Tellus (1994), 46B, 123–131
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TELLUS
ISSN 0280–6509

Coherence between seasonal variation in satellite-derived cloud optical depth and boundary layer CCN concentrations at a mid-latitude Southern Hemisphere station

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(Manuscript received 10 August 1993; in final form 22 October 1993)

ABSTRACT

A 7 year record of satellite-derived cloud optical depth for marine stratus clouds upwind of the Cape Grim Baseline Station is found to have a seasonal cycle in phase with that apparent in long-term (10 year) records of CCN concentration measured in marine boundary layer air. The amplitude and phase of the mean seasonal cycle in cloud optical depth was reproduced reasonably well by a semi-quantitative cloud model initialised with observed monthly mean CCN size distributions. While this coherence does not prove a causal relationship, it is consistent with the hypothesis that emissions of reduced sulfur gases by oceanic phytoplankton can influence cloud radiative transfer properties via an influence on CCN concentrations.

1. Introduction

Charlson et al. (1987) have suggested the possibility of biological regulation of climate as a result of variations in emissions of dimethyl sulfide (DMS) by oceanic phytoplankton. DMS is regarded as the gaseous precursor of sulphate aerosol that makes up the principal constituent of cloud condensation nuclei (CCN) in the remote marine atmospheric boundary layer (MABL). Since at low concentrations CCN are thought to control the number of water droplets (N) in clouds, which in turn influences the albedo of clouds at visible wavelengths, Charlson et al. (1987) hypothesise a possible climate feedback loop connecting oceanic DMS emissions, via CCN, with the radiative transfer properties of marine stratus cloud. DMS-production is linked to sea surface temperature, which suggests that one way to establish the link between DMS, CCN, cloud microphysical and radiative properties is to look for their seasonal variations.

Field measurements to assess the various biological, chemical and physical connections implicit in the Charlson et al. (1987) hypothesis have on a number of occasions yielded ambiguous results, a fact that Bates et al. (1987) ascribe to the difficulty in finding high correlations over short measurement periods between species having multiple source and sink pathways and widely varying lifetimes. However using multi-year data records with each data point averaging over time periods considerably longer than one day (i.e.,, using weekly and monthly averaged data) coherent relationships have been evident between atmospheric aerosol parameters and DMS data obtained at Cape Grim (144° E, 41° S), in Southern Ocean air. Specifically, the Cape Grim DMS , aerosol and rainwater methanesulfonate (MSA) data, some going back to 1976, all show distinct annual cycles in concentration with similar phases (Ayers et al., 1986; Bigg et al., 1984; Gras, 1989; Gras, 1990; Ayers and Ivey, 1990; Ayers and Gras, 1991; Ayers et al., 1991). The phase of these cycles, which exhibit mid-summer maximum and mid-winter minimum, reflects the expected seasonality in the

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biological processes (which have a summer maximum in productivity) that drive the seasonal cycle in DMS emissions from the ocean surface (Bates et al., 1987; Erickson et al., 1990).

This coherent seasonal behaviour is known to be geographically widespread over the oceans of the southern hemisphere, since similar summer maxima/winter minima have been observed in levels of aerosol sulfate and MSA at Norfolk Island (167.59°E, 29.05°S), in CN measurements at Macquarie Island (158.58°E, 54.29°S) and at Mawson on the Antarctic continent (62.53°E, 67.36°S), in DMS, SO2 and rainwater sulfate concentrations at Amsterdam Island (77.40°E, 37.55°S), and in surface water DMS concentrations at Davis (79.00°E, 68.58°S), on the Antarctic coast (Savoie et al., 1989; Pitaud et al., 1992; Wagenbach et al., 1988; Nguyen et al., 1992; Gibson et al., 1990; Prospero et al., 1991).

The existence of such a consistent pattern involving aerosol chemical and physical parameters and the gaseous aerosol precursors has lent considerable support to that part of the Charlson et al. (1987) hypothesis linking emissions of DMS with aerosol sulfate, CN and CCN concentrations. However this pattern also offers the possibility of evaluating other parts of the hypothesis by using the observed seasonal variation in CCN concentration as a model for CCN variations induced by climate change.

According to the Charlson et al. (1987) hypothesis the observed seasonal cycle in CCN measured in marine air masses at Cape Grim should produce a measurable change in cloud droplet numbers, and thus cloud optical depth. It is worth noting that quite apart from the logic of the arguments put by Charlson et al. (1987) airborne data were obtained in the southern hemisphere in modified marine clouds as long as two decades ago confirming the theoretically expected tight relationship between sub-cloud CCN concentration and cloud droplet concentration near cloud base (Twomey and Warner, 1967).

The approach adopted here is to seek evidence of seasonal variation in optical depth of marine stratus cloud that, on the basis the Charlson et al. (1987) hypothesis, should result from the observed factor of 2 seasonal change in CCN at Cape Grim reported by Gras (1990). Accordingly, a record of satellite derived cloud optical depth is presented that appears to exhibit the same seasonal cycle as ground-based observations of CCN at Cape Grim. A model of cloud droplet growth on CCN is then used to verify that the expected seasonal variation in cloud droplet number concentration is sufficient to largely explain the phase and amplitude of the optical depth signal.

2. Satellite-derived cloud optical depth

Fig. 1 shows a time series of observed, monthly averaged cloud optical depth between 1984 and 1990. These optical depths are derived from the International Satellite Cloud Climatology Project (ISCCP) C2 data set (Rossow and Shiffer, 1991). The chosen site for optical depth determination is the gridpoint of complete oceanic coverage (41°25′S, 141°25′E) closest to Cape Grim. Although noisy, the optical depth time series shows a seasonal cycle with peaks in the months of December and January. Fig. 1 includes the concurrent time series of Cape Grim data for CCN active at 0.23%, a value that is typical for marine stratus clouds (Pruppacher and Klett, 1978). Both records in Fig. 1 show a trend towards lower values, amounting to a 15% reduction in optical depth, and a 20% reduction in CCN over the 7-year period.

The optical depth retrieval procedure is based on a simulation of the measured radiance at the satellite under cloudy conditions using a model atmosphere in which ozone absorption, Rayleigh scattering and reflection at the ocean surface are taken into account (Rossow et al., 1989a; Rossow and Lacis, 1990; Rossow et al., 1989b). The retrievals are averaged over a 2.5° latitude-longitude grid and represent a selection of data with optical depths between 3.5 and 125, and cloud top pressures higher than 680 hPa. This classification selects only clouds that are likely to be in the MABL (although this is not certain), and excludes very thin clouds for which the optical depth retrieval may be prone to error.

Both data records shown in Fig. 1 were detrended by subtracting linear fits from the data and spectra were computed using a Fast Fourier Transform analysis (Fig. 2). The 12-month harmonic waves in the optical depth and CCN time series were in phase peaking in January, with a confidence level of 98%, 99% respectively, indicating that the spectral peaks were evidence of

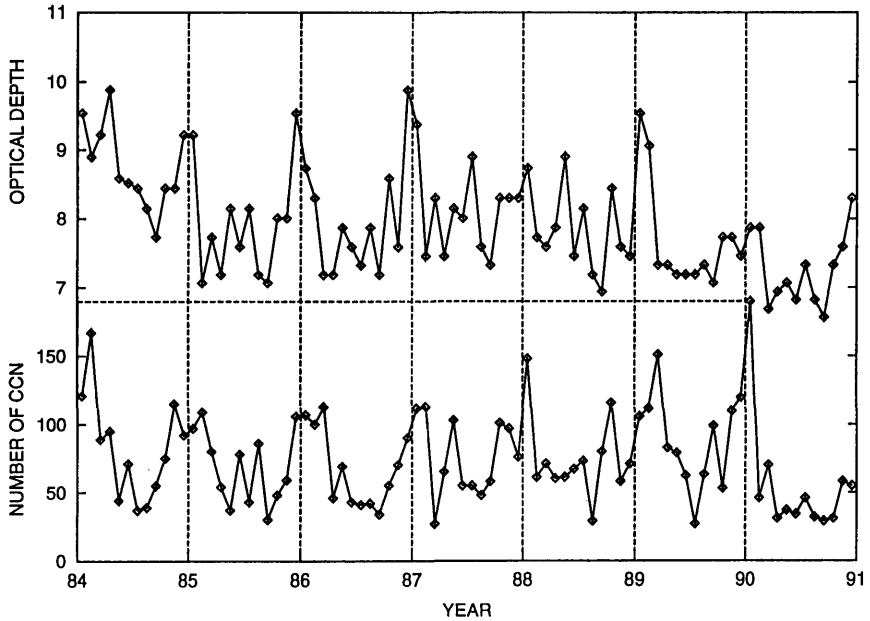

Fig. 1. Monthly average values of the stratus cloud optical depth (upper time series), derived from the ISCCP-C2 data set. Data are averaged over a 2.5° by 2.5° latitude-longitude region centered at 41°25′S, 141°25′E. Shown below is the time series of CCN at 0.23% supersaturation observed at Cape Grim (40°41′S, 144°41′E) over the same time period.

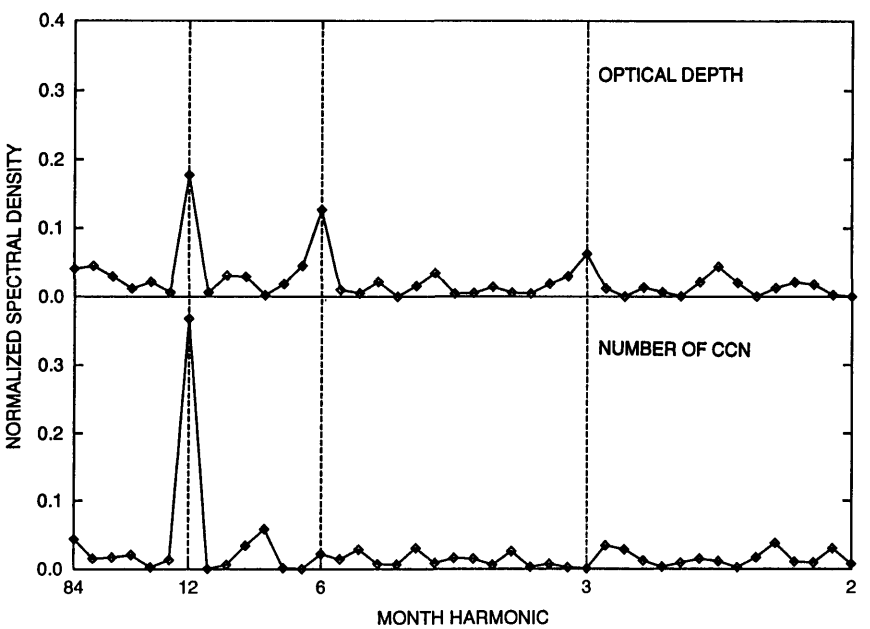

Fig. 2. Periodograms of monthly averaged values of the stratus cloud optical depth, derived from the ISCCP-C2 data set (upper segment) and CCN concentration (lower segment). Data were detrended using a linear mean square estimation, and cosine tapered at 5% of the interval before the Fourier transform was performed. The 12-, 6- and 3-month harmonics are emphasized.

real periodic signals (Fisher, 1929). The 6 month harmonic wave in the optical depth time series had a confidence level of 93%, and peaked in January and July. The three month harmonic wave is not significant although it appears to be slightly elevated above the remainder of the harmonics.

3. Predicted seasonal cycle in optical depth

At the Cape Grim observatory the CCN measurements are made at 100 m altitude, whereas previous airborne field work of Cape Grim in both winter and summertime periods indicates that cloud base is typically at 500–1000 m altitude (Mossop, 1985). This difference in altitude is not expected to affect our considerations, since the marine boundary layer was usually found to be well mixed to cloud base, which allows us to assume that the Cape Grim CCN measurements at 100 m provide a good approximation to the actual CCN concentrations near cloud base. Objective evidence in support of this assumption is provided by 22 vertical profiles of CN concentration data taken over a 13 month period in 1981–1982, and reported by Bigg et al. (1984). The correlation coefficient between CN concentration at a given height and at Cape Grim (100 m) was close to 1 throughout the sub-cloud layer, leading Bigg et al. (1984) to conclude that "below cloud base, normally 500 to 1000 m, the atmosphere is well mixed". Despite this evidence recent studies of the structure of the lower marine boundary layer in tropical regions suggest that the temperature gradient is often weakly stable while the vertical gradient in moisture is slightly negative (Betts and Albrecht, 1987), so that small departures from the well-mixed condition are relatively common. Nevertheless, in absence of additional evidence on the thermodynamic structure of the boundary layer over the Southern Ocean we believe that the assumption that the PBL is well-mixed is realistic especially in view of the high wind speed conditions that are often encountered there.

On this basis, mean monthly surface CCN data were used as input to a closed parcel adiabatic model of cloud droplet growth (Ayers and Larson, 1991) to predict seasonally varying cloud droplet concentrations, from which to predict cloud optical depth. Size distributions of CCN used as input to the cloud model were determined from the concentrations of CCN active at 5 supersaturations between 0.23 and 1.2% and the concentration of particles with radii larger than 0.05 μm measured with a PMS ASASP-X particle size spectrometer. Mean size distributions were derived from measurements made continuously between January 1991 and June 1992. For particles in the CCN size range the size distribution is bimodal with one mode (ammonium-bisulfate) centred around 0.07–0.1 μm radius and a larger mode (sodium chloride) centred around 0.3 μm radius (Gras, 1993). The amplitude of the 0.07 μm radius mode has a similar seasonal variation to observed CCN concentrations and is attributed to non-sea salt sulphate derived from DMS. The mode at 0.3 μm is attributed to wind produced sea salt (Gras, 1993).

The model runs were initiated at 500 m altitude and 99% relative humidity at monthly mean temperatures derived from the long-term Cape Grim database, which shows that at 100 m the annual temperature range is modest (9.5°C–14.3°C), as would be expected for air marine air masses in which sea surface temperature (which varies only a few degrees seasonally) must be the prime determinant of boundary layer air temperature. Model runs were terminated at a cloud depth of 250 m. As a result of the small seasonal cycle in air temperature the seasonal variation in predicted liquid water content at 250 m above model cloud base was small, ranging from 0.45 gm^{-3} in July to 0.51 gm^{-3} in January. Since updraft speed is not variable in the model, sensitivity to this parameter was investigated by carrying out for each month a series of model runs with updraft speed varied between 0.1 and 0.3 ms^{-1}. Across all model runs (a total of 48: 12 months × 4 updraft speeds) the maximum supersaturation calculated varied between 0.15% (December, at 0.1 ms^{-1},) and 0.35% (July, at 0.3 ms^{-1}). Table 1 lists the predicted droplet number concentrations and effective radii as a function of month and updraft speed. Using the droplet concentration data generated by the cloud model, cloud optical depth may be predicted as a function of month and updraft speed by using the approximation

$$\tau = \alpha A_T^{2/3} N^{1/3} h^{5/3}, \quad (1)$$

where the known parameter α is constant, A_T the vertical lapse rate of adiabatic liquid water con-

tent, which is a function of temperature and therefore known from the Cape Grim records, and h is cloud depth. Here it is assumed that the liquid water content changes linearly from cloud base to cloud top. The optical depth can be calculated by assuming h to be constant and adjustable so that the yearly averaged predicted cloud optical depth matches the yearly averaged observed optical depth. The validity of the assumption that cloud depth is constant will be discussed below.

4. Discussion

4.1. Results

Mean monthly observed values of the optical depth were derived from the time series of optical depth shown in Fig. 1 by subtracting a linear fit from the data, averaging the monthly differences over the seven years, and adding the result to the value of the linear fit at the end of the record to closely match the average optical depth near the one and a half year period for which the size distribution of CCN data was available.

Fig. 3 shows the observed and computed monthly averaged optical depths. The monthly mean computed values plotted are for 0.2 ms^{-1} updraft speed, with the limits shown about each point reflecting the modelled 0.1 and 0.3 ms^{-1} extremes. From the figure it is apparent that within the given uncertainties the phase and amplitude of optical depths predicted from the aerosol concentration are clearly similar to those in the observed optical depth cycle. Some caution is necessary when making this comparison, as only CCN observations from Cape Grim made in "baseline" conditions (air from purely maritime sources) occurring 40% of the time, with wind directions between 190° and 280°, were included in the CCN data input to the cloud model for prediction of the seasonal cycle in optical depth. For non-baseline conditions additional CCN from continental sources will also affect cloud optical depth. The frequency of occurrence of purely maritime con-

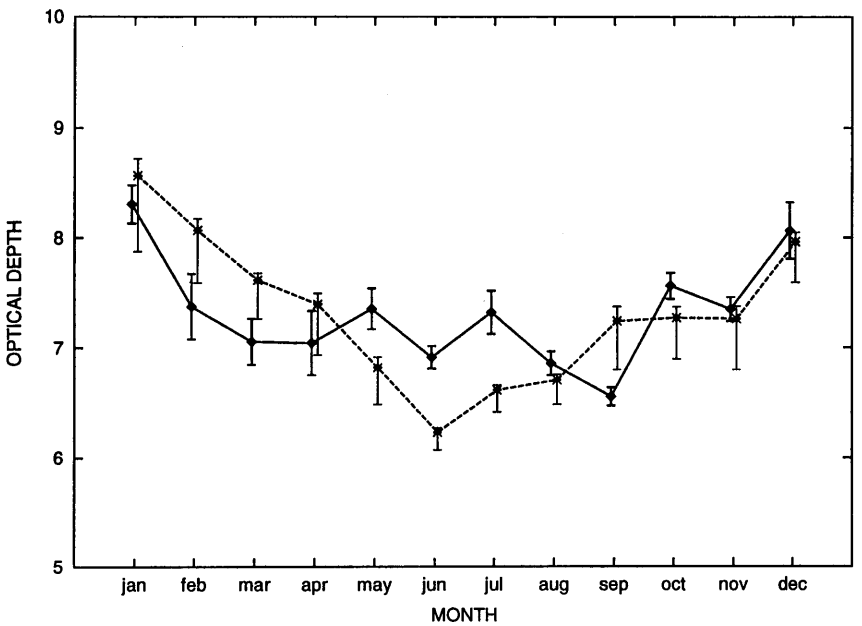

Fig. 3. Seasonal cycle in optical depth (1984-1990). Solid line represents the observations derived from the detrended data set shown in Fig. 1, and averaged over the seven year period. The error bars represent the standard deviation of the mean over the seven years. The broken line represents the optical depth modeled using a 0.2 ms^{-1} updraft speed. The error bars on this line represent the uncertainties in optical depth calculated by varying the updraft speed between 0.1 ms^{-1} and 0.3 ms^{-1}. Modeled droplet number concentration using the 0.2 ms^{-1} updraft speed ranged from 90 cm^{-3} in January to 35 cm^{-3} in July.

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ditions (i.e., unmodified by continental aerosol plumes) over the open ocean at the location of optical depth retrievals upwind of Cape Grim is unknown. However based on the Cape Grim wind direction records and daily trajectory analysis it is estimated to occur 70% of the time.

4.2. *Sensitivity analysis*

There are a number of uncertainties in our results which are due to the optical depth retrieval procedure and model assumptions. We discuss these uncertainties in turn to assess their influence on the results.

4.2.1. Surface reflectivity. The optical depth retrieval is sensitive to variations in surface reflectivity which is a function of the solar angle. Although the geometry of sun-satellite angles is such that sunglint conditions are not expected we can estimate the errors in optical depth due to uncertainty in surface reflectivity by using an analytic expression for the cloud reflection function (King, 1987):

$$R(\tau;\mu,\mu_0,\phi)=R_\infty(\mu,\mu_0,\phi)-\frac{4(1-A_g)\,K(\mu)\,K(\mu_0)}{3(1-A_g)(1-g)(\tau+2q_0)+4A_g}, \quad (2)$$

where μ, μ_0 are the cosines of sun and satellite zenith angles, A_g surface albedo, R_∞ is the reflection function of a semi-infinite atmosphere, g is the asymmetry parameter ($=0.85$), $K(\mu)$ is the escape function and q_0 is the (constant) extrapolation length both of which are further discussed by King (1987). For a given R, i.e., $\partial R=0$, we can calculate the perturbation in τ necessary to cancel perturbations in A_g exactly. We find that

$$\partial\tau=-\frac{4}{3}\frac{A_g}{(1-g)(1-A_g)^2}\frac{\partial A_g}{A_g}. \quad (3)$$

Rossow et al. (1989) claim that $\partial A_g/A_g<0.06$ even under conditions of high turbidity, so that with the estimate $A_g=0.03$ we find that $\partial\tau=0.02$. Hence the error in optical depth due to perturbations in surface reflectivity is negligible.

4.2.2. Cloud scattering properties. The retrieval of optical depth is performed by assuming a droplet size distribution with effective radius constant at 10 μm (Rossow et al., 1989). This assumption is unlikely to hold in stratocumulus clouds resulting in uncertainties in optical depth retrievals. The effective radius close to cloud top is probably slightly higher than assumed in the retrieval process varying seasonally between 10 μm in summer to 15 μm in winter (see Table 1). Seasonal variations in effective radius will correspond to seasonal variations in the scattering phase function necessary to compute the radiance emergent from the cloud which in turn is required to perform an optical depth retrieval. The question is whether these seasonal variations in effective radius might account for the seasonal variations in retrieved optical depth.

It is possible to estimate this error using eq. (2). The analysis involves estimating the perturbations in τ due to perturbation in the escape function K and asymmetry parameter g under the assumption $\partial R=0$. Assume $\partial K(\mu)=\partial K(\mu_0)$. We find that:

$$\partial\tau=\frac{\tau}{1-g}\partial g+\left[2\tau+\frac{4q_0(1-g)}{1-g}+\frac{8}{3}\frac{A_g}{1-A_g}\frac{1}{1-g}\right]\frac{\partial K}{K} \quad (4)$$

The first term on the right-hand side of this equation involves a strong sensitivity of τ to seasonal changes in the asymmetry parameter. Slingo and Schrecker (1982) found that g is a weak

Table 1. *Seasonal variation in modeled droplet number concentration and cloud droplet effective radius as a function of parcel updraft speed at cloud base*

Updraft	0.10	0.15	0.20	0.30	0.10	0.15	0.20	0.30
Jan.	72	83	92	97	11.7	11.0	10.7	10.5
Feb.	64	72	77	80	12.2	11.7	11.5	11.3
Mar.	56	62	64	66	12.7	12.3	12.1	12.0
Apr.	49	55	59	61	13.1	12.6	12.3	12.2
May	40	45	46	48	13.9	13.4	13.2	13.1
Jun.	33	34	35	36	14.4	14.2	14.1	14.1
Jul.	39	41	42	43	13.5	13.3	13.2	13.1
Aug.	40	42	44	45	13.4	13.2	13.1	13.0
Sep.	46	52	55	59	13.1	12.7	12.4	12.1
Oct.	48	53	56	58	13.0	12.6	12.3	12.2
Nov.	46	52	56	59	13.3	12.8	12.5	12.3
Dec.	64	71	74	76	12.1	11.6	11.5	11.3

The 4 left columns represent modeled droplet number concentration (in cm^{-3}); the 4 right columns represent cloud droplet effective radius (in μm).

increasing function of the effective radius of a size distribution. Hence, it is expected that for winter time conditions g will be larger than for summer conditions. Eq. (4) shows that this could introduce an apparent signal in retrieved optical depth which is in phase with the seasonal variation in CCN and which is entirely due to changes in cloud droplet effective radius but not due to actual changes in the optical depth. We calculated the asymmetry parameter for a range of droplet size distribution with effective radius ranging from 4 to 30 μm for visible wavelengths. Given the calculated variation in r_{eff} from summer to winter from 10 μm to 15 μm (see Table 1), we found $\partial g = 0.007$. Hence the apparent signal in τ in phase with the observed variation in CCN is no more than 0.37.

For the second term in eq. (4) we assume $\partial K/K = 0.01$ based on King (1983, 1987) who found little variation of K for various size distributions. Using $(1-g)\,q_0 = 0.712$ (King, 1987) we find $\partial\tau = 0.35$ due to K only.

We therefore conclude that the optical depth seasonal signal evident in the data set is larger than the uncertainty in the retrieved optical depth due to uncertainty in assigning a proper phase function. However, a small part of the signal (0.37) could in principle be due to improper specification of the asymmetry parameter in the retrieval procedure.

4.2.3. Glaciation. Cloud top temperatures were derived from the ISCCP-C2 data set for the same region for which the optical depth record was shown earlier. They do not drop below the level of about -5°C needed for widespread glaciation. Therefore, a seasonal cycle in optical depth resulting from change in phase of the cloud elements (i.e., glaciation in winter) can be ruled out.

4.2.4. Cloud depth. Our calculations have been performed assuming that the cloud depth is constant throughout the seasons. Since optical depth is sensitive to variations in h, the question arises whether a seasonal variation in cloud depth could be responsible for the seasonal variation in optical depth. Little or no data exist on seasonal variability in marine stratocumulus cloud depth. Nevertheless, it is possible to postulate on theoretical grounds that seasonal variations in cloud depth if present are probably 180° out of phase with those necessary to explain our results.

We used a "jump" model of the marine uniformly mixed stratocumulus covered boundary layer (Nicholls and Turton, 1986) to simulate the seasonal variation in cloud depth. Our results show that clouds are deeper in winter than in summer for two reasons. (1) Solar absorption (which is less in winter than in summer) partly offsets cloud top cooling responsible for cloud top entrainment so that cloud top is higher in winter than in summer; (2) Cloud and subcloud layer are colder in winter than in summer, so that cloud base is lower in winter than in summer. Both effects result in a deeper winter cloud. Our findings are in agreement with Turton and Nicholls (1987) who used an extension of the Nicholls and Turton model to allow for daytime decoupling of the cloud layer from the surface.

Our conclusion is therefore that a seasonal cycle in cloud depth is likely but of opposite phase to the cycle in optical depth. The occurrence of 2 12-month cycles of opposite phase but of unequal strength could appear as a six-month harmonic in spectral analysis. Therefore, we suggest that it is probably the cause of the six-month harmonic (slight winter maximum) evident in the satellite optical depths that was not reproduced by our cloud model. Clearly, more research is necessary to investigate the seasonal variation in cloud depth.

4.2.5. Calibration drift. The trend towards lower optical depths as evident from the time series in Fig. 1 corresponds to a 4% reduction in cloud top reflectance over the seven year period. 35% of this reduction can be explained by the reduction of 20% in CCN concentration observed at Cape Grim in the same time period (Gras, 1993), resulting in a trend towards lower modelled droplet number concentration. The remainder of the observed trend towards lower optical depth is probably caused by calibration drift of the radiometer (Klein and Hartmann, 1993).

4.2.6. Updraft speed. The number of cloud droplets generated on CCN is sensitive to the updraft speed assumed at cloud base. The higher the updraft speed, the more cloud droplets are formed. However, the number of CCN that can be activated is limited. Table 1 indicates that for the winter months there is little difference between the calculated droplet number concentrations for updraft speeds ranging from 0.15 to 0.30 ms^{-1}; hence there will be little differences between calculated optical depth. Fig. 3 shows error bars on the calculated optical depth which corresponds to a variation in updraft speed between 0.1 ms^{-1}

and 0.3 ms^{-1}. The asymmetry in upper and lower bound in optical depth reflects the non-linearities inherent in the relationship between particle size, activation supersaturation and ambient size distribution.

5. Conclusion

An inspection of ISCCP-C2 data set records at other Southern Ocean locations indicates that seasonal cycles in stratus optical depth are ubiquitous. For many locations, but not all, seasonal peaks are in phase with the annual cycle in DMS. Phase shifts of the peaks in optical depth to other months were found for locations dominated by seasonal winds of continental origin, and for high latitude locations where seasonal shifts in exposure to east-west cyclone passage significantly affects cloud depth. Only in the absence of continental and synoptic scale influences can the hypothesised link between DMS, CCN and maritime cloud optical depth signals be expected to be in phase.

Despite the need to be cautious, having considered the various possible complications we believe that the essential feature of our findings remains: the observed seasonal cycle in optical depth over the Southern Ocean near Cape Grim appears to be explicable largely by variations in cloud droplet concentration, induced by observed changes in CCN concentration, although the prominence of the six month harmonic wave in the optical thickness record and the results of our "jump" model calculation imply that other factors are influential as well.

Because of the sensitivity of cloud optical depth on cloud depth future work should include an investigation of the seasonal influences on cloud depth of factors such as precipitation, large scale divergence and upper atmospheric stability.

6. Acknowledgements

We thank Dr. D. O'Brien for his review and discussions on the accuracy of the optical depth calculations, Dr. A. Prata for his review and discussion on spectral analysis, and Dr. P. Turner for assistance in accessing the ISCCP data set.

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