Comments on "Oceanic Aerosol Levels Deduced from Measurements of the Electrical Conductivity of the Atmosphere"

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1. Introduction

The geographic distribution and the chronology of the concentration of atmospheric aerosols are a vital research interest. It is necessary to know the geographic distribution of the aerosol concentration in order to estimate the relative strengths of continental and marine sources both natural and anthropogenic, and to identify the meteorological regimes which act as aerosol sinks. With knowledge of the geographic distribution and the history of aerosols at some remote areas, one might estimate the role of particles in maintaining the chemical balance of the atmosphere and evaluate the possibility of climate modification by aerosols resulting from fossil fuel combustion or agriculture. Cobb has approached this problem by measuring electrical conductivity of the atmosphere, along several oceanic routes and at Mauna Loa, and has related these conductivity measurements to aerosol concentrations. His results are very creditable especially with respect to the limited number of data points available.

2. Theoretical background

The electrical conductivity of the atmosphere has been used at many locations in the past 60 years to provide, among other parameters, an index of the level of suspended particulates. With instrumentation available for measuring the number concentration of aerosols directly, one should be rather careful in overemphasizing the value of conductivity measurements. Conductivity λ is defined by

$$\lambda = nek, \tag{1}$$

where n is the number density of ions, e the elementary charge, and k the mobility of ions. Here, n and k are variables that depend on the atmospheric composition as well as on variations in the rate of ionization. The mobility of ions is, in part, determined by their nature, which is established by the gaseous composition of the atmosphere at the time and place of measurement. Of great influence are trace gases (Mohnen, 1971). As far as the ion density is concerned, it is governed by the well-known Schweidler (1919) expression

$$\frac{dn}{dt} = q - \alpha n^2 - \int_{R\min}^{R\max} \beta(R) f(R) dR, \tag{2}$$

where q is the rate of ionization (ion pairs formed per cm^3 and sec), α the recombination coefficient, $\beta(R)$ the attachment coefficient of small ions onto aerosol particles of size R, and $f(R)$ the atmospheric aerosol size distribution function, with

$$Z = \int_{R\min}^{R\max} f(R) dR,$$

the total number of particles per unit volume.

As we can see, it is the ion-annihilation process

$$\frac{dn}{dt} = -n \int_{R\min}^{R\max} \beta(R) f(R) dR \tag{3}$$

that allows us to deduce information on atmospheric particle concentrations (Mohnen, 1971). After making some obvious assumptions, we have [from Eq. (3)]

$$\frac{dn}{dt} = -n\overline{\beta(R)}Z = -\text{constant} \times n\bar{R}Z. \tag{3a}$$

Thus, from conductivity measurements we can deduce only changes of the product of average size ($\bar{R}$) and total number concentration Z.

The other approach to measurement of the total number concentration of the atmospheric aerosol is the condensation nucleus counter, of the several types described by Aitken (1923), Scholz (1932), Nolan and Pollak (1946) or Rich (1955). The latter photoelectric types are well suited to performing frequent observations at remote stations or aboard ship, although they must be calibrated with an absolute counter before use. Although the absolute accuracy of these detectors is rather poor (around $\pm 30\%$ of the concentration observed), this accuracy is sufficient to determine meteorological variability in aerosol concentration (Hogan, 1968). The condensation nucleus counter is capable of determining the total aerosol concentration in maritime air during nearly all ambient meteorological conditions, while the conductivity detector can provide the aerosol concentration only in fair weather conditions.

A program to measure the geographic distribution of aerosol concentrations over the North Atlantic was begun by the authors in cooperation with the State University Maritime College in 1966 (Hogan *et al.*, 1967). This program has continued to date along the

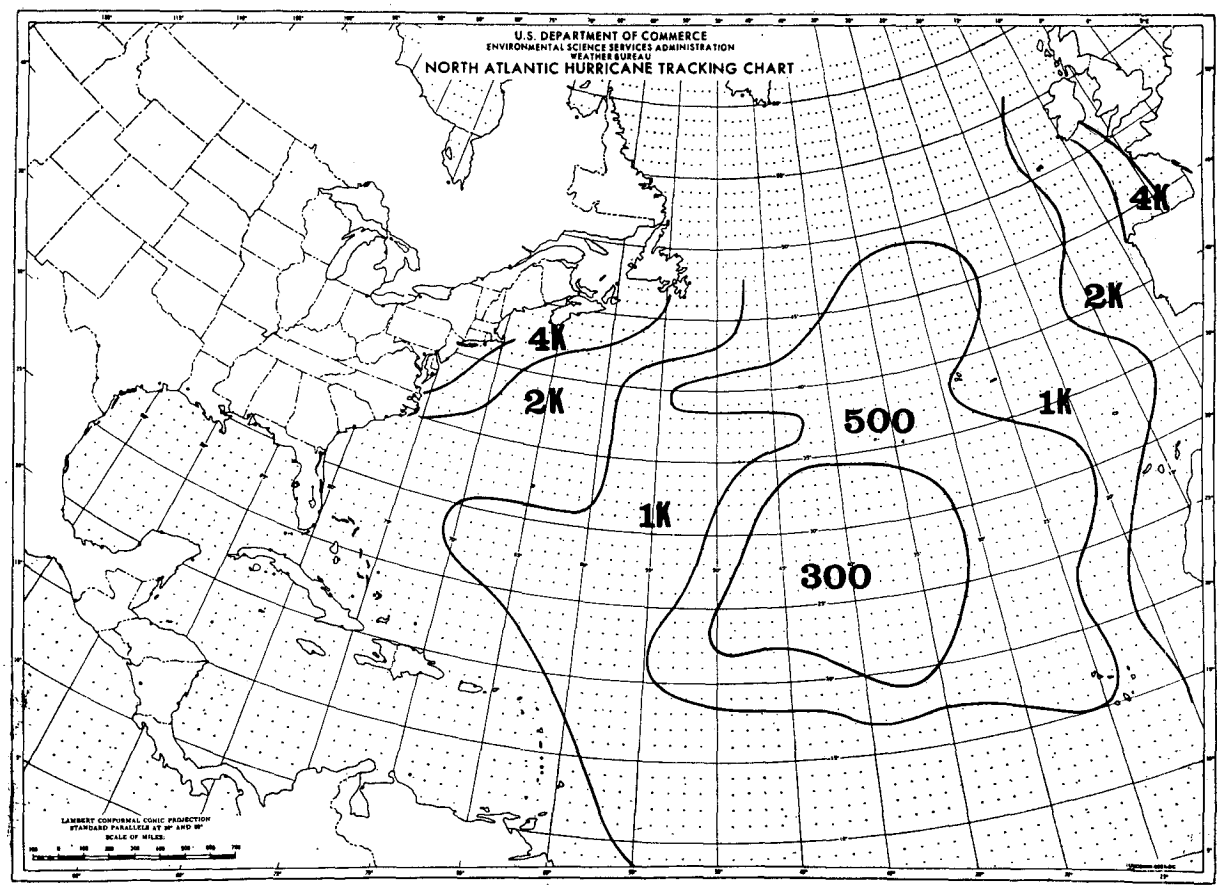

FIG. 1. Mean Aitken nuclei concentrations observed over the North Atlantic during the period 1966–71. Observations are most frequent along 40N, and along the major great circle trade routes. The highest mean values are found in the regions generally characterized by persistent high pressure systems. The lowest are found in mid ocean.

Maritime College's training route in the North Atlantic. Additional measurements were begun over a larger area of the oceans in 1970, with observations made aboard several merchant ships and extended to include several oceanographic ships, island and mountain stations in 1972. Because of the wide spread in the data, we have refrained from publishing the results until a comprehensive analysis of the seasonal, geographic and meteorological variation of aerosol concentrations could be completed. We present some preliminary data to illustrate our point.

3. Aerosol concentrations observed over the North Atlantic

The route followed by the State University Maritime College on training cruises generally follows latitude 40N across the Atlantic. One round trip was made along this route from June through August, 1966, and two were made during the summer months since 1968. Additional data are available from the authors' experiments aboard F. S. *Meteor* in 1971, from cooperative measurements made aboard *American Argosy* in 1970–72, the *African Crescent* in 1969–70, and the *Moor MacCape* in 1971–72. Fixed-point data were also obtained at Ocean Station Echo in November and December, 1971. An analysis of the average value of aerosol concentration observed over the North Atlantic by these observers between 1966 and 1971 is offered as Fig. 1. It is unmistakable that, on the average, strong continental influence is observed well west of Europe and east of North America and in the Bermuda and Azores high pressure regions. These continentally influenced concentrations are often found to extend across the Atlantic at 40N in fair weather; during stormy conditions, concentrations of less than 300 cm^{-3} are often found a few miles offshore (Hogan *et al.*, 1972). Recent data obtained at Sable Island, Nova Scotia, through the kind cooperation of Environment Canada, show that these low concentrations are occasionally found there, with winds from the open Atlantic. Two additional figures showing the extremes of Aitken nuclei concentrations observed over the Atlantic during 1966–71 are also included. The low levels found at Sable Island and relatively near shore and the high values found far at sea indicate the presence of extremely variable transport and removal processes.

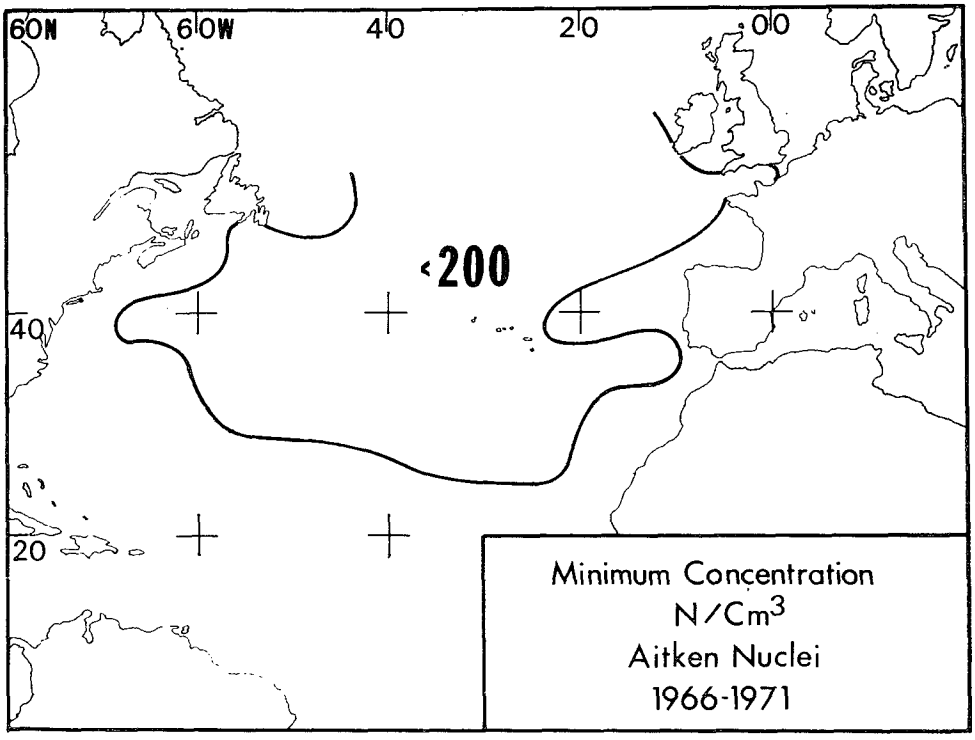

FIG. 2. The area where Aitken concentrations at, or below, the threshold of detection of the portable photoelectric condensation nucleus counter have been observed since 1966. The minimum measured concentration can then be considered to be less than 200 cm^{-3} in this area.

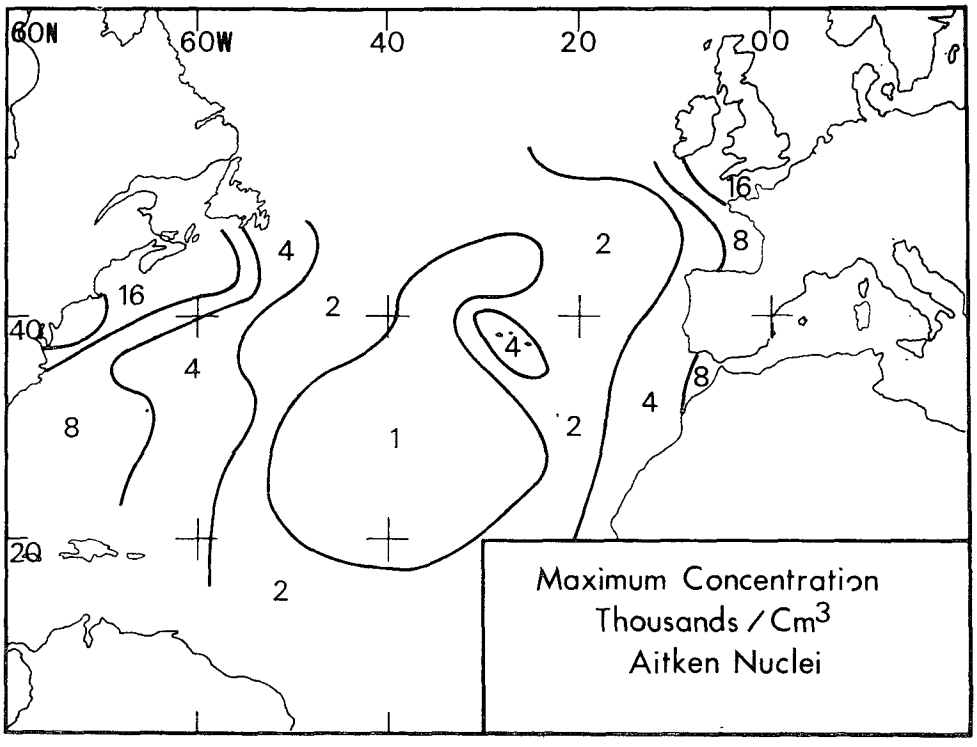

FIG. 3. The maximum Aitken concentration observed over the North Atlantic during 1966–71. A comparison of Figs. 2 and 3 illustrates the extreme variability with meteorological phenomena over many areas of the Atlantic.

TABLE 1. Aitken nucleus concentrations (cm^{-3}) observed over the North Atlantic in the area bounded by 30–45N, 30–60W.

Date*	Average	Highest	Lowest	Source and remarks
1909–27	500–1000			Calculated from *Carnegie* conductivity data by Shiratori (1934).
1929 W	490	2060	2	Wigand, as quoted by Landsberg (1938); includes data 40–50N, 5–63W.
1929 E	800	4100	62	
1934	950	1450	150	Landsberg (1938); most frequently quoted average data from 41–47N, 34–68W.
1951 Jul		1920	1220	Hess (1951); data observed north of 45N not included
1951 Aug		675	505	
1951 Dec		782	387	Parkinson (1952); data from 31–35N, 62–67W
1962 May–Jun	650	1000+	100	Gunn (1964); Baltimore Hamburg great circle
1966 E Jun–Jul	585	1200	200	Hogan *et al.* (1967)
1966 W Jul–Aug	650	1400	200	
1968 E Jun	499	750	230	State University of New York Maritime College, unpublished; all data bounded by 30–45N, 30–60W
1968 W Jul	673	1475	295	
1968 E Jul	527	1375	200	
1968 W Aug	870	1850	375	
1969 Jun	293	980	<200	
1969 Jul	310	980	<200	
1969 Aug	368	1500	<200	
1970 E Jun	429	1100	200	
1970 W Jul	553	1400	240	
1970 E Jul	553	1300	240	
1970 W Aug	454	850	280	
1971 Jun	1115	3000	350	
1971 E Jul	1105	2250	250	
1971 W Jul	630	1600	350	
1971 Aug	691	1650	380	

* W, westbound; E, eastbound.

Based on data analyses an arbitrary "uncontaminated zone" has been defined as the area bounded by 30–45N and 30–60W, and a chronological record of observations in this area has been prepared. This chronology is based primarily on Maritime College data and published historic data; the values are shown in Table 1.

The overall average value found in this area during 1966–71 was 641 cm^{-3}, and the averages in this table seem to oscillate about this value. When one examines the values obtained in consecutive crossings in, for example, 1968 or 1971, the short-term variability is so great as to make any set of observations made on a single or pair of transects unrepresentative of "normal" conditions.

Additional data were obtained from Ocean Station Delta (35N, 48W) during November and December of 1971, through the kind cooperation of J. Dew of NOAA, North Atlantic Weather Project. An average concentration of 562 cm^{-3} was obtained during this period, a close approximation of the long-term mean. A small diurnal tendency was present in the Aitken concentration:

Time				
GMT	0000	0600	1200	1800
LST	2048	0248	0848	1448
Average concentration	626	520	525	582

A similar diurnal tendency has been found in the data obtained along 40N in summer; if this tendency is real it may be indicative of gas-to-particle conversion processes in the marine atmosphere.

4. Aerosol concentrations over the Pacific

Cobb (1973) and Cobb and Wells (1970) have stated that while conductivities in the North Atlantic east of the United States and over the North Pacific east of Japan have decreased (implying an increase in aerosol concentration due to increased fossil fuel combustion), the conductivities measured in the eastern South Pacific have remained constant. These conclusions are again based on recent single-data cruises.

The authors have obtained cooperative Aitken nuclei counts from the merchant ships *Mobil Energy*, *American Argosy*, *American Legacy*, *Moor MacCape*, *Austral Patriot* and *Austral Pilot*, over several great circle routes since 1970. Additional data have been obtained by the oceanographic ships *Robert Conrad*, *Thomas Washington*, and *Nella Dan* in less temperate waters. Beginning in 1972, fixed-point data have been obtained at Pitcairn Island (130W, 26S) and at the north shore of Oahu.

The tongue of continental aerosol reported by Cobb has been found east of Japan on several occasions; however, analogous to the North Atlantic situation east of the United States, it is often absent, and mid-ocean Aitken nuclei concentrations (~200–300 cm^{-3}) are often found very near Tokyo Bay. In general, the concentrations obtained over the temperate Pacific seem to be nearly as variable as those over the North Atlantic.

The most interesting information from the Pacific has come from Pitcairn Island (Hogan and Christian,

manuscript in preparation). A distinct seasonal trend was found to exist during calendar year 1972, with a mean value of 403 cm^{-3} occurring from January through June, and a mean value of 591 cm^{-3} from July through December. This seasonal change is concurrent with the buildup of the 30S high and with increased precipitation at Pitcairn. This seasonal trend may be due to chance, as it is but one year's data; however, such a large seasonal trend or short-term trend, if it is that, should be accounted for in benchmark data.

It is very interesting to note that only rarely does the Aitken nuclei concentration on Pitcairn fall to the low values (<200 cm^{-3}) frequently observed (Blanchard and Syzdek, 1972) concurrent with trade winds at Hawaii. Pitcairn rarely observes Aitken concentrations >1000 cm^{-3} (only seven times during calendar year 1972) and values <200 cm^{-3} are almost equally rare. Very similar concentrations are reported along the Panama-New Zealand great circle route, which is tangent to Pitcairn.

The variations in shipboard data and the seasonality of the Pitcairn data preclude presenting a map of aerosol concentrations over the Pacific similar to that shown for the Atlantic in Fig. 1. Recent data supplied to the authors by J. Warburton from Antarctica and U. Radok from Macquarie Island allow us to construct a rough latitudinal variation for the Pacific area (Table 2).

Previous experiments at Hawaii (Blanchard and Syzdek, 1972), the west coast of the United States (Hogan, 1968), and the west coast of the British Isles (O'Connor, 1966; Aitken, 1923) have led us to believe that "good, pure, maritime air" should contain 200–300 cm^{-3}. However, these observations show that maritime concentrations often exceed those found over the continents in remote locations.

The concentration of Aitken nuclei over the seas is governed by transport, removal, and atmospheric chemical processes. The transport may be at levels well above the surface and potential particles may be transported as precursor gases rather than particles, with the ultimate particles being formed as a result of ion-molecule or oxidation reactions between convecting or subsiding layers.

The dominating removal mechanism for near-surface aerosols over the North Atlantic seems to be stratus decks and fog. The cloud drops apparently collect the smaller particles by Brownian coagulation and reduce the number concentration to that approaching the number of cloud drops. This accounts for the low concentrations found in the Roaring 40's and near shore in the North Atlantic. The absence of low clouds, fog and stratus may, in part, explain the relatively high concentrations found in clear subsiding air in the tropics.

The authors' network of aerosol observers is supplying frequent data from island, continental and shipboard stations. Some data are now available from almost every area in the North Pacific and for much of the tropical South Pacific. Data are now available from the North Atlantic for nearly every calendar month and most hours of local standard time. The authors are continuing to analyze these data with respect to determining the existence and magnitude of diurnal and seasonal variations. An additional analysis is being carried out to evaluate the idea of subsiding air in high pressure systems as a maritime aerosol source and storm systems as aerosol sinks.

TABLE 2. Latitudinal variation of Aitken nuclei concentration (cm^{-3}) for the Pacific.

Latitude	Average concentration	Source and remarks
~90S*	<200	J. Warburton, DRI; single-season data
~70–90S*	200–1000	Readings above 1000 cm^{-3} may reflect contamination at McMurdo
40S–60S	<200	I. Allison and U. Radok, University of Melbourne, observations from *Nella Dan*, single-cruise data.
20S–40S	200–700	Farrell Lines; *Thomas Washington*, Scripps; C. Owen, ASRC; T. Christian
20S–30N	<200–500	C. Owen, ARSC; F. Guenthner; *Thomas Washington*, Scripps; *Moor MacCape*, Farrell Lines.
30N–50N	200–1000	*American Argosy*, *American Legacy*, *Moor MacCape*, T. Egelston

* Continental site.

Aerosol concentrations are as variable as any other meteorological parameter. An aerosol climatology must be a result of frequent observations at intervals which will allow determination of diurnal and seasonal changes before a secular change can be inferred. Cobb may be correct in his observations that no secular change in aerosol concentrations has occurred at Mauna Loa and over the Pacific. However, the presence of a change in electrical conductivity on a few fair days cannot uniquely support a conclusion that a secular change in aerosol concentrations has occurred in the air near the surface of the North Atlantic.

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