

## Cloud Condensation Nuclei and Ship Tracks

JAMES G. HUDSON

*Division of Atmospheric Sciences, Desert Research Institute, University and Community College System of Nevada,  
Reno, Nevada*

TIMOTHY J. GARRETT, PETER V. HOBBS, AND SCOTT R. STRADER

*Department of Atmospheric Sciences, University of Washington, Seattle, Washington*

YONGHONG XIE AND SEONG SOO YUM

*Division of Atmospheric Sciences, Desert Research Institute, University and Community College System of Nevada,  
Reno, Nevada*

(Manuscript received 17 September 1996, in final form 3 December 1997)

### ABSTRACT

Enhancements of droplet concentrations in clouds affected by four ships were fairly accurately predicted from ship emission factors and plume and background cloud condensation nucleus (CCN) spectra. Ship exhausts thus accounted for the increased droplet concentrations in these "ship tracks." Derived supersaturations were typical of marine stratus clouds, although there was evidence of some lowering of supersaturations in some ship tracks closer to the ships where CCN and droplet concentrations were very high.

Systematic differences were measured in the emission rates of CCN for different engines and fuels. Diesel engines burning low-grade marine fuel oil produced order of magnitude higher CCN emissions than turbine engines burning higher-grade fuel. Consequently, diesel ships burning low-grade fuel were responsible for nearly all of the observed ship track clouds. There is some evidence that fuel type is a better predictor of ship track potential than engine type.

### 1. Introduction

As part of the endeavor to determine the immediate cause of ship tracks (Durkee et al. 2000) it is important to determine whether, and which of, the particles emitted by ships are responsible for the formation of the enhanced cloud droplet concentrations that constitute ship tracks. The water solubility of a particle, as determined by cloud condensation nucleus (CCN) measurements, is key to this determination because this characterizes particles according to the likelihood of cloud droplet nucleation (e.g., Twomey and Squires 1959). Many studies have demonstrated the connection between CCN spectra and cloud droplet spectra (e.g., Justo 1966; Radke and Hobbs 1969; Warner 1969). In general, those particles that have lower critical supersaturations ( $S_c$ ) are more likely to produce cloud droplets (e.g., Hudson 1984); there is also a tendency for larger cloud droplets to form on larger (lower  $S_c$ ) CCN (Hudson and Rogers 1986;

Twohy and Hudson 1995). Concentrations of CCN and cloud droplets can be compared if it is assumed that the lower  $S_c$  nuclei will preferentially produce cloud droplets. Thus, an integration of the cumulative CCN spectrum up to the  $S_c$  that results in a match of CCN number concentration with cloud droplet number concentration ( $N_d$ ) reveals the effective supersaturation ( $S_{\text{eff}}$ ) in a cloud. Comparisons between CCN and cloud droplets amount to a comparison of cloud formation in the controlled environment of a cloud chamber with cloud formation in the real atmosphere.

Here we explore the relationships between the CCN emitted by ships and  $N_d$ . We use the measurements of particle source strengths from ships (Hobbs et al. 2000) and a Gaussian plume diffusion model to predict enhancements of  $N_d$  due to the CCN emitted by ships. These predictions are made and compared with measurements for a series of cloud penetrations at various distances from four ships. We explore reasons for discrepancies between the predictions and measurements. We then characterize various ship engines and fuels according to their relative CCN production; this helps predict the likelihood of a ship to produce a ship track in

---

*Corresponding author address:* Dr. James G. Hudson, DRI-DAS,  
2215 Raggio Pkwy., Reno, NV 89512-1095.  
E-mail: hudson@dri.edu

marine stratiform clouds under prescribed ambient conditions.

## 2. Experimental

The Desert Research Institute (DRI) CCN spectrometer (Hudson 1989) was on the University of Washington (UW) C-131A aircraft during the Monterey Area Ship Track (MAST) experiment (Hobbs et al. 2000). It produced a nearly continuous record of the CCN spectrum between 0.02% and 1% supersaturation with about 20 channels of resolution. This detailed CCN spectrum was recorded at a rate of 1–0.1 Hz throughout most of the C-131 MAST flights. These measurements were complemented by measurements of the total number of particles by a TSI 3760, which has a lower size detection limit of about 0.02- $\mu\text{m}$  diameter (Hobbs et al. 2000). Cloud droplet concentrations ( $N_d$ ) were measured using a PMS Forward Scattering Spectrometer Probe (FSSP-100) aboard the C-131 (Hobbs et al. 2000).

Most of the out-of-cloud measurements of the ship plumes were made so close to the ships that the particle concentrations were very high. Such measurements were not ideal for the CCN–droplet comparisons for two reasons. 1) The out-of-cloud plume measurements and many of the cloud measurements were spatially separated; several processes (i.e., coagulation and gas-to-particle conversion) could change CCN spectra between these locations over time periods as long as 2 h (see Figs. 2–4). 2) The high particle concentrations (tens of thousands  $\text{cm}^{-3}$ ) close to the ships often challenged CCN measurement accuracy. The main cause of inaccuracies for high-concentration CCN measurements is the depletion of water vapor within the cloud chamber, which lowers the supersaturation. This difficulty was minimized by using small sample flow rates, by calibrating the instrument with high particle concentrations, and by relying on plume measurements with lower CCN concentrations.

## 3. Analysis methods

The primary goal is to compare measured enhancements of cloud droplet concentrations ( $\Delta N_d$ ) in ship tracks with predictions of  $\Delta N_d$  that are based on CCN measurements in plumes close to the ships that caused the ship tracks. Observed  $\Delta N_d$  were determined from the difference in the average  $N_d$  measured within the ship tracks (perturbed clouds) compared with the average  $N_d$  measured in adjacent background clouds. These  $\Delta N_d$  values were determined from several horizontal flight passes that were generally done perpendicular to the length of the ship tracks. Several of these passes were made at various distances from the ship. In general,  $\Delta N_d$  decreased with distance from a ship because of the horizontal spreading of the plume with distance from its source. Hobbs et al. (2000) used this method to compare predictions of condensation nucleus

(CN) concentrations with the sum of measurements of interstitial CN and cloud droplets. To apply this method of analysis the source strength of the CN was first determined for each ship. Then the plume was assumed to spread out and to dilute linearly with the background air; the height of the boundary layer was assumed to remain constant. Thus, the plume was assumed to spread or dilute only in the horizontal dimension perpendicular to the length of the plume. This width of the plume was determined from CN measurements, and from the assumption of a Gaussian shape. We found, nevertheless, that the assumed shape of the plume is much less important than the width as long as it is determined consistently. Wider plumes at greater distances from the ships, were more diluted with background aerosol. Hobbs et al. (2000) verified this procedure by finding good agreement between predictions of total aerosol concentrations—based only on the dilution of the CN concentrations—and measurements of  $\Delta N_d$  plus interstitial CN concentrations. These comparisons showed a general decrease in  $\Delta N_d$  and interstitial CN with distance from a ship.

Here we employ the same Gaussian diffusion model, along with some other factors, to make predictions of  $\Delta N_d$  alone. We refer to these predictions of  $\Delta N_d$  as  $\Delta N_{\text{CCN}}$ . Two factors in addition to those used by Hobbs et al. (2000) are needed to make these predictions: 1) CCN spectra that can be attributed to production by the ships ( $\text{CCN}_p$ ) and 2) the effective supersaturation ( $S_{\text{eff}}$ ) of the clouds. We also need to assume 1) that  $S_{\text{eff}}$  is the same for all clouds in each case study—background and ship tracks—which also implies homogeneity of background CCN and  $N_d$ , and 2) that the ratio of plume CCN concentrations at  $S_{\text{eff}}$  to the plume CN concentrations is constant throughout each case study. These assumptions will later be tested and discussed. These comparisons of excess cloud droplet concentrations from measurements ( $\Delta N_d$ ) and predictions ( $\Delta N_{\text{CCN}}$ ) constitute what has come to be known as “closure” experiments.

The prediction of  $\Delta N_{\text{CCN}}$  began with a determination of  $S_{\text{eff}}$  for each case study. Next, the ratio of the concentration of CCN at  $S_{\text{eff}}$  to the CN concentration was determined for out-of-cloud plumes close to the ships. Finally, the same Gaussian plume model prediction of CN in each ship track, used by Hobbs et al. (2000), was applied, with the multiplication by the CCN/CN ratio. This predicts  $\Delta N_{\text{CCN}}$  for each ship track. Hobbs et al. (2000) used above-background CN concentrations in a ship plume and a Gaussian plume model to predict the increase, above background, of the sum of interstitial aerosol and  $N_d$  downwind in a ship track. These predictions were based on the source strength of the ship, which was determined from CN and  $\text{CO}_2$  measurements near the ships (Hobbs et al. 2000). The apportionment of excess CN between interstitial aerosol and cloud droplets in the ship tracks was not addressed by Hobbs et al. (2000).

The background CCN measurements ( $\text{CCN}_b$ ) at each

TABLE 1. CCN/CN concentrations ( $\pm$  standard deviations) grouped according to ship engine and fuel types.

Date (1994) and ship name	Number of plumes measured	CCN/CN at 0.2% $S_c$	CCN/CN at 1.0% $S_c$
1 Jun, <i>Monterrey</i>	3	0.022 $\pm$ 0.004	0.151 $\pm$ 0.042
*8 Jun, <i>Fremo Scorpilus</i>	3	0.016 $\pm$ 0.004	0.098 $\pm$ 0.009
*27 Jun, <i>Taihe</i>	4	0.037 $\pm$ 0.013	0.156 $\pm$ 0.030
*29 Jun, <i>Star Livorno</i>	3	0.138 $\pm$ 0.008	0.231 $\pm$ 0.039
29 Jun, <i>Hanjin Barcelona</i>	2	0.072 $\pm$ 0.036	0.174 $\pm$ 0.079
*30 Jun, NYK <i>Sunrise</i>	2	0.061 $\pm$ 0.006	0.187 $\pm$ 0.009
All of above ships (namely, diesels using marine fuel oil)	17	0.058 $\pm$ 0.041	0.166 $\pm$ 0.040
21 Jun, <i>Kansas City</i>	2	0.006 $\pm$ 0.005	0.023 $\pm$ 0.008
27 Jun, <i>Mt. Vernon</i>	3	0.012 $\pm$ 0.009	0.104 $\pm$ 0.086
Above two ships (namely, steam turbines using navy distillate fuel)	5	0.009 $\pm$ 0.003	0.064 $\pm$ 0.040
9 Jun, <i>Copeland</i> (gas turbine using navy distillate fuel)	7	0.004 $\pm$ 0.005	0.022 $\pm$ 0.020
12 Jun, <i>Safeguard</i> (diesel using navy distillate fuel)	1	0.0011	0.018
1, 9, 12 Jun, unknown ships	7	0.022 $\pm$ 0.019	0.113 $\pm$ 0.070

\*Used for CCN–cloud droplet comparisons in ship tracks.

$S_c$  were subtracted from the corresponding CCN measurements within the ship plume close to the ships so that the contribution to the CCN of the ship effluents alone could be determined ( $CCN_p$ ). This subtraction was necessary because any plume that was wide enough to be measured with the aircraft was always significantly diluted by background air. Measurements of the undiluted plume, which only exists within the smokestack, were not possible. CCN measurements within clouds could not be used because of possible droplet splashing (which often causes spurious counts; i.e., Hudson and Frisbie 1991) and the difficulty of sampling CCN within cloud droplets.

The value of  $S_{eff}$  was determined by matching  $N_d$  of background clouds to the  $S_c$  that yielded the same cumulative concentration for  $CCN_B$ . Five to 10 measurements, each of 1–2-min duration, of the value of  $CCN_B$  out of cloud within the boundary layer were obtained during each of the four flights analyzed here (Table 1). We had to assume that the CCN concentrations within the boundary layer outside of the ship plumes were homogeneous for each case study. Examples of these spectra, each of which consisted of several records of a few seconds, are shown in Fig. 1. Figures 1a and 1b illustrate typical polluted and clean background concentrations encountered during MAST.  $CCN_B$  was subtracted from the CCN concentrations measured in the plume to determine the contribution of the CCN from the ships alone ( $CCN_p$ ), which are shown in Figs. 1c and 1d.

The high particle concentrations in some of the plumes closest to the ships could cause erroneously low measurements of CCN/CN ratios because high concentrations of growing droplets can lower the supersaturation in diffusion cloud chambers (Hudson and Squires 1973). This could result in some droplets not growing

large enough to be put into the correct droplet size (nucleus  $S_c$ ) channels (Hudson 1989). However, laboratory calibrations with known aerosols at the concentration ranges encountered in the ship plumes demonstrated that this should have been a problem only for the very highest CCN concentrations at the lowest  $S_c$  values. Channel shifting was minimized by using concentrations for the calibrations that were similar to those encountered in the plumes. Fortunately, there was also more than one plume penetration for each of the four cases analyzed here (Table 1). For the sake of accuracy and consistency, the lower particle concentrations measured in each case study were used in the analysis. In some cases, there appeared to be a trend of higher CCN/CN ratios for the lower concentration plume measurements. Nonetheless, the consistency in CCN/CN ratios, as expressed by the standard deviations in Table 1 (columns 3 and 4), was so good that this problem seemed to be minimal anyway.

#### 4. Results

The third column of Table 2 lists  $N_d$  in background clouds adjacent to the various ship tracks. The variabilities (minimum, mean, maximum) displayed here are a combination of the standard deviations of the mean  $N_d$  measured in the different background clouds, the average standard deviations of the 1-s data intervals of the different cloud penetrations, and the range in the mean values of  $N_d$  for the various cloud penetrations on each flight. An amalgamation of these three different ways of expressing the variations in  $N_d$  was used to estimate representative variations in  $S_{eff}$  (minimum is mean minus variation, and maximum is mean plus variation); these  $S_{eff}$  values are shown in the fourth column of Table 2. Here,  $S_{eff}$  was determined by matching each  $N_d$  in column 3 with  $CCN_B$  (e.g., Hudson 1984). The

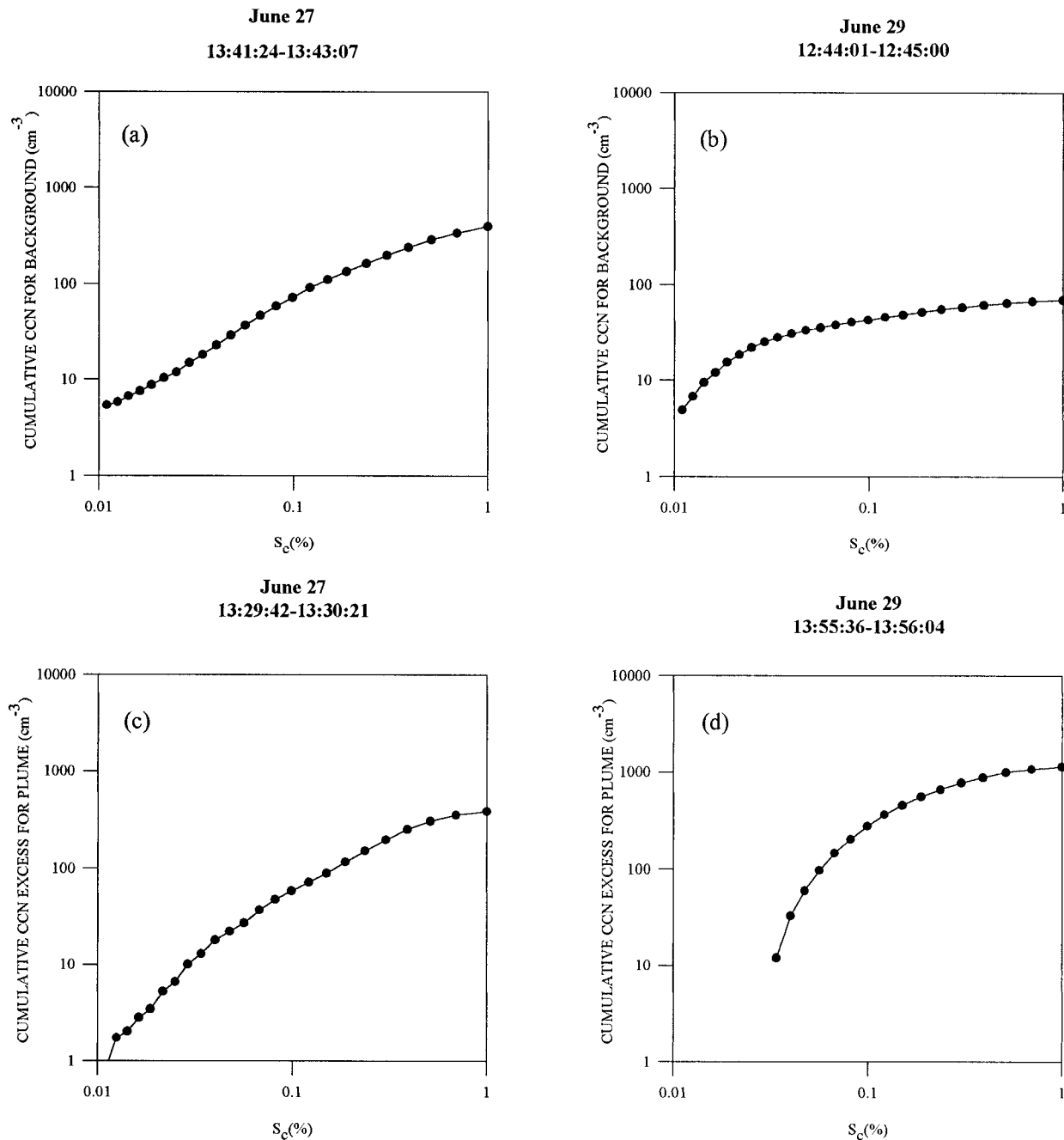


FIG. 1. Examples of cumulative CCN spectra. (a) and (b) are for background air ( $\text{CCN}_B$ ) for dirty and clean, respectively; (c) and (d) are for ship exhausts ( $\text{CCN}_P$ ) with the background contribution ( $\text{CCN}_B$ ) removed.

fifth column shows the CCN concentrations at  $S_{\text{eff}}$  in the plume of the ship ( $\text{CCN}_P$ ). This is the difference between the measured concentration at  $S_{\text{eff}}$  in the exhaust plume and the measured concentration at  $S_{\text{eff}}$  for  $\text{CCN}_B$  (e.g., Figs. 1a,b).

The sixth column in Table 2 shows the CN concentration for that same ship plume; this also has the background CN concentration subtracted from the CN concentration measured in the plume. The seventh column

shows the ratio of concentrations of CCN at  $S_{\text{eff}}$  to CN in the plume [ $\text{CCN}(S_{\text{eff}})/\text{CN}$ ]. When this is multiplied by the emission factor given in Eq. (3) of Hobbs et al. (2000), a prediction of the increase in cloud droplet concentration due to the ship exhaust ( $\Delta N_{\text{CCN}}$ ) is obtained. Figure 2 shows the ratio of this prediction ( $\Delta N_{\text{CCN}}$ ) to the measured enhancement in cloud droplet concentration ( $\Delta N_d$ ) in the ship tracks (solid points). The eighth column in Table 2 shows the averages of the

TABLE 2. Characterization of ship tracks. Here,  $N_d$  is ambient cloud droplet concentration,  $S_{eff}$  the effective cloud supersaturation,  $CCN_p$  the CCN concentration from the ship,  $CCN_b$  the total particle concentration from the ship exhaust only,  $\Delta N_d$  the measured excess droplet concentration in the ship track,  $\Delta N_{CCN}$  the predicted CCN concentration from the Gaussian plume model (column 8) and from the cloud droplet growth model of Robinson (1984)(column 11), and  $W$  the updraft velocity.

1	2	3	4	5	6	7	8	9	10	11
Date and ship name	$N_d$ (cm <sup>-3</sup> )	$S_{eff}$ (%)	$CCN_p$ (cm <sup>-3</sup> )	$CCN_p$ (cm <sup>-3</sup> ) at $S_{eff}$	$CCN_p$ (cm <sup>-3</sup> )	$CCN_p$ (at $S_{eff}$ )/ $CCN_p$	$(\Delta N_{CCN}/\Delta N_d)_{GPM}$ (Gaussian plume model)	$W$ (cm s <sup>-1</sup> )	$S_{eff}$ (%)	$(\Delta N_{CCN}/\Delta N_d)_{DGM}$ (cloud droplet growth model)
8 Jun, <i>Fremo Scorpis</i>	Min	0.03	3	3	1419	0.002	0.034	5.8	0.19	0.034
	Mean	0.18	28	28		0.02	0.34	16.9		0.33
	Max	0.42	99	99		0.07	1.17	38.6		1.15
27 Jun, <i>Taitie</i>	Min	0.19	118	118	2232	0.053	1.18	14.9	0.24	1.28
	Mean	0.21	135	135		0.061	1.36	18		
	Max	0.24	155	155		0.069	1.55	21.8		
29 Jun, <i>Star Livorno</i>	Min	0.07	153	153	4001	0.038	0.32	9.7	0.19	1.04
	Mean	0.19	582	582		0.142	1.24	31.1		
	Max	0.63	1075	1075		0.27	2.30	51.5		
30 Jun, <i>NYK Sunrise</i>	Min	0.90	1325	1325	7548	0.175	1.52	100	0.69	1.38
	Mean	1.0								
	Max									

ratios of the predicted to the observed excess droplet concentrations displayed in Fig. 2 (this is the average of all of the ship tracks in each case study). Column 8 of Table 3 completes the analysis using the Gaussian plume diffusion model.

We now investigate the assumption of constant  $S_{eff}$ , in particular by testing the assumption that  $S_{eff}$  is the same in the background clouds as in the ship tracks. This was done by employing the condensational cloud droplet growth computer model of Robinson (1984). This model uses as input the CCN concentrations in several  $S_c$  bins (~15 over the entire CN spectrum from 0.01% to 1%), the updraft velocity ( $W$ ), pressure, and temperature to predict cloud droplet spectra as a function of time (and thus distance above cloud base). The model was run until a distinct dichotomy was found between the unactivated haze droplets and the activated cloud droplets ( $N_d$ ) (i.e., Hudson and Svensson 1995; Hudson and Yum 1997). This model is adiabatic; it does not consider any entrainment of outside air into the cloud, nor losses of droplets due to coalescence or precipitation.

The first step was to run this droplet growth model iteratively on  $CCN_b$  with various constant  $W$  until a match was found between the  $N_d$  from this model and the measured  $N_d$  in column 3 of Table 2. The  $W$  values that produced these  $N_d$  matches are shown in the ninth column of Table 2. The finite resolution of the CCN spectrometer limited the number of  $S_c$  bins so that exact matches of predicted  $N_d$  with measured  $N_d$  in column 3 of Table 2 were not always possible.

The model was then run with the same  $W$  but with the linear combination of  $CCN_p$  and  $CCN_b$  that was the input to each ship track according to the width of the ship's plume at that location. This spectrum was obtained by diluting  $CCN_p$  according to the width of the plume for each ship track and adding  $CCN_b$ . When the model was run with these appropriate input CCN spectra, predictions of  $N_d$  were determined for each ship track cloud. When the background  $N_d$  was subtracted from this predicted  $N_d$ , a second prediction,  $\Delta N_{CCN}$ , was obtained; the ratios of this  $\Delta N_{CCN}$  to the measured  $\Delta N_d$  are plotted as the open circles in Fig. 2. In most cases, these ratios were identical to those derived using the Gaussian diffusion plume (solid points). In those cases where these ratios differed, it was because the high concentrations in the less diluted ship tracks reduced  $S_{eff}$ . Figure 3 shows how this  $S_{eff}$  compares with  $S_{eff}$  in the background clouds. The  $S_{eff}$  for the background clouds is also displayed in column 10 of Table 2. Figure 3 shows that  $S_{eff}$  is lower when the cloud condensation droplet growth model is used for some of the less diluted tracks. The 11th column of Table 2 shows the average agreement between predictions of  $\Delta N_{CCN}$  using the droplet growth model and the measured values of  $\Delta N_d$ . These values were usually lower than the ratios in column 8 of Table 2.

With the exception of the underprediction for the *Fre-*



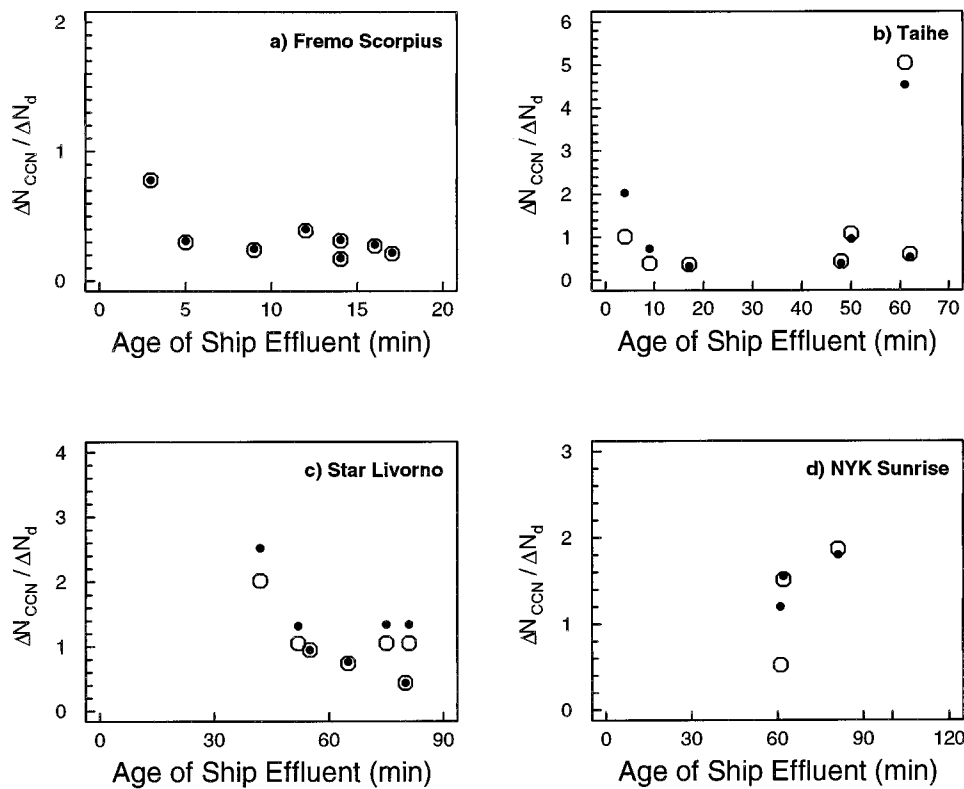


FIG. 2. Ratio of predicted excess droplet concentrations ( $\Delta N_{\text{CCN}}$ ) in ship tracks to measured excess droplet concentrations ( $\Delta N_d$ ). Solid points are for the Gaussian plume model (dilution only). Open circles are for the adiabatic cloud model (Robinson 1984), which allows for lowering of  $S_{\text{eff}}$ .

*mo Scorpious* on 8 June (mean ratio of prediction to measurements of 0.34 or 0.33), the mean predictions of  $\Delta N_{\text{CCN}}$  are higher than the measured values of  $\Delta N_d$ . It should be noted that the CN concentration was also considerably underpredicted for 8 June—0.6 average ratio of predictions to measurements (Hobbs et al. 2000). The CCN/CN ratio for the *Fremo Scorpious* was the lowest of the diesel-powered ships burning marine fuel oil, as shown both by Table 1 and by Hobbs et al. (2000). Moreover, the mean size of the dried particles from this ship was smaller than some other diesel ships burning low-grade fuel (Hobbs et al. 2000). The overall average agreement between these four predictions ( $\Delta N_{\text{CCN}}$ ) and measurements ( $\Delta N_d$ ) of excess droplet concentrations was 1.1 for the Gaussian diffusion plume

model and 1.0 for the cloud droplet condensation growth model. The latter model accounts for any lowering of  $S_{\text{eff}}$  in the ship tracks. In view of the uncertainties pointed out by Hobbs et al. (2000), the results, though perhaps fortuitous, support the view that the enhancements in droplet concentrations in ship tracks ( $\Delta N_d$ ) are due to CCN emitted by ships.

Figure 4 shows how the measured  $\Delta N_d$  decreased with the age of the plume, and Fig. 5 shows the same for  $\Delta N_{\text{CCN}}$  (predicted from Gaussian plume model) with age. Table 3 quantifies the slopes of the linear regressions of these relationships showing that the reductions with distance are greater for the Gaussian diffusion model (column 3) than for the measurements (column 2). The last column of Table 3 shows that the condensational

TABLE 3. Slopes of linear regressions of measured excess droplet concentrations ( $\Delta N_d$ ) and predicted excess droplet concentrations ( $\Delta N_{\text{CCN}}$ ) vs age of ship track.

Date and ship name	Measured (see Fig. 4)	Predicted from Gaussian diffusional plume model (see Fig. 5)	Predicted from cloud droplet condensation growth model (see Fig. 2)
8 Jun, <i>Fremo Scorpious</i>	-1.58	-1.82	-1.84
27 Jun, <i>Taihe</i>	-2.12	-2.76	-1.00
29 Jun, <i>Star Livorno</i>	-0.079	-2.58	-2.10
30 Jun, <i>NYK Sunrise</i>	-1.57	-1.30	-0.99
Average	-1.34	-2.11	-1.48

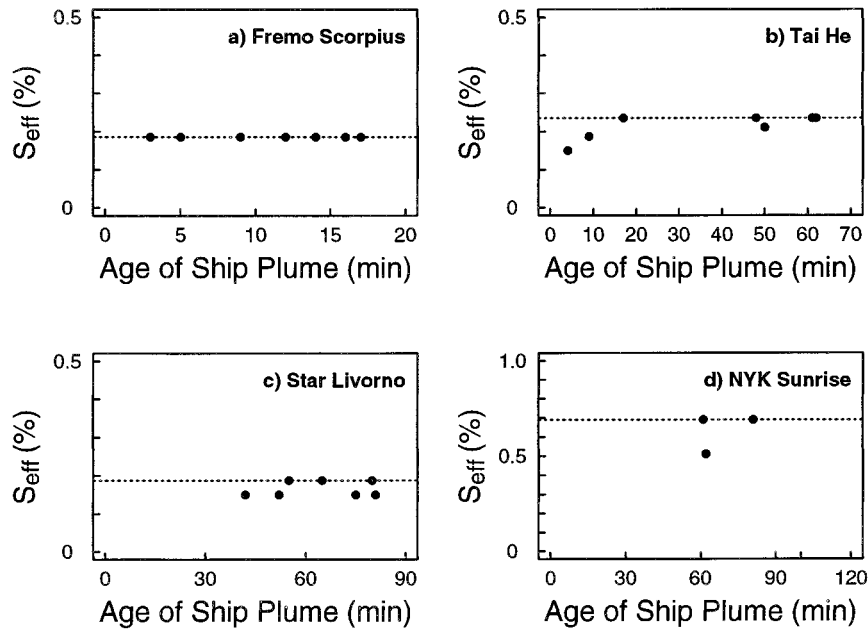


FIG. 3. Effective supersaturation ( $S_{eff}$ ) in ship tracks vs plume age using the adiabatic cloud model. The dashed line is  $S_{eff}$  for the background clouds.

cloud droplet growth model predicts a lower slope of  $\Delta N_{CCN}$  versus age, which is more in line with the observations (column 2 of Table 3). The lower slopes shown in column 4 than in column 3 of Table 3 are a direct result of the lowering of  $S_{eff}$ , and thus of a relative lowering of  $\Delta N_{CCN}$  values in the less diluted ship tracks; this decreases the slope of  $\Delta N_{CCN}$  versus plume/track age.

Table 4 illustrates these points more clearly where, instead of plume age, a more direct indicator of plume/track dilution, namely, the width of the ship track, is used for the independent variable. Here three of the four cases produce a very close match of the predicted to the observed slope. These results suggest that one of the explanations given by Ferek et al. (1998) for the apparently greater than expected persistence of excess

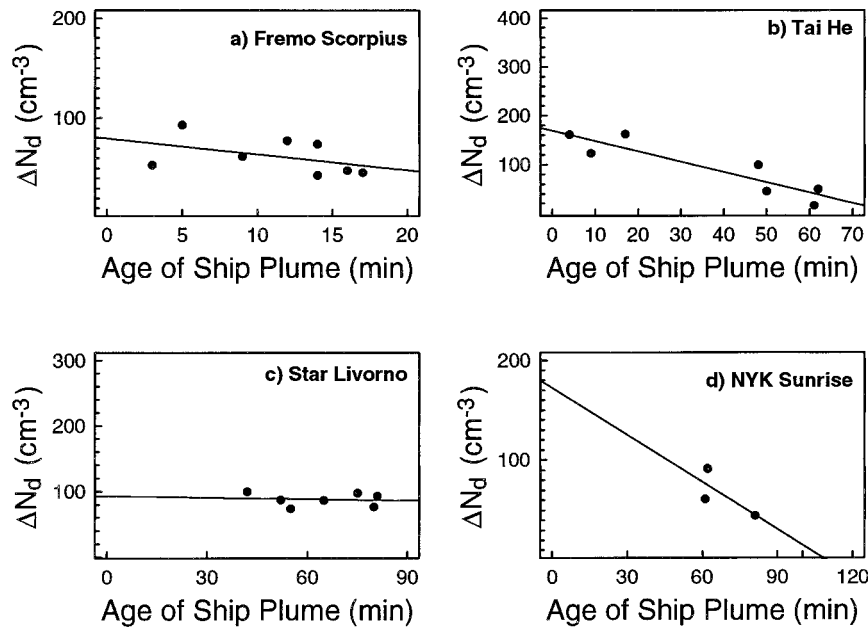


FIG. 4. Measured excess cloud droplet concentrations ( $\Delta N_d$ ) in the ship tracks vs age of ship plume.

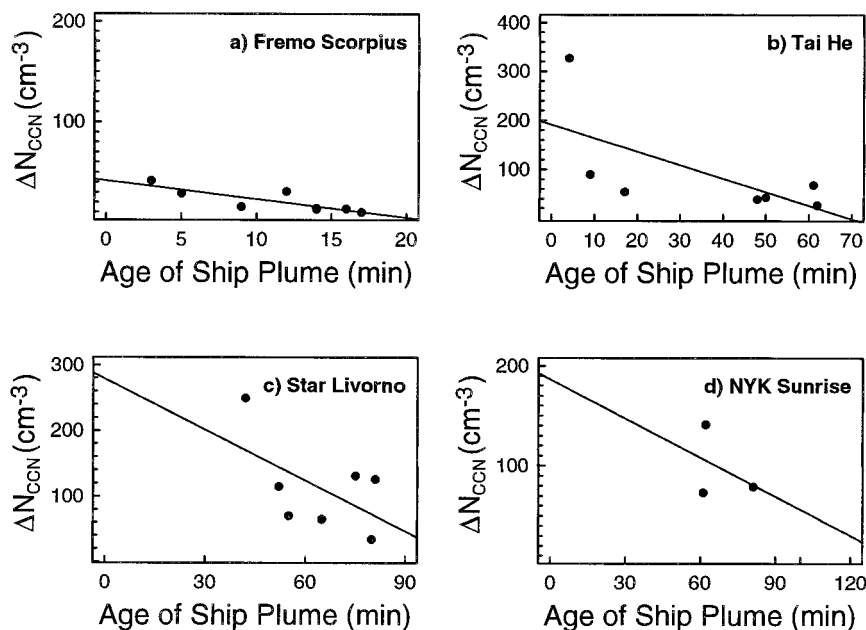


FIG. 5. Predicted excess cloud droplet concentrations ( $\Delta N_{\text{CCN}}$ ) vs age of ship plume using the Gaussian plume model, which does not include any lowering of  $S_{\text{eff}}$ .

droplet concentrations in ship tracks, namely, the increase in  $S_{\text{eff}}$  downwind of the ships, may be correct. The  $S_{\text{eff}}$  is predicted to be higher for lower values of CCN and  $\Delta N_{\text{CCN}}$ , which are produced at greater distances from ships due to the diffusion of the ship effluents. The fact that even the more detailed droplet growth model still predicts a greater slope than observed suggests that there may still be some enhancement, with time, of CCN either by chemical or physical processes (e.g., coagulation), which increase the CCN concentrations downwind (in opposition to the effect of dilution). This latter effect was also discussed by Ferek et al. (1998).

## 5. Particle volatility

To obtain some information on particle composition, volatility measurements were made of the CCN and CN in the ambient air and in the ship plumes using the method described by Hudson and Da (1996). All of the volatility measurements described here were made out of cloud. A composite of the volatility measurements of the background aerosol is presented in Fig. 6 and, for 1 June only, in Fig. 7. In agreement with the results of Hudson and Da (1996) the present results show that most of the background CCN had a fractionation temperature characteristic of ammonium sulfate or ammonium bisulfate ( $\sim 200^\circ\text{C}$ ). Figure 8 shows that the volatility measurements within the plumes of diesel ships using marine fuel oil were nearly identical to those in the background air. Although most of the latter measurements were made on the plume from one ship, the *Monterrey* (Table 1), there were partial (at a limited

number of temperatures) measurements from other ships that were consistent with these results. Due to the lack of knowledge of volatility temperatures for organic compounds, we cannot test the suggestions of Hobbs et al. (2000) that these substances contribute to the CCN in ship plumes. All we can say is that these volatility measurements suggest that ammonium sulfate (or bisulfate) could be the predominant component of the aerosol in both the background air and in the ship effluents.

## 6. Discussion and conclusions

The results presented in this paper show that the excess droplet concentrations in ship tracks can be fairly accurately predicted with a simple model of the diffusion of the effluents emitted by a ship's engine. More accurate predictions are made with a cloud droplet condensational growth model, which allows for lower  $S_{\text{eff}}$  in the ship tracks when CCN concentrations are high.

The cloud droplet condensational growth model used in this paper requires as input the condensation coefficient for water. This is the fraction of molecules that are on a path toward the droplet that actually stick to the droplet. The value of the condensation coefficient is uncertain (Mozurkewich 1986). We used a value of 0.036, which has often been used in cloud physics (e.g., Pruppacher and Klett 1980; Rogers and Yau 1989), based on Chodes et al. (1974). When a condensation coefficient of 1.0, as suggested by Leitch et al. (1986), was used for one of the flights, only one value of  $S_{\text{eff}}$  was further decreased. A higher condensation coefficient (e.g., 1.0) would cause the droplets to grow faster



TABLE 4. Slopes of linear regressions of excess droplet concentrations in the ship tracks vs width of the ship tracks.

Date and ship name	Measured	Predicted from Gaussian plume model	Predicted from cloud condensation growth model
8 Jun, <i>Fremo Scorpis</i>	-0.021	-0.022	-0.022
27 Jun, <i>Taihe</i>	-0.017	-0.046	-0.020
29 Jun, <i>Star Livorno</i>	-0.0056	-0.041	-0.030
30 Jun, <i>NYK Sunrise</i>	-0.017	-0.030	-0.018
Average	-0.015	-0.035	-0.022

and thus reduce the maximum supersaturation. However, this enhanced growth rate of droplets did not further reduce  $S_{\text{eff}}$  in most of the ship tracks studied here. Use of the higher condensation coefficient slightly increased the deduced updraft velocity. On the other hand, if the condensation coefficient were lower than 0.036, it would reduce the rate of droplet growth and allow higher cloud supersaturations. A lower condensation coefficient would then tend to mitigate the reductions of  $S_{\text{eff}}$  due to high  $N_d$ , which would bring the predictions of the cloud droplet growth model closer to the predictions of the Gaussian plume model (which assumes constant  $S_{\text{eff}}$ ). The fact that better agreement was found between the predictions and the measurements for higher condensation coefficients suggests, at least, that the condensation coefficient is probably not significantly lower than 0.036. If it were much lower than 0.036, then there would be less lowering of  $S_{\text{eff}}$  in the more concentrated ship tracks closer to the ships. If that were the case, the less than predicted decrease in  $\Delta N_d$  with increasing distance from the ships would be more difficult to explain.

The inferred  $S_{\text{eff}}$ , which was usually  $\sim 0.2\%$  (see columns 4 and 10 of Table 2), is similar to previous estimates for ambient eastern Pacific stratus clouds (e.g., Hudson 1983; Hudson and Svensson 1995; Hudson and Yum 1997); the implied updraft velocities ( $W$ , column 9 of Table 2) are also typical of stratus clouds (e.g., Curry 1986; Hudson and Svensson 1995; Hudson and Yum 1997). The exception is the flight of 30 June, when the inferred  $S_{\text{eff}}$  was above 1%, which was the highest  $S_c$  that was measured in this study with an inferred  $W$  of  $100 \text{ cm s}^{-1}$ .

On average, about 10% of the particles from the ship exhausts were found to act as CCN (at  $S_{\text{eff}}$ )—average of column 7 in Table 2 for the mean. This is in close agreement with the 12% value derived independently by Hobbs et al. (2000) from the ratio of  $\Delta N_d$  to the increase in total aerosol in each ship track. Since  $S_{\text{eff}}$  was about 0.2% for three of the four flights listed in Table 2, in Table 1 we have listed the CCN/CN ratios measured in the plumes for a supersaturation of 0.2%. Table 1 also shows the CCN/CN ratio for 1%  $S_c$ , which is generally an upper limit of  $S_{\text{eff}}$  (e.g., Rogers and Yau 1989) and the highest  $S_c$  measured in MAST. Table 1

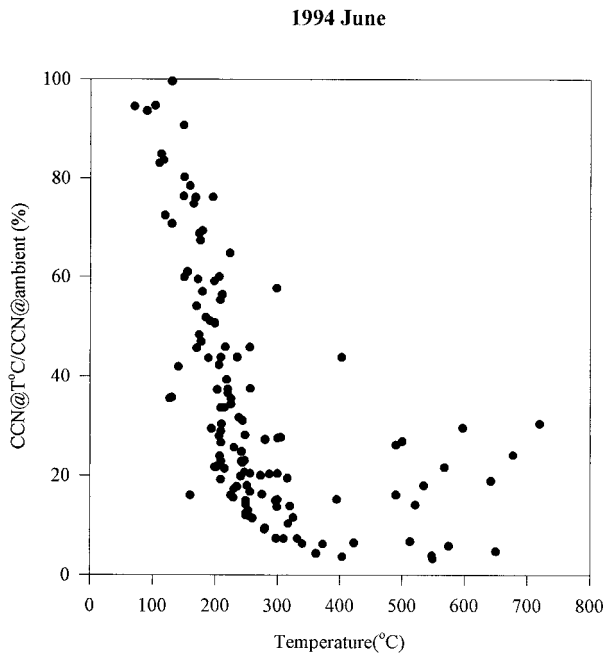


FIG. 6. Composite of all CCN (at 1%  $S_c$ ) volatility measurements in background air. Measurements of heated samples were normalized to near-simultaneous measurements of unheated samples.

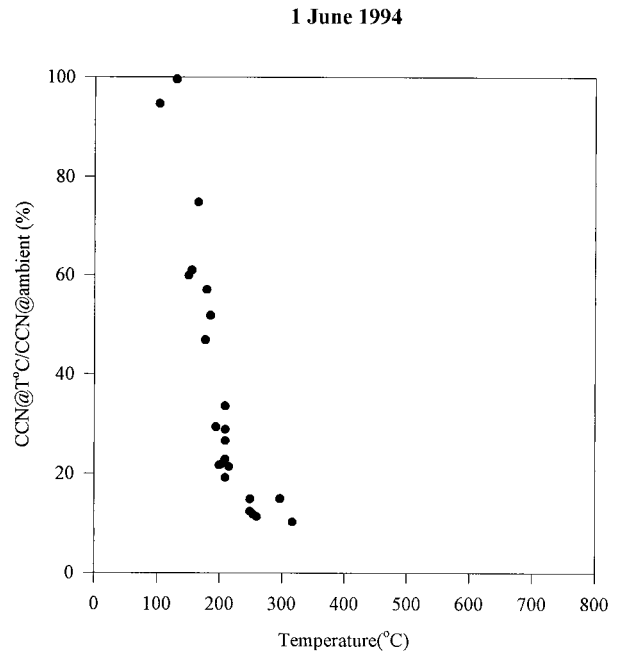


FIG. 7. As Fig. 6 but for data obtained on 1 Jun.

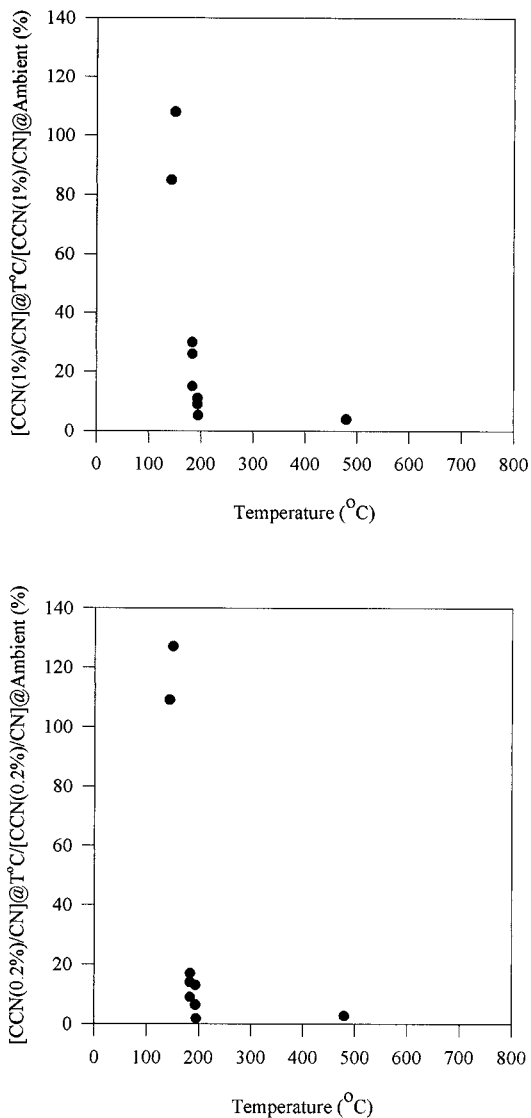


FIG. 8. As Fig. 6 but for measurements within ship plumes and CCN at 1%  $S_c$ . Except for the point at 470°C, which was made on the *NYK Sunrise*, all of the measurements were made on the *Monterrey*. The data are further normalized to the ambient CN concentrations because the concentrations within the plumes were not constant.

shows substantial differences in CCN/CN ratios for different ships, with engine type and/or fuel type appearing to affect the value of this ratio. Clearly, turbine engines using navy distillate fuel emit fewer CCN than diesel engines using marine fuel oil.

The results listed in Table 1 are consistent with the hypothesis of Hobbs et al. (2000) that fuel type is a better predictor than engine type of whether ship tracks will form in a given environment. For example the *Safeguard*, which had diesel engines but used navy distillate fuel, had CCN/CN ratios similar (even lower) than those of the turbine-powered ships. However, the evidence presented here that favors fuel type over engine type is

based on measurements of the plume from just one diesel-engine ship that used the higher-grade fuel.

Although there was variability in the plume measurements from a given ship, and there was variability among ships with similar fuels and engines, these variabilities were not enough to blur the clear differences in CCN/CN ratios between the different engines and/or fuels. The diesel engines burning marine fuel oil had order of magnitude higher CCN/CN ratios (Table 1). The much lower CCN/CN ratios for the turbine-powered ships using navy distillate fuel are probably the reason that these ships were never observed to produce ship tracks in MAST. Although the navy-distillate-fueled *Safeguard* produced a weak ship track, it did so only in very clean air.

The CCN/CN ratios are also consistent with the dry particle measurements described by Hobbs et al. (2000). The lower CCN/CN particle ratios for the distillate-fueled exhausts are consistent with the smaller particle sizes that were measured in those exhaust plumes. The larger particles in diesel plumes were consistent with the higher CCN/CN ratios found in those plumes.

If CN fluxes were similar for the variously powered and fueled ships, as seemed to be the case (Hobbs et al. 2000), then we might expect  $\Delta N_d$  to be an order of magnitude less for the navy-distillate-fueled ships compared to the ships burning marine fuel oil. The measured  $\Delta N_d$  in the ship tracks produced by diesel-powered ships using marine fuel oil (Table 1) ranged from 40 to 160  $\text{cm}^{-3}$  (Fig. 4) [there was only one case lower than 40  $\text{cm}^{-3}$ : 15  $\text{cm}^{-3}$  (Fig. 4b)]. The background  $N_d$  values were about 50  $\text{cm}^{-3}$  for two of those flights and 150  $\text{cm}^{-3}$  for the other two flights (column 3 of Table 2). Any one of the  $\Delta N_d$  values in the ship tracks is distinguishable from the lower values of  $N_d$  in the background clouds (80%–320% differences) because even for the higher background  $N_d$  values, the  $\Delta N_d$  values in ship tracks were 27%–107% above the mean background  $N_d$ . This is still distinguishable from the standard deviations of the 1-s background  $N_d$ , which ranged from 7% to 23% of the background mean  $N_d$ . There were some higher standard deviations on 8 June, a clean background day. We have inferred that  $\Delta N_{\text{CCN}}$  would be an order of magnitude lower, thus 4–16  $\text{cm}^{-3}$ , for the navy-distillate-fueled ships because of their order of magnitude lower CCN/CN ratios. For a background cloud with  $N_d$  of 50  $\text{cm}^{-3}$  this is an 8%–32% perturbation, which is generally greater than the standard deviation of the mean value of  $N_d$  in the background cloud (7%–23%). However, for the higher background  $N_d$  values, this  $\Delta N_{\text{CCN}}$  represents a perturbation of only 3%–10%, which is less than the measured variability in  $N_d$  in the background clouds (7%–23%), making it difficult to produce a detectable ship track.

The above discussion explains why the turbine-powered ships using navy distillate fuel (*Copeland*, *Kansas City*, and *Mt. Vernon*) did not produce ship tracks when the background clouds had high  $N_d$  values ( $N_d > 150$

$\text{cm}^{-3}$ ), while the *Safeguard* did produce weak ship tracks with background  $N_d$  values of 12–40  $\text{cm}^{-3}$ . Conversely, we suggest that turbine ships could produce ship tracks in cleaner background clouds ( $N_d < 50 \text{ cm}^{-3}$ ), and that the *Safeguard* could not produce ship tracks when background  $N_d$  were higher than 150  $\text{cm}^{-3}$ , conditions that were sometimes encountered in MAST.

The fluxes of CCN from the ships can be put into perspective by comparing them with fluxes from an urban area (Frisbie and Hudson 1993). First, the average CN flux from the ships was about 1% of that from the city of Denver, Colorado— $10^{16} \text{ s}^{-1}$  (Hobbs et al. 2000) versus  $10^{18} \text{ s}^{-1}$  (Frisbie and Hudson 1993). The CCN/CN ratios at high  $S_c$  for the diesel ships burning low-grade fuel were remarkably similar to results for the city of Denver—16.6% for  $S_c = 1\%$  (MAST) and 17% for  $S_c = 0.9\%$  (Denver); thus, the flux of CCN with  $S_c = 1\%$  from the ships was about 1% of that from Denver. However, for  $S_c = 0.2\%$ , these ships produced significantly higher CCN/CN ratios than Denver—5.8% versus 1.4%. Thus, the flux of CCN from dirty ships (diesel with low-grade fuel) at typical stratus cloud supersaturations (0.2%) was about 4% of that from the city of Denver. On the other hand, the CCN/CN ratio for the cleaner-burning turbine-powered and/or higher-grade fuels was significantly lower for  $S_c = 1\%$  (Table 1), but only somewhat lower than Denver at  $S_c$  values typical of marine stratus clouds (0.2%, Table 1). Thus, urban emissions should have a relative effect on stratus clouds that is intermediate between those of the two types of ship effluents discussed here, even though the total output of particles from a ship is much smaller than that of a large urban area.

*Acknowledgments.* We thank Ronald Ferek and other members of the UW C-131A crew for help in collecting data. This research was supported by ONR Grants N00014-93-1 and N00014-94-1-0339 to the UW and DRI, respectively.

#### REFERENCES

- Chodes, N., J. Warner, and A. Gagim, 1974: A determination of the condensation coefficient of water from the growth rate of small cloud droplets. *J. Atmos. Sci.*, **31**, 1351–1357.
- Curry, J. A., 1986: Interactions among turbulence, radiation and microphysics in arctic stratus clouds. *J. Atmos. Sci.*, **43**, 90–106.
- Durkee, P. A., and Coauthors, 2000: The impact of ship-produced aerosols on the microstructure and albedo of warm marine stratocumulus clouds: A test of MAST hypotheses Ii and Iii. *J. Atmos. Sci.*, **57**, 2554–2569.
- Ferek, R. J., D. A. Hegg, P. V. Hobbs, P. Durkee, and K. Nielsen, 1998: Measurements of ship-induced tracks in clouds off the Washington coast. *J. Geophys. Res.*, **103**, 23 199–23 206.
- Frisbie, P. R., and J. G. Hudson, 1993: Urban cloud condensation nuclei spectral flux. *J. Appl. Meteor.*, **32**, 666–676.
- Hobbs, P. V., and Coauthors, 2000: Emissions from ships with respect to their effects on clouds. *J. Atmos. Sci.*, **57**, 2570–2590.
- Hudson, J. G., 1983: Effects of CCN on stratus clouds. *J. Atmos. Sci.*, **40**, 480–486.
- , 1984: CCN measurements within clouds. *J. Climate Appl. Meteor.*, **23**, 42–51.
- , 1989: An instantaneous CCN spectrometer. *J. Atmos. Oceanic Technol.*, **6**, 1055–1065.
- , and P. Squires, 1973: Evaluation of a recording continuous cloud nucleus counter. *J. Appl. Meteor.*, **12**, 175–183.
- , and C. F. Rogers, 1986: Relationship between critical supersaturation and cloud droplet size: Implications for cloud mixing processes. *J. Atmos. Sci.*, **43**, 2341–2359.
- , and P. R. Frisbie, 1991: Cloud condensation nuclei near marine stratus. *J. Geophys. Res.*, **96** (D11), 20 795–20 808.
- , and G. Svensson, 1995: Cloud microphysical relationships in California marine stratus. *J. Appl. Meteor.*, **34**, 2655–2666.
- , and X. Da, 1996: Volatility and size of cloud condensation nuclei. *J. Geophys. Res.*, **101**, 4435–4442.
- , and S. S. Yum, 1997: Droplet spectral broadening in marine stratus. *J. Atmos. Sci.*, **54**, 2642–2654.
- Jiusto, J. E., 1966: Maritime concentration of condensation nuclei. *J. Rech. Atmos.*, **2**, 245–250.
- Leitch, W. R., J. W. Strapp, G. A. Isaac, and J. G. Hudson, 1986: Cloud droplet nucleation and cloud scavenging of aerosol sulphate in polluted atmospheres. *Tellus*, **38B**, 328–344.
- Mozurkewich, M., 1986: Aerosol growth and the condensation coefficient for water: A review. *Aerosol Sci. Technol.*, **5**, 223–236.
- Pruppacher, H. R., and J. D. Klett, 1980: *Microphysics of Clouds and Precipitation*. D. Reidel, 714 pp.
- Radke, L. F., and P. V. Hobbs, 1969: Measurement of cloud condensation nuclei, light scattering coefficient, sodium-containing particles, and Aitken nuclei in the Olympic Mountains of Washington. *J. Atmos. Sci.*, **26**, 281–288.
- Robinson, N. F., 1984: The efficient numerical calculation of condensational cloud drop growth. *J. Atmos. Sci.*, **41**, 697–700.
- Rogers, R. R., and M. K. Yau, 1989: *A Short Course in Cloud Physics*. Pergamon Press, 293 pp.
- Twohy, C. H., and J. G. Hudson, 1995: Cloud condensation nuclei spectra within maritime cumulus cloud droplets. *J. Appl. Meteor.*, **34**, 815–833.
- Twomey, S., and P. Squires, 1959: The influence of cloud nucleus population on the microstructure and stability of convective clouds. *Tellus*, **11**, 408–411.
- Warner, J., 1969: The microstructure of cumulus cloud. Part II: The effect on droplet size distribution of the cloud nucleus spectrum and updraft velocity. *J. Atmos. Sci.*, **26**, 1272–1282.