Particles in the Lower Troposphere over the High Plains of the United States. Part I: Size Distributions, Elemental Compositions and Morphologies

PETER V. HOBBS, DAVID A. BOWDLE* AND LAWRENCE F. RADKE

Atmospheric Sciences Department, University of Washington, Seattle 98195

(Manuscript received 27 April 1984, in final form 8 July 1985)

ABSTRACT

Airborne measurements are presented of particle size distributions obtained during spring and summer at various locations in the High Plains of the United States. Two main particle modes are apparent: an accumulation mode between particle diameters (D_p) of 0.1–1 μm and a coarse particle mode between D_p = 10–20 μm. Near urban/industrial sources, a weak nucleation mode was detected at $D_p < 0.1$ μm. The accumulation mode was affected primarily by the type of airmass; recent involvement in precipitation was associated with significantly reduced particle concentrations in this mode. The coarse particle mode, which contained mostly local soil particles, was controlled primarily by mesoscale and convective-scale processes. Large NaCl particles occasionally reached deep into the interior of the High Plains. The presence of relatively high concentrations of particles with $D_p \geq 10$ μm has important implications for precipitation processes.

1. Introduction

This is the first in a series of three papers concerned with measurements and interpretations of airborne particles in the lower troposphere over the High Plains of the United States. The data to be described and analyzed consist of particle size distributions and the elemental compositions and morphologies of the particles (this paper), cloud condensation nuclei (CCN) (to be presented in Part II, Hobbs *et al.*, 1985), and ice nuclei (to be presented in Part III, Bowdle *et al.*, 1985).

The measurements were obtained during the late springs and summers of 1975 and 1976. The 1975 measurements were obtained from 19–25 June on a round-trip flight between Great Falls, Montana, and Big Spring, Texas, from 20–26 July at Miles City, Montana, from 24–30 August at Goodland, Kansas, and from 3–9 August at Big Spring, Texas. In 1976 measurements were obtained over a period of six weeks (15 June–30 July) in the vicinity of Miles City.

The specific purpose of this study was to obtain information on those particles that play a role in cloud and precipitation processes. However, the results to be presented provide more general information on the atmospheric aerosol in the High Plains, including the sources and sinks of various types and sizes of particles and the large- and small-scale processes that modify the concentrations and distributions of particles in the atmosphere.

Although the data set that has been analyzed is quite extensive (~125 particle size spectra, ~270 cloud condensation nucleus spectra, ~150 ice nucleus spectra, and ~300 slide samples on which morphology and elemental composition analyses were carried out), in view of the large areal extent of the High Plains, and the potentially large variety of processes that can affect atmospheric aerosol, even this study must be considered exploratory rather than definitive.

Sections 2 and 3 of this paper provide general information relevant to all three papers in this series. Specific discussion of the particle size measurements begins with section 4.

2. Locations of the measurements

The High Plains of the United States is a broad (~300–500 km wide from east to west), relatively flat grassland region that extends from Texas in the south to Montana in the north and includes all or portions of the states of Texas, New Mexico, Oklahoma, Colorado, Kansas, Wyoming, South Dakota, Nebraska, Montana and North Dakota. The location, soil and climate of the High Plains restrict it primarily to agricultural usage, although it has generally low rainfall. The population density is low; urban centers are relatively small and widely scattered. Regional industrial development in the mid-1970s revolved primarily around the petrochemical industry in Texas and Oklahoma, with oil wells and local industries scattered over the rest of the region.

Miles City, Montana, is located in the northern portion of the High Plains, deep in the continental interior of the United States. It is ~2250 km from the Gulf of Mexico and ~250 km east of the Rocky Mountains. Its distance from the Gulf prevents all but the most

* Present affiliation: Universities Space Research Association Visiting Scientist, NASA Marshall Flight Center, Hunstville, AL 35812.

energetic Gulf storms from reaching the site. Pacific storm systems lose much of their intensity and precipitable water before they reach Miles City. Nevertheless, eastern Montana receives much of its rainfall from large-scale synoptic systems and mesoscale cumulus convection in modified Pacific airmasses.

Goodland, Kansas, is located ~1300 km from the Gulf of Mexico and ~250 km east of the Rocky Mountains. The Rockies effectively isolate Goodland from the influence of Pacific storm systems. However, this site is near enough to the Gulf, separated only by the gradually rising terrain of the Plains States, that low-level convergence of warm, moist air from the Gulf produces summer thunderstorms that account for much of the annual rainfall of the region.

Big Spring, Texas, is situated in the southern portion of the High Plains, ~500 km from the Gulf of Mexico. Its rainfall is generated primarily by strong line convergence in subtropical airmasses.

3. Flight paths and procedures

All of the measurements to be described were obtained aboard the University of Washington's B-23 research aircraft. Aircraft flight paths were designed to obtain information on synoptic-scale, mesoscale, and convective-scale variations in the concentrations and compositions of the atmospheric aerosol.

To obtain information on the synoptic-scale and mesoscale variations, a round trip survey flight was flown between Great Falls, Montana, and Big Spring, Texas. To investigate smaller-scale variations, more detailed studies were carried out in the vicinity of Miles City, Montana, Goodland, Kansas, and Big Spring, Texas. The locations of these sites and the flight tracks between Great Falls and Big Spring are shown in Fig. 1.

The survey flight in 1975 was designed to obtain information on particles below cloud base or, in the absence of clouds, in the mixing layer (~0–2 km AGL). On each day a horizontal flight leg was flown at ~1 km AGL from ~0900–1100 local daylight time (LDT) and, in the afternoon (~1300–1500 LDT), a horizontal leg was flown at ~1.4 km AGL (Fig. 2). Large cities, major highways, and other potential sources of aerosol, while not scrupulously avoided, were usually skirted. Measurements of CCN spectra and the compositional properties of large (>5 μm) particles, were collected every 20 or 30 min in regions that appeared to be unaffected by strong local sources of particles; all other measurements were continuous or semicontinuous.

A typical flight pattern for the more intensive studies at Miles City, Goodland, and Big Spring in 1975 is illustrated in Fig. 3. These flights were designed to provide information on the vertical profiles of particles; hence, measurements were made at three or more constant altitudes between ground level and cloud base. If small or medium sized cumulus clouds were present,

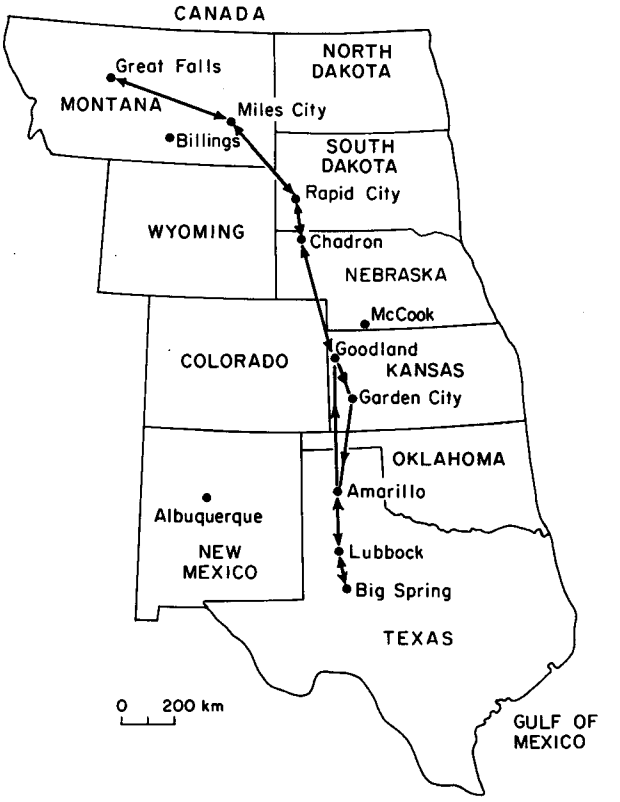

FIG. 1. Locations of the research flights in the High Plains. The route for the survey flight between Great Falls, Montana, and Big Spring, Texas, is shown by the arrowed line. Intensive airborne studies were carried out in the vicinity of Miles City, Montana; Goodland, Kansas; and Big Spring, Texas.

additional measurements were made above the mixing layer and several cloud penetrations were made at midcloud level. In 1976 at Miles City, particle measurements were generally obtained during the afternoon at altitudes of 1.2, 1.8, 2.4, 3.0 and 3.7 km MSL.

Measurements were obtained in a wide variety of

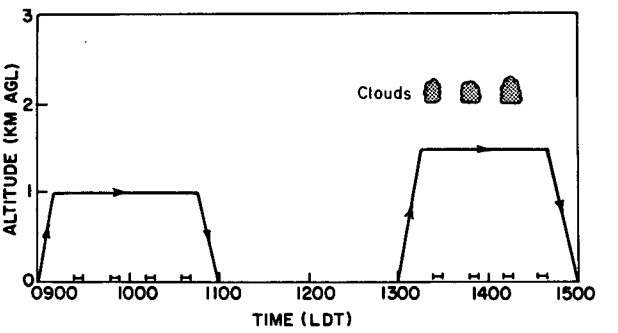

FIG. 2. Schematic to illustrate a typical daily flight track (arrowed line) on the survey flight. The markers just above the time axis represent the periods when batch samples of large particles were collected by direct impaction and cloud condensation nucleus spectra were measured.

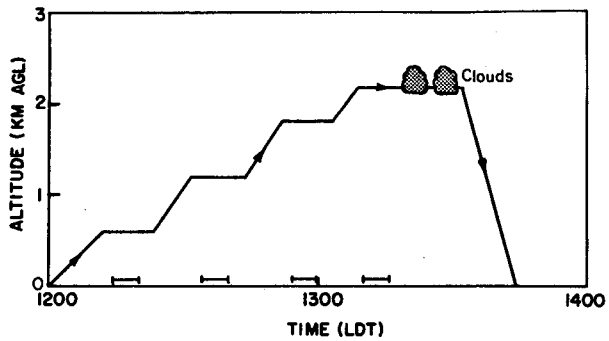

FIG. 3. As in Fig. 2 but for intensive studies in the vicinity of Miles City, Goodland and Big Spring in 1975.

conditions, ranging from prefrontal to postfrontal synoptic situations, and in continental and maritime airmasses. Clear skies or scattered fair weather cumulus clouds dominated the local weather, but a few large cumulus, thunderstorms, squall lines, duststorms, and even a funnel cloud, were encountered.

Details of the conditions under which each individual data set was collected, and listings of all the data, may be found in Hobbs *et al.* (1978).

4. Measurement facilities and data reduction

In this section, brief descriptions are given of the instrumentation and techniques used to obtain the measurements described in this paper. More detailed descriptions may be found in Hobbs *et al.* (1978).

The size spectra of particles from ~0.01–45 μm in diameter were measured semicontinuously, and displayed immediately in the aircraft by integrating measurements from an electrical aerosol analyzer (EAA), two Royco optical particle counters (OPC), and a Particle Measuring Systems' (PMS) axially scattering spectrometer (ASSP).

The EAA, which has been described by Liu *et al.* (1974), was used to measure the size spectra of particles from ~0.01–0.36 μm diameter in eight equal logarithmic size ranges. The instrument was calibrated in the manner described by Liu and Pui (1975). Laboratory tests on homogeneous aerosol samples showed that the concentrations measured with this instrument are stable to within a factor of ~1.8.

Particles with diameters from ~0.3–12 μm were sized in 16 equal logarithmic size ranges and counted with a Royco Model 202, 90° light-scattering, OPC (Zinky, 1962). The size discrimination of the instrument was calibrated using spheres of polystyrene latex and styrene divinylbenzene of known sizes and indices of refraction (Whitby and Liu, 1968). A Royco Model 225, forward-light scattering, OPC was used to measure the size spectra of particles from ~3–40 μm in diameter. The sampling inlet of the commercial instrument was modified to give ~50% collection efficiency for 40 μm diameter particles. Calibration was carried out with glass spheres.

The ASSP, which detects light scattered in the near forward direction from a He–Ne laser, was used to measure particles ~1.4–45 μm in diameter in 15 equal linear size intervals (Knollenberg, 1981). Unlike the instruments described above, the ASSP measures particles *in situ* as they pass through a sampling port in the airstream outside of the aircraft. The size discrimination of the ASSP was calibrated in the same manner as the OPCs.

To improve the sampling statistics for large particles, as well as to obtain particles for compositional analyses, particles were collected by direct impaction on pairs of glass slides (7 mm and 14 mm wide × 76 mm long), coated with a thin (10 μm) layer of silicone grease. Circular carbon planchets, 14 mm in diameter, were glued to the 14 mm wide slides and coated with vaseline. One slide of each width was attached to a steel bar 1.2 m in length. This assembly was deployed into the free airstream outside of the aircraft for periods of 3–7 min. After exposure, the slides were stored in a dust-free box at low relative humidity (<50%).

The particles collected on the slides were subsequently sized and counted from microphotographs taken of the slides at low relative humidity. The number concentrations in the air of particles of various sizes were then deduced using the collision efficiencies for a ribbon (Noll and Pilat, 1970). The low collision efficiencies for small particles limit this technique to particles > 5 μm diameter.

The particles collected on the carbon planchets were photographed under a scanning electron microscope. Elements within the particles having an atomic number ⩾ 16 were determined by energy dispersive analysis of X-rays (EDAX).

Number concentration functions, $dN/d(\log D_p)$, where N is the number concentration (cm^{-3}) of particles with diameters $\geqslant D_p$, were calculated from the particle size distribution measurements. For discrete size intervals bounded by measured particle diameters D_i and D_{i+1}, D_p is the geometric mean of the size interval:

$$D_p(i) = (D_i \times D_{i+1})^{1/2}. \tag{1}$$

Particle surface area and volume distributions were determined from the expressions:

$$\frac{dS}{d(\log D_p)} = \pi D_p^2 \left[\frac{dN}{d(\log D_p)}\right], \tag{2}$$

$$\frac{dV}{d(\log D_p)} = \frac{\pi}{6} D_p^3 \left[\frac{dN}{d(\log D_p)}\right], \tag{3}$$

where S and V are the surface area ($\mu m^2\ cm^{-3}$) and volume ($\mu m^3\ cm^{-3}$) concentrations of particles with diameters $\geqslant D_p$.

Two techniques were used to measure the integral properties of the aerosol. Cumulative number concen-

trations of particles in the air with $D_p > 0.01$ μm (*i.e.* Aitken nucleus counts) were measured continuously with a rapid expansion counter. Also, the light-scattering coefficient (b_{sp}) of the total particle population, integrated over scattering angles from 10° to 170°, was measured continuously with an integrating nephelometer (Charlson *et al.*, 1969). The fine particle mass concentration ($D_p < 3$ μm) can be estimated from b_{sp} (Waggoner and Weiss, 1980). The nephelometer was calibrated using molecular (i.e., Rayleigh scattering from particle-free dry air and Freon 12.

Information on airmass origins was obtained using the layer-averaged terrain-following trajectory model of Heffter (1973).

5. Particle size distributions

a. Overview

Shown in Fig. 4 are the ranges in the values of the number, surface area and volume distributions of the particles measured within the mixing layer over the High Plains during the entire period of the 1975 field study.

The variability in the number concentration of particles (defined as the ratio of the maximum to minimum concentration at each D_p) is shown by the dash-dot line in Fig. 4a. For particles with $D_p \leqslant 0.1$ μm the variabilities shown in Fig. 4a are minimum values, since the concentration of particles may have fallen below the detectability of the EAA. The variability in this size range appeared to be related to variations in the strength of ground sources of Aitken nuclei, the age of the particle population, and the presence or absence of rain. The relatively high variability between $D_p \simeq 0.1$–1.0 μm was due primarily to the removal of these particles by nucleation scavenging (i.e., particles serving as cloud condensation nuclei) followed by their gradual reappearance from gas-to-particle conversion and coagulation. The variability around $D_p \simeq 5$ μm was caused primarily by variations in surface windspeed. Variability for $D_p > 15$ μm was due, in part, to statistical uncertainties in particle counting and to variations in the strength of convective mixing in the boundary layer.

The prominent shoulders and inflection points on the envelope of the particle number distributions do not fit a conventional Junge distribution very well. These features correspond, respectively, to maxima and minima in the particle surface area and volume distributions (Fig. 4b, c). The latter two distributions thus provide more insight into the nature and properties of the particles than does the number distribution. The surface and volume distributions were generally bimodal, with an *accumulation mode* centered between $D_p \simeq 0.1$–1 μm and a *coarse particle mode* between $D_p \simeq 10$–20 μm. Other modes were occasionally detected in other size ranges. For example, near industrial centers, and other sources of combustion products, a weak *nucleation mode* at $D_p < 0.1$ μm appeared. However, with airmass aging, the nucleation mode rapidly di-

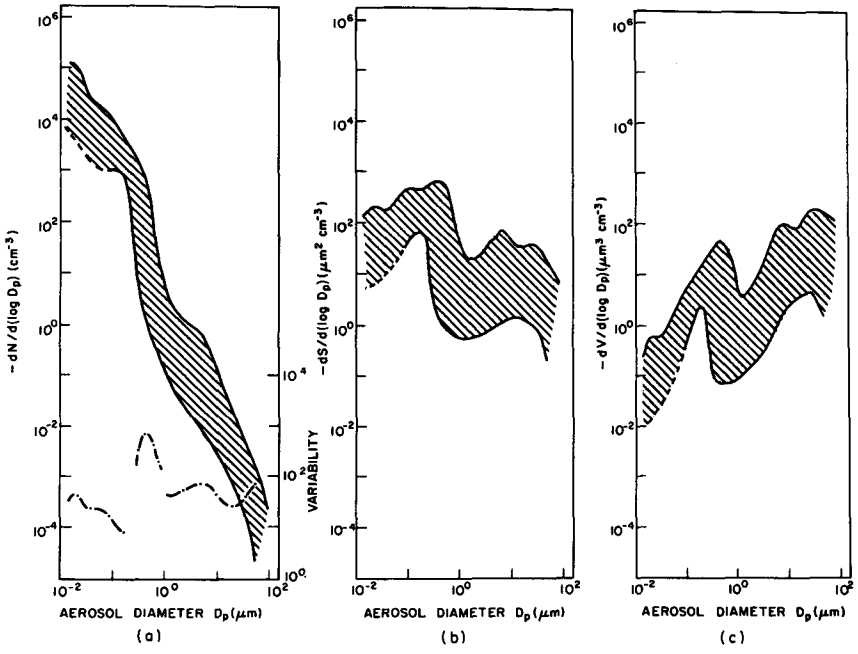

FIG. 4. Envelopes (shaded areas) for all measurements of particle size distributions obtained within the mixing layer (~1–3 km MSL) over the High Plains in 1975. (a) Particle number concentrations and variability (dash-dot line), (b) particle surface areas, and (c) particle volumes.

minished, presumably due to coagulation with particles in the accumulation mode.

Since most of the surface area of the particles is contained in the accumulation mode, particles in this size range dominate visible light scattering, as well as surface chemistry and adsorption phenomena. Most of the particle volume (and therefore mass) is contained in the coarse particle mode. This mode was strongest during periods of vigorous convective mixing below fair-weather subsidence inversions. Near precipitating cloud systems, the total particle volume loading within the mixing layer reached values as low as 2–4 μm^3 cm^{-3}. On clear days, with strong convective mixing, the particle volume loading reached values as high as 75 μm^3 cm^{-3}. About 24 h transit time downwind from a duststorm, values as high as 125 μm^3 cm^{-3} were measured; in the fringes of the duststorm the volume loading reached ~250 μm^3 cm^{-3} (see below). Assuming a soil particle density of 2.5 g cm^{-3}, the total particle mass loadings (= volume loading × particle density) for these conditions were: ~5–10 μg m^{-3} near rain, ~190 μg m^{-3} in strong convective mixing, ~310 μg m^{-3} downwind from the duststorm, and ~625 μg m^{-3} in the fringes of the duststorm.

b. *Some individual data sets*

Envelopes of the particle number, surface area and volume distributions measured on the survey flight and at the three individual sites (Miles City, Goodland and Big Spring) in 1975 are shown in Figs. 5–8. The envelopes of the particle distributions measured on the survey flight (Fig. 5) essentially contain the envelopes of the measurements at the three individual sites (Figs. 6, 7 and 8).

Below 0.3 μm diameter the minimum measurable concentrations were determined by the lower detection limit of the EAA; they therefore show only minor differences between the four sets of measurements (Figs. 5a, 6a, 7a, 8a). Below 0.1 μm diameter, the maximum particle concentrations measured during the survey flight were somewhat higher than those obtained at the three individual field sites due to the influence of scattered local sources of particles along the survey flight track. Similarly, maximum concentrations in this size range were higher at Big Spring and Goodland than at Miles City, due to the higher level of industrial activity near the former two sites. Indeed, the 1975 measurements at Miles City failed to reveal the existence of any significant localized sources of submicron particles within at least 100 km to the west, north and east of the city.

The significance of the differences in particle measurements between 0.2–0.6 μm diameter in the survey flight and at the three individual field sites is best seen in the surface and volume distributions (Fig. 5b, c, Fig. 6b, c, Fig. 7b, c, Fig. 8b, c). In this size range, the maximum particle surface area concentrations and the diameters of the peaks in the accumulation modes were quite similar for the measurements obtained at the three individual field sites, but these parameter values were significantly higher than the corresponding values from the survey flight. Measurements of the particle light-scattering coefficient, which is dominated by par-

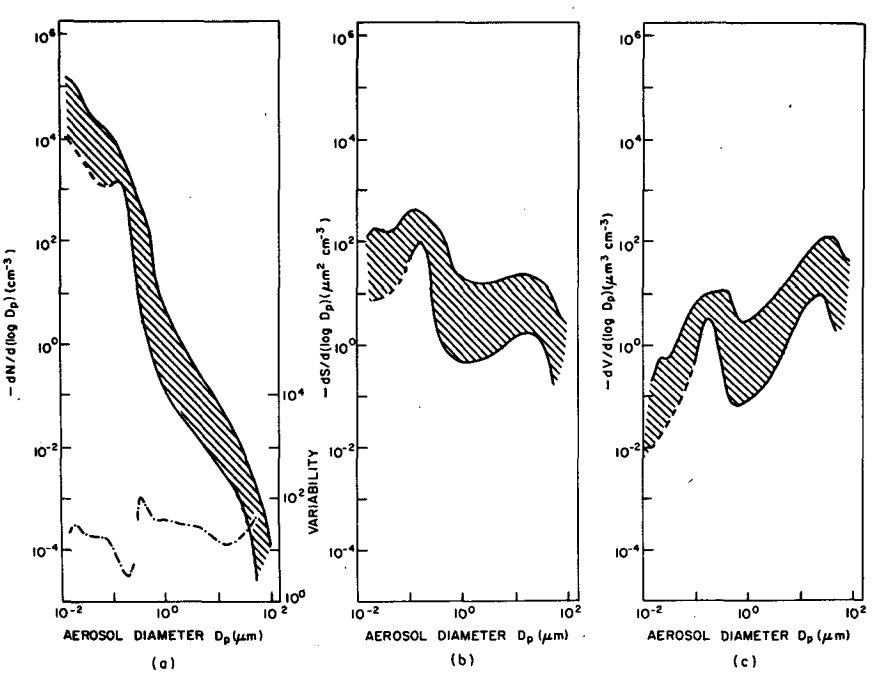

FIG. 5. As in Fig. 4 but for measurements obtained on the survey flight in 1975.

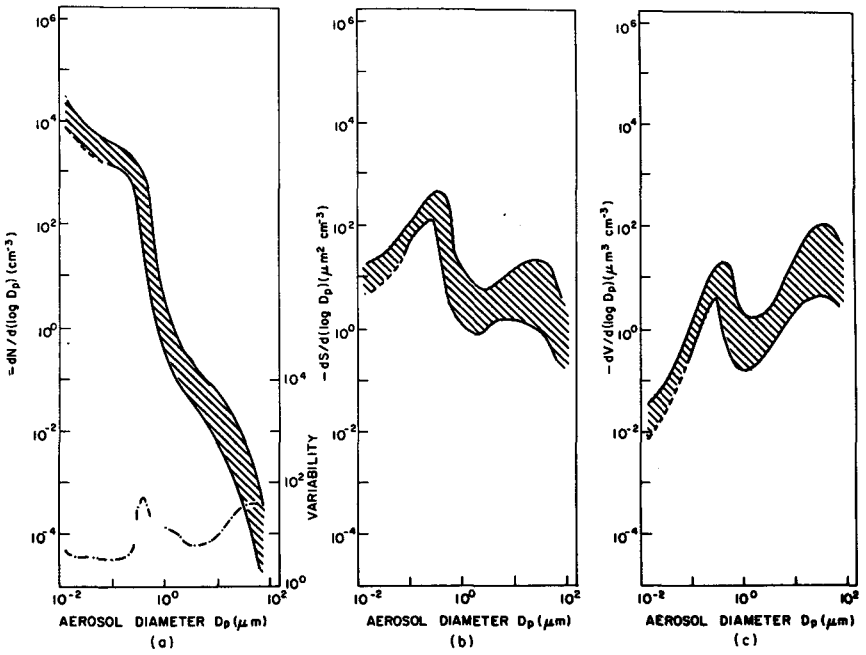

FIG. 6. As in Fig. 4 but for measurements at Miles City in 1975.

ticles from 0.2–0.6 μm in diameter (Waggoner and Weiss, 1980) were also rather low during the survey flight.

The measurements described heretofore can be explained by postulating that the maximum concentrations of submicron particles at the three field sites occurred in aged aerosol systems with a well-developed accumulation mode, while the maximum particle concentrations on the survey flight occurred in relatively young aerosol systems. The low concentrations of 0.2–0.6 μm particles near rainy regions during the survey flight allowed self-coagulation of newly formed Aitken nuclei, rather than their coagulation with preexisting aerosol in the accumulation mode. The maximum

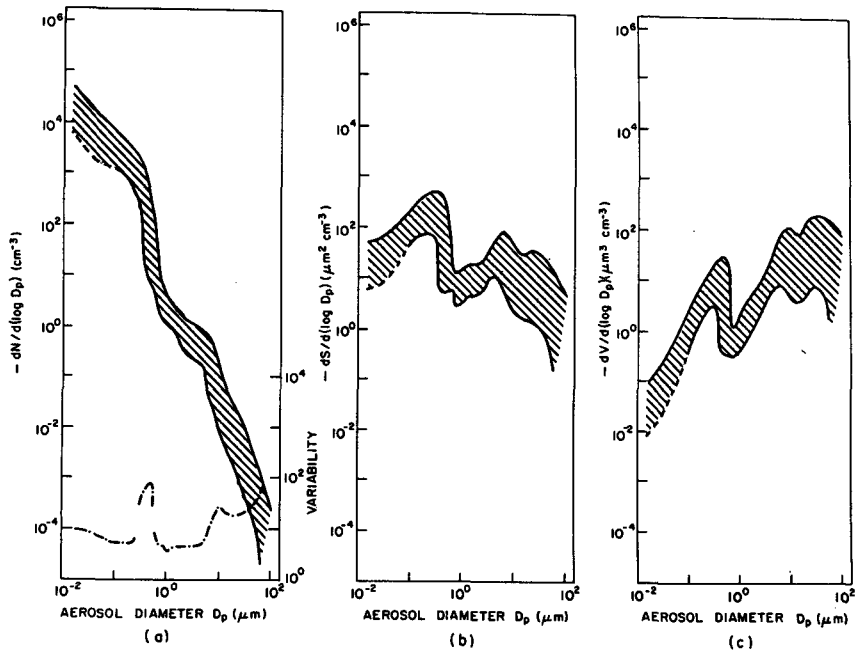

FIG. 7. As in Fig. 4 but for measurements at Goodland in 1975.

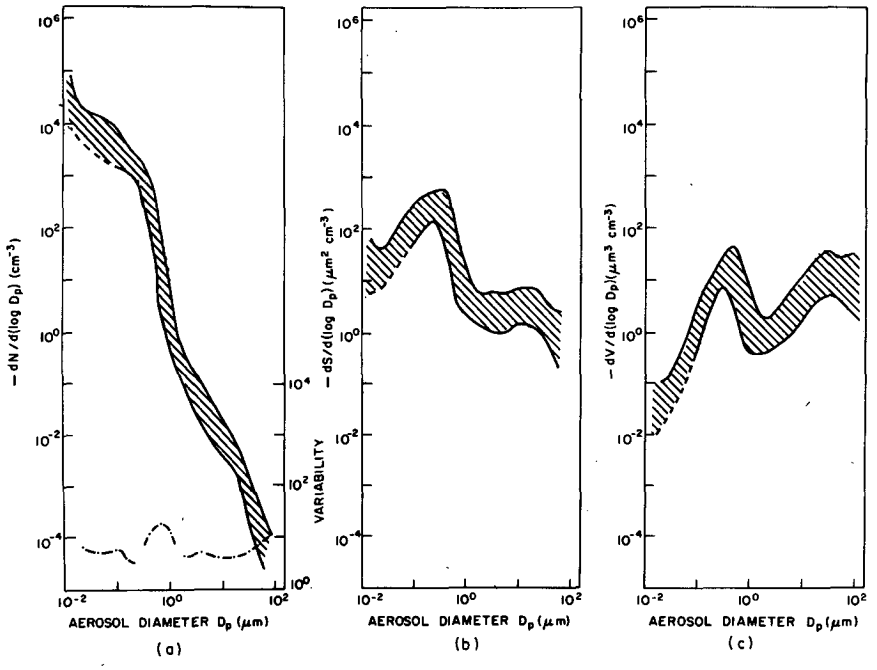

FIG. 8. As in Fig. 4 but for measurements at Big Spring in 1975.

concentrations were somewhat higher in Big Spring than in Goodland. This could be due to local aerosol sources in the Big Spring area and to the extremely dry conditions and long residence times during the Big Spring study.

The differences in the minimum concentrations of submicron particles were most likely due to differences in the frequency, intensity, and proximity of precipitation. The survey flight was dominated by a slow-moving cold front associated with large thunderstorms and rapidly moving squall lines. The intense precipitation and widespread postfrontal subsidence associated with these strong convective systems effectively removed the submicron particles and replaced aerosol-laden airmasses with cleaner air from higher altitudes. During the Miles City and Goodland studies, weaker frontal systems with more scattered convective activity were located quite near the field sites. These systems partially removed the submicron particles. During the study at Big Spring, frontal systems were never closer than a few hundred kilometers and the aerosol had aged for at least two days. The effect of aging in the Big Spring measurements is clearly evident in the particle number distribution shown in Fig. 8a, where the entire envelope of the measurements of submicron particles is shifted to higher concentrations.

Differences in the size distributions of particles > 1.0 μm diameter were probably caused by a combination of variations in ground conditions, surface windspeed, strength of convective mixing, altitude of the measurements, and the frequency and intensity of precipitation. Perhaps the most remarkable feature of the particle measurements > 1.0 μm diameter is contained in the measurements at Goodland (see Fig. 7). The concentrations of 5 μm diameter particles measured at Goodland in 1975 were as much as an order of magnitude higher than those measured during the survey flight or at Miles City or Big Spring in 1975. Even the minimum concentrations of particles with 1.0 μm $< D_p <$ 10 μm at Goodland were comparable to the maximum concentrations measured on the survey flight or at the other two sites. An explanation for the anomalously high concentrations at Goodland is given as follows.

Figure 9 shows measurements of particle size distributions obtained at about 1.05 km AGL in a small duststorm between Chadron, Scottsbluff, and Alliance, Nebraska, on 24 August 1975 during the passage of a dry cold front. Although the concentrations of particles with $D_p < 0.7$ μm and $D_p > 20$–30 μm in the duststorm were well within the entire range of concentrations obtained during the 1975 studies (Fig. 4), the concentrations of particles with $D_p = 0.7$–10 μm were the highest we encountered in 1975. These results show that the duststorm was a significant source of 0.7–10 μm diameter particles.

Air trajectory analysis revealed that the airmass that traversed the region affected by the duststorm passed over Goodland the following day. Airborne measurements at Goodland that day showed a sharp increase in dust content as the afternoon progressed. The frontal system that generated this duststorm remained over the central High Plains throughout the week, reversed direction once, and passed over the Goodland site three

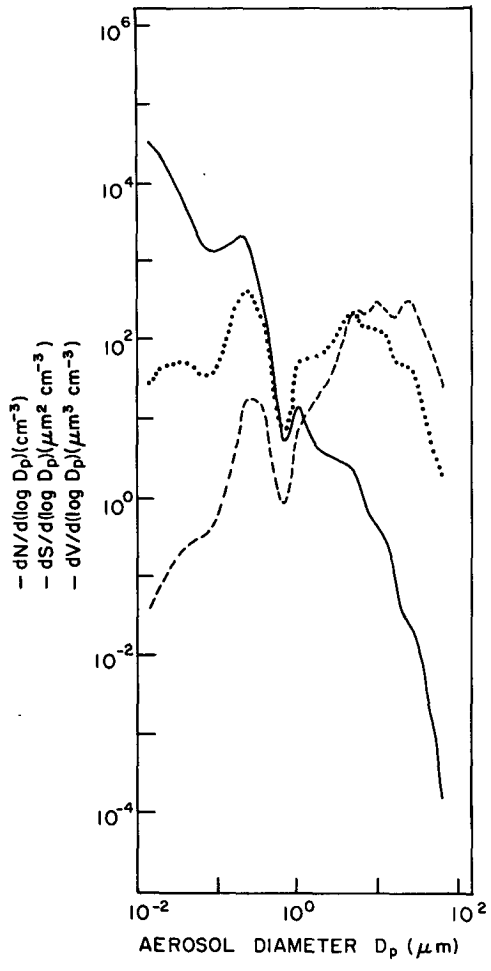

FIG. 9. Measurements of particle size distributions in the fringes of a duststorm at 1.05 AGL over western Nebraska on 24 August 1975. The solid, dotted and dashed lines show the particle number, surface area and volume distributions, respectively.

times. Trajectory analyses suggested that the low-level circulations generated by the oscillations of this frontal system apparently kept the dust-laden airmass, which had traversed the duststorm, over the Goodland area for the next two days (25–26 August 1975). The second passage of the front was associated with high surface winds which were observed to generate a duststorm near Fort Morgan, Colorado, on 27 August and several large dust devils and small duststorms near Goodland on 28 August. Comparisons of the measurements made in the duststorm (Fig. 9) with those made at Goodland (Fig. 7) reveal obvious similarities in the shapes of the particle size distributions and the locations of the shoulders and peaks of the distributions.

These results strongly suggest that the enhancement in the concentration of 5 μm particles at Goodland was due to the suspended component of wind-blown soil dust. This suspended dust was also associated with at least a factor of 2 increase in ice nucleus concentrations at −20°C (Bowdle *et al.*, 1985). However, even in the fringes of the duststorm, the concentrations of submicron particles were too low to significantly affect the concentrations of cloud condensation nuclei between 0.2 and 1.5 supersaturation (Hobbs *et al.*, 1985).

6. Elemental compositions and morphologies of the particles

a. Overview

A total of 185 slide samples (68 from 1975 and 117 from 1976) that had been collected over the High Plains at altitudes of ~1.2, 2.4 and 3.7 km MSL were analyzed to determine the morphologies and elemental compositions of the particles.

The particles were labeled as siliceous, calcareous, soluble salt, or miscellaneous mineral, depending on whether the most abundant mineral substance in the particle was, respectively, silicon (Si), calcium (Ca), a soluble salt (e.g., NaCl or KCl), or some other mineral substance such as aluminum (Al) or iron (Fe). When no mineral content could be detected above the EDAX noise level, the particle was labeled nonmineral (most likely organic or carbon-based compounds or substrate artifacts).

For each field study period (except the survey flight, for which EDAX data are not available), the number of particles in each compositional category was tabulated for each of seven diameter intervals: 1–2, 2–4, 4–6, 6–10, 10–20, 20–40 and 40–60 μm. These measurements were then used to determine normalized size distributions of the sampled particles $[-(1/n_T)(dn/d(\log D_p)]$, where n_T is the total number of sampled particles). The results are shown by the histograms in Fig. 10. Particles with $D_p < 5$ μm are underestimated due to biases in both sampling and analysis.

The size-segregated compositional data can be related to the true atmospheric particle size distributions (see section 5) through the percentage contribution that particles in a given compositional category make to the total number of particles in a given size interval. These frequency percentage contributions are shown by the graphs in Fig. 10. The percentage contribution of all particles in a given composition category to the total number of particles examined with D_p = 1–60 μm are shown in Table 1. Siliceous particles were generally the most abundant, followed by nonmineral particles and calcareous particles.

Siliceous and calcareous particles were more abundant in 1975 than in 1976, while soluble salt and nonmineral particles were more abundant in 1976 than in 1975. During 1975, siliceous particles were most abundant at Goodland, and least abundant at Miles City and Big Spring. Nonmineral particles were most abundant at Miles City and least abundant at Big Spring and Goodland. Calcareous particles were found only rarely at Miles City or Goodland, but quite regularly at Big Spring. The percentages of siliceous particles

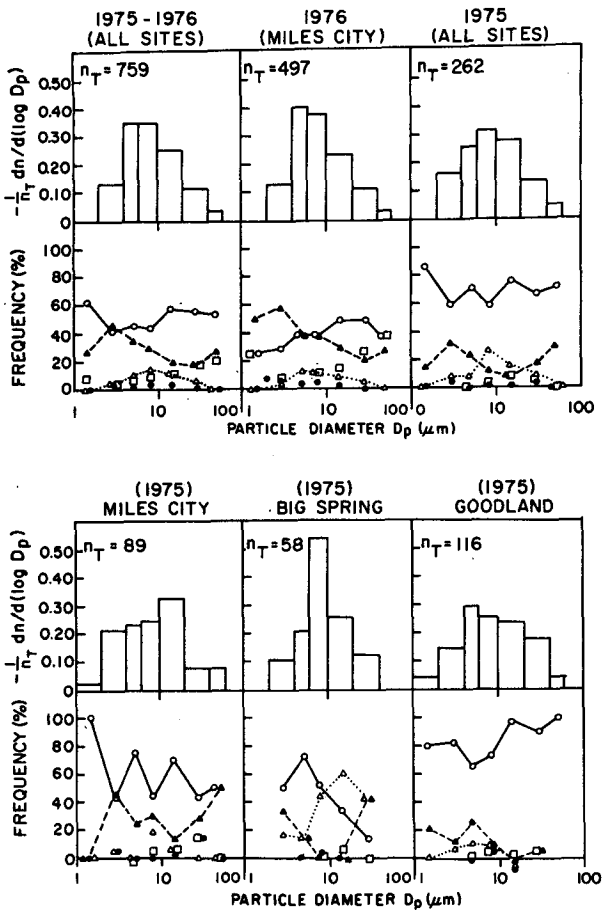

FIG. 10. Normalized size distributions (histograms) and size-segregated percentages of types of particles (graphs). Type of particles in the graphs are indicated by: siliceous, open circles; calcareous, open triangles; soluble salt, squares; miscellaneous mineral, solid circles; nonmineral, solid triangles. The total number of particles in each sample is n_T.

with D_p < 6–10 μm at Miles City, Big Spring and Goodland were quite high (~80%) and similar from one site to another. On the other hand, calcareous particles accounted for a very large fraction of the particles with D_p > 10 μm at Big Spring, while siliceous particles dominated this size range at Miles City and Goodland. These differences reflect the high calcium content of the Texas soils near Big Spring and the more clayey and sandy content of the soils near Miles City and Goodland.

b. Particle composition and morphology

Figures 11 and 12 depict some typical particles and their elemental compositions.

Figure 11a shows particles sampled at 3.66 km MSL, between 55 and 90 km NE of Miles City, at 1206–1214 MDT on 5 July 1976. The sample was collected in an airmass that had originated over the Rocky Mountains to the west and southwest of Miles City. Particles labeled 1 and 4 in Fig. 11a were calcareous but contained some Si, Al and Mg; particles 2 and 5 were siliceous but they also contained Al and Mg; particle 3 contained only a small amount of mineral, consisting of Si, Fe and Mg, with traces of Al.

Figure 11b shows a small clay particle ~ 3.4 μm long consisting of Si and Al with traces of Na. This particle was collected at 3.05 km MSL, between 25 and 45 km northeast of Miles City, at 1544–1551 MDT on 30 July 1976, in an airmass that had originated over the Rocky Mountains to the west and southwest of Miles City. Notice the plate-like structure of the primary particle and the presence of attached plates with much smaller dimensions (<1 μm long).

Figures 11c, d show a larger clay particle (~7 μm long) consisting of Si, Al and Ca, with lesser amounts of K and Fe, and traces of S and Ti. This particle was collected at 1.83 km MSL, 50 km SE of Big Spring, at 1406–1413 CDT on 7 August 1975, in an airmass that had originated over southern Texas. Notice again the layered plate-like structure of the primary particle and the presence of secondary particles with dimensions similar to those of the primary particle in Fig. 11b.

Figures 11e, f show a very large particle (~26.5 μm long) consisting primarily of S and Si with somewhat lesser amounts of Ca, Al, K, Mn and Fe. This particle was obtained at 1.22 km MSL, between 30 and 65 km SW of Miles City, at 1207–1216 MDT on 2 July 1976, in an airmass that had originated over the arid Wyoming Plains to the south and southwest of Miles City. Again, the layered structure of the primary particle is evident, as are secondary structural features of similar size to those shown in Fig. 11b, d.

The three particles in Fig. 11 are typical of many of the mineral particles that were collected, including the prominent presence of S and other potentially soluble substances in the larger particles (D_p ~ 15–30 μm). These results suggest that a large fraction of the at-

TABLE 1. Comparison of composition of particles with D_p = 1–60 μm obtained in the various field study periods over the High Plains. The numbers are the percentage contributions of the stated particle type to the total number of particles measured during the indicated study period.

	Field study period					
Particle type	Overall (1975, 1976)	1976	1975	Miles City 1975	Goodland 1975	Big Spring 1975
Siliceous	49	40	66	58	83	45
Calcareous	10	8	13	7	4	41
Salt	10	14	3	4	3	2
Miscellaneous materials	2	2	2	3	1	2
Nonmineral	29	36	16	28	9	10
Total number of particles	759	497	263	89	116	58

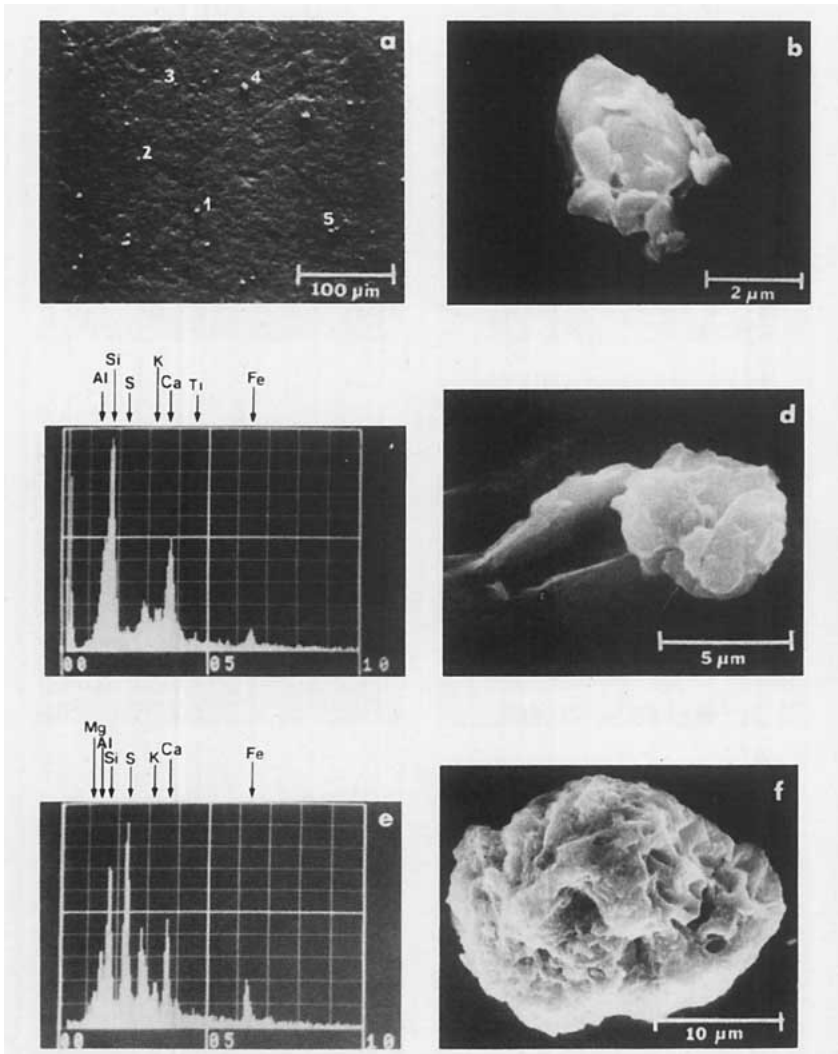

FIG. 11. Analyses of typical large particles using scanning electron microscope with EDAX. The conditions under which the particles were collected are described in the text.

mospheric particles between ~1–30 μm in diameter over the High Plains consist of small clay particles and large aggregates of these primary clay particles.

Some of the airborne particles were considerably different from those described above. For example, Fig. 12a, b depict a small particle (~5.5 μm long) consisting primarily of Si, with lesser amounts of Al, Ca and Na, and traces of S, K and Fe. This particle was sampled at 3.66 km MSL, between 60 and 90 km southeast of Miles City, at 1427–1434 MDT on 28 June 1976, in an airmass that had originated over the Rocky Mountains to the northwest of Miles City. Notice the very flat laminar structure of this particle and the absence of any adhering particles.

Figures 12c, d show a much larger particle (~20 μm long) consisting of Ca and Al with traces of K, S and Fe. This particle was collected at 1.22 km MSL to the west of Miles City at 1140–1145 MDT on 24 August 1978, during high winds with low clouds and light stratiform rain, in an airmass that had originated over the Rocky Mountains to the northwest of Miles City. This calcareous particle appears to be a single large particle with smaller particles adhering to its surface.

A completely different type of particle, shown in Fig. 12e, f, consists primarily of Ca with lesser amounts of Si and Al. This particle was also collected on 24 August 1978 but at 1.8 km MSL, to the southeast of Miles City, and at 1451–1456 MDT. This 50 μm particle shows little trace of a plate-like or laminar structure, but is instead very loose and "spongy" in appearance.

Particles similar in appearance to those in Figs. 11 and 12 were found between 1 and 7 m above the ground during a duststorm near Big Spring (Gillette and

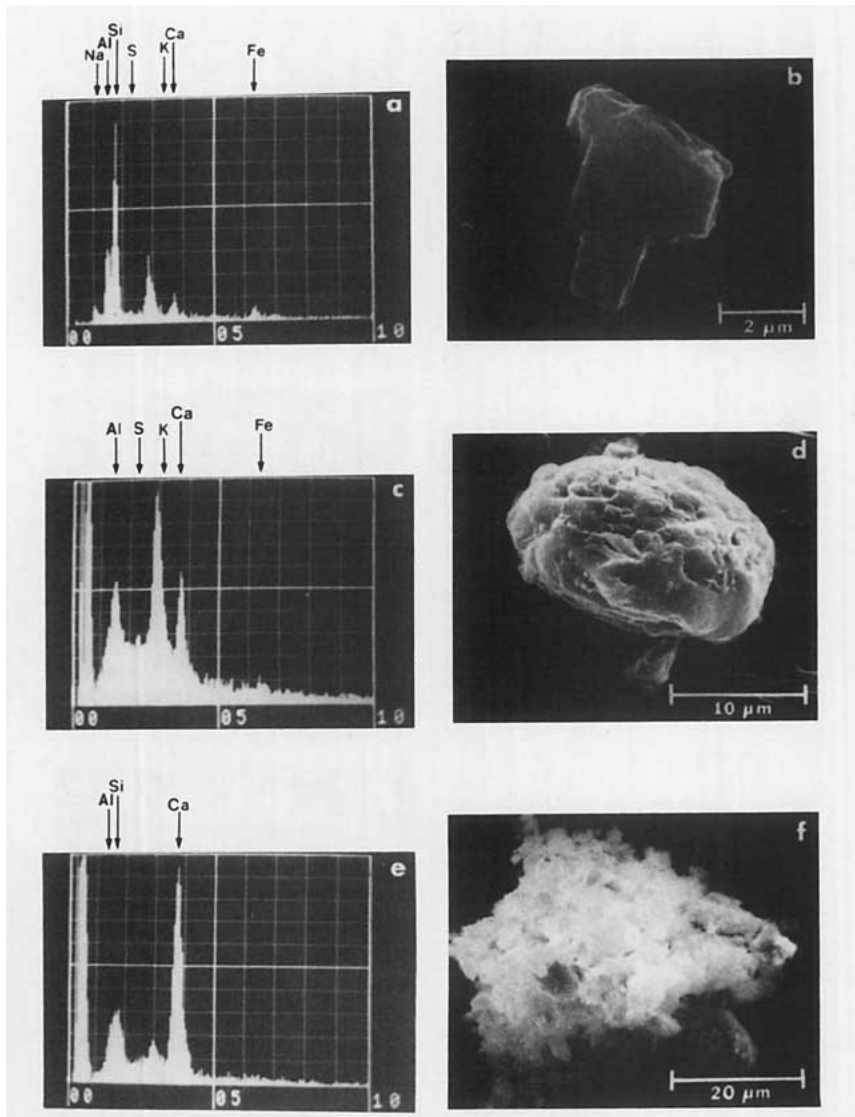

FIG. 12. As in Fig. 11.

Walker, 1977). These authors suggested that the primary small clay particles had originally adhered to the surface of large quartz grains in the parent soils, that the small particles had become dislodged by "sandblasting" during high winds, and that the clay particles then formed easily fragmented aggregates. However, we found both small clay particles and large aggregates during hot, dry, clear weather with light winds and vigorous convective mixing. This suggests that the aggregates were already present in the parent soil and that duststorms are not necessary to suspend small clay particles in the atmosphere. However, we occasionally collected clusters of particles of such appearance and orientation that they could have been loosely aggregated in the atmosphere, as suggested by Gillette and Walker, or perhaps in an evaporating raindrop, and then broken apart by the impact with the collection surface.

c. Soluble components

Particles of NaCl were detected by EDAX in several samples collected over the Southern High Plains between Texas and Kansas in 1975 in a prefrontal airmass that had recently originated over the Gulf of Mexico. A 17 μm NaCl particle was also collected near Miles City on 22 June 1976, at 3.22 km MSL in clear air between layers of St and As clouds. The airmass apparently originated on 19 or 20 June in a region of light winds off the southern California coast, passed over a region of strong surface winds (including the Great Salt Lake area of Northern Utah) on 21 June,

and arrived at Miles City on 22 June without having encountered any rainy regions in transit. It is likely that the NaCl particle collected in Miles City originated in the strong winds over the Great Salt Lake area, because other quite large soil particles were found in the same sample. In a third case, a 2 μm NaCl particle was collected near Miles City on 15 July 1976 at 3.05 km MSL in fair weather. The airmass in which this particle was collected apparently originated over the Pacific Ocean off the coast of western Canada on 11 or 12 July, passed over the Canadian Rocky Mountains, and arrived over Miles City on 15 July without encountering any rain in transit. In this case, the fallspeed of the salt particle was small enough to have survived a three- or four-day transit in clear weather to Miles City; therefore, it could have originated over the Pacific Ocean. In this connection we note that Byers *et al.* (1957) found that in the lower Mississippi Valley and in Illinois, large chloride particles were more numerous above the surface than at the ground.

Figure 13 shows a sample of soil particles collected in clear weather at 68% ambient relative humidity at 1.05 km AGL near the Yellowstone River 30 km northeast of Miles City in an airmass that had originated several days before over the Canadian Great Plains. About one-third of the particles are surrounded by "haloes." Elemental analysis of one halo showed that it contained Si and Cl with a trace of S, while the parent particle contained Si and Al with traces of Fe and K. Other "haloed" parent particles in this sample consisted of soil materials, some pollen grains and occasionally other biological material. If an EDAX analysis was carried out on the parent particle first, the surrounding halo generally vanished; subsequent elemental analysis of the area formerly covered by the halo showed no traces of elemental constituents above background noise levels. This shows that the materials comprising the haloes were volatized by the heat from the scanning beam of the electron microscope.

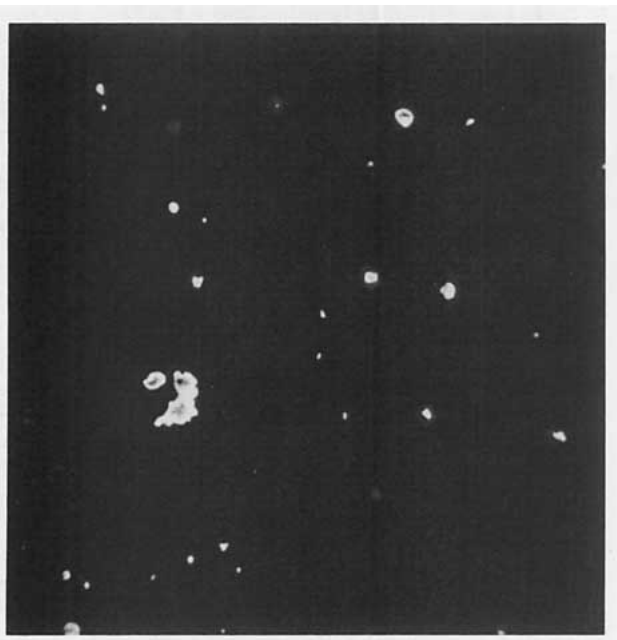

FIG. 13. Scanning electron microscope photograph illustrating "haloes" surrounding large soil particles.

Frank and Lodge (1967) and Bigg *et al.* (1971) postulated that haloes of the type shown in Fig. 13 are associated with sulfate compounds (*e.g.*, sulfuric acid or ammonium sulfate). Such compounds acquire a layer of liquid water even at moderate ambient relative humidities. The outer limits of the halo are thought to define the impact area of the resultant droplet, which evaporates after impact leaving a residue and the parent particle. Examination of Fig. 13 shows that the ratio of halo dimensions to parent particle dimensions ranges from about 2, for particles larger than 15 μm in diameter, to 10 or more for particles less than a few micrometers in diameter. These results indicate that trace quantities of water-soluble material on the surface of small, otherwise insoluble, dust particles can significantly increase the particle size in high relative humidity conditions. In 1975, such "coated" particles occurred in 37% of all samples, with concentrations high enough to affect the development of precipitation (0.1 to 3 L^{-1}). The observance of these haloes around a variety of organic and inorganic particles suggests that the coating was deposited on the particles during their residence in the atmosphere, perhaps by coagulation with sub-micron aerosol. This mechanism is consistent with the frequent observance of coated particles in the generally dry weather of 1975, and their total absence in the more rainy weather in 1976.

7. Discussion and summary

The measurements presented in this paper provide information on the size distributions and general nature of atmospheric particles over the High Plains. Two main modes were identified in the particle surface area and volume distributions: an accumulation mode (between $D_p \simeq 0.1$–1 μm) and a coarse particle mode (between $D_p \simeq 10$–20 μm). Near industrial sources, a weak nucleation mode ($D_p < 0.1$ μm) was also detected. Most of the surface area of the particles was contained in the accumulation mode and most of the volume in the coarse particle mode.

The main factor affecting the concentrations of particles in the accumulation mode was the nature of the airmass and its recent life history of involvement in convective cloud systems. Variations with time of day, location, and altitude (within the mixing layer) appeared less important. The highest concentrations of particles in the accumulation mode were obtained in airmasses that were more than two day's transit time from organized precipitation systems. This is consistent with the suggestion of Willeke and Whitby (1975) that

particles in the accumulation mode are generated by the coagulation of Aitken nuclei. The lowest concentrations of particles in the accumulation mode (which were similar to those characteristic of maritime air) were measured in airmasses that had a high probability of recent involvement with precipitation. We believe this was due to their removal by cloud nucleation scavenging and to the replacement of aerosol-laden airmasses by cleaner air descending from higher altitudes after passages of storms.

Particles in the coarse particle mode were generally controlled by mesoscale or shorter-range transport. Hence, their compositions and concentrations in the mixing layer were determined primarily by the local soil cover, surface wind strength, and convective mixing. Near Big Spring, these particles were primarily calcareous, while near Miles City, and Goodland, they were primarily siliceous. The large siliceous particles frequently consisted of cemented aggregates of numerous small clay platelets.

On several occasions, large salt particles were detected deep in the interior of the High Plains. Depending on the airflow and the particle size, it appears that these salt particles can originate from the Gulf of Mexico, the Pacific Ocean or the Great Salt Lake.

The concentrations of 10 μm particles in the lower troposphere over the High Plains ranged from $\sim$0.001 to 0.09 cm^{-3}, and particles with diameters of 50 μm were often present in measurable concentrations. The presence of such large particles at cloud levels is contrary to some traditional views (e.g., Ludlam, 1980). If such particles are ingested into growing clouds, they can produce a "tail" of large drops in the droplet size distribution that will speed precipitation development by condensation and coalescence (see, for example, Johnson, 1982). This effect will be enhanced if the particles are coated with even trace amounts of soluble material. The cloud-active nature of the particles in the High Plains is treated explicitly in Parts II (Hobbs *et al.,* 1985) and III (Bowdle *et al.,* 1985) of this series of papers.

Finally, we note that proposals have been made to measure atmospheric winds remotely by means of doppler lidars aboard satellites, using atmospheric particles as targets. It has been estimated that if the concentration of particles with diameters between $\sim$2–4 μm exceeds 10^3 m^{-3}, sufficient backscattering should be present to obtain accurate wind measurements by this means (Hall, 1983). Even the lower envelope of our measurements of summertime particle concentrations in the lower troposphere over the High Plains exceeds this value by an order of magnitude.

Acknowledgments. We wish to thank J. Heffter of the NOAA Environmental Research Laboratories for air trajectory analyses. This study was supported in part by the Division of Atmospheric Water Resources Research, Bureau of Reclamation, Department of Interior. It was written while one of us (P.V.H.) was a Visiting Scientist at the Meteorological Office Radar Research Laboratory, Malvern, England, and an Alexander von Humboldt Visiting Professor at the Fraunhofer-Institute für Atmosphärische Umweltforschung, Garmish-Partenkirchen, FRG.

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