

A CLIMATOLOGY OF SOLID ATMOSPHERIC CONTAMINANTS AT ASPENDALE, NEAR MELBOURNE, VICTORIA, 1966 TO 1971

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ABSTRACT

Air concentrations of solid matter near the surface have been recorded on a routine weekly basis since 1966. These allow documentation of a gradual increase until late 1968, followed by a general decrease. Overall, the values found are lower than usually reported in urban situations. Particulate concentrations are found to be highest in conditions of light winds from the industrial areas located to the north and northwest. A correlation is found with mean maximum temperature, but no rainfall effect is found. Week-to-week dust concentrations are found to vary statistically by about 20% (one standard deviation) and on a monthly basis this figure reduces to about 10%. The particle mass spectrum is found to maximize at about 25 μm diameter, giving a mean sedimentation velocity of about 1.3 cm s^{-1} which is supported by dust-fall considerations. The average residence time of the dust observed at Aspendale appears to be of the order of 11 hours.

INTRODUCTION

Aspendale, an outer suburb of Melbourne, is located about 30 km southeast of the city, on the shore of Port Phillip Bay (Fig 1). As part of a more extensive program studying atmospheric radioactivity, solid matter concentrations in air near the surface have been routinely measured since 1966. In this period, Aspendale has changed in character from a lightly populated beach area to a more densely settled residential zone. In housing density, it is now comparable to many recently developed urban areas.

In one important aspect, the experiment described here differs from most other similar studies. Since the CSIRO Division of Atmospheric Physics located at Aspendale is primarily engaged in atmospheric research, a large number of meteorological parameters are measured on a routine basis. Thus the present data can be compared with a large quantity of atmospheric information.

However, because of the location of Aspendale, little long-term correlation with most meteorological parameters would be expected. Most of the variance should be attributable to such considerations as, for example, traffic density on the nearby major highway and the proportion of the exposure time for which pollution sources were upwind. These, and other similar criteria, are obviously difficult to quantify. Thus the experimental site at Aspendale is far from ideal for studies of air pollution.

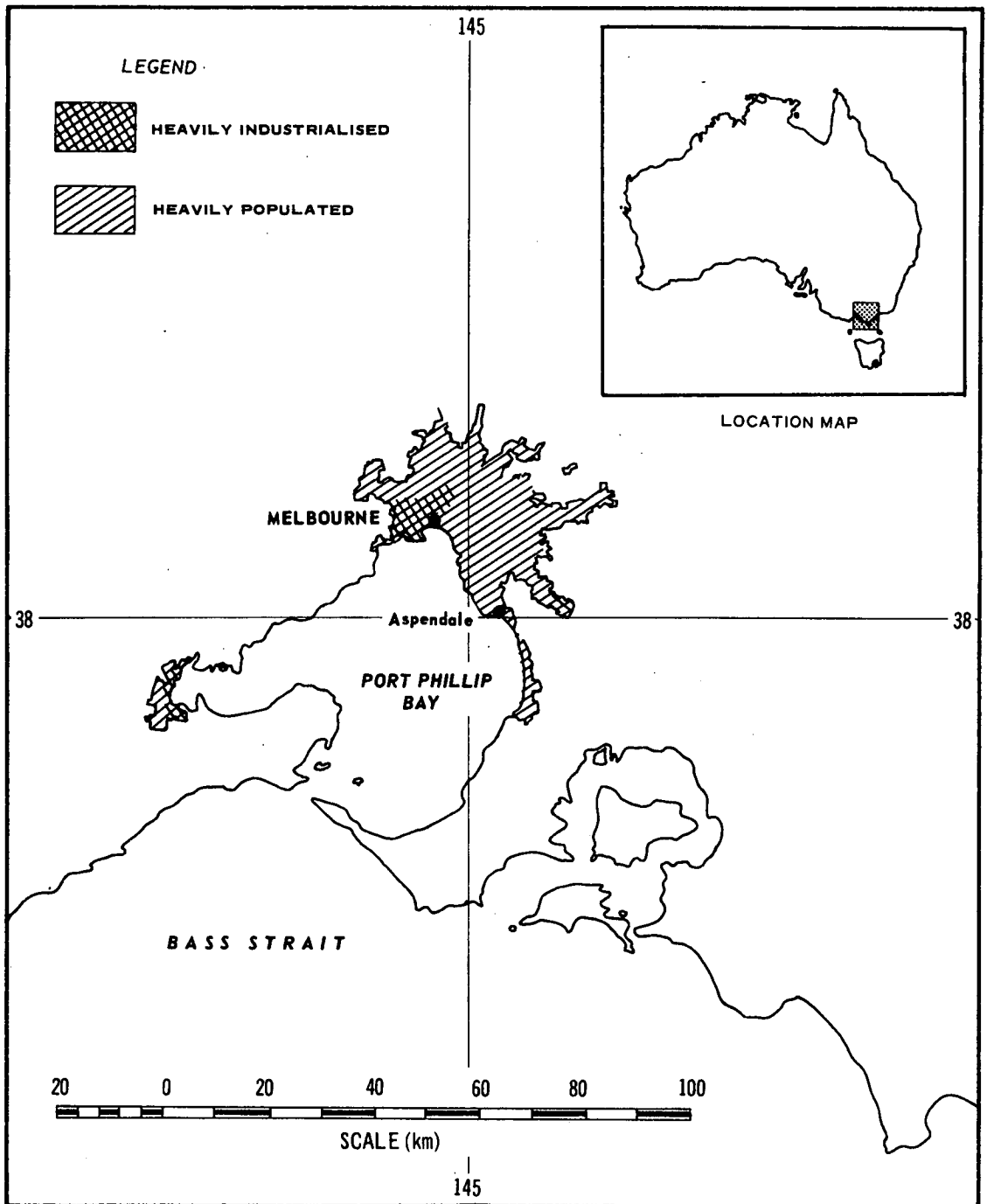


Fig 1 Map of Port Phillip Bay and surrounding areas showing the main residential and industrial zones.

The data are reported here from the viewpoint of long-term changes in the local air associated with the changing residential nature of the area and the general expansion of industry. A search for possible meteorological effects, including such obvious parameters as rainfall, temperature and wind velocity is also discussed.

DATA ACQUISITION

Air is drawn through highly efficient polyester filters (exposed at a height of 1.5 m) at a sampling rate of about $150 \text{ m}^3 \text{ hr}^{-1}$. Filters are normally changed every Monday and after collection are allowed to come into equilibrium with standard conditions of temperature and humidity. Comparison of the weight after collection with that prior to exposure yields the time averaged particulate loading of the atmosphere near the surface.

The measurements of dust concentration are probably accurate to about 5%, and to a large extent this error results from our inability to determine such high sampling rates to a greater degree of certainty. Routine meteorological measurements are made at the same site.

COMPOSITION OF SAMPLES

The filter material employed effectively retains particles larger than about $0.2 \mu\text{m}$ diameter. However, it is obvious that some upper limit on the size of particles sampled must exist, since abnormally large particles will not be drawn into the sampling system. The velocity at which air is sampled is more than 2 m s^{-1} , much greater than the sedimentation rate of most particles. Hence the system may be considered relatively efficient.

A microscopic study of particles from exposed filters shows that in conditions of moderate dust content some particles in excess of $100 \mu\text{m}$ are detected. Between about 1 and $100 \mu\text{m}$, the slope of the size distribution obtained is $n = 3.5$ in $dN/dr = C.r^{-n}$ (in conventional notation), a somewhat smaller value than is usually quoted for this exponent. Stern (1968) quotes a value of about 4, and other workers have found that even higher values apply to the distributions of larger particles. Thus, there is certainly no evidence that the measurements made at Aspendale are low because of lower filtering efficiencies for larger particles.

The same particle size study indicates that the mass spectrum found at Aspendale peaks at about $25 \mu\text{m}$ diameter in average conditions, in complete agreement with Chepil (1957) who studied suspended dust near the surface. Large particles are irregular in shape and are clearly of combustion origin. Particles less than about $5 \mu\text{m}$ diameter tend to be spherical, and some of these may be sea salt from the nearby bay. The relatively small size of these particles allows some confidence to be associated with the assumption that the particulate concentrations reported here predominantly reflect the influence of man-made pollution.

THE SECULAR CHANGE

The time trend of the atmospheric particulate data presented as quarterly averages is illustrated in Fig 2. A peak in the long-term average concentrations is seen to have occurred in the third quarter of 1968, preceded by a general increase since the commencement of observations two years previously. Between 1968 and 1971, concentrations have generally decreased. (Subsequent results, not used here, have indicated a return to the earlier higher levels.)

It is possible that the early increase in concentrations resulted from the increase in local population density. Since the measurements were made at a low height, they might reflect the influence of sources in the near vicinity. Accepting

this assumption (about which later considerations will prove important), it is difficult to explain the subsequent decrease in concentrations, although two possibilities appear reasonable. Windblown dust from the ground surface might well decrease as the surface re-adjusts to its new surroundings after a period of building activity (*ie*, as plants cover previously bared soil). Also it is conceivable that the decrease in particulate concentrations between 1968 and 1970 might have resulted from the decreasing popularity of coal and wood as energy sources and the increasing efforts made by industry to minimize pollution. It is not possible to draw specific conclusions concerning this aspect, and the above considerations are presented here merely to illustrate the complexities involved in interpreting such time trends. There is no evidence of an annual cycle modulating the time curve of Fig 2.

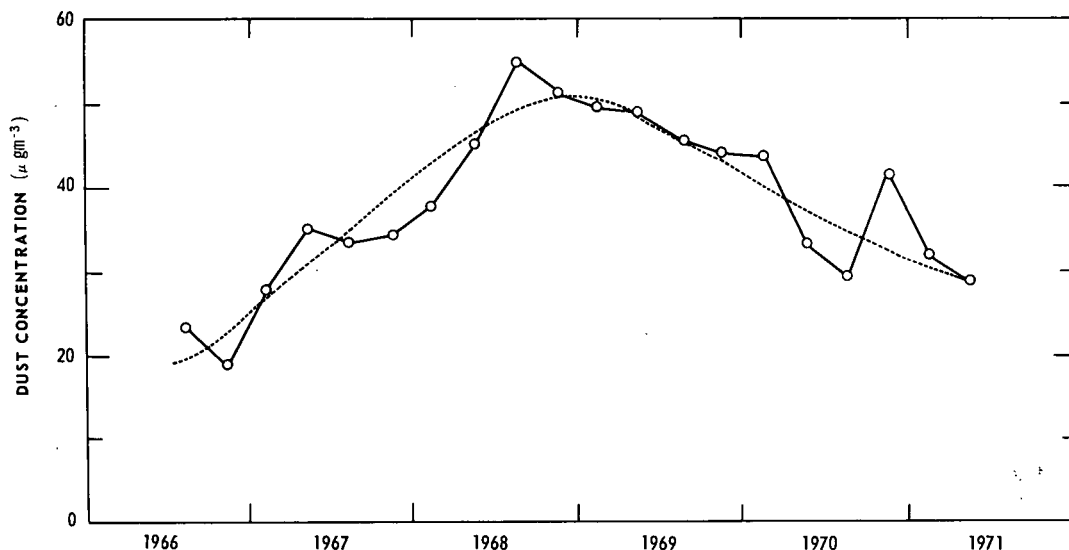


Fig 2 Total particulate concentrations measured at Aspendale, 1966 to 1971.

In general, the concentrations found at Aspendale are lower than those usually reported for urban environments. That this is, in part, a result of the proximity of large bodies of water will become apparent later. For comparison, Stern (1968) quotes urban values for the USA which are more than double those reported here, while the US Department of Health, Education and Welfare (1969) reports between 60 and 200 $\mu\text{g m}^{-3}$ for urban areas and between 10 and 60 $\mu\text{g m}^{-3}$ for non-urban areas (annual means). In European non-urban areas, Junge (1963) gives particulate concentrations of about 70 $\mu\text{g m}^{-3}$, higher than the peak value during 1968 at Aspendale.

The dotted line drawn in Fig 2 is the smoothed variation in dust concentration. Scatter about this line is indicative of changes in meteorological parameters and in the source and sink functions. Information on the short-term variability in particulate concentrations is of some physical importance, since it allows estimation of the probability of exceeding preset dangerous levels.

A statistical study of the data obtained during the period when there was no significant time trend (the 45 week period centred on November, 1968) shows that the standard deviation of the particulate concentrations decreases with increasing sampling time. In accordance with statistical theory, a power law relationship is found in which the observed standard deviations (σ) are closely proportional to the

inverse square root of the sampling time. One week samples give $\sigma = 22.0\%$, but when the one week samples are combined into three week periods σ is reduced to 12.6% and if nine week averages are constructed σ becomes 7.9%. Compatible with these values is the standard deviation associated with daily measurements, which from the small quantity of data available over a different period is found to be 40.4%.

The variations which contribute to these standard deviations result from changes in sources and sinks, and from atmospheric effects. An investigation of these, using the weekly averaged data and looking for those situations liable to give rise to abnormally high or low particulate concentrations, is studied below.

RELATIONSHIPS WITH ATMOSPHERIC VARIABLES

Probably the most important atmospheric parameter contributing to the variability in any near-surface pollutant is wind velocity. Its effect in a shore situation such as Aspendale must be even greater than at an inland site, since not only do the conventional considerations of ventilation, surface scavenging, atmospheric mixing and inversion height apply, but also the wind direction is of major importance. It is important to recognise that conventional models (such as that of Llewelyn, 1971) refer to pollution *within* a city, whereas the present study is of particulate concentrations at a seaside location in the environs of a large industrial complex. It is on this basis that the following analysis heavily weights consideration of extreme cases, rather than of the more commonly considered parameters contributing to an "Air Pollution Potential".

Some daily dust measurements made in 1970 confirm the importance of wind direction: significantly greater particulate levels are found when the wind is blowing from the industrial areas to the north and northwest. These data are included in a separate report (Galbally and Goodman, 1972).

Time trends contribute much to the variance in particulate concentrations - this is shown in Fig 2. Consequently, the analysis presented here will be concerned with normalised variables (X'_i , Y'_i , etc) obtained from the corresponding weekly averages (X_i , Y_i , etc) by dividing each value by the five week running mean centred on it. That is

$$X'_i = \frac{5X_i}{X_{i-2} + X_{i-1} + X_i + X_{i+1} + X_{i+2}}$$

by which procedure long-term changes are effectively removed. By normalising in this way, the earlier considerations regarding the statistical variability of the data suggest that more than 50% of the variance in weekly samples is retained.

Correlations performed on data corrected in this manner show that the only meteorological variable which significantly affects the weekly averaged dust data is the averaged maximum temperature (correlation coefficient 0.35 from 243 data pairs). No relationship is found with wind speed, rainfall, or minimum temperature, even when lag correlations are considered. The lack of a significant wind speed effect confirms, to some extent, the earlier suggestion that mixing depth might be of minor importance in the present case. On the other hand, the finding of a significant correlation with mean maximum temperature tends to confirm the importance of wind direction, since the highest temperatures will be associated with over-land trajectories.

It is possible that the use of a conventional correlation technique might cause some effects to be overlooked. For example, statistical noise might swamp the possible occurrence of high particulate concentrations in periods of abnormal wind speeds, or when there is little or no rainfall. In Table 1, the results of an analysis of extreme values are given: for each meteorological variable, the ten lowest and ten highest normalised values are selected, and the corresponding normalised dust concentrations are calculated.

The data in Table 1 suggest that high concentration levels tend to occur in conditions of

- . abnormally low wind speed
- . abnormally high maximum temperature
- . abnormally high temperature range

Average temperature range is included in the variable list to allow a comparison with the predictions of conventional air pollution models, which generally associate high concentrations with low ventilation rates. In this context, temperature range can be considered as an indicator of the maximum height to which the lower atmosphere is mixed during daytime, and hence of the depth of the boundary layer over which pollutants can be dispersed. However, a negative correlation between particulate concentration and temperature range would result, which is not supported by the present data. We must conclude that the effect found is probably related to wind direction considerations rather than the conventional mixing depth approach, even though the wind speed relationship found above tends to confirm the predictions of the model.

Table 1 An analysis of extremes, showing the effects on the normalised dust concentration of abnormally high and low values of five meteorological parameters. In constructing this table, the ten highest and ten lowest values of each variable are selected and average normalised dust concentrations are calculated in each class

	High Extreme		Low Extreme	
	Mean	Dust	Mean	Dust
Rainfall	3.65	1.11 ± 0.10	0.00*	0.98 ± 0.06*
Wind Speed	1.41	0.94 ± 0.07	0.51	1.17 ± 0.06
Max Temp	1.23	1.25 ± 0.10	0.80	0.85 ± 0.09
Min Temp	1.34	1.16 ± 0.09	0.59	1.07 ± 0.09
Temp Range	1.52	1.34 ± 0.09	0.58	0.91 ± 0.08

* 28 values contribute to the zero-rainfall average

A further consideration is of interest, namely the extremes of particulate concentrations themselves. These are listed in Table 2, together with normalised values of the other parameters under consideration. The data are shown in full to illustrate the complexity of the problem. For example, while the two highest dust levels are associated with low rainfall periods, so are the two lowest values. On the whole, the values listed in Table 2 confirm the previous conclusions.

Table 2 Extremes of normalised particulate concentrations and values of other parameters averaged over the same period. X'_1 to X'_6 are, respectively, particulate concentration, rainfall, wind speed, mean maximum temperature, mean minimum temperature and the temperature range (maximum minus minimum). \bar{U} and θ define the vector mean wind, while \bar{u} is the average wind speed. Velocities are in m s^{-1} and θ is in degrees. The month of each observation is identified numerically (starting with January as 1) to give an indication of the seasonal dependence

X'_1	X'_2	X'_3	X'_4	X'_5	X'_6	\bar{U}	θ	\bar{u}	Month	\bar{u}/\bar{U}
HIGH VALUES										
2.03	0.05	0.93	1.24	1.09	1.40	1.19	283	2.88	11	2.4
2.02	0.15	1.29	0.98	0.93	1.03	1.28	205	3.36	2	2.6
1.89	0.85	1.19	1.11	1.09	1.12	0.83	279	3.32	10	4.0
1.75	2.04	0.87	1.10	1.12	1.07	3.15	335	4.85	9	1.5
1.69	3.28	1.02	1.14	1.26	1.00	0.99	276	3.02	2	3.0
1.61	0.39	0.96	0.96	1.02	0.90	2.63	285	4.28	9	1.6
1.57	2.04	0.87	1.10	1.12	1.07	1.12	168	2.59	2	2.3
1.54	0.02	0.97	1.05	1.08	1.02	0.77	200	1.85	8	2.4
1.51	3.64	1.08	0.91	1.05	0.76	1.09	244	3.08	4	2.8
1.48	0.27	0.77	1.08	1.09	1.07	0.64	137	1.61	2	2.5
LOW VALUES										
0.39	0.00	0.74	0.94	0.98	0.89	1.11	161	1.20	2	1.1
0.41	0.00	1.10	0.75	0.78	0.72	2.26	200	2.94	2	1.3
0.45	1.84	1.11	0.95	1.08	0.83	3.40	295	4.50	8	1.3
0.46	2.22	0.82	1.00	1.06	0.96	2.86	220	3.48	9	1.2
0.48	1.84	1.09	0.83	0.66	0.98	1.04	230	1.72	12	1.7
0.50	1.14	1.24	0.96	1.06	0.83	1.47	198	4.20	10	2.9
0.50	0.60	0.93	0.97	0.97	0.97	1.41	160	2.30	11	1.6
0.51	0.00	0.83	0.94	0.70	1.11	1.62	320	2.90	9	1.8
0.51	1.27	1.05	1.02	1.11	0.86	4.40	300	4.84	6	1.1
0.55	1.96	1.22	0.84	1.09	0.58	2.41	165	4.44	11	1.8

WIND DIRECTION EFFECTS

The map of the surroundings of Aspendale (Fig 1) shows industrial areas located to the north and northwest and major residential zones to the north and north-east. In conditions when the wind is consistently from these directions, high particulate concentrations may be expected. For this reason, wind information is also listed in Table 2. Vector mean winds are calculated from Dines anemometer traces, evaluated at six hour intervals.

Consider, for example, the lowest normalised particulate concentration recorded. This occurred when the winds were, on the average, from 161° , with little variation in wind direction through the week as is evidenced by the extremely low ratio of the average wind speed to the vector mean wind. This ratio tends to one for winds of constant direction, and increases as the winds become more variable. Table 2 shows that, in general, low particulate concentrations are associated with periods of low wind variability.

In every case, the high dust levels listed are associated with either high wind direction variability or with mean directions from one of the surrounding industrial areas. However, the present definition of wind direction variability is not sufficiently precise to allow specific conclusions regarding the importance of upwind pollution sources. To obtain better information, it seems preferable to consider the proportion of time for which particular sources were upwind, and to look for correlations with the measured dust concentrations.

On the basis of the six hourly Dines anemometer determinations of the wind speed and direction, the proportion of each sampling period for which conditions were calm (T_c) and that for which the wind was from a particular direction (T_{θ}) were calculated^c for the data list of Table 2. Partial correlation coefficients were then calculated between the dust concentrations and T_c and T_{θ} . The results of this analysis are listed in Table 3. It is immediately obvious that southerly wind components lead to lower dust concentrations with relatively little effect of calm conditions, while in northerly winds the concentrations are high, particularly if winds are light. A more thorough investigation of the wind direction effect shows that dust concentrations are most sensitive to winds from between 300° and 20° , when about 60% of the total variance is accounted for by the two parameters considered. Reference to Fig 1 shows that this range of wind directions is precisely that for which there are large industrial or residential areas upwind of Aspendale, and hence it must be concluded that these account for a large proportion of the particulate variance observed. It is of interest that there is no evidence of any increase associated with winds from the industrial complex due west of Aspendale.

Table 3 The effects of wind direction and of periods of calm on weekly particulate concentrations at Aspendale. Partial correlation coefficients are given as a measure of the effect of winds from specific directions on concentrations when that of calm conditions is removed ($R(\theta)$) and of the complementary correlation ($R(c)$) between concentrations and the incidence of calm after allowance for the effect of wind direction. The variables used as measures of calm and of winds from specified directions are the proportions of the sampling time for which these conditions existed. Data are listed in Table 2, and the 5% significance level is at 0.46

	$R(\theta)$	$R(c)$	Proportion of variance accounted for
Northerly components	0.59	0.74	0.90
Southerly components	-0.63	0.45	0.60
Winds from between			
260° and 320°	0.33	0.43	0.29
280° and 340°	0.29	0.41	0.25
300° and 360°	0.52	0.55	0.57
320° and 20°	0.54	0.56	0.61
340° and 40°	0.35	0.43	0.31

Thus, whereas it was suggested above that some of the airborne dust observed at Aspendale might be of extremely local origin, it now appears that a large proportion can be related directly to the existence of sources to the north and north-west.

THE DEPOSITION OF PARTICLES

The concentrations measured at Aspendale can be compared with dust deposition measurements made in the Melbourne area (reported by Le Roy, 1971) to obtain the average sedimentation rate and the mean residence time. From the published dustfall survey, the rate of particulate deposition at the surface varied from 2.6 to 18.5 tons per square mile (6.8 to 48.8 t/km²) per month during 1970. The low value was obtained in a residential urban area, the high value in the central commercial section of the city of Melbourne. No measurement reported is applicable to a bayside situation such as Aspendale. Assuming that the average 1970 dustfall applicable to the present data amounted to about 3.0 tons per square mile (7.9 t/km²) per month, and that the average atmospheric concentration near the surface was 35 µg m⁻³ (as indicated by Fig 2), a sedimentation velocity of about 1.3 cm s⁻¹ is obtained.

In the section, composition of samples, the mass spectrum of particles collected at Aspendale was found to peak at about 25 µm diameter, the larger particles having the appearance of ash. Assuming that the density of these particles is 0.75 g cm⁻³ (the value commonly employed for timber ash), then the sedimentation rate predicted by Stokes' Law is about 1.4 cm s⁻¹. The close agreement between this result and the value deduced from the dustfall considerations allows some confidence to be associated with the data.

Knowledge of the average depth of the mixing layer allows the above sedimentation velocities to be interpreted as mean residence times for particles in the air. During daytime, the atmosphere is well mixed up to an average height of about 1 km, but at night a much lower value is appropriate. If it is assumed that the overall average mixing depth is about 500 m, then the sedimentation velocity of 1.3 cm s⁻¹ implies a mean residence time of 11 hours. Clearly, nearer to sources of particulate pollution larger particles will be present whose greater sedimentation velocities will result in a shorter residence time.

The value of the residence time obtained here is obviously an approximation, and little weight should be placed upon it. It is felt that much greater reliance can be associated with the sedimentation rate calculation, particularly considering the agreement with the earlier particle size discussion.

CONCLUSIONS

In general, the air concentrations of particles at Aspendale are low. These low values result, in part, from the geographical location of the observing site, which is some distance away from the major sources of particulate pollution. After reaching a maximum in 1968, the particulate levels at Aspendale have shown a general decrease through 1970. The cause of the decrease is not certain, but it might result from local (residential or land reclaiming) effects, from improvements in industrial pollution control, or from changes in the long term weather pattern.

Abnormally high particulate concentrations are likely to occur in periods when winds are light, especially from the industrial areas. Over long averaging periods, wind variability and maximum daily temperature appear to be important. No significant rainfall effect is found.

The standard deviation of any single average dust measurement decreases with increasing sampling time (as must be expected even with random data), dropping from about 20% for a weekly average to about 10% for a monthly.

A conventional particle size distribution is found, with the peak of the mass spectrum being at about 25 μm diameter. The sedimentation rate predicted for particles of this diameter agrees with that deduced from dustfall measurements, about 1.3 cm s^{-1} . The average residence time for particles of the type studied here appears to be of the order of 11 hours.

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