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The vertical distribution of the concentration of sea salt in the marine atmosphere near Hawaii

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ABSTRACT

The vertical distribution of the sea-salt concentration has been obtained in the lowest 800 m of the atmosphere over the sea windward of Oahu, Hawaii, for eight days in October and November of 1981. Although the sea-salt profiles varied from day to day, reflecting changes in wind speed, temperature structure and other parameters, a marked salt concentration gradient was nearly always observed between about 19 and 30 m. The average sea-salt concentration at 19 m, about 22 $\mu g\ m^{-3}$, decreased to about half that value, 11 $\mu g\ m^{-3}$, at 30 m. From there, the salt concentration decreased slowly to an average of about 5 $\mu g\ m^{-3}$ at 600 m above the sea.

1. Introduction

Early work on the vertical gradient of sea-salt concentration over the sea was done by Woodcock (1953, 1962), Lodge (1955), and Durbin and White (1961), but little has been done in the past 20 years, despite an increased awareness of the rôle of sea-salt particles as carriers of organic material and heavy metals.

To learn more about vertical salt gradients, and especially how they might vary from day to day, we made 20 flights 5 to 10 km windward of Oahu, Hawaii, in October and November, 1981. On each flight, 12 samples were obtained at altitudes between about 1000 m and 30.5 m (100 feet). Since the pilot did not deem it prudent to fly any lower, and we wanted to know something of the gradient in the lowest 30 m, we also got salt samples at an altitude of 19 m from the top of a tower on the windward shore. This tower has been used extensively for marine aerosol sampling (Barger and Garrett, 1970; Blanchard and Syzdek, 1972; Hoffman et al., 1972). The tower sampling, which did not begin until after the ninth flight, but was done for eight of the remaining flights, revealed some interesting and unexpected salt gradients in the lowest 30 m. They form the basis for this paper.

These gradients must be known before a complete understanding of the cycling of salt through the atmosphere is obtained. Estimates of the amount of salt cycled each year range from 10^9 tons (Eriksson, 1959, 1960) to 10^{10} tons (Blanchard, 1963). However they were based on sea-salt data obtained at altitudes of 600 to 800 m (Woodcock, 1953). Surely the amount of salt cycled through the lowest 20 m, where salt concentrations are higher than they are at 600 m, is much more than this. But what fraction of this low-level airborne salt is cycled up through 100, 200, or 2000 m (Petrenchuk, 1980) will remain unknown until more data on vertical salt gradients are obtained. Some of the data are provided in this paper.

2. Determining the sea-salt concentration

2.1. *Aircraft samples*

All aircraft samples were obtained by collecting sea-salt particles on small glass slides exposed for 30 s from a Cessna aircraft flying at 40 m s^{-1}. The

slides (about 20 mm long, half of which length was 1 mm and the other half 3 mm wide) were exposed to the air with a sampling device developed by Woodcock (1952). Twelve slides were exposed on each flight. A remaining slide, brought along on the flight but never exposed, was used as a control. In the laboratory, the salt was washed from a known area of each slide by carefully moving it into the top of a drop of distilled water placed on a small, inverted glass beaker. The drop made a contact angle of 90° with the beaker. This was accomplished by treating the glass with a silicon agent. The drop (usually 500 μl in volume) was analyzed for its sodium concentration by flame atomic adsorption spectroscopy. The sea-salt concentration was obtained by multiplying the sodium concentration by 3.25, the ratio of sea salt to sodium in seawater.

Knowing the amount of salt collected, the size of the slide, the exposure time and aircraft speed, and the collection efficiency, E, for the salt particles, we calculated the salt concentration. Although it is impossible, without a knowledge of the size distribution of the particles collected, to make exact calculations of E, we have estimated E by using assumed particle distributions. Particle-size distributions observed earlier by Woodcock (unpublished) for salt concentrations of about 3 and 9 μg m^{-3} were used to calculate E for relative humidities between 65 and 90%. E increases with salt loading and relative humidity. Our calculations, based on the Langmuir and Blodgett (1961) collection efficiency data, show that as the relative humidity goes from 65 to 90%, E goes from 83 to 88%. It conceivably could be some 2% higher for salt loads around 10 μg m^{-3} and 2% lower for salt loads around 1 μg m^{-3}, but this is small compared to the variations in concentration we observed. Consequently, corrections for E between 0.83 and 0.88 were applied, depending on relative humidity, to every slide that was exposed. At humidities of less than about 75%, there is a possibility that the salt particles were not supersaturated droplets but had undergone a phase change to become primarily sea salt. We assumed this did not happen. If it did, then E would be 7 or 8% less than we calculated it to be.

2.2. *Tower samples*

Sea salt was collected from the air as it flowed by the tower by exposing vertically-mounted platinum wires of 254 μm diameter and 10 cm long (Blanchard and Syzdek, 1972). All wire exposures were for 10 min. The wires were washed off in a known amount of distilled water which was analyzed for sodium in the same manner as the aircraft samples. Again, multiplication by 3.25 gave the sea-salt concentration in the water. Air flow past the wires (meters of air in 10 min) was monitored with a cup anemometer mounted about 0.5 m away. For all samples, the average air speed was between 5 and 8 m s^{-1}. Calculation of the salt concentration in the air was similar to that for the glass slides. On average, about 4 km of air flowed by the wires during exposure, giving a swept-out volume of 0.1 m^3. In contrast, the slides exposed from the aircraft swept out about half that amount while moving through 1.2 km of air.

As with the glass slides, E for the wires could not be exactly determined because we did not know the salt particle-size distribution at the time of sampling. But a reasonable estimate of E can be made. We knew from experience that the salt in the air at tower height (19 m) would most likely be in the range of 10 to 30 μg m^{-3}. We calculated E for wind speeds from 5 to 8 m s^{-1} and for humidities from 65 to 80%, for two salt particle distributions. One had been observed by Woodcock (unpublished) for 9 μg m^{-3} and the other was a log-normal distribution for 29 μg m^{-3} that appeared reasonable to McDonald et al. (1982). Considering the variation of wind speed and humidity at the tower, we calculated that E was in the range of 0.73 to 0.82. An average of 0.78 was used for all samples. Thus the maximum error in E for a given sample is no more than 5%.

Did the salt on the wires come from the surf zone along the shore? This is highly unlikely in the present case. First, some years ago one of us (AHW) made tests at night atop the tower to see if salt particles from the nearby surf could be detected in the beam of a flashlight. They could not, though they were clearly visible at the base of the tower. Second, the tower was only about 35 m from the center of the relatively inactive surf zone (see Fig. 1 in Blanchard and Syzdek (1972)). With the winds observed during sampling, 5 to 9 m s^{-1}, updrafts averaging 2.7 and 5 m s^{-1}, respectively, are needed to carry the surf-produced salt to the sampling wires. This is very unlikely, especially on four of the days when the temperature was nearly isothermal in the lowest 30 m.

All temperatures, both from the tower and the aircraft, were read from the same wet and dry bulb mercury thermometers. The aircraft temperatures were obtained by AHW at the same time that DCB was collecting the salt sample. They were reduced by 0.6 °C to correct for aerodynamic heating (Vonnegut, 1950). The thermometers were calibrated against a precision thermometer where errors were known. The maximum error was only 0.1 °C over the range observed in this work.

3. Results

The vertical gradient of salt was determined on four days in October and four in November 1981. The gradients for 21, 22, 26, and 29 October are shown in Fig. 1. In Fig. 2 are the gradients for 9, 11, 12, and 16 November. Wet and dry bulb temperature and relative humidity are also shown. The aircraft samples were obtained in mid-morning, generally between 1000 and 1030 HST. Sampling always proceeded from the highest to the lowest elevation. At the start of sampling, the Beaufort wind force was estimated by observing the whitecap coverage. After the last sample was obtained, at 30.5 m (100 feet), the wind direction was determined from the plane's compass by having the pilot fly directly into the wind. A correction for compass variation (11 degrees east) gave the true wind direction. The tower sampling was carried out three to four hours after that from the aircraft.

On 8, 9, 12 and 16 November, salt samples were obtained not only at 19 m but also at 12 or 14 m altitude. Sampling was essentially simultaneous at the two altitudes; 80% of the 10-min sampling time was common to both samples. Figs. 3 and 4 show these data plus the 30.5 and 61 m altitude samples

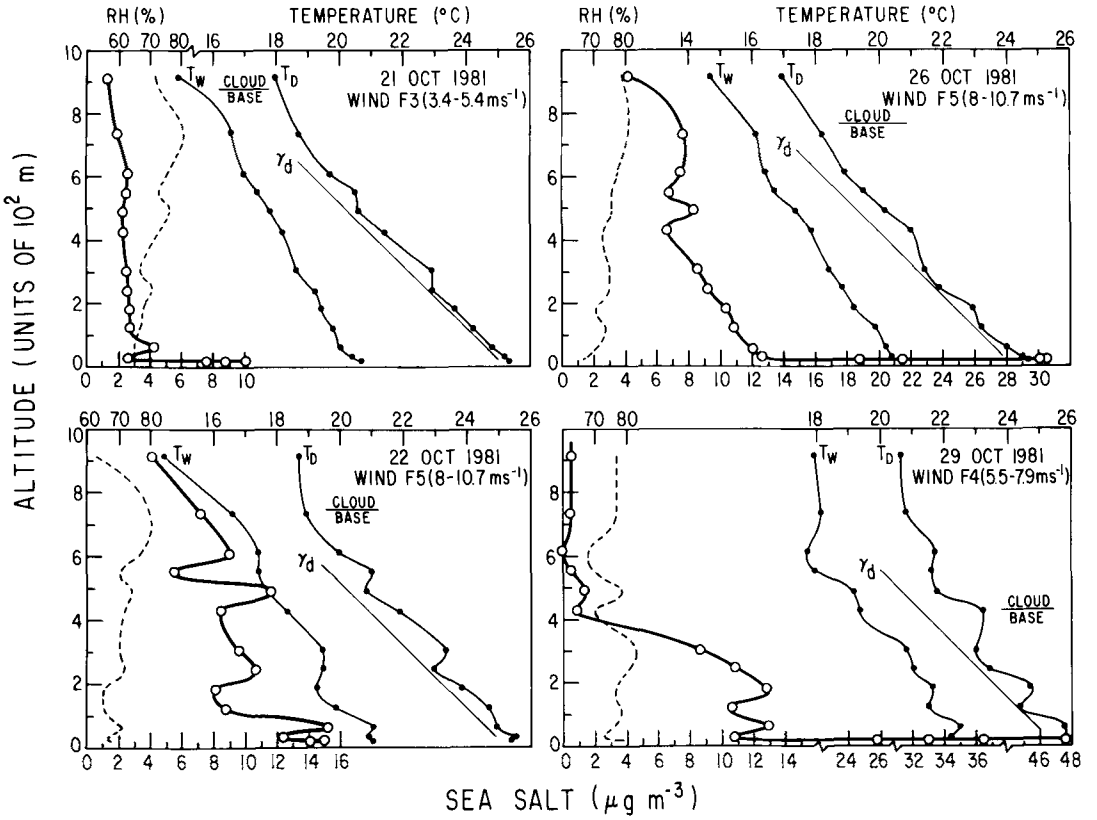

Fig. 1. Vertical profiles of sea-salt concentration, wet and dry bulb temperature, and relative humidity over the sea windward of Oahu, Hawaii, for 4 days in October, 1981.

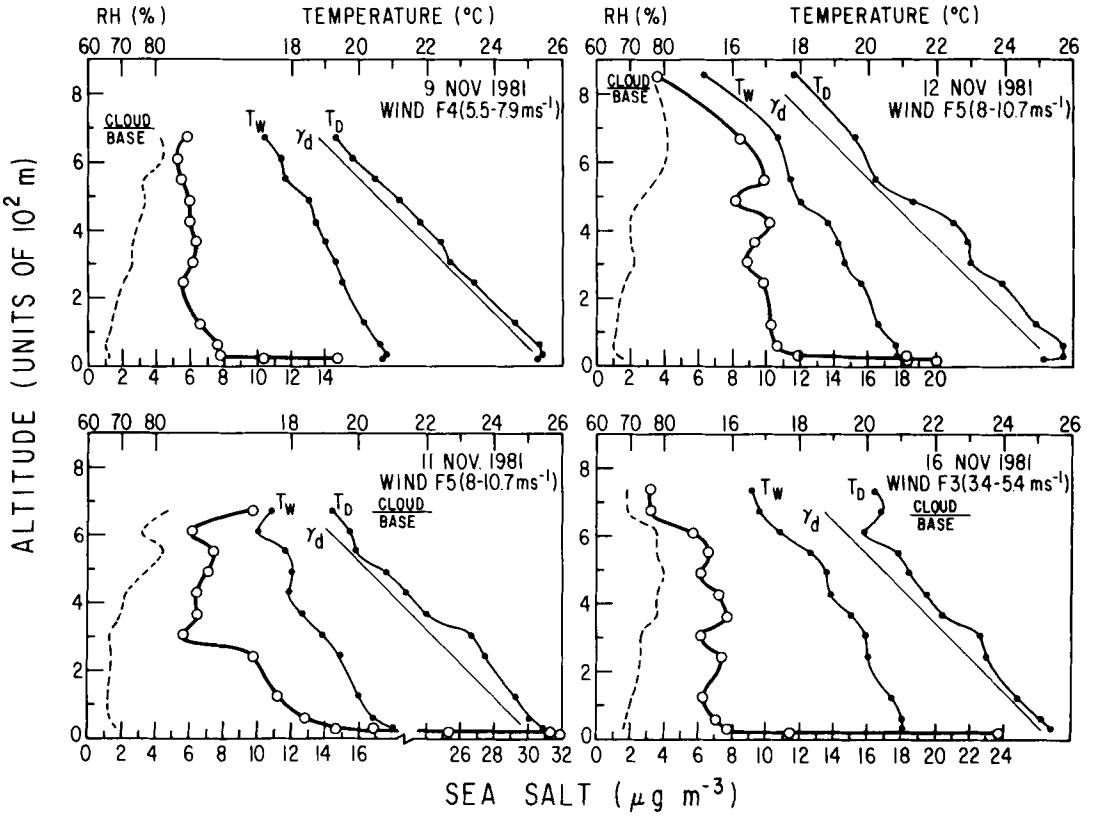

Fig. 2. Vertical profiles of sea-salt concentration, wet and dry bulb temperature, and relative humidity over the sea windward of Oahu, Hawaii, for 4 days in November, 1981.

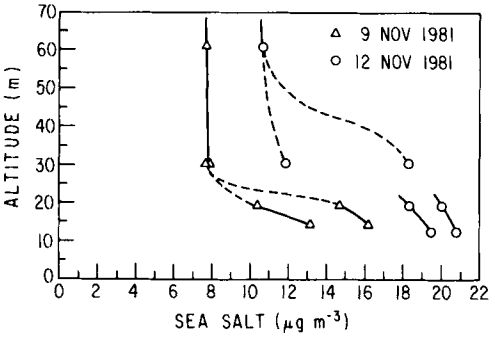

Fig. 3. Vertical profiles of sea-salt concentration in the lowest 60 m over the sea on 9 and 12 November, 1981.

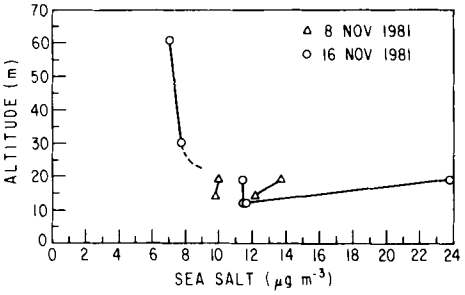

Fig. 4. Vertical profiles of sea-salt concentration in the lowest 60 m over the sea on 8 and 16 November, 1981.

obtained from the aircraft. On 8 November there were no aircraft data.

Fig. 5 shows an average of the data from the eight flights plus the tower data at 19 m. Each point is an average of 8 samples, with the exception of that at 610 m, which includes only seven. The average salt concentration at other sampling altitudes (183, 365 and >610 m) is not shown, since only 4 samples were obtained at each altitude. However, their inclusion would not significantly change Fig. 5.

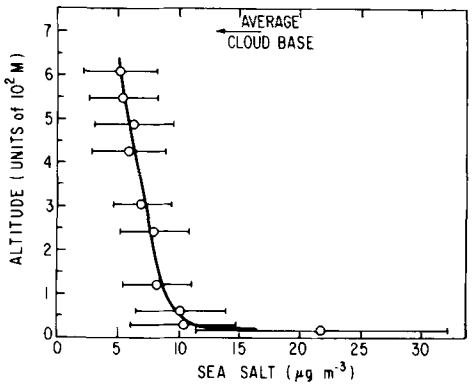

Fig. 5. Average vertical distribution of sea-salt concentration ($\bar{\chi} \pm \sigma$) near Oahu, Hawaii, for 8 days in October and November, 1981.

4. Discussion

The subtropical, marine boundary layer consists predominantly of two main layers, with a transition layer near the top of the lower layer (Malkus, 1958; Nicholls and LeMone, 1980). The lower or subcloud layer, from 500 to 1000 m thick, is generally well mixed and characterized by an adiabatic lapse rate. The lapse rate tends to become stable in the shallow transition layer that leads into the stable cloud layer. Most of our samples were obtained in the subcloud layer where the winds, temperature, and humidity are usually very steady, especially during the summer months. However, we were sampling late in the fall when the trades are disturbed by fronts passing Hawaii from the north. The weather on some of the days was far from the typical summer tradewind weather.

4.1. *Sea salt in the subcloud layer*

21 October (Fig. 1) was an exceptionally clear day with few showers. The winds were from 040°. The lapse rate was roughly adiabatic throughout most of the subcloud layer but became stable in the transition layer. A satellite photo showed no disturbed weather near the islands. Little change in salt concentration was found at higher atitudes, but a marked three-fold increase, from less than 3 to an average of about 9 $\mu g\ m^{-3}$, was found between 30.5 and 19 m. The relative humidity increased, as it did in most of the soundings, from <70% at 19 m to 75 or 80% at cloud base altitude.

The following day, 22 October, was again relatively clear though the winds had increased to F5 from 080°. Thus, we see an increase in salt; at each sampling altitude it is several times that of the preceding day. This is the only day in which a strong salt gradient between 30.5 and 19 m is not apparent.

On 26 October, moderate tradewinds (F5 from 060°) had become well-established over the islands. There were numerous shower clouds and several large convective systems that produced heavy rains and winds. As expected, the salt gradient between 19 and 30.5 m was large. The four samples at 19 m were taken about 10 min apart. The rapid change in salt concentration from about 19 to 30 $\mu g\ m^{-3}$ appears to be characteristic of the air near the sea. At 30.5 m, the salt concentration had dropped dramatically to less than 13 $\mu g\ m^{-3}$.

29 October was unusual in that the cloud base at 430 m was the lowest of any of the 8 days. Winds were moderate at F4 from 065°. Clouds were numerous and many so small that when flying in the cloud layer we had difficulty in avoiding them. The temperature structure was nearly isothermal in the lowest 60 m, possibly caused by a warm air mass moving over a colder sea. The temperature at nearly all altitudes was warmer than that on the other days.

The altitude variation of salt was unusual in that the 19 m concentrations averaged the highest of the 8 days while that in the cloud layer was the lowest. The high salt concentration at 19 m may in part be caused by the stability in the lowest 60 m preventing salt from being mixed upward. As in all the sampling at 19 m, rapid time variations were seen. Only 5 min elapsed between the end of the 26 $\mu g\ m^{-3}$ sample and the start of the 48 $\mu g\ m^{-3}$ sample.

From 250 m up through the transition layer to cloud base, salt concentrations dropped rapidly to about 1 $\mu g\ m^{-3}$, remaining very low up through the cloud layer. The marked decrease in the transition layer may have been caused by the temperature inversion. A final point of interest is that in the cloud layer, the relative humidity paralleled the salt concentration, a correlation that usually was not found during the flights. Consequently, we do not think that one caused the other but that both are controlled by a third factor not yet known.

On 9 November (Fig. 2) small cumulus clouds over the sea were capped with a layer of stratus. Winds were F4 from 070°. The temperature was nearly isothermal in the lowest 60 m (possibly for

the same reason as on 29 October). The now-familiar marked decrease in salt concentration appears as we go from 19 to 30.5 m. On this day we obtained for the first time two salt samples at 30.5 m, the second one about 5 min after completing the work out over the sea and about 300 m offshore from the tower. There was no significant difference between the two samples.

11 November had brisk tradewinds (F5 from 075°) and many small showers capped by an extensive stratus layer. A large rain system just offshore forced us to fly about 20 km upwind to get the salt samples. Even there we found many small clouds and showers. As on 9 November we got a second sample at 30.5 m, just offshore from the tower (17 μg m^{-3}) and as soon as possible after getting the 30.5 m sample 20 km at sea (15 μg m^{-3}).

The salt concentration decreased rapidly with altitude in the lowest 250 m, where the air was stable, but increased at higher altitudes where the lapse rate showed neutral stability. We appear to be dealing with two different air layers that have different histories, and thus different salt concentrations.

On 12 November, a ridge of high pressure to the northeast of the islands produced strong trade winds (F5 from 060°), many small cumulus clouds, mostly non-raining, but none of the stratus associated with the prior two salt soundings. The temperature sounding showed an inversion in the lowest 60 m. As in earlier flights, after a salt sample of 12 μg m^{-3} was obtained at 30.5 m at sea, a second sample was taken near the tower. This was higher, about 18 μg m^{-3}, and about the same as the two samples at 19 m. It seems clear that on occasions the relatively high salt concentrations commonly found at 19 m can penetrate upward to 30 m and higher.

The final salt sounding, on 16 November, was made the day after a cold front had passed over the islands from the north. We observed many small cumulus but no higher clouds. There was no rain. The winds were F3 from 090°. The cloud layer, with a relative humidity of 69%, was as dry as that of any of the other flights. Two samples 20 min apart at 19 m showed the usual variation (11 and 24 μg m^{-3}).

The difficulty in trying to explain vertical salt gradients in the subcloud layer by using only local conditions (winds, temperature, humidity, etc.) was recognized years ago (Woodcock, 1953; Eriksson, 1959; Blanchard, 1963). Since the residence time of salt particles can vary from minutes to days, depending on particle size, the past history of the air mass being studied must be known. A complete understanding of salt gradients is unlikely to be obtained until an air mass is followed for several days over the sea, during which time salt gradients, cloud structure, winds, temperature and humidity profiles, and sea surface conditions are measured.

4.2. *Sea salt in the lowest 30 m*

After completion of the sampling in October (Fig. 1), it was apparent that very large salt concentration gradients existed in the lowest 30 m. To learn more about the gradient, we got samples at two elevations on the tower on 4 days. On 9 November (Fig. 3), both aircraft samples at 30.5 m (one obtained far offshore, the other close by the tower) gave about 8 μg m^{-3}, about the same as was found at 60 m. Samples at 14.5 and 19 m (each set obtained nearly simultaneously, i.e., five of the 10-minute sampling time common to both samples, but with 15 min between the two sets) clearly showed a gradient. On 12 November, with the lowest tower sample obtained at 12.6 m, the gradient again shows up on both data sets at the tower. The highest of two different salt concentrations at 30.5 m, about 18 μg m^{-3}, was probably obtained in parcels of salt-laden air that had penetrated upward to at least 30 m altitude.

It may be no coincidence that the decrease in salt concentration with altitude between about 12 and 19 m is associated with stability in the lowest 60 m. On 9 November, we had an isothermal condition and on 12 November a temperature inversion existed (Fig. 2). Such was not the case on 16 November, and probably was not so on 8 November, where the salt concentration was either constant or increased with height along the tower (Fig. 4). On 16 November, the atmosphere was neutrally stable in the lower 150 m (Fig. 2). Salt concentrations obtained nearly simultaneously (seven of the 10-minute sampling time common to both samples) were about 11.5 μg m^{-3} at 12.6 m but nearly 24 μg m^{-3} at 19 m. Twenty min later, the sampling was repeated under the same conditions, and salt concentrations at both altitudes were 11.5 μg m^{-3}, the same as found earlier at 12.6 m.

On 8 November, no aircraft flight was made, so

we have no temperature soundings. However, from the variation of salt concentration with height in the lowest 20 m, we conclude that the atmosphere, like that on 16 November, was not stable in the lowest 100 m. Two sets of salt samples, each obtained nearly simultaneously, showed that the salt concentration at 19 m was as much as or slightly more than that at 14.5 m.

Both Figs. 3 and 4 suggest that the salt gradient between 10 and 20 m is less than that between 20 and 30 m. Though we made no measurements below 10 m, it seems clear that the salt gradient must again increase as we approach the surface of the sea. In the lowest meter we are in the layer of direct influence of large jet drops (Blanchard and Woodcock, 1980; Wu, 1979) where salt concentrations can exceed 1000 $\mu g\ m^{-3}$ (Monahan, 1968).

4.3 *Average height variation of sea salt*

Since in all but one of the 8 cases shown in Figs. 1 and 2 there is a rapid decrease in salt concentration from 19 to 30.5 m, and in general the salt decreases with height after that, we thought it of interest to plot a curve of the average height variation of salt. It is shown in Fig. 5 where it is clear that on average, the salt concentration decreases by half, from about 22 to 11 $\mu g\ m^{-3}$, as height increases from 19 to 31.5 m. The decrease with height continues, but much more slowly. This curve must not be considered an average for the tradewind region near Hawaii, but an average *only* for the 8 soundings of Figs. 1 and 2. The average wind speed was 7.4 $m\ s^{-1}$. The one aspect of Fig. 5 that we believe applicable to most situations in the Hawaiian tradewinds, and probably to other tradewind regions as well, is the marked salt concentration gradient in the lowest 50 m.

Inasmuch as the salt concentrations at 19 m and less were obtained in the afternoon and several hours after those obtained at higher altitudes, it might be argued that the concentrations are always higher in the afternoon (perhaps because of strong sea breezes), and if we had obtained the low-level concentrations in the morning, we might have found them compatible with those found at 30.5 and 61 m from the aircraft. We consider this unlikely, because, unlike other larger islands, the winds crossing the windward shore of Oahu at the location of the tower show no significant change from morning to afternoon. This was determined by one of us (AHW) after examination of 262 days of wind speed data obtained at the tower in 1968 and 1969.

The average decrease of about 1 $\mu g\ m^{-3}$ per meter altitude shown in Fig. 5 between 19 and 30 m is less than the 3 to 4 $\mu g\ m^{-3}\ m^{-1}$ found in two of the soundings made by Durbin and White (1961) over the English Channel. Our largest gradient, found on 29 October (based on the highest salt concentration shown in Fig. 1, about 47 $\mu g\ m^{-3}$), is 3.3 $\mu g\ m^{-3}\ m^{-1}$.

These gradients are probably caused by very large salt particles (>10 μm radius when equilibrated at a relative humidity of 70%, and about twice that size when they leave the sea as droplets of seawater) that are so difficult to sample with cascade impactors (McDonald et al., 1982). These large particles, which contribute significantly to the salt concentration and even more so to the rate of dry deposition (McDonald et al., 1982), are controlled primarily by gravitational forces and little by turbulence and consequently do not penetrate much above an altitude of 20 m.

5. Conclusion

On each of 8 days, gradients in salt concentration averaging about 1 $\mu g\ m^{-3}\ m^{-1}$ have been found between heights of about 20 and 30 m over the sea windward of Oahu, Hawaii. The origin of such marked gradients is not known, but is probably caused by large salt particles with a short residence time that do not rise much above 20 m over the sea. This could be confirmed by making measurements of the sea-salt particle-size distribution. In future work, it would be best not to combine data from two sampling platforms, but to obtain the gradient of the particle-size distribution either by using a tower where 30 m sampling heights can be obtained or by using an aircraft capable of making long salt sampling runs at heights as low as 20 m.

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