on other phosphates such as ribonucleoside triphosphates or *p*-nitrophenyl phosphate.

We have also found an ecto–adenosine triphosphatase and an ecto–*p*-nitrophenyl phosphatase on these cells (*15*), and others have reported the hydrolysis of added substrates by intact cells of other types (*16*). The existence and characteristics of such ecto-enzymes are of interest in the study of the interaction between cells and their external environment, although the function of such enzymes is still obscure. The ecto–adenosine monophosphatase described above provides a rigorous marker for the plasma membrane of guinea pig polymorphonuclear leukocytes. It remains to be established whether or not the 5′-nucleotidases of other mammalian cells are also ecto-enzymes.

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The Size of Suspended Particle Matter in Air

We agree with Lee (*1*) that the size distribution of particles suspended in the air is important. However, we disagree with several of his conclusions. His data, obtained with an Anderson cascade impactor operated at five times the design flow rate, are plotted exclusively on logarithmic-probability coordinates. A log normal distribution was assumed, the points were fitted to a straight line, and mass median diameters were calculated.

Examination of the graphs presented shows that from 50 to 85 percent of the collected aerosol mass was found on the first stage and the after filter, and was therefore not sized. However, even though only 15 to 50 percent of the collected mass was sized, Lee assumed that the size distributions could be approximated in all cases with a log normal distribution. Examination of Lee's figure 3 indicates that the data for Cincinnati, Ohio, and Washington, D.C., would be fitted better by a curve than by a straight line.

Whitby *et al.* have presented evidence that the mass distribution of atmospheric aerosol is usually bimodal with one mode occurring below 1 μm and another in the 5- to 15-μm range (*2*). If a reasonable curve is drawn through Lee's points, and the resulting data are transformed into a graph of $\Delta m/\Delta \log D_p$ on a linear scale against log D_p, it is seen from Fig. 1 that the Lee data may also generate a bimodal distribution. Thus, we believe that in general the mass distribution of atmospheric aerosols is not log normal. In particular, we believe that Lee's data should not be used as evidence for a single mode log normal distribution.

Obtaining mass median diameters by extrapolation of the data on a log normal plot to sizes below the last data point leads to some unusually low mass median diameters, such as 0.02 μm for

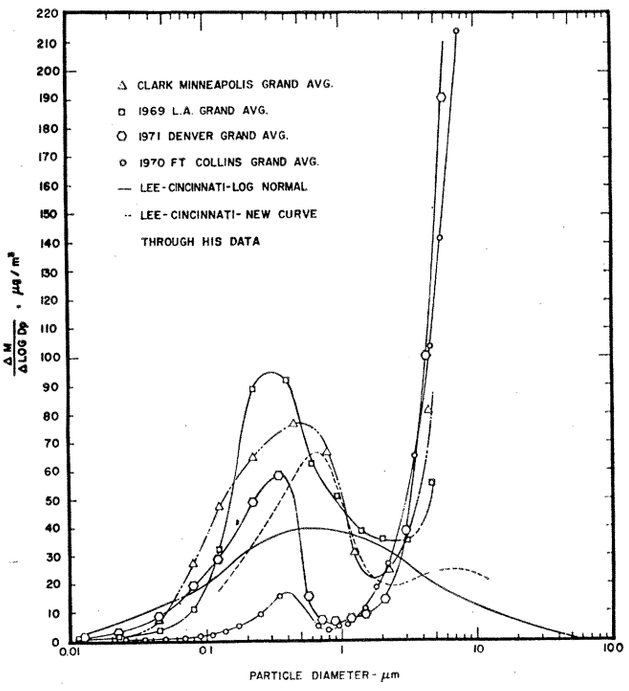

Fig. 1. Mass distribution graph of the grand averages from four cities as measured with the Minnesota aerosol analyzing system compared with Lee's six-city log normal size distribution average. The dashed line shows the Cincinnati average distribution redrawn so that the curve goes through the actual points measured rather than those obtained on the assumption that it is log normal. The bimodal nature of the distributions is produced, in contrast to the broad unimodal distribution resulting from Lee's assumption that the distributions are all log normal.

the Eskadalemuir Observatory shown in Lee's table 2. Lee's average mass median diameters for urban sites are lower than those found by other workers with Anderson impactors operated at lower flow rates (*3*).

We question whether the overall geometric standard deviations of actual atmospheric aerosols are as broad as those presented by Lee. Whitby *et al.* have found that there is usually less than a few percent of mass below 0.1 μm (*2*). Thus, geometric standard deviations obtained from the data in the 0.5- to 3.5-μm range are probably too high by as much as a factor of 2 or 3.

Lee also states that ". . . large particles scatter more light than the predominantly submicrometer-sized aerosols present in urban air." Others (*4*) have found that on the order of 90 percent of visibility degradation (caused by light scattering) is caused by the 0.1- to 1.0-μm diameter particles. On the basis of optical scatter per mass, the 0.1- to 1.0-μm decade is about five times more efficient than the 1.0- to 10-μm decade of particle size. It is the submicrometer particles that usually determine the degree of visibility in urban air. For Lee's inversion aerosols, mass median diameter (MMD) of 1.99 and 1.32 μm, the particles below 1.0 μm account for 70 and 79 percent, respectively, of the calculated light scattering refractive index (wavelength = 1.5 and 550 nm) (*5*).

In addition, since the two modes of the bimodal distribution have different sources, control decisions based on a single log normal distribution may then be ineffective. Apart from the interpretation of the data, there are several experimental problems:

1) The calibration of the sampling device was not discussed. Both the impactor characteristics and the large size cutoff (caused by curved flow stream lines entering the sampler housing and going into the first stage orifices) should be experimentally tested.

2) The errors in the method were not considered, nor was their influence on the results mentioned. These include—in addition to calibration problems—particle bounce, fracture, reintrainment, and volatilization in the low pressure of the after filter. The problems of particle bounce and reintrainment are known to increase when the flow rate is increased above the 1–cubic foot per meter design flow rate (*6*); Lee used 5 cfm.

3) The cascade impactor data were not compared to data from any other work or from other methods. Such comparisons are necessary in view of the limitations and biases of any given method of measurement.

4) We think it is useful to consistently plot aerosol size distribution data (such as number, area, volume, and mass) on several different sets of coordinates. These different graphing approaches are necessary to provide insight into the relevant integral properties of the system, such as total volume, surface area, and light scattering.

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Whitby *et al.* (*1*) have brought out a number of worthwhile criticisms which require comment. It should be emphasized, however, that size distributions presented in my article (*2*) were based on mass measurements while size distributions referenced by Whitby *et al.* were calculated from particle count measurements. Because count methods measure volatile particles which are lost with mass measurement techniques, it is difficult to relate mass size distributions to number size distributions.

The National Air Surveillance Network (NASN) cascade impactor used in our studies was designed to have a particle capture velocity equivalent to the high volume sampler which is EPA's reference method for measuring airborne "suspended particulate matter" (*3*). The advantages of cascade impactors to determine mass distributions are that the size fractionated aerosol is physically collected for subsequent chemical analysis, each unit is relatively low in cost, and their ruggedness and simplicity of operation make them especially amenable for field monitoring use. Impactors, as well as other sizing methods, suffer from inherent well-known problems, discussed by Ranz and Wong (*4*), including calibration inaccuracies, inlet collection efficiency, loss of particles on the walls, and particle bounce off.

A theoretical calibration of the NASN impactor based on the theory of Mercer (*5*) and Ranz and Wong (*4*) was preferred to an experimental calibration with laboratory-generated aerosols because laboratory aerosols differ from aerosols suspended in ambient air. Ordinarily polystyrene latex spheres or liquid generated aerosols are used for laboratory calibration. However, polystyrene latex spheres, which act like "ping pong balls," can bounce off impaction surfaces and also vary in diameter size up to $\pm$ 10 percent (*6*). Similarly, liquid generated aerosols may splatter on a collection surface or shatter if in a crystalline state and be reentrained. Microscopic examination reveals the irregular shape of ambient aerosols, which are "sticky"—being derived in part from incomplete fuel combustion of carbonaceous material (*7*).

The samples we collected with the NASN impactor showed no indication of bounce off as evidenced by "trailing" around the impacted material. I believe that the "sticky" nature of ambient particles, which contain from 16 to 20 percent total carbon (*7*), accounts for the adhesion on the uncoated aluminum collection surface. This brings up the point of a coated versus an uncoated collection surface. Although some data are in disagreement, Lundgren has reported (*8*) no difference in collection efficiency between coated and uncoated impaction surfaces. Hu (*9*) has suggested the use of glass fiber filters as an impaction surface to reduce reentrainment; however, his tests were made with polystyrene latex spheres and, in my opinion, he overlooks the possibility of retaining irregularly shaped particles by "filtration" as the airstream penetrates the porous surface, thereby biasing the distribution toward a larger particle size.

Expressing a particle size distribution as a log normal function (plotting the logarithm of the size as a function of the cumulative percentage mass $\leq$ the

stated size on a probability scale) has been the most widely used and generally accepted method of treating aerosol data since the 1930's (*10*). The major advantage of a log normal plot is that the size distribution can be directly correlated with lung desposition (*11*) based on the mass aerodynamic (Stokes) equivalent diameters. The log-normal size curves presented in my article can be used to assess the aerosol fraction which can penetrate and be retained in various portions of the respiratory system. However, there are many other ways to plot size data depending on what the investigator wants to determine.

As Whitby *et al.* point out I have used extrapolated data to obtain mass median diameter (MMD) values in some cases. (It is interesting that Whitby *et al.* have also yielded to the same temptation in their figure by extrapolating some of my replotted data beyond the upper size limit of about 4 μm in diameter.) I have reported (*12*) that some uncertainty may be associated with an MMD value derived by extrapolation and have pointed out that "values for the average percent of the particle mass $\leq$ 1 μm diameter . . . interpolated directly from the particle size distribution curves . . . provide a more accurate picture of the particle size than does the MMD, a value often obtained by extrapolating the distribution curve" (*13*).

The only other large body of mass size distribution data has been published by Lundgren (*8, 14*). His values published for both the MMD's and the geometric standard deviation are in good agreement with my findings (*1*). Patterson's work (*15*) referred to in the letter by Whitby *et al.* presents size data that are somewhat higher in MMD values than the findings presented by Lundgren and my group. The difference may be attributed to the presence of a nearby emission source, a higher particle capture velocity than the high volume sampler, or weighing problems. (A particulate concentration of 100 μg/m^3 sampled at 1 cubic foot per minute and fractionated over seven stages would amount to only 0.57 mg per stage, provided that there was equal loading, and represents an extremely small amount of material to weigh accurately.) Why there should be a relation between concentration and size of ambient particulate matter as indicated by Patterson is not clear.

Since my article was published, evidence has been accumulating that aerosols may be distributed in more than one mode. Expressing the distribution as single mode log-normal may be an oversimplification, although previous work by Lundgren (*8, 14*) and Lee (*2, 13*) indicated a log-normal plot well approximated the data obtained from available impactors. Treating four- and five-stage impactor data in any more depth than I have reported probably exceeds the limitations of these devices; that is, the size resolution is too poor to define subtle differences in the distribution. An eight-stage impactor system recently developed by Lundgren (*16*) apparently has sufficient resolution to characterize bimodality on a mass basis. This is encouraging since the article (*2*) cites the need for better instruments which would provide more effective size fractionation with better resolution of particle sizes than presently available devices.

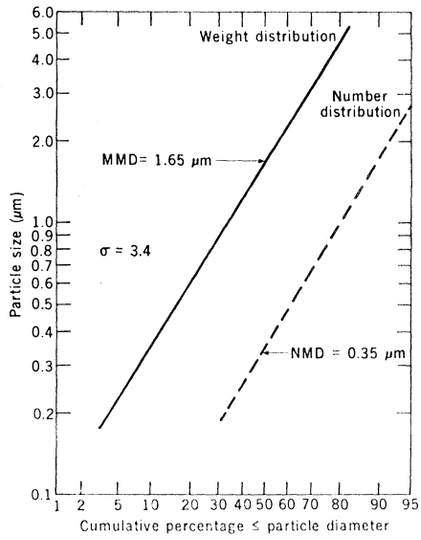

Fig. 1. Composite weight and transformed number distribution of cascade impactor samples collected in Cincinnati, Ohio, on 4 and 21 April 1971, during temperature inversions (*13*).

Visibility reduction is an optical effect of particles. In general, particles that have an optical size of about 0.2 to 1.6 μm diameter on a number distribution basis are most effective in scattering light. In Fig. 1, I have replotted a composite of the inversion data (*2*) both as a mass distribution and a calculated number distribution (*17*), recognizing that volatile particles are probably not accounted for in the transformation. On a mass basis, the MMD is 1.65 μm and constitutes the reference to "large" particles in my article, but on a number basis the MMD is 0.35 μm, which is precisely in the effective scattering range.

Aerosols are difficult to measure and represent the most complicated air pollutant to characterize. Increasing attention from researchers should result in a better understanding of aerosol sources, atmospheric interactions, and methods of control. Of particular importance, in my view, is the need to characterize the chemical composition of suspended particulate matter as a function of size for assessing the inhalation health hazard. Cascade impactors are especially suitable for collecting sufficient quantities of size fractionated materials for chemical analysis.

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Radiocarbon Dates for Earliest Domesticated Animals from Europe and the Near East

In entitling our article "Earliest radiocarbon dates for domesticated animals," we followed the practice of keeping titles concise and brief (*1*) even though Bökönyi, Braidwood, and Read have taken exception to this (*2*). We then specified that "Europe is added to the Near East as another early center of domestication" (*1*, p. 235). Thus we indicated clearly at the outset which