in a relatively short time.

EXPERIMENTAL

The oxyhydrogen torch method (CD&F Method CW-9-8-3) for combustion of blood has been described in our report [6]. Total fluorine was determined as fluoride in the combusted analyte with an ion-selective electrode [4]. Inorganic fluoride in blood was determined by an analyte addition method we developed [5].

TABLE I

ORGANIC FLUORINE IN RAT BLOOD

Exposure Conc. of [redacted] (mg/m^3)	ppm Organic Fluorine in Blood*			
	Initial	2-Weeks Recovery	6-Weeks Recovery	12-Weeks Recovery
0	2.0	0.9	0.9	0.4
7.6	10.2	4.0	1.8	0.6
58.0	42.0	13.0	7.2	2.2
480.0	147.0	40.5	16.6	4.4

*Average values for blood samples of five rats.

TABLE II

SORPTION OF [redacted] IN RAT BLOOD: THE COEFFICIENT k_t AND THE EXPONENT n

Recovery time (weeks)	k_t	n
0	2.87	0.64
2	1.31	0.56
6	0.67	0.53

TABLE III

KINETICS OF FLUORINE ELIMINATION FROM RAT BLOOD

Recovery Period (weeks)	Fluorochemical	Rate Coefficient k_t	Exponent z
0-2	[redacted]	0.36	1.10
2-6	(inhalation)	0.17	1.03
6-12		0.16	1.07
		Average	1.07
0-2	Perfluorooctanoic	0.73	0.92
2-6	acid (feeding)	0.33	1.03
		Average	0.98

TABLE IV

RATE COEFFICIENTS AS A FUNCTION OF TIME

Recovery Period (weeks)	Average Recovery Time (weeks)	Square Root of Average Recovery Time	Rate Coefficients			
			[redacted]		Perfluorooctanoic Acid	
			Found	Calc.*	Found	Calc.*
0-2	1	1	0.36	-	0.73	-
2-6	4	2	0.17	0.18	0.33	0.37
6-12	9	3	0.16	0.12		

* Calculated by the equation (8).

TABLE V

INORGANIC (IONIC) FLUORINE IN RAT BLOOD

[redacted] Exposure Conc. In Air (mg./m^3)	Recovery Time (Weeks)	Organic Fluorine (ppm)	Inorganic Fluorine (ppm)	Rat No.
0	0	1.4	0.016	376 840
0	2	0.7	0.025	376 849
0	2	1.1	0.019	376 835
0	2	0.7	0.007	376 841
480	0	140.0	0.055	376 854
480	0	112.0	0.034	376 828
480	0	168.0	0.063	376 879
480	2	41.2	0.020	376 858

FIGURE 1
CONCENTRATION OF ORGANIC FLUORINE IN BLOOD AS AN
EXPONENTIAL FUNCTION OF [redacted] CONCENTRATION IN AIR

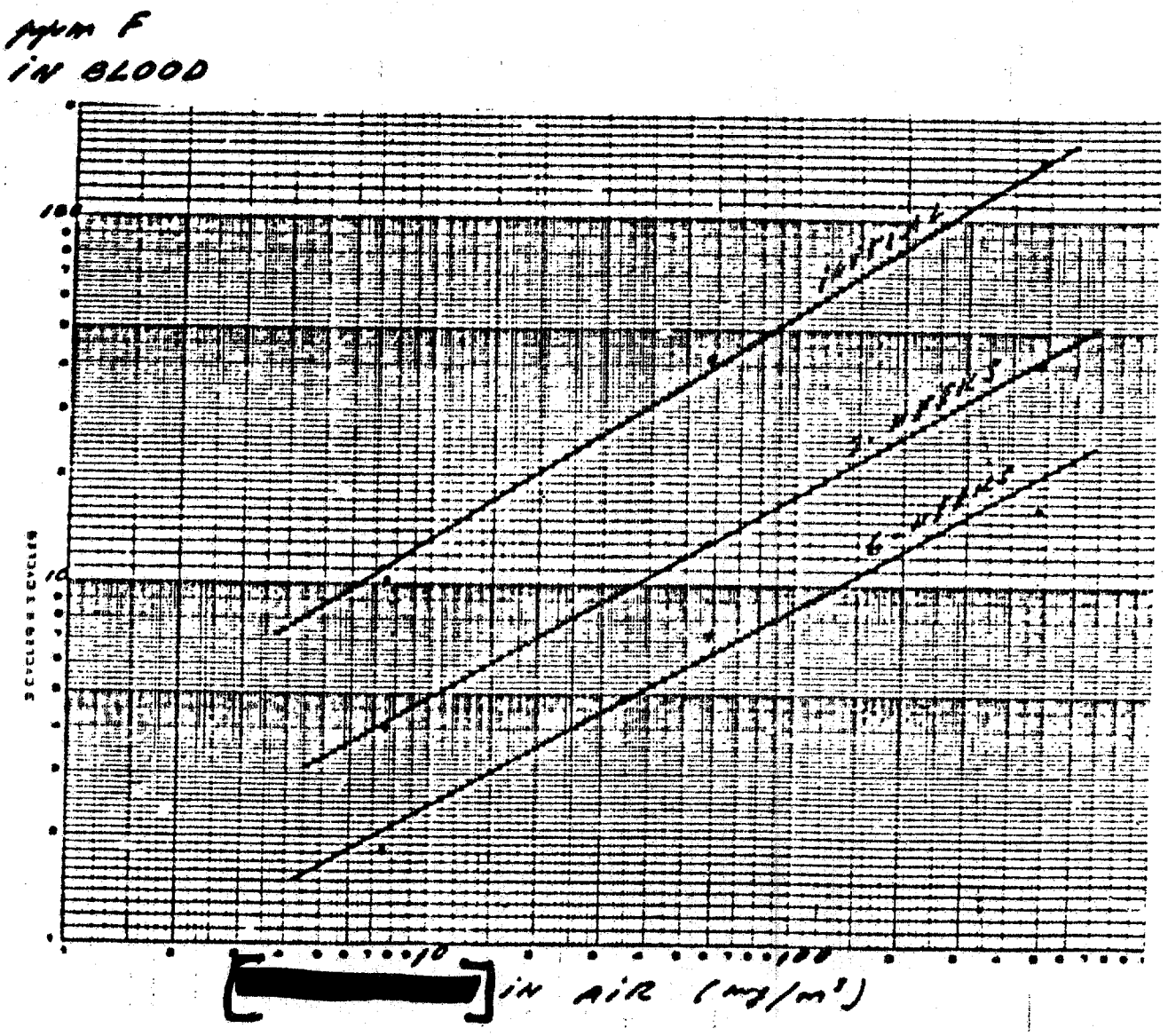

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FIGURE 2

CONCENTRATION OF ORGANIC FLUORINE IN BLOOD VS. THE SQUARE ROOT OF [redacted] CONCENTRATION IN AIR.

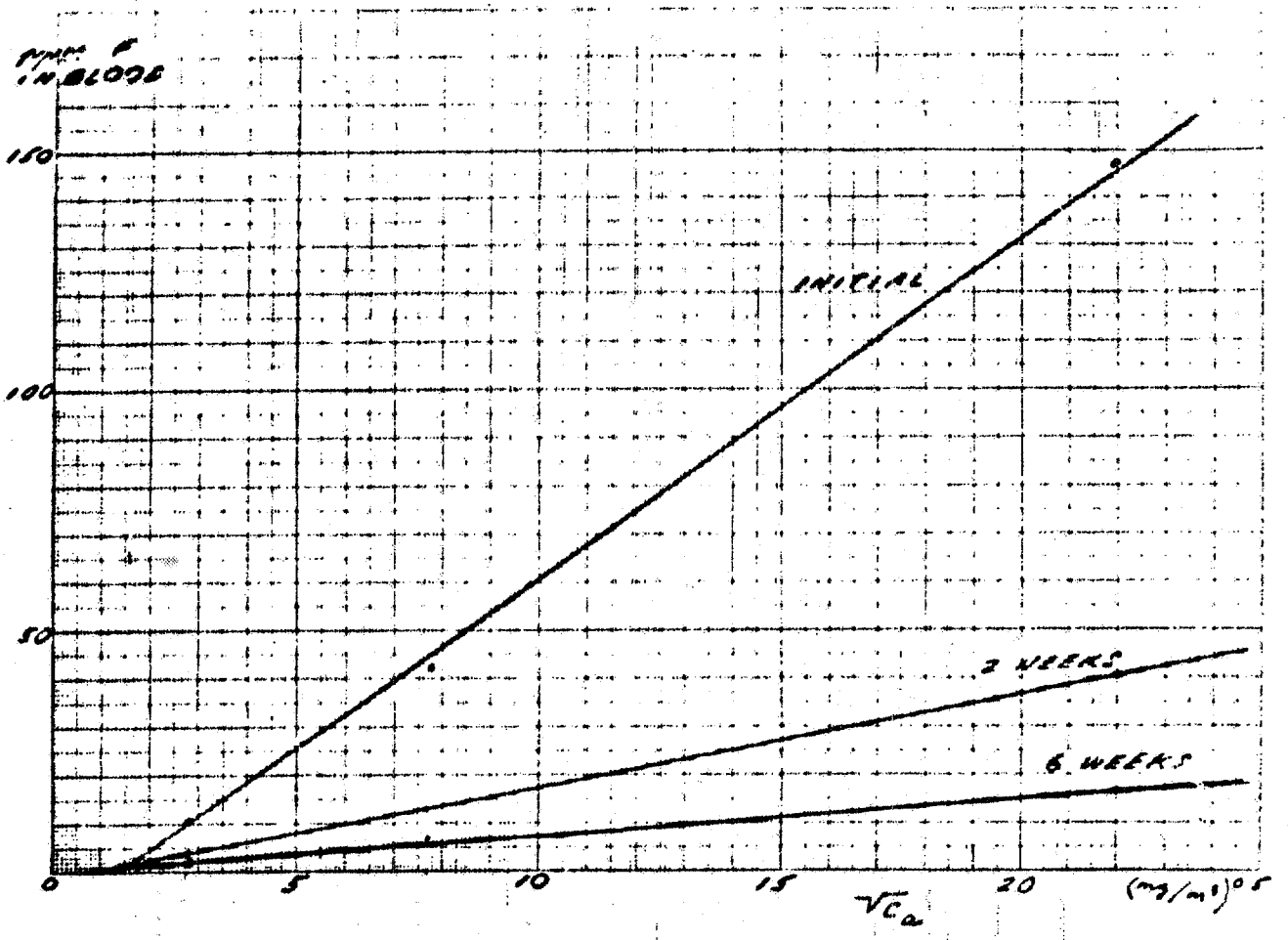


FIGURE 3
A FIRST-ORDER PLOT OF PERFLUOROOCTANOIC ACID
ELIMINATION FROM BLOOD

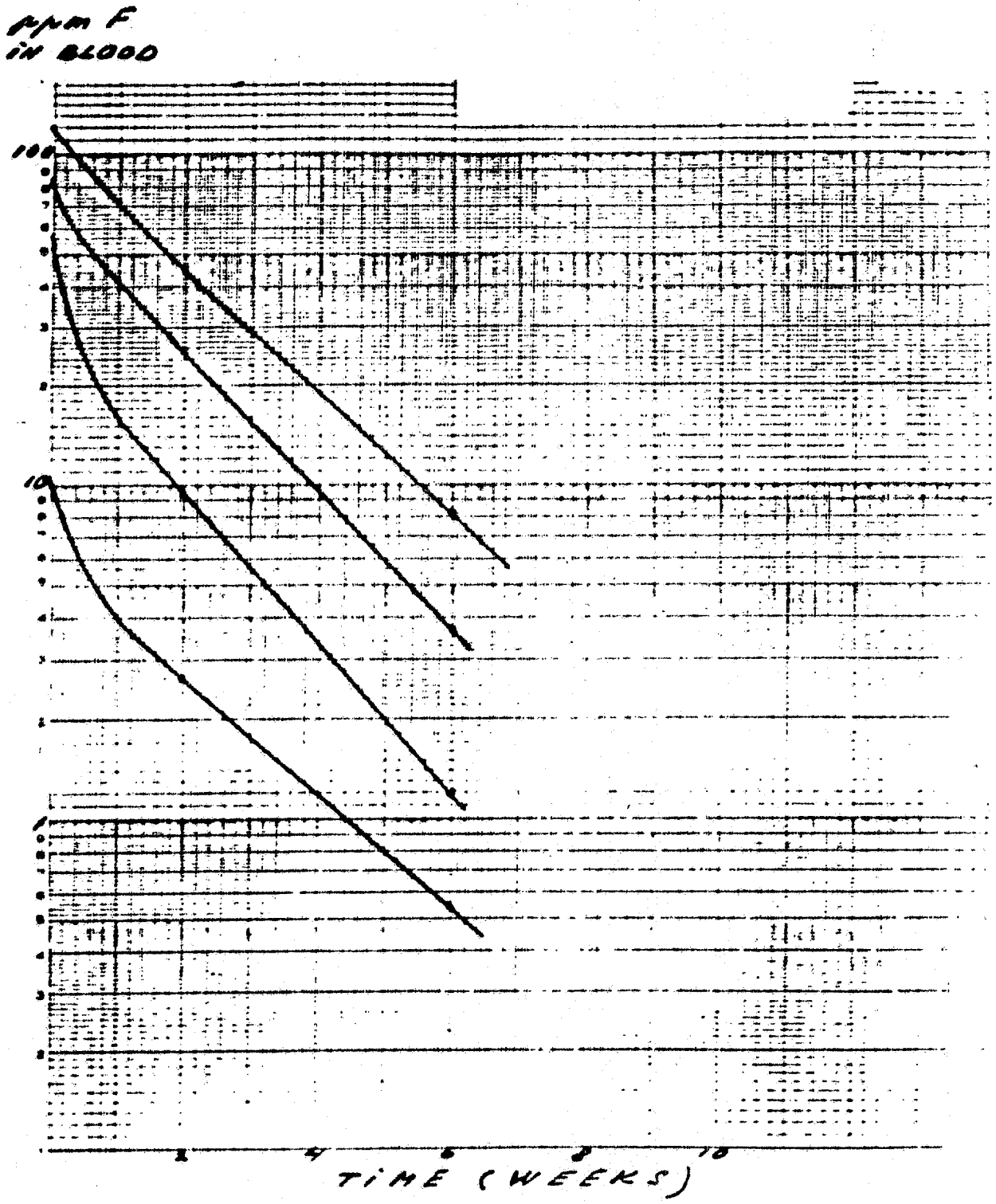


FIGURE 4

A FIRST-ORDER PLOT OF [redacted] ELIMINATION FROM BLOOD

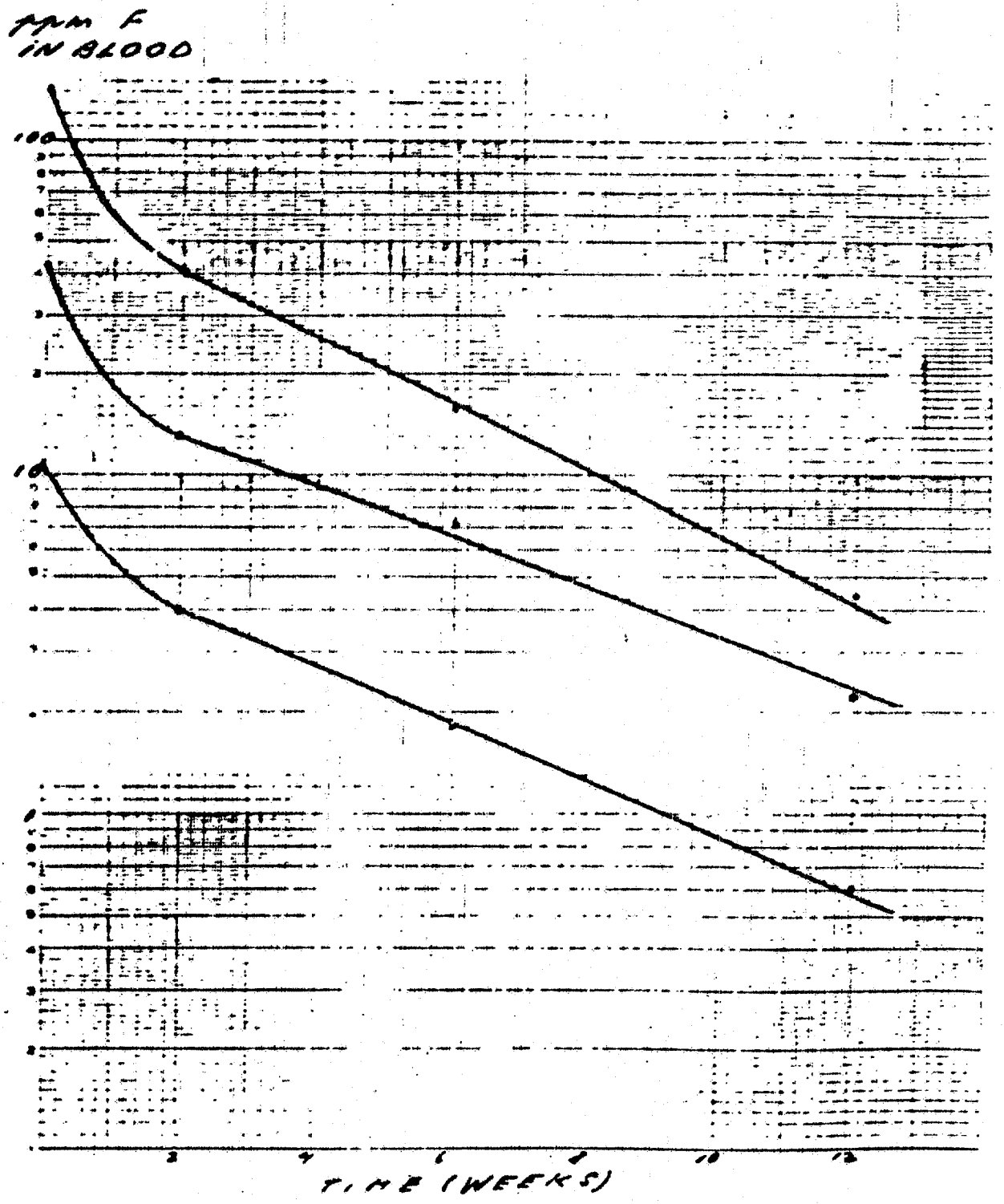


FIGURE 5
THE ELIMINATION RATE OF [redacted]
AS A FUNCTION OF ITS CONCENTRATION IN BLOOD

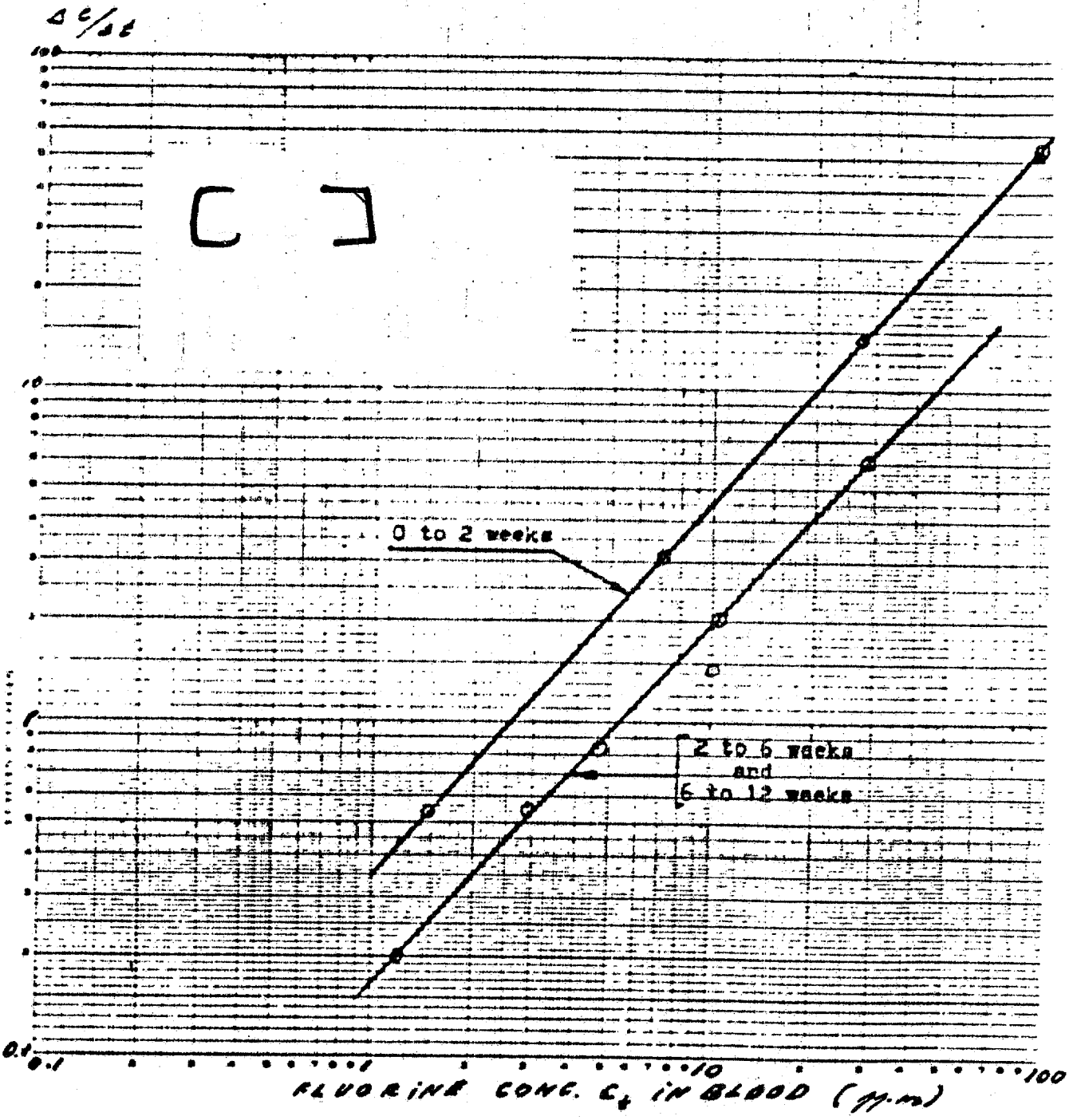


FIGURE 6
THE ELIMINATION RATE OF PERFLUOROOCTANOIC ACID
AS A FUNCTION OF ITS CONCENTRATION IN BLOOD.

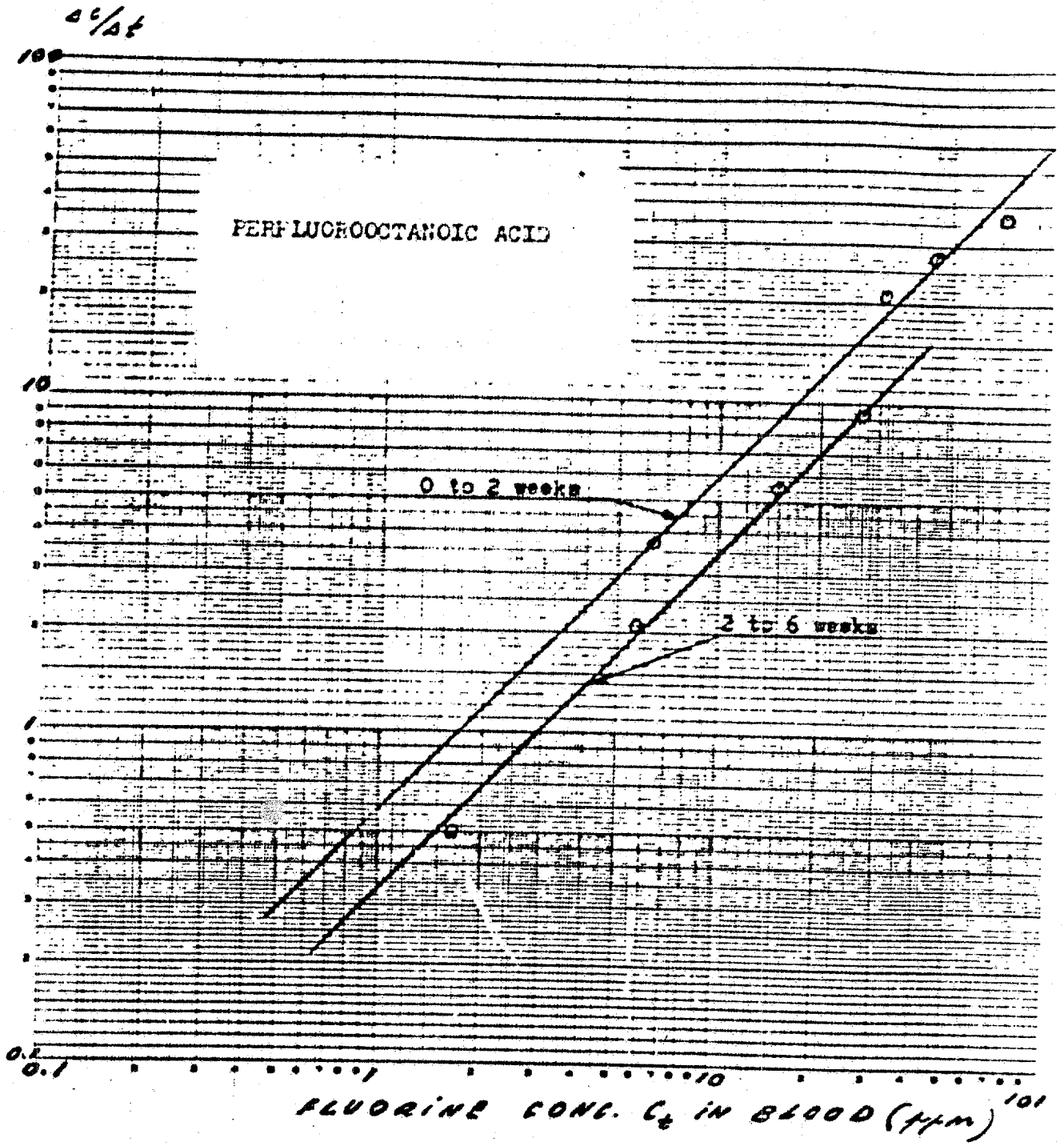
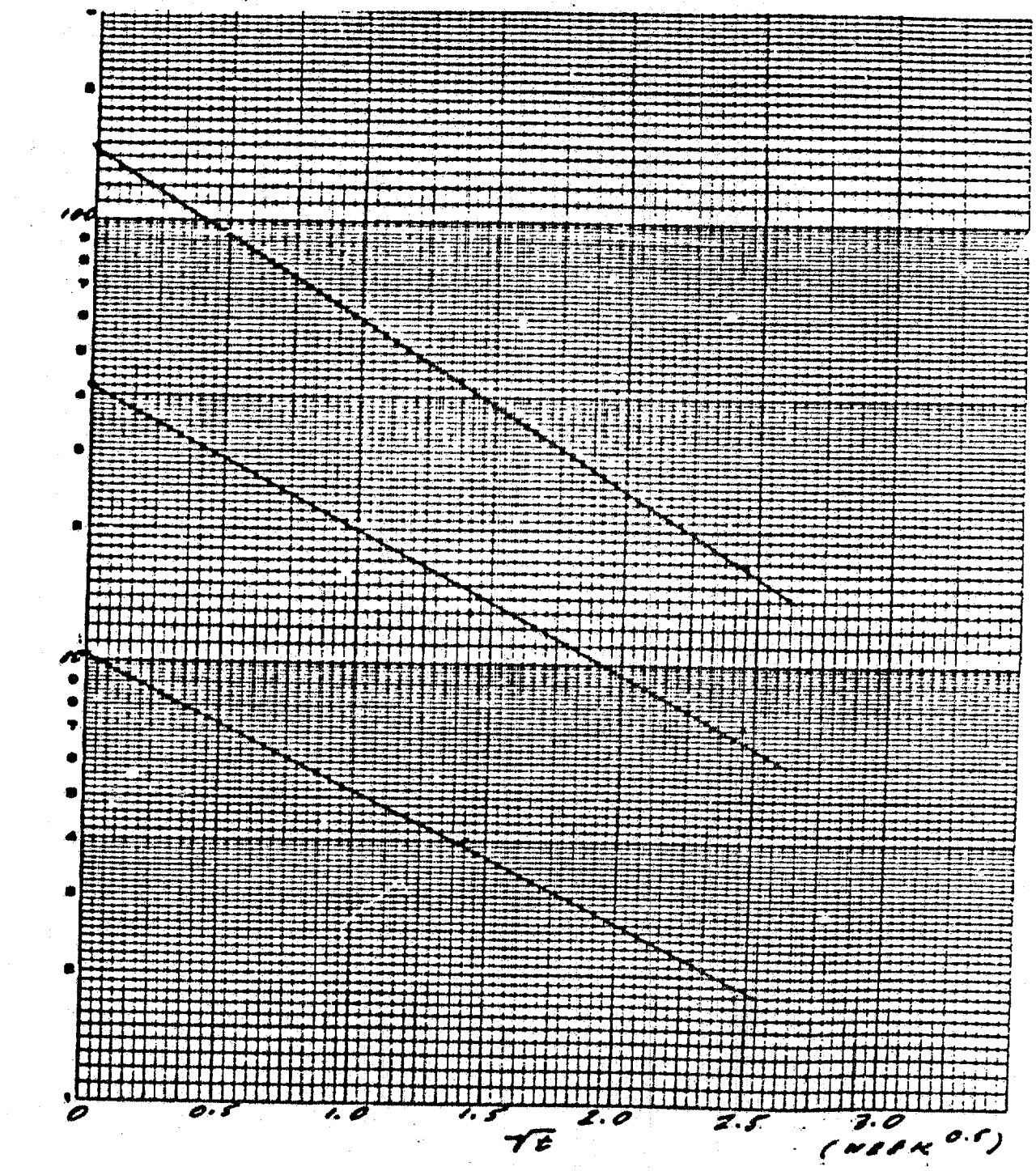


FIGURE 7

[redacted] RELATED FLUORINE IN BLOOD AS A FUNCTION OF TIME



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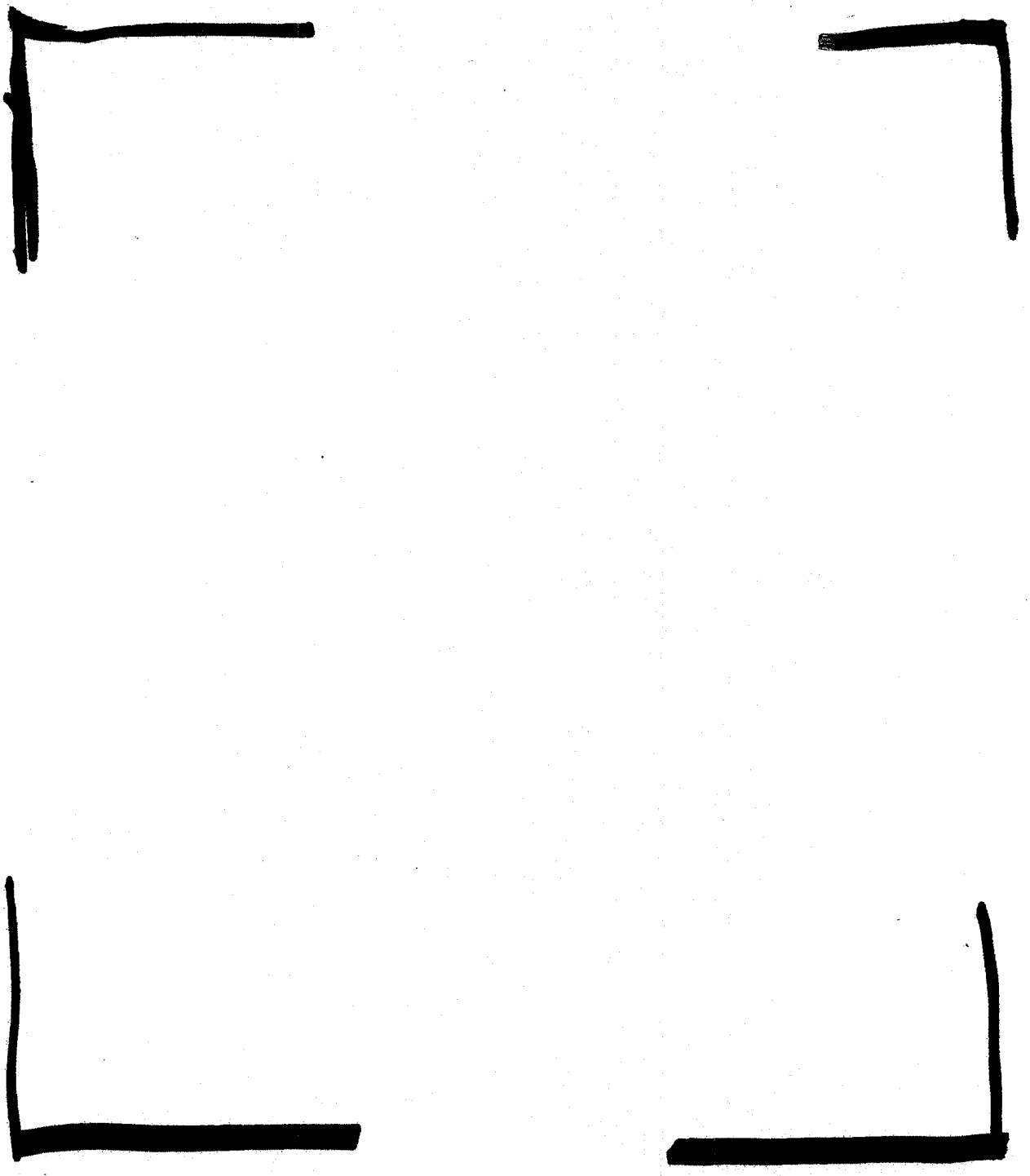

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Erik Kissa

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5-6		Central Report Index
7-10		Jackson Laboratory Files
11	Research Supervisor	H. F. Wyles

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