

reflection coefficients of 20 percent and 2 percent, respectively. Yet in powder form both materials have very high albedos because the volume-scattered rays are weakly absorbed.

If the coatings were metallic they could decrease *R* appreciably by absorption; however, apparently they are nonmetallic. Only strong, wavelength-dependent, volume absorption of the refracted rays can account for the lunar spectrum. Hapke *et al.* (4) have suggested that the large absorbing power of grains of lunar fines is primarily due to a vapor-deposited component of the soil, while Conel and Nash (5) and Adams and McCord (6) have emphasized the importance of shock-melted glass.

The following additional considerations are relevant to the discussion: (i) Bibring *et al.* state that the coatings recrystallize at 800°C. However, heating lunar fines in a vacuum to 900°C causes a negligible change in albedo (4). (ii) The calculated spectrum of the coated interface (1) has a minimum at 3500 Å, below which the reflectivity rises again. However, the spectra of lunar soils continue to decrease below 3000 Å (4). (iii) The indexes of refraction used by Bibring *et al.* are 1.80

and 1.98, whereas values of the order of 1.5 to 1.7 would be much more representative of the major components of lunar soil (anorthite, augite, olivine, ilmenite, and glass).

The interesting correlation between albedo and fraction of grains coated (1) is undoubtedly due to the fact that both phenomena are associated with exposure at the surface of the moon.

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References and Notes

1. J. Bibring, J. Duraud, L. Durrieu, C. Jouret, M. Maurette, R. Meunier, *Science* **175**, 753 (1972); J. Borg, M. Maurette, L. Durrieu, C. Jouret, *Geochim. Cosmochim. Acta* **3** (Suppl. 2), 2027 (1971).
2. H. Hecht, in *Modern Aspects of Reflectance Spectroscopy*, W. Wendlandt, Ed. (Plenum, New York, 1968), p. 1.
3. J. Geake, A. Dollfus, G. Garlick, W. Lamb, G. Walker, G. Steigman, C. Titulaer, *Geochim. Cosmochim. Acta* **3** (Suppl. 1), 2127 (1970).
4. B. Hapke, A. Cohen, W. Cassidy, E. Wells, *ibid.*, p. 2199; B. Hapke, W. Cassidy, E. Wells, paper presented at the Second Lunar Science Conference, Houston, Texas (1971).
5. J. Conel and D. Nash, *Geochim. Cosmochim. Acta* **3** (Suppl. 1), 2013 (1970).
6. J. Adams and T. McCord, *ibid.* **3** (Suppl. 2), 2183 (1971).

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Mercury and Lead in the Greenland Ice Sheet:

A Reexamination of the Data

It is imperative that the natural background content of mercury in the environment be determined. Examination of the Greenland ice sheet offers the opportunity to determine if the concentration of mercury in the atmosphere correlates with human industrial activity (1). Weiss *et al.* have recently reported such a measurement (2). Figure 1 is a plot of the data given in their table 1. The sites were identified by reference to the original work for which the samples were collected (3, 4). The pre- and post-1952 data are from different sites with no overlap in dates to make possible an intersite calibration. Weiss *et al.* conclude from their data that there has been a recent increase in the mercury content of the atmosphere. This conclusion is not unambiguously justified.

Indeed, I propose that the only justifiable conclusion is that no trend is apparent in either the pre- or post-1952 data (although seasonal variations are revealed in the latter). Since in 1952 there was a change of sample collec-

tion sites, the post-1952 data are larger than all the pre-1952 data, and, since there is no overlap in dates between the two sites, it seems very likely that there is a systematic difference between these sites, perhaps the result of vehicular contamination (3). It is enlightening to refer to the original work on these samples in which an effort was made to measure the time dependence of lead in the atmosphere (3).

Table 1. Density of air and mercury in the atmosphere as a function of altitude.

Altitude (km)	Density		
	Air (g/m ³)		Mercury (calc.) (ng/m ³)
	Obs.*	Calc.	
0	1226	1226	1.00
1	1112	1121	0.45
2	1007	972	.20
3	909	865	.09
4	819	770	.04
5	736	686	.02
10	413	384	
20	88	120	

* For a U.S. standard atmosphere. For example, see (1).

The data for the lead content of the parent samples are shown in Fig. 2 and were obtained from table 7 of Murozumi *et al.* (3). In contrast to the data on mercury, a trend is clearly apparent in the pre-1946 lead data, but again interpretation of later data is open to debate. One could argue for a smooth curve from 800 B.C. right up to 1965, or argue, along with Murozumi *et al.* that between the 1933 and 1946 points there is a rapid increase, perhaps due to the increased use of lead additives in gasolines. However, examination of the data shows that there is a third alternative: There could very well be a systematic difference between pre- and post-1952 sites with no rapid increase around 1940. In either case, the burden of interpretation falls upon the 1946 point since its relative largeness appears to fit smoothly with the post-1952 data. In the absence of an actual date overlap between the sites, this smooth fit could contribute confidence that the sites yield consistent data. In the investigation of Murozumi *et al.* (3) the sea salt content of the snow was also measured and the time series for magnesium shows no discrepancy among the several sites. However, even though the arduous sample collection procedure involved many safeguards against contamination, Murozumi *et al.* stated (with respect to the post-1952 sites) that "drifted winter snow from regions contaminated by vehicle traverse may provide the greatest source of locally contaminated snow in samples from the virgin trench site. There can be no question but that some of the winter snows we collected drifted in from great distances away. It is emphasized that dilution effects, after a few tens of km, should make snow contaminated by vehicle traverse indistinguishable from background snow in this region" (emphasis mine). It is worrisome that contamination by exhaust from gasoline-burning vehicles would contribute to an elevation of both the lead and mercury contents of the atmosphere. The validity of the mercury data depends on one's confidence in the compatibility of the data from the two sites, and this confidence rests upon the validity of the lead data, which in turn depends almost entirely upon the single data point of 1946.

Because the time dependence of both the lead and mercury contents in the atmosphere have immediate implications for public health policy, I suggest that, at the very least, additional samples be collected spanning the time from 1930 to 1970 from a single site, and prefer-

ably two sample series from two separate sites. The description of the data collection (3) and the analysis procedures (2) indicates that this is no trivial suggestion, but, in view of the implications, we cannot afford to do less.

The calculation of Weiss *et al.* of the flux of mercury to the atmosphere by outgassing of the earth's crust (1.5×10^{11} g/year) appears to be too large by at least a factor of 10. Although they deliberately sought to keep their calculation on the conservative side by assuming the lowest possible value for the average amount of mercury in the air and a uniformly dense atmosphere at standard temperature and pressure, their calculation is an overestimate because it assumes a uniform concentration of mercury in the air. A simple approximation of the density ρ of the atmosphere as a function of altitude is the still-air, isothermal "law of atmospheres" (5):

$$\rho(y) = \rho(0) \exp(-mgy/kT)$$

where m is the mass of a single gas molecule, g is the acceleration of gravity, k is the Boltzmann constant, T is the absolute temperature, $\rho(0)$ is the density of the atmosphere at the earth's surface, and y is the altitude above the earth's surface. If this law is applied to the partial pressure of mercury vapor, the exponential falloff will be characterized by a distance 6.9 times smaller than that for air ($6.9 = 201/29$, because mercury has a molecular weight of 201 whereas air has a "molecular weight" of 29). Table 1 illustrates this point.

If the total mass M of mercury in the atmosphere is estimated from

$$M = \int_0^{5 \text{ km}} 4\pi \rho(0) e^{-mgy/kT} (R_0 + y)^2 dy$$

where R_0 is the radius of the earth, and if we use the same value for $\rho(0)$ as Weiss *et al.* used, namely, $\rho(0) = 1$ ng/m³, the atmospheric burden of mercury is 6×10^8 g. Even this is an overestimate because the sea covers 70 percent of the earth's surface and the density of mercury in the air above the sea is lower by about a factor of 2 than that over the land (6). Refining this calculation by a weighted average over the land and sea yields the estimate for the amount of mercury in the atmosphere, 4×10^8 g. This value is lower by a factor of 10 than the estimate of Weiss *et al.*, who further calculated the flux from the land to the atmosphere by assuming rains at 10-day intervals that

completely cleanse the air of mercury. This last calculation is erroneous since it presumes that none of the mercury precipitated revolatilizes and reenters the atmosphere. Thus, even though this assumption is known to yield an overestimate, if it is applied to my estimate of the mass of mercury in the atmosphere, the flux of mercury to the atmosphere is 1.5×10^{10} g/year. This value is to be compared with the mercury flux to the atmosphere from the burning

of fossil fuels. Weiss *et al.* quote an upper limit of 1.6×10^9 g/year, and examination of their source [table 1 in (7)] shows that this value derives entirely from the contribution of oil. This number includes an arbitrary reduction by a factor of 10 to allow for uncertainty in the amount of mercury left behind in the ash as compared to the amount vaporized (7), although other authors (8) hold that very little of the mercury is trapped with the fly ash.

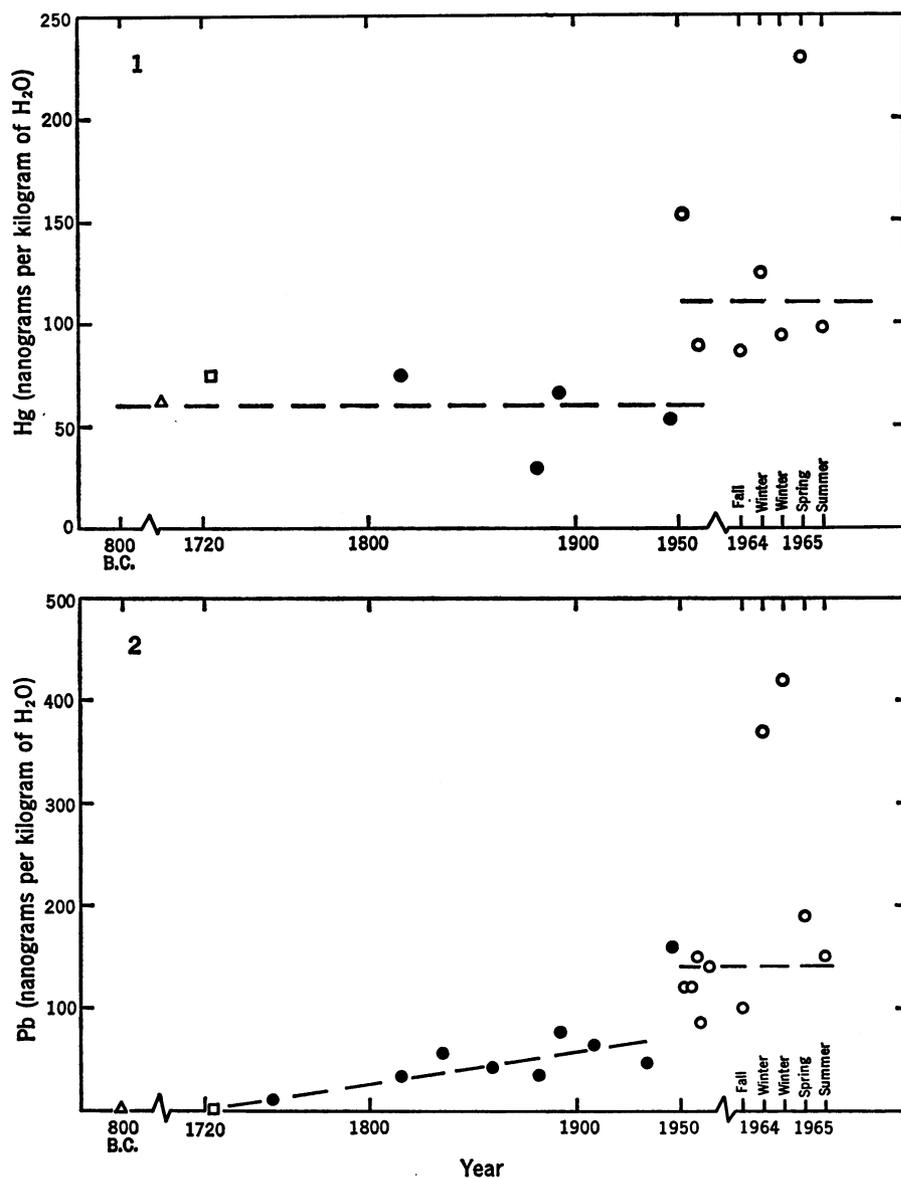


Fig. 1 (top). Plot of the mercury concentration in polar snow packs as reported in table 1 of Weiss *et al.* (2). Different sites of sample origin are indicated by distinct symbols; ●, inclined shaft at Camp Century; ○, virgin trench site. The dashed lines represent, roughly, an interpretation of the data different from that advanced by Weiss *et al.* The discontinuity in the dashed line may be attributable to a systematic difference in the two sites. Note that the 1964 and 1965 data are presented on an expanded horizontal scale to display seasonal variations. Fig. 2 (bottom). Plot of the lead concentration in polar snow packs as reported in table 7 of Murozumi *et al.* (3). Symbols used are the same as those in Fig. 1. The dashed lines represent, roughly, an interpretation of the data different from that advanced by Murozumi *et al.* The discontinuity in the dashed line may be attributable to a systematic difference in the two sites. The single point at 1946 plays an important role in establishing confidence in the consistency of the two sites.

Joensuu has estimated that a conservative value for the flux from coal alone is 3×10^9 g/year (9). Thus the total value for both fossil fuels could be about 5×10^9 g/year. This same estimate was obtained by Hammond who assumed a mercury concentration of 1 part per million in all 5×10^{15} g of all fossil fuels consumed each year (10). Consequently, the burning of fossil fuels may contribute as much as one-third of the total mass of mercury in the atmosphere. Clearly, the conclusion of Weiss *et al.* that industrial activity does not contribute an amount of mercury to the atmosphere comparable to that produced by the natural outgassing of the crust is not justified.

One cannot have very much confidence in any of these estimates. All the data show wide variations, and the measurements of the mercury content in the atmosphere are so poorly documented that there is no way to judge the extent to which the mercury measured is in its elemental form or the amount that is adsorbed on particulates. If a substantial fraction is adsorbed on particulates, the estimates calculated here on the basis of the exponential law of atmospheres are also incorrect. The entire question of the amounts, sources, and time distribution of mercury in the atmosphere remains an open and compelling question.

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References and Notes

1. E. M. Dickson, *BioScience* 21, 450 (1971).
2. H. V. Weiss, M. Koide, E. D. Goldberg, *Science* 174, 692 (1971).
3. M. Murozumi, T. J. Chow, C. C. Patterson, *Geochim. Cosmochim. Acta* 33, 1247 (1969).
4. For a further discussion of the data reported in (3), see also J. Tinker [*New Sci.* 52, 49 (1971)] and M. K. Williams (*ibid.*, p. 235).
5. R. P. Feynman, R. B. Leighton, M. Sands, *Lectures on Physics* (Addison-Wesley, Reading, Mass., 1963), vol. 1, chap. 40, p. 1.
6. *U.S. Geol. Surv. Prof. Pap.* 713 (1970).
7. K. K. Bertine and E. D. Goldberg, *Science* 173, 233 (1971).
8. L. Dunlop, *Chem. Eng. News* 49, 22 (5 July 1971).
9. O. I. Joensuu, *Science* 172, 1027 (1971).
10. A. L. Hammond, *ibid.* 171, 788 (1971).
11. S. L. Hess, *Introduction to Theoretical Meteorology* (Holt, Rinehart & Winston, New York, 1959), p. 85.
12. I thank R. M. Cotts and A. N. Garroway for useful discussions.

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Dickson questions our interpretations of the lead and mercury concentrations in the Greenland ice sheet (1, 2), concentrations which appear to us to have increased in recent years. Our concern is with the validity of the assumptions upon which his criticism is based.

1) "... there is no overlap in dates between the two sites, it seems very likely that there is a systematic difference between these sites, perhaps the result of vehicular contamination." This interpretation is in direct contradiction to the data presented by Murozumi *et al.* (1). The following information is abstracted from that report. The virgin site was 80 km upwind (east-south-east) from Camp Century, which, in turn, was 210 km upwind (east) from Thule. Within the 1964-1965 annual cycle, lead concentrations at the virgin site were three times higher in winter than in summer snows, although (except at Thule) there was no vehicular traffic during the winter months. Winter snows did not breach the hoarfrost layer of the autumn of 1964. The 1965 spring snow layer was analyzed for lead at the virgin site, at the periphery of Camp Century, and at a location between the two sites. The results of these analyses, 0.19, 0.27, and 0.20 $\mu\text{g}/\text{kg}$, respectively, indicate that the Camp Century and virgin collecting sites were linked together by horizontally extensive layers of snow that contained approximately uniform ambient concentrations of lead for given time horizons. Moreover, these studies show that at a distance of only 1 km from the source of local exhaust, lead concentrations are elevated only about 50 percent above the ambient background. They further show from a material balance standpoint that local exhaust effects even from Thule in the winter, should the wind occasionally blow from that direction, would be completely undetectable above the ambient background. Thus, the uppermost firn strata samples collected were free from contamination by local vehicular exhaust, and overlap did exist between the different sites at which older and younger samples were collected.

2) "... their calculation is an overestimate because it assumes a uniform concentration of mercury in the air. A simple approximation of the density ρ of the atmosphere as a function of altitude is the still-air, isothermal 'law of atmospheres'" (3). Dickson has transformed a model of an isothermal atmosphere into a law. The following was abstracted from the same reference used by Dickson (3): [The fractionation of such gases as nitrogen and oxygen as a result of gravitational attraction] "... does not really happen in our own atmosphere, at least at reasonable heights, because there is so much agitation which mixes the gases back together again. It is not an isothermal

atmosphere." If our calculation is an overestimate, or possibly an underestimate, it is not for this reason cited by Dickson.

3) "This last calculation is erroneous since it presumes that none of the mercury precipitated revolatilizes and re-enters the atmosphere." There is some merit to the revolatilization argument, but it is limited to that mercury which falls back to land. For that which falls to the sea surface, which constitutes two-thirds of the earth's surface, the argument loses its force because volatilization of dissolved species of mercury would most probably involve a small fraction of that present. However, a correction factor of 1/3 applied to the highest of three estimates, which span nearly an order of magnitude, is not of great significance to our arguments.

4) Finally, Dickson attempts to increase the estimates of the amount of mercury mobilized during the burning of fossil fuels by combining poorly known numbers. The petroleum value he cites is described in the following way (4): "This Hg value may be unrepresentative of petroleum in general. It is the only published value for Hg in fuel oils of which we are aware. The samples came from the Cymric oil field of California, an area near known Hg deposits." Furthermore, we believe that the mobilization value of 3×10^9 g/year attributable to Joensuu (5) should be reduced since no correction was made for the mercury retained in the bottom ash or incorporated in coke. Nonetheless, to enlarge or diminish poorly known values is of little avail in understanding man's involvement in the disposition of mercury about the earth's surface.

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References

1. M. Murozumi, T. J. Chow, C. C. Patterson, *Geochim. Cosmochim. Acta* 33, 1247 (1969).
 2. H. V. Weiss, M. Koide, E. D. Goldberg, *Science* 174, 962 (1971).
 3. R. P. Feynman, R. B. Leighton, M. Sands, *Lectures on Physics* (Addison-Wesley, Reading, Mass., 1963), vol. 1, chap. 40, p. 1.
 4. K. K. Bertine and E. D. Goldberg, *Science* 173, 233 (1971).
 5. O. I. Joensuu, *ibid.* 172, 1027 (1971).
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