

Pacific. Few storms have the intensity to penetrate as far as San Diego; thus the annual rainfall averages only 10 inches per year. We have collected rainfall at the La Jolla station and analyzed it for lead during the past 3 years. Lead concentrations range from 3 to over 300 μg per liter of rainwater, and there is an inverse relationship between the amounts of rainfall and the lead concentration. The average lead contents in La Jolla rainwater is about 40 $\mu\text{g}/\text{liter}$. Lazrus *et al.* (12) reported an average of 36 μg of lead per liter for a U.S. nationwide sampling network. They concluded that the lead content in rainwater at various cities is correlated with the gasoline consumption there. According to the U.S. Public Health Service drinking water standard, the "grounds for rejection" limit for drinking water is 50 μg of lead per liter (13).

In the San Diego region the amount of dust falling was 72 and 113 mg/m^2 per day for the fourth quarter of 1969 for our La Jolla and downtown San Diego stations, respectively. About two-thirds of the La Jolla dust consists of water-soluble matter primarily composed of sea salts. The gross lead content of the La Jolla dust is 0.09 percent, which corresponds to a lead precipitate of 65 $\mu\text{g}/\text{m}^2$ per day. San Diego dust, about one-third of which consists of soluble matter, contained 0.84 percent lead, equivalent to 950 $\mu\text{g}/\text{m}^2$ per day. On the basis of a comparison of the lead contents in the La Jolla and downtown San Diego dust (0.09 and 0.84 percent, respectively) with those of the lead aerosols in the suspended matter (0.68 and 4.62 percent, as given in Table 1), it is clear that only a fraction of the lead aerosols precipitate out near the source of emissions. The remainder of the lead aerosols are carried in the air currents and spread around the globe (14).

A portion of these lead pollutants have undoubtedly been incorporated into plants and animals (15). Long-term increases in atmospheric lead concentration will result in predictably higher concentrations of lead in the blood of the exposed populations (16). Because of the well-known toxicity of lead, the health hazard of increasing lead pollutants in the environment cannot be ignored.

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References and Notes

1. R. A. Kehoe *et al.*, *Arch. Environ. Health* 8, 202 (1964); C. C. Patterson, *ibid.* 11, 344 (1965); State of California, *Lead in the Environment and Its Effects on Humans* (Air Resources Board, Sacramento, California, 1967); U.S. Department of Commerce, *The Automobile and Air Pollution* (U.S. Government Printing Office, Washington, D.C., 1967).
2. U.S. Department of Health, Education, and Welfare, *Survey of Lead in the Atmosphere of Three Urban Communities* [Public Health Serv. Publ. No. 999-AP-12 (1965)].
3. A. P. Altshuler, *Anal. Chem.* 41, No. 5, 1R (1969).
4. T. J. Chow, *J. Water Pollut. Contr. Fed.* 40, 399 (1968).
5. —, J. L. Earl, C. F. Bennett, *Environ. Sci. Technol.* 3, 737 (1969).
6. *Community Air Quality Guide, Lead* (American Industrial Hygiene Association, Detroit, 1969).
7. J. R. Goldsmith, *J. Air Pollut. Contr. Ass.* 19, 714 (1969).
8. T. J. Chow and C. C. Patterson, *Geochim. Cosmochim. Acta* 26, 263 (1962).
9. J. S. Brown, *Science* 147, 502 (1965).
10. T. J. Chow and M. S. Johnstone, *Science* 147, 502 (1965).
11. T. J. Chow and J. L. Earl, *Chem. Geol.* 6, 43 (1970).
12. A. L. Lazrus, E. Lorange, J. P. Lodge, Jr., *Environ. Sci. Technol.* 4, 55 (1970).
13. U.S. Department of Health, Education, and Welfare, *Public Health Service Drinking Water Standards* [Public Health Serv. Publ. No. 956 (1962)].
14. M. Murozumi, T. J. Chow, C. C. Patterson, *Geochim. Cosmochim. Acta* 33, 1247 (1969).
15. T. J. Chow, *Nature* 225, 295 (1970); H. L. Motto, R. H. Daines, D. M. Chilko, C. K. Motto, *Environ. Sci. Technol.* 4, 231 (1970).
16. J. R. Goldsmith and A. C. Hexter, *Science* 158, 132 (1967).
17. We thank C. Snyder for technical assistance. Supported by the PHS grant No. 5-R01-AP-00875 and the Office of Naval Research contract No. Nonr 2216(23). This paper was presented at the Symposium on Geochemical Atmospheric Constituents of the American Chemical Society National Meeting in Houston, 25 February 1970.

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Gadolinium: Distribution between Aqueous and Silicate Phases

Abstract. Studies of the partition of trivalent gadolinium between aqueous and silicate phases have been made at temperatures from 800° to 900°C and at pressures from 500 to 1000 bars. Constant values for the distribution coefficients for forsterite, enstatite, and diopside were obtained over a concentration range from 0.6 part per billion to 4 parts per thousand in the solid phases. Ratios of silicate crystal-aqueous phase distribution coefficients to silicate melt-aqueous phase distribution coefficients are close to the values for silicate crystal-silicate melt distribution coefficients estimated from natural systems. The free ion activity of trivalent gadolinium in the silicate melts appears to be less than one-hundredth of its value for aqueous phases of the same concentration.

The number of thermodynamic parameters that would have to be evaluated in order to predict the behavior of a trace element during solidification of a natural silicate liquid is prohibitive. A promising approach is to measure distribution coefficients for trace elements in the hope that they will be constant over a useful variety of silicate compositions. Such seems to be the case for the rare earth elements since estimates of their distribution coefficients lie within a narrow range. These estimates are derived from studies of several natural systems which include phenocrysts and host matrices treated as equilibrium solid-liquid pairs (1), ultramafic rocks and basalts treated as partial melt-residue pairs (2), and minerals and estimated residual liquids from the Skaergaard intrusion (3). We have obtained laboratory values for distribution coefficients of Gd that are similar to those found for natural systems. Our measurements also show a strong affinity of silicate liquids for that element.

Most of the cations that are essential components of common silicate

minerals are mono- or divalent. Entry of Gd^{3+} into one of the silicate structural sites, therefore, requires charge compensation. In natural silicate melts the concentrations of both major cations and possible compensating anions presumably exceed that of Gd by several orders of magnitude. If so, no conceivable distribution of the Gd between the solid phase and its parent liquid will significantly affect the activities of those ions in either phase. The equilibrium distribution of Gd will then be given by the ratio of the activities of that element for the two phases according to Eq. 1

$$K = \frac{A(\text{Gd}^{3+}, \text{solid solution})}{A(\text{Gd}^{3+}, \text{liquid})} \quad (1)$$

where K is a constant.

In this paper we define a distribution coefficient D as the ratio at equilibrium of the bulk concentration of the trace element in the solid to its bulk concentration in the liquid. This distribution coefficient is related to the constant K as follows, where γ represents the activity coefficient of Gd^{3+}

$$K = \frac{\gamma(\text{Gd}^{3+}, \text{solid solution})}{\gamma(\text{Gd}^{3+}, \text{liquid})} D \quad (2)$$