was the lack of introduction of foreign flavors along with AETS when this was handled in water solution. Although AETS is not presently included in a list of approved food additives, the results are encouraging enough to warrant experimentation on a larger scale.

ACKNOWLEDGMENTS

The authors thank H. E. Vettel and his staff for carrying out the pilot plant operations, and B. S. Logan for conducting the taste panel sessions.

LITERATURE CITED

Ellis, G. P., Advan. Carbohyd. Chem. 14, 63 (1959).
Ferretti, A., J. Agr. Food Chem. 21, 939 (1973).
Field, L., Ferretti, A., Crenshaw, R. R., Owen, T. C., J. Med. Chem. 7, 39 (1964).
Field, L., Owen, T. C., Crenshaw, R. R., Bryan, A. W., J. Amer. Chem. Soc. 83, 4414 (1961).
Hutton, J. T., Patton, S., J. Dairy Sci. 35, 699 (1952).
Josephson, D. V., J. Agr. Food Chem. 2, 1182 (1954).
Keith, E. S., Powers, J. J., J. Food Sci. 33, 213 (1968).
Leviton, A., Anderson, H. A., Vettel, H. E., Vestal, J. H., J. Dairy Sci. 46, 310 (1963).
Liming, N. E., J. Dairy Sci. 49, 628 (1966).
Maillard, L. C., C. R. Acad. Sci. 154, 66 (1912).
Patton, S., J. Agr. Food Chem. 6, 132 (1958).
Schönberg, A., Moubacher, R., Chem. Rev. 50, 261 (1952).

Aldo Ferretti[*1]
Locke F. Edmondson
Frederick W. Douglas, Jr.

Dairy Products Laboratory
Eastern Regional Research Center
Agricultural Research Service
U. S. Department of Agriculture
Philadelphia, Pennsylvania 19118
[1] Present address: Lipid Nutrition Laboratory
Agricultural Research Service
U. S. Department of Agriculture
Beltsville, Md. 20705

Radiation-Induced Dechlorination of Chloral Hydrate and 1,1,1-Trichloro-2,2-bis(p-chlorophenyl)ethane (DDT)

Chloral hydrate and DDT, present in low concentrations in aqueous media, are dechlorinated by moderate doses of γ-radiation. Oxygen and relatively high concentrations of organic material reduce, but do not completely inhibit, chloral hydrate dechlorination.

Concern about pollution of natural waters by persistent organochlorine pesticides has prompted studies of means to destroy these substances in aqueous media. Exposure to ionizing radiation is one way of achieving this since organic halogen compounds are generally rather susceptible to radiation-induced breakdown (Spinks and Woods, 1964), and Sherman and his colleagues (Sherman et al., 1971; Evans et al., 1971) have shown that solutions of DDT in 2-propanol can be dechlorinated by a free-radical chain process initiated by γ-radiation. Radiation-initiated chain dechlorination of organic chlorine compounds has also been observed in aqueous solution (Woods and Spinks, 1960), although little is known about the optimum conditions for complete dechlorination or of the effect of irradiating very dilute solutions. We have therefore investigated the ^{60}Co γ-ray induced dechlorination of dilute aqueous solutions of chloral hydrate, $Cl_3CCH(OH)_2$, which resembles DDT in having a $-CCl_3$ group and has the advantage of being water soluble, and of DDT, which was irradiated as a dispersion on particles of silica gel (Cab-O-sil M5, Cabot Corporation, Boston, Mass.) suspended in water.

Initial yields of acid products from the irradiation of aqueous chloral hydrate solutions are plotted in Figure 1 for air-saturated and oxygen-free (helium purged) solutions. Acid yields are based on conductivity measurements assuming the product to be HCl since preliminary titration with base and with mercuric nitrate (Clarke, 1950) showed HCl to be the major acid product. Dechlorination is initiated by radicals (e_{aq}^-, H, OH) formed with a combined G value (radicals per 100 eV of energy absorbed) of approximately 6 by irradiation of the solvent. The yield of acid is dependent upon solute concentration (Figure 1) and dose rate (Table I) and is highest under conditions conducive of a radical-initiated chain reaction,

i.e. relatively high solute concentration and low dose rate. Oxygen promotes the release of chloride at the higher chloral hydrate concentrations, probably by adding to the intermediate organic radicals to give readily hydrolyzed peroxy radicals, $-Cl_2O_2$. However, with the more dilute solutions oxygen exhibits an inhibitory effect since it competes with the solute for hydrated electrons, forming the rather unreactive species O_2^-. Nevertheless, the absorbed dose necessary to release all the chlorine present in 10^{-5} and 10^{-4} M solutions of chloral hydrate as inorganic chloride (Figure 2) was not markedly different in solutions saturated with air or oxygen or rendered oxygen-free by purging with nitrogen or helium (Table II). The presence of other organic material interfered with, but did not completely inhibit, the release of chloride; for example, 10^{-3} M acetone and 10^{-3} M methanol reduced G(acid) from 10^{-4} M air-saturated chloral hydrate solutions from 8.1 to 2.5 and 2.2, respectively (dose rate 1.2×10^{17} eV g^{-1} min^{-1}). Thus it appears that dechlorination of chloral hydrate is possible under adverse conditions such as irradiation of a dilute solution in the presence of oxygen and a large excess of other organic material.

Initial G(acid) values for the irradiation of DDT on silica gel suspensions are included in Figure 1 and the effect of continuous irradiation on such a suspension is shown in Figure 2. Although virtually complete dechlorination can be achieved it is clearly less efficient than with the chloral hydrate solutions. This is to be expected since (i) the larger DDT molecule can suffer radical attack (e.g., on the aromatic ring systems) without release of chloride, (ii) chlorine attached to the aromatic rings is not as readily released as aliphatic chlorine, and (iii) the molecules of DDT will be less exposed to radicals generated in the solvent than the chloral hydrate molecules in the present system. Indeed, the fact that dechlorination occurs at all,

Table I. Initial G(Acid) Values for γ-Irradiated Aqueous Solutions of Chloral Hydrate

Dose rate,[a] 10^{15} eV g^{-1} min^{-1}	Air-saturated solutions		Oxygen-free solutions	
	0.1 M	10^{-5} M	0.1 M	10^{-5} M
110	54	2.5	14	3.6
8.1			13	5.4
0.93	170	3.1	28	9.6

[a] Measured by ferrous sulfate dosimetry.
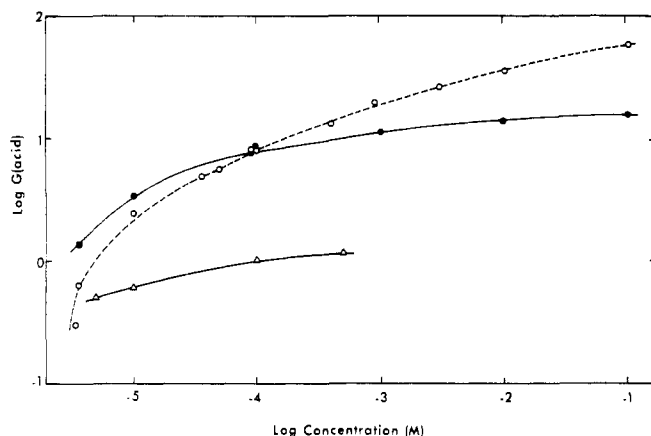
Figure 1. Effect of solute concentration upon initial G(acid) for aqueous solutions irradiated with γ-radiation at a dose rate of 1.1×10^{17} eV g^{-1} min^{-1}: (- - O - -) air-saturated chloral hydrate solutions; (-●-) oxygen-free chloral hydrate solutions; (—△—) suspension of DDT on silica gel in air-saturated solution.
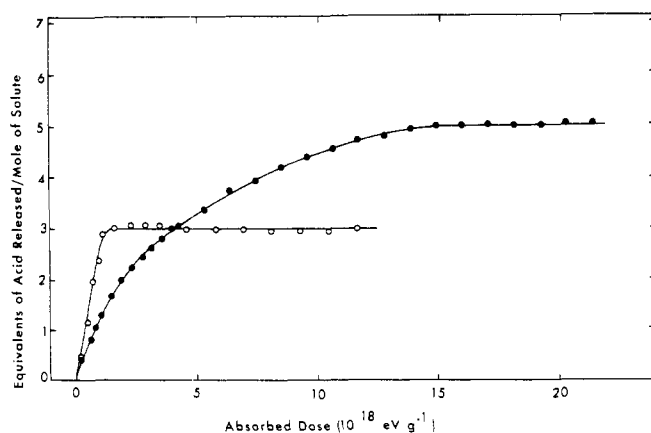
Figure 2. Acid released upon γ-irradiation. Dose rate 1.1 × 10^{17} eV g^{-1} min^{-1}: (- - O - -) air-saturated 10^{-5} M chloral hydrate solution; (—●—) air-saturated aqueous suspension of DDT (10^{-5} M) on silica gel.

Table II. Absorbed Dose of γ-Radiation Necessary to Completely Dechlorinate Chloral Hydrate in Aqueous Solution and DDT on Silica Gel in Aqueous Suspension[a]

Concn, M	Dose to plateau (cf. Fig 2), 10^{18} eV g^{-1}		
	Air-saturated solutions	Oxygen-saturated solutions	Oxygen-free solutions
Chloral hydrate			
10^{-3}	28.6		
10^{-4}	2.8	2.8	2.8
10^{-5}	1.0	0.7	0.7
DDT			
10^{-5}	15	6.5	210
5×10^{-6}	4.5	4.5	9.5

[a] Dose rate 1.1×10^{-17} eV g^{-1} min^{-1}.

particularly in air or oxygen saturated solution, indicates that it can be initiated by O_2^- or by organic radicals such as those produced by the reaction of OH with DDT.

Extraction of the organic material from a 10^{-5} M DDT suspension which had been exposed to 3×10^{18} eV g^{-1} γ-radiation (yielding 2.7 equiv of acid/mol of DDT originally present) and analysis of the extract by gas chromatography showed that ~25% of the original DDT remained and that DDD and DDE, degradation products of DDT in which a CCl_2 group replaces the CCl_3 group originally present, were absent. This suggests that dechlorination proceeds with the simultaneous loss of all three chlorine atoms from the CCl_3 group.

The radiation doses used in the present investigation (2–15×10^{18} eV g^{-1}) are smaller than those required to condition or sterilize sewage ($\sim 10^{20}$ eV g^{-1}) (Ballantine and Miller, 1970) so that radiation treatment of sewage will probably dechlorinate any organic chlorine compounds present in small amounts.

LITERATURE CITED

Ballantine, D. S., Miller, L. A., Chem. Eng. Progr. Symp. Ser. No. 104, 66, 187 (1970).
Clarke, F. E., Anal. Chem. 22, 553 (1950).
Evans, R., Nesyto, E., Radlowski, C., Sherman, W. V., J. Phys. Chem. 75, 2762 (1971).
Sherman, W. V., Evans, R., Nesyto, E., Radlowski, C., Nature (London) 232, 118 (1971).
Spinks, J. W. T., Woods, R. J., "An Introduction to Radiation Chemistry," Wiley, New York, N. Y., 1964.
Woods, R. J., Spinks, J. W. T., Can. J. Chem. 38, 77 (1960).

R. J. Woods*
S. Akhtar

Department of Chemistry and Chemical Engineering
University of Saskatchewan, Saskatoon
Saskatchewan, Canada