tap water and fed Chlamydomonas reinhardi tap water and ted Chlamydomonas reinhardi and Ankistrodesmus falcatus, easily assimilated green algae that are natural, high-quality foods of Daphnia [D. W. Schindler, J. Anim. Ecol. 37, 369 (1968); F. B. Taub and A. M. Dollar, Lim-nol. Oceanogr. 13, 607 (1968); D. Arnold, ibid, 16, 906 (1971)]. Algae were cultured axenically in Woods Hole medium [J. Stein, Ed., Phycolog-ical Matheds (Plonum Now York, 1973) with In woods hole medium [3, stem, Ed., *Phycological Methods* (Plenum, New York, 1973)] with glycylglycine buffer and without silicate. They were saturation-labeled by adding 20 μc of NaH³CO₃ in 1 m lof sterile distilled water to 50 ml of log growth phase culture 2 days before use. Algae were saturation-labeled in the same way with $K_2H^{33}PO_4$ for autoradiographic stud ies. Experimental feeding suspensions were pre-pared by centrifuging, washing, and resuspending algae in the appropriate experimental feeding medium at the desired algal concentration. Concentrations were determined as cell counts and dry weight. Feeding and assimilation rate deter minations were made adapting procedures of D. W. Schindler (cited above), J. E. Schindler [J. Anim. Ecol. 40, 589 (1971)], and D. Arnold (cited above). Adult D. magna were sorted and certed above). Adult D. magna were sorted and equivalent-sized animals were acclimated to ex-perimental conditions for 2 hours before use. Samples of D. magna (20 to 30) were pipetted into 300-ml bottles filled with aquarium water that had been filtered through $0.45 - \mu m$ HA Millipore filters; the concentration of ¹⁴C-labeled algae was at least 0.1 mg (dry weight) per millili-ter. This is well above the incipient limiting concentration at which feeding rates are maximal and are independent of food concentration. Bottles were rotated at 1 rev/min to keep animals and food in suspension. Feeding rates were determined by removing animals after 10 min-utes, rinsing for 1 minute in unlabeled algae, and utes, rinsing for 1 minute in unlabeled algae, and immobilizing them in boiling water. Feeding pe-riods of up to 1 hour, used by previous workers, exceed the gut passage time for most *Daphnia* species. This experimental error yields er-roneous, low feeding rates and excessively high calculated assimilation efficiencies. Radio-activity ingested per animal and activity per milliliter of feeding suspension were determined by liquid scintillation (D. Heisey and K. G. by liquid scintillation (D. Heisey and K. G Porter, in preparation). Assimilation rates were determined by removing animals after 1 hour and allowing them to clear their guts of radio-active algae for 1 hour in a suspension of unlabeled algae. Radioactivity incorporated and re tained in the animals was determined by liquid scintillation. The relation between this net value and absolute assimilation measurements is dis-cussed by W. Lampert [Verh. Int. Ver. Theor. Angew. Limnol. 19, 2913 (1975)]. Assimilation efficiencies were calculated as 100 × (dpm incorporated per animal per hour)/(dpm ingested per animal per hour), where dpm is disintegrations per minute.

- per minute. Daphnia magna were fed a suspension of $5 \times 10^{9} (12 \mu m)$ plastic beads per milliliter for 1 hour to flush their guts. They were then pipetted into 300-ml light or dark bottles of feeding suspen-sion containing 2.5×10^{3} cells per milliliter each of labeled A. falcatus and unlabeled S. schroe-teri. One hour of feeding on a rotating plankton wheal (1 ray(min) allowed on pinale to fill their wheel (1 rev/min) allowed animals to fill their guts but not produce or reingest significant mounts of feces
- Methods were adapted from G. W. Fuhs and E Methods were adapted from G. W. Fuhs and E. Canelli, Limnol. Oceanogr. 15, 962 (1970); M. L. Brock and L. D. Brock, Mitt. Int. Ver. The-or. Angew. Limnol. 15, 1 (1968); H. Rogers, Techniques of Autoradiography (Elsevier, Am-sterdam, ed. 2, 1973). Kodak NTB-3 emulsion was used. Incubation times for ¹⁴C and ³³P slides were 32 and 59 days, respectively.
 B. H. Ketchum, J. Cell Comp. Physiol. 13, 373 (1939); G. P. Fitzgerald, J. Phycol. 6, 239 (1970).
 Daphnia magna were fed saturation-labeled A.
- 10. Daphnia magna were fed saturation-labeled A. falcatus for 1 week before use. Their guts were flushed of algal remains (7) and they were fed unlabeled, log growth phase S. schroeteri (5 > 10^3 colonies per milliliter) for 1 hour. Animal were then transferred to a suspension of plastic
- beads to aid in gut evacuation.
 Daphnia magna were fed a suspension of S. schroeteri. Feces were collected and streaked schroeteri. Feces were collected and streaked onto five sterile plates of 2 percent agar (Difco-Bacto) in distilled water. Five plates of algae from the same culture, but which had not been fed to D. magna, were also made. Cells were visually monitored at ×400. Values in the text are the means \pm standard deviations for five replicate plates and represent an increase over 24 hours not the accurate for the means resulting the means result 24 hours, not the population parameter r. 12. Equivalent amounts of S. schroeteri in station
- ary growth phase were placed in five tubes with 10 ml of algal culture medium (6) deficient in nitrogen, phosphorus, and vitamins and in five

tubes with 10 ml of deficient medium in which D magna swam (two animals per milliliter) for 2 hours before use. To each tube 0.5 μ c of NaH¹⁴CO₃ was added and all the tubes were incubated at 20°C for 2 hours in cool white fluorescent lighting (100 microeinstein m⁻² sec⁻¹). Carbon-14 fixation rates in deficient and Danhuig-enriched medium ware 554 ± 124 Sec $^{\circ}$). Carbon 14 match rates $^{\circ}$ 12 Daphnia-enriched medium were 554 ± 124 dpm/ml and $12,596 \pm 982$ dpm/ml, respectively. Porter, Am. Sci., in press

J. F. Haney, Arch. Hydrobiol. **72**, 87 (1973). G. E. Hutchinson, Am. Sci. **3**, 269 (1973). Low pH and the CO₂ supplied during gut passage may also stimulate green algal growth [J. Shapiro, Science 179, 382 (1973); ibid. 182, 306 (1973); J.

C. Cohen, *ibid.*, p. 306]. I thank R. K. Trench for advice on experimental methods. R. K. Trench, J. Porter, S. Ohlhorst, D. Janzen, C. Yocum, J. Korstad, D. S. Weth-ey, and D. Titman provided valuable comments on the neuronaire L. Ukwitte D. M. 16. on the manuscript L. Hewitt, D. Heisey, T. Nerad, and K. Belen lent technical assistance. Research was supported by NSF grant BMS 75-11893

3 February 1976; revised 16 April 1976

Properties of the Background Global Aerosol and Their Effects on Climate

Abstract. Properties of the aerosols above Hawaii, Alaska, and the South Pole are derived from sun photometry at several wavelengths. The mass loading of aerosol material is several milligrams per square meter. At the South Pole the mean particle radius is 0.04 micrometer; at Hawaii in March 1975 there was a thin volcanic layer with a mean particle radius of 0.1 micrometer. The aerosols cause heating of the earth-atmosphere system at the poles and cooling at low latitudes.

Minute particles suspended in the earth's atmosphere (the atmospheric aerosols) interact with the atmospheric radiation field and potentially can affect climate by increasing or decreasing the radiation that passes into or out of the earth-atmosphere system (1). The aerosol influence on the heat balance interests scientists mainly because of the possibility that fluctuations in the aerosol concentrations may be responsible for climatic changes. For instance, there is evidence of a correlation between dust layers found in ice cores and the temperature (as determined from oxygen isotopic analyses) at the time the layers were deposited, with dusty periods corresponding to lower temperatures (2)

The earth's aerosol load is known to undergo natural variations due to occasional injections of ash and gas into the stratosphere by volcanic eruptions (3)and to variations in surface sources, such as changes in vegetation or changes in the size of desert areas. Of particular interest, however, is the possibility that the global aerosol load is also significantly altered by human activity, with effects ranging from pollution of the stratosphere by supersonic transports to industrial process pollution. In view of this, it is important to establish a quantitative measure of so-called baseline data on aerosols to use as a reference against which future changes can be gauged.

In this report I discuss the background aerosol parameters at the Mauna Loa Observatory (MLO), Hawaii, and the South Pole, as derived from precision multiwavelength measurements of the total vertical optical atmospheric transmission made by using the sun as a standard

source of irradiance over the wavelength interval $400 < \lambda < 1000$ nm. The measurements were made at the South Pole in December 1974 and at MLO in March 1975. In addition, selected data acquired in Alaska are discussed.

The total vertical optical extinction of sunlight in the earth's atmosphere is conveniently expressed in terms of an optical depth, $\tau_{\rm T}$, and is due to molecular or Rayleigh scattering, $\tau_{\rm R}$; gaseous absorption, $\tau_{\rm G}$, and scattering and absorption by the atmospheric aerosols, τ_A . The latter term is of interest here and was derived by subtracting tabulated values of $\tau_{\rm B}$ and values of $\tau_{\rm G} = \tau O_3 + \tau NO_2(4, 5)$.

The aerosol optical depth curves (aerosol extinction spectra) as a function of wavelength were used to derive aerosol parameters by referring to curves calculated from Mie theory, assuming that the aerosols (i) are spherical with an index of refraction n = 1.5 + i0.002 and (ii) are distributed by size according to a modified gamma size distribution function defined by the relationship dn/dr = $a r^2 e^{-br}$, where dn/dr is the number density of aerosol particles and r is the particle radius (6). With appropriate values chosen for the coefficients a and b, the modified gamma size distribution function defined by the relationship dn/dr = $a r^2 e^{-br}$, where dn/dr is the number density of aerosol particles and r is the particle radius (6). With appropriate values chosen for the coefficients a and b, the modified gamma distribution function can closely simulate actual tropospheric or stratospheric aerosol size distributions derived from direct sampling techniques. Although the fit of the modified gamma distribution function may not always be perfect, when a and b are properly chosen it probably represents a reasonable approximation to the actual vertically integrated aerosol size distribution function.

The results of least-squares fitting of the aerosol extinction spectrum to the modified gamma distribution function (7) are expressed here in terms of Γ , the columnar aerosol mass loading (assuming an aerosol material mass density of 1.0 g cm⁻³), and the radius value at which dn/dr is maximum, $r_{\rm m} = 2/b$. Average values of these two parameters as observed at various times and places and their observed ranges are presented in Table 1.

Table 1 shows that the minimum aerosol loading occurs at the South Pole station (SPS) at a mean value of about 2.3 mg m⁻²; the particle concentration is greatest at $r = 0.04 \ \mu$ m. The same value of Γ is occasionally also approached in interior Alaska but, somewhat surprisingly, the mass loading over the frozen Arctic Ocean near Barrow has always been observed to be larger than this by a factor of 2 to 10, possibly because of the formation of ice crystals due to open leads in the pack ice or the carry-over of anthropogenic pollutants into the Arctic basin from central Europe.

The Antarctica data were taken at SPS in early austral summer 1974–1975 during a time when the stratospheric turbidity was at a minimum (8) (Mount Fuego in Guatemala had erupted 2 months earlier, but most likely its effects had not reached Antarctica at the time of the measurement), and therefore the listed values of aerosol parameters should be close to the absolute minimum aerosol loading found on the earth.

Hogan (9) reports that the surface aerosol concentration at SPS is about 50 cm⁻³. Assuming that this value represents a typical aerosol concentration in the troposphere, we conclude that the tropospheric aerosol density is less than 0.3×10^8 cm⁻², compared to a total column number density (inferred from the optical extinction measurements) of 1.1×10^8 cm⁻². This comparison, although rough, suggests that most (about 70 percent) of the aerosol load over SPS is probably contained in the stratosphere. This is supported by a comparison of the calculated number of stratospheric particles with radii larger than 0.15 μ m with the vertically integrated counting data from optical measurements from balloon reported by Hofmann et al. (10); both of these imply that there are about 0.8×10^6 aerosol particles with $r > 1.6 \,\mu m$ per square centimeter. The good agreement found may be partly fortuitous since the method used to obtain the aerosol size distribution 25 JUNE 1976

Table 1. Average aerosol parameters deduced from optical atmospheric extinction measurements. Abbreviations: Γ , mass of aerosol material in a vertical column (assuming density = 1 g cm⁻³); r_m , radius where dn/dr is maximum; N, number of particles (in millions) in a vertical column; and τ , aerosol extinction optical depth (referred to base *e*) evaluated at $\lambda = 550$ nm. Error limits are calculated for uncertainties in n_r , which may be between 1.33 (water) and 1.55 (silicates); measurement errors are also taken into account. A double-peaked modified gamma distribution function is used at Mauna Loa: D1 is the background Aitken aerosol and D2 is the volcanic aerosol layer at 20 km.

Location	Γ (mg m ⁻²)	r _m (µm)	$N \times 10^{-6}$ (cm ⁻²)	τ
South Pole station (90°S)	2.3 + 0.9 - 0.7	0.04 ± 0.01	114 + 47 - 33	0.011 ± 0.002
Mauna Loa Observatory (19.5°N)				
DI	$2.3 + 0.9 \\ - 0.7$	0.04 ± 0.01	114 + 47 - 33	0.011 ± 0.002
D2	1.6 + 0.9 - 0.7	0.09 ± 0.02	7.4 + 3.0 - 2.1	0.012 ± 0.002
Alaska				
Near Fairbanks (64.8°N)	7.3 + 3 - 2	0.09 ± 0.02	48 + 20 - 14	0.055 ± 0.002
Barrow (71.3°N)	9.6 + 4 - 3	0.07 ± 0.02	76 + 31 - 22	0.075 ± 0.022

function from optical measurements of atmospheric extinction is most accurate for particles in the size range $0.1 < r < 0.5 \,\mu\text{m}$ and is much less accurate for particles in the Aitken size range. Uncertainties in the particle size distribution function at the small-particle end (Aitken particles) can seriously affect estimates of particle number density, although such uncertainties will be of little consequence for computed radiative fluxes.

The size distributions of aerosols above SPS in December 1974 and above MLO in March 1975 are very different. From the assumed modified gamma distribution, at SPS the peak number density concentration, dn/dr_m , occurs at about $r_{\rm m} = 0.04 \ \mu {\rm m}$, while at MLO the peak occurs at about 0.09 µm. The difference in $r_{\rm m}$ at SPS and MLO is puzzling, especially since the average particle size is smaller in the Antarctic than it is at MLO. One would expect the opposite, since the Antarctic aerosols (which are probably first generated at low latitudes) should take a longer time to reach Antarctica, and continual coagulation of the Aitken particles would increase the mean particle size.

At the time of the MLO measurements (March 1975) there was a thin layer of particulates near the tropopause, evidenced by a colorful volcanic twilight and by lidar measurements made at MLO, which probably resulted from the eruption of either Mount Fuego in Guatemala or Ngauruhoe volcano in New Zealand. This suggested that the aerosol size distribution near MLO might be more reasonably given by a double-peaked modified gamma distribution, with one distribution to represent the normal intervolcanic conditions and another superim-

posed distribution to represent the particles in the volcanic layer. To obtain the optical characteristic of the volcanic stratospheric layer, I assumed that the Aitken nucleus load at MLO was the same as that at SPS and subtracted its optical depth from the total observed aerosol optical depth at MLO. The resultant curve of $\tau(\lambda)$ was then used to obtain an estimate of the columnar aerosol properties of the thin stratospheric layer above MLO. This method yielded a columnar number concentration (for the volcanic laver) of 7.4 \times 10⁶ cm⁻², with a mean radius of 0.09 μ m and columnar mass loading of 1.6 mg m⁻². This layer is probably composed of sulfate particles (11) and exists in addition to the hypothesized much thicker layer of Aitken nuclei of nonvolcanic origin. The inferences about the properties of the sulfate layer are consistent with the twilight and lidar observations.

What is the cause of the substantial count of Aitken nuclei at MLO and SPS? Smoluchowsky's coagulation theory (12) predicts that the particle size distribution observed at SPS ($r_m \approx 0.04 \ \mu m$, $n \approx 1000$ cm⁻³) is less than about 2 weeks old. Although stratospheric meridional transport processes are understood poorly, it is estimated that it takes on the order of months for air to diffuse from the equatorial zones into the polar regions. I thus surmise that the Aitken particles above Antarctica are likely formed in the subpolar latitude zones. Perhaps they are formed by homogeneous nucleation of trace gases or by oxidation of organic terpenes by stratospheric O₃. Terpenes are introduced into the atmosphere by vegetation and in the equatorial zones are carried aloft in the Hadley cells, where they could then diffuse poleward, eventually

estimated by using the theory developed by Chylek and Coakley (13). The value of the imaginary index of refraction for the aerosol material was estimated to be $0.000 < n_i < 0.010$ by comparing monochromatic measurements of the zenith sky radiance with radiance values calculated for several values of assumed n_i . The value $n_i = 0.002$ employed in the heating and cooling calculations indicate that the aerosols cause a slight heating (averaged over the year) of the earth-atmosphere system in the polar regions of about 0.06°C (assuming radiative equilibrium and neglecting climatic feedback mechanisms) and a cooling in the low latitude zones of about 0.2°C. These figures are for the background aerosols, far from significant surface sources. If the tropospheric aerosols over the continents, and especially the aerosols over the deserts, were included, the low-latitude cooling would probably increase. Indeed, estimates of desert surface albedos and desert aerosol loads, based on turbidity measurements made in northern Kenya, suggest that additional cooling is likely to result from the continental aerosol sources.

numerical calculations are These somewhat tentative since they are based on a rough determination of the imaginary component of the index of refraction n_i . Additional information is needed on n_i . In the meantime, calculations for various possible values of n_i indicate that the major conclusions reached here always hold (heating at the poles, cooling near the equator), although the numerical values could change upward or downward within a factor of 2 or so, depending on the actual value of n_i .

The increased cooling caused by the presence of the aerosol column over the equatorial regions and the slight aerosolinduced polar heating decrease the heat contrast between pole and equator, and this will act to slightly alter the atmospheric circulation (compared to an aerosol-free atmosphere) and may lead to a net cooling in the polar regions due to decreased advective heat transport, especially during the polar night. Ultimately, to derive the actual climatic impact of the aerosols, one must incorporate the aerosol parameters into a climate model that takes account of the various climatic feedback mechanisms and infrared heating terms. Until this is done, it can be stated roughly that the aerosols act to slightly heat the polar regions and cool the equatorial regions.

GLENN E. SHAW

Geophysical Institute, University of Alaska, Fairbanks 99701

References and Notes

- 1. Report of the Study of Man's Impact on Climate, Inadvertent Climate Modification (MIT Press, Cambridge, Mass., 1971), sections 8.2 to 8.7
- L. Thompson, W. L. Hamilton, C. Bull, Ant-arct. J. U.S. 8 (No. 6), 340 (1973). Although the evidence of correlations between dusty periods and cool temperatures suggests volcanic dust effects, there are other possible explanations for instance, changes in the circulation patterns accompanying cool periods could have led to increased windblown dust near the Antarctic coastline. Thus, the evidence relating global cooling to dusty periods in the paleoclimatic records suggests, but does not prove, that vol-
- canic dust veils cause climatic change.D. Diermendjian, Volcanic Dust Effects D. Dichmengian, *voicanic Dust Effects—A Critical Survey* (Report R-886-ARPA, Rand Corp., Santa Monica, Calif., 1971).
 G. E. Shaw, J. A. Reagan, B. M. Herman, J. *Appl. Meteorol.* 12, 374 (1973).
 J. F. Noxon, *Science* 189, 547 (1975).

- 6. D. Deirmendjian, Electromagnetic Scattering on Spherical Polydispersions (American Else-vier, New York, 1969).
- 7.
- M. A. Box and S. Lo, preprint (School of Physics, University of Melbourne, 1975).
 P. B. Russell, W. Viezee, R. D. Hake, Jr., *Lidar Measurements of the Stratospheric Aerosol* (Project 4019 report, Stanford Research Institute Strategied Cov^[5], 1976). 8.
- (Project 4019 report, Stanford Research Institute, Stanford, Calif., 1975).
 9. A. W. Hogan, J. Appl. Meteorol. 14, 550 (1975).
 10. D. J. Hofmann, J. M. Rosen, G. L. Olson, Antarct, J. U.S. 10 (No. 4), 189 (1975).
 11. R. D. Cadle and G. W. Grams, Rev. Geophys.
- R. D. Cadle and G. W. Grams, *Rev. Geophys.* Space Phys. **13**, 475 (1975).
 N. A. Fuchs, *The Mechanics of Aerosols* (Pergamon, Oxford, 1964), pp. 288–352.
 P. Chylek and J. A. Coakley, Jr., in *Climate of the Arctic*, G. Weller and S. Bowling, Eds. (Geophysical Institute, University of Alaska, Fairbanks, 1973), pp. 159–165.
 Supported by NSF grant OPP73-05829-AO2 and State of Alaska funds.

5 January 1976; revised 6 April 1976

Metamict Mineral Alteration: An Implication for Radioactive Waste Disposal

Abstract. Pervasive alteration is a common feature of metamict minerals. Previous studies of metamict niobium-tantalum-titanium oxides and electron microprobe analyses of altered areas in metamict yttrialite document the leaching of uranium, thorium, and rare earth elements. These data suggest that glasses may not provide a stable medium for radioactive waste disposal.

A recurring theme in discussions of radioactive waste disposal is the possibility of immobilizing the radioactive wastes in a silicate or borate glass. The stability of

Table 1. Electron microprobe analyses of unaltered and altered zones in metamict yttrialite from the Rode Ranch pegmatite, Central Mineral Region, Texas. The unaltered material was analyzed by B. Bruun and B. Jensen, Minerologisk Museum, Oslo (9). Values for the altered material are averages from 15 analyses by R.C.E. Abbreviation: ND, not determined.

Metal oxide	Unaltered	Altered
SiO	27.35	26.95
Fe ₃ O ₃	2.64	12.02*
FeO	0.10	{ 3.02*
MnO	1.68	1.64
MgO	0.05	0.05
CaO	0.89	1.64
U ₂ O ₂	2.09	1.56
ThO ₂	11.98	10.67
La ₃ O ₂	0.30	0.25
CeO ₂	1.33	1.28
Pr ₂ O ₂	0.27	0.30
Nd ₂ O ₂	2.30	2.10
Sm ₂ O ₃	1.79	1.61
Gd ₂ O ₃	3.11	3.05
Tb ₂ O ₃	0.56	0.50
Dv ₂ O ₃	3.78	3.64
Ho ₂ O ₃	0.68	0.58
Er ₂ O ₃	2.30	2.12
Tm ₂ O ₃	0.43	0.32
Yb_2O_3	2.54	2.11
Lu_2O_3	1.13	1.10
Y_2O_3	29.14	28.21
H ₂ O	3.84	ND
Total	100.28	92.70

*Oxidation state not determined by microprobe anal-. calculated as Fe₂C

such a glass over long periods of time (for example, more than 100 years) is an important consideration, and one that is difficult to evaluate from laboratory experiments of necessarily more restricted time periods. One possible method of evaluating the stability of such glasses is to examine metamict minerals for alteration effects.

Metamict minerals are naturally occurring materials that were once crystalline but, because of later effects, are now amorphous (1). Although the exact mechanism for the transition from crystalline to metamict state is poorly understood, radiation damage from constituent uranium and thorium nuclides must be critical to the process (2-4). Thus metamict minerals provide an ideal model for studying glasses that have suffered radiation damage over geologic periods of time.

The most detailed studies of alteration of metamict minerals have been of complex Nb-Ta-Ti oxides (5, 6). The alteration is of two types: (i) primary or hydrothermal and (ii) secondary or weath-Previously reported electron ering. microprobe analyses of eight AB₂O₆type Nb-Ta-Ti oxides [A = rare earth]element (REE), Fe2, Mn, Ca, Th, U, or Pb; B = Nb, Ta, Ti, or Fe^{3+}] document the effect of primary and secondary alteration (6). With primary alteration there is a consistent increase in Ca, generally a decrease in U and Th, a decrease in the REE, a slight decrease in B-site cations, and an increase in structural and absorbed water. Secondary alteration is

SCIENCE, VOL. 192

1336