

# P1.3 An Analysis of Ice Crystal Number Concentration Versus Aerosol Number Concentration and Supersaturation During FIRE.ACE

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## 1. INTRODUCTION

The purpose of this work is to analyze relationships between ice crystal number concentration ( $N_i$ ), aerosol number concentration ( $N_a$ ), cloud condensation nuclei number concentration ( $N_{CN}$ ), and supersaturation ( $S_i$ ) with respect to ice. Formulation of precipitation processes in numerical models would be improved by a better understanding of the relationships between these parameters.

Szyrmer and Zawadzki (1997) estimated that ice nuclei (IN) from anthropogenic sources could contribute to climatic change. However, at the present time, this connection has not been established, and the relationship between man-made pollution and the IN concentration remains unclear. Mossop (1985) showed that  $N_i$  were generally less than  $100 \text{ l}^{-1}$  at  $T > -30^\circ\text{C}$  with many concentrations exceeding those of Fletcher (1962). Mossop and Ono (1969) stated that larger  $N_i$  values ( $10$ – $100 \text{ l}^{-1}$ ) appeared to occur at warmer  $T \sim -5^\circ\text{C}$  and that there could be  $10^3$  times as many  $N_i$  as expected on the basis of ice nucleus measurements. DeMott et al. (1982) suggest that in zones of rapid uplift, there may be higher  $S_i$  that causes ice nuclei to be activated. Mossop (1970) stated that nucleation processes are time dependent and many more ice nuclei would be activated in natural clouds that last tens of minutes compared to laboratory clouds which typically last 2-3 minutes.

These studies suggest that nucleation of ice crystals is not easily parameterized. Meyers et al (1992) parameterized ice crystal concentrations using  $S_i$  and  $T$ . Gultepe et al. (2001) showed that  $N_i$  in precipitation sizes ( $>1000 \mu\text{m}$ ) could be parameterized as a function of  $T$ . However there was no apparent correlation for sizes less than  $1000 \mu\text{m}$ . In the present work, the observations collected with a Convair-580 during the First International Satellite Cloud Climatology Project (ISCCP) Regional Experiment-Arctic Cloud Experiment (FIRE.ACE) will be analyzed to better understand relationships among  $N_i$ ,  $N_a$ , and  $S_i$ . The ice nuclei concentration ( $N_{IN}$ ) cannot be equal to  $N_i$ , but to first order it is assumed that  $N_{IN} \sim N_i$  (Rogers and Vali, 1987).  $N_i$  is compared to  $N_a$  ( $N_{CN}$ ) to test the assumption that  $N_{IN}$  will increase with an

increase in  $N_a$  ( $N_{CN}$ ). It should be recognized that processes such as ice multiplication, along with temperature and supersaturation dependencies would confound any attempts to relate  $N_i$  with  $N_a$ . Because of a large particle's many nucleation sites,  $N_i$  can also be compared with aerosol surface area, but it will not be considered here.

## 2. OBSERVATIONS

The observations were gathered during FIRE.ACE which took place over the Arctic Ocean during April of 1998, and details on aircraft observations can be found in Gultepe et al. (2001, 2002). For this study, data collected during research flights over 12 days were analyzed.

The averages of  $N_i$ ,  $N_a$ , and  $S_i$  over  $5^\circ\text{C}$  intervals were calculated from the measurements of the Particle Measuring Systems (PMS) 2-Dimensional Cloud (2DC) probe at sizes from  $25$  to  $800 \mu\text{m}$ , 2-D Precipitation (2DP) probe at sizes from  $200 \mu\text{m}$  to  $6400 \mu\text{m}$ , Particle Cavity Aerosol Spectral Probe (PCASP) at sizes between  $0.135$  and  $3 \mu\text{m}$ , and the Li-Cor infrared gas analyzer (Li-Cor 6262  $\text{CO}_2/\text{H}_2\text{O}$ ) frost point measurements, respectively. The Li-Cor measurements can be more accurate than EGG dew-point temperature ( $T_d$ ) measurements at cold temperatures. Its accuracy for  $T_d$  is about  $\pm 0.2^\circ\text{C}$  (Li-Cor Inc). The  $N_a$  measurements with the the PCASP at sizes  $>0.8 \mu\text{m}$  are affected by ice crystals where the uncertainty is about 10-50 particles per  $\text{cm}^{-3}$  (10-15%), and it is not considered in the calculations. The 2D-C probe measurements are not accurate at sizes less than  $100 \mu\text{m}$ , and are discounted. Ice crystal number concentration from the Forward Spectral Scattering Probe (FSSP) ( $N_{if}$ ) measurements at sizes  $<100 \mu\text{m}$  can sometimes be used to estimate  $N_i$ . The standard and extended size ranges for the PMS FSSP were  $3$ - $45 \mu\text{m}$  and  $5$ - $95 \mu\text{m}$ , respectively. Arnott et al. (2000) suggested that FSSP measurements in glaciated clouds can be used for  $N_i$  measurements.  $N_{if}$  was also used for a qualitative analysis of a relationship between  $N_i$  and  $N_a$ . The condensation nuclei ( $N_{CN}$ ) for sizes  $>0.005 \mu\text{m}$  were measured with a TSI-3025 Particle counter.

## 3. METHOD

The Rosemount icing detector (RID) voltage signal, hot-wire probe and PMS measurements are used to identify glaciated regions (Cober et al. 2001). Liquid water content (LWC) and total water content (TWC) are obtained from the Nevzorov probe (Korolev et al. 1998).

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Observations were averaged over 10-s intervals.  $N_i$  from the 2DC (2DP) probe are determined for sizes larger than 100 (1000)  $\mu\text{m}$  as  $N_{\text{IDC}}$  ( $N_{\text{IDP}}$ ).  $N_{\text{if}}$  for sizes  $<100 \mu\text{m}$  are determined with the FSSP probes. Details on the rationale for using  $N_i$  from the FSSP probe can be found in Gultepe et al. (2001).  $N_{\text{if}}$  and CN number concentrations are also analyzed as a function of RHi using 1-s data.

Time series of the measurements are analyzed to better understand temporal variability in the microphysical parameters. Also, the parameters are averaged over 5°C intervals to show the relationships between  $N_i$  (or  $N_a$ ) and T that are commonly used in large-scale models.

## 4. RESULTS

### 4.1 1-s Observations and profiles

Fig. 1 shows a time history of a flight in glaciated clouds on 22 April 1998. The panels from bottom to top show T, RID voltage,  $N_{\text{ifs}}$  (diameter  $<45 \mu\text{m}$ ) and  $N_{\text{ife}}$  ( $<95 \mu\text{m}$ ),  $N_a$  and  $N_{\text{CN}}$ , and IWC(TWC-LWC), respectively. The  $N_a$  is less than  $100 \text{ cm}^{-3}$  at about  $-2^\circ\text{C}$ , but it increases to  $1200 \text{ cm}^{-3}$  at  $-20^\circ\text{C}$ . When the aircraft reaches the  $-20^\circ\text{C}$  level, the IWC has values of  $0.05 \text{ g m}^{-3}$ . The TWC (or IWC) is usually less than  $0.05 \text{ g m}^{-3}$ . Throughout the climb, IWC is about  $0.03 \pm 0.01 \text{ g m}^{-3}$ . In this case,  $N_{\text{if}}$  increased with increasing  $N_a$  and decreasing T where RHi was 100%. The RID values indicate that supercooled droplets were present for two short time periods.  $N_{\text{if}}$  reached values of more than  $4000 \text{ l}^{-1}$  and were generally correlated with  $N_a$ . The data points of  $N_i$  found at  $\text{RHi} < 100\%$  are likely related to particles falling from higher levels.

$N_a$  and  $N_{\text{ife}}$  versus T are shown in Figs. 2 and 3, respectively. The median, 5%, and 95% values did not show a clear trend with T. Fig. 4 shows 1-s data of  $N_a$  and  $N_{\text{CN}}$  for the entire project. Although, both parameters reached large values at about  $\text{RHi} = 100\%$ , there was no significant change in the mean and median  $N_a$  with changing of  $\text{RHi}$ . Overall, the mean and median  $N_{\text{CN}}$  increased gradually with increasing RHi except at  $\text{RHi} > 110\%$ . Because of the size range of the probes,  $N_{\text{cn}}$  was higher than  $N_a$ . Fig. 5 indicates that  $N_{\text{ife}}$  also increased toward  $\text{RHi} = 100\%$  value, and it reached a maximum at about  $S_i = 10\%$ . Fig. 6a, which shows  $N_{\text{IDC}}$  versus RHi, indicates that the median  $N_i$  (also 90% values) increase with increasing  $\text{RHi}$ . Fig. 6b shows  $N_{\text{IDC}}$  versus  $N_a$  and  $N_{\text{CN}}$ . Median and 90% values of  $N_{\text{IDC}}$  increased slowly with increasing  $N_{\text{CN}}$  from  $200 \text{ cm}^{-3}$  to  $800 \text{ cm}^{-3}$ , with most of the data points scattered below  $5 \text{ l}^{-1}$ .

### 4.2 Summary of Results

The  $N_{\text{ife}}$  versus  $S_i$  and  $N_a$  (or  $N_{\text{CN}}$ ) showed some correlations, but these relationships were likely affected by the uncertainties related to  $N_{\text{if}}$ ,  $S_i$ , and  $N_a$ . Overall,  $N_{\text{if}}$  was approximately 1 order of magnitude larger than  $N_{\text{IDC}}$ . Lawson et al. (2001) using a cloud particle imaging (CPI) probe measurements showed that high concentration of small ice crystals (sizes  $<100 \mu\text{m}$ ) can exist in the ice clouds. Their results are consistent with those of the

present work. Neither  $N_{\text{ife}}$  or  $N_a$  showed a trend with T (Figs. 3 and 2).  $N_{\text{cn}}$  measurements indicated the same lack of dependence (not shown). Both  $N_{\text{ife}}$  and  $N_a$  ( $N_{\text{CN}}$ ) increase with increasing RHi.

$N_{\text{IDC}}$  increased with increasing  $N_a$  and RHi but there are still large uncertainties related to ice particles with sizes larger than  $100 \mu\text{m}$ . Also, it is expected that an uncertainty in RHi can be about 20%, causing further complications in the analysis.  $N_i$  values at low RHi levels are likely related to falling particles from high levels and the uncertainty in RHi.

## 5. DISCUSSION AND CONCLUSION

Gultepe et al. (2001) showed that  $N_{\text{IDC}} > 100 \mu\text{m}$  did not have a relationship with T but they found that  $N_{\text{IDP}} (>1000 \mu\text{m})$  has a relationship with T for data collected in several Canadian field projects. Although  $S_i$  is not included in their parameterizations, Figs. 5 and 6 suggest that  $N_i$  values at sizes  $>100 \mu\text{m}$  is correlated with  $S_i$  and consistent with earlier studies.

Overall, the variability and uncertainties related to  $N_i$  and  $N_a$  ( $N_{\text{CN}}$ ) indicate that the results in the present work need to be verified using additional data sets.  $N_i$  may increase with increasing  $N_a$ , as shown in Figs. 6. This suggests that aerosols from either natural or anthropogenic sources can affect ice microphysical parameters. A better measurement of  $N_i$  at small size ranges is a necessity to better understand ice crystal nucleation processes.

Supersaturation with respect to ice is an important parameter affecting  $N_i$  formation (Fig. 5 and 6a), and it needs to be measured accurately in order to develop an appropriate parameterization. Dew-point measurements obtained from EGG hygrometers in the earlier studies were used to develop parameterized equations. Those equations need to be verified with new data sets.

Other data sets from the Canadian field projects can also be used for a further understanding of the  $N_i$  versus  $N_a$  and  $S_i$  relationships. The uncertainties in the data from most of the instruments used in this study pose very difficult problems. Hopefully, with further analysis, these can be overcome. Some new instrumentation can also be tested in the future. For example, a new Small Ice Detector (SID) as described by Field et al. (2000) can be used in future field projects to help estimate  $N_i$  at small sizes.

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## 6. REFERENCES

- Arnott, W. P., D. Mitchell, C. Schmitt, D. Kingsmill, and D. Ivanova, 2000: Analysis of the FSSP performance for measurement of small crystal spectra in cirrus. 13<sup>th</sup> international Conf. On Clouds and Precipitation, 14-18 Aug. 2000, Reno, Nevada, 191-193.
- Cober, S.G., G.A. Isaac, A.V. Korolev, and J.W. Strapp, 2001: Assessing cloud-phase conditions. *J. Appl. Meteor.*, 40,1967-1983.
- DeMott, P. J., W. G. Finnegan, and L. O. Grant, 1982: A study of ice crystal formation in a stable stratified orographic cloud. Preprints, Cloud Phys. Conf., Chicago, IL, 488-490.
- Field, P. R., R. J. Cotton, and D. Johnson, 2000: Ice nucleation in orographic wave clouds 13<sup>th</sup> international Conf. On Clouds and Precipitation, 14-18 Aug. 2000, Reno, Nevada, 33-35.
- Fletcher, N. H., 1962: *Physics of Rain Clouds*. Cambridge University Press, 386 pp.
- Gultepe, I., G. A. Isaac, and S. G. Cober, 2001: Ice crystal number concentration versus T in mid-latitude and Arctic clouds. *Inter. J. Climate*, 21, 1281-1302.
- Gultepe, I., and G. Isaac, 2002: Air mass origin effect on cloud microphysical characteristics during FIRE.ACE. *J. Geophys. Res.*, in Press.
- Korolev, A.V., J.W. Strapp, G.A. Isaac, and A. Nevzorov, 1998: The Nevzorov airborne hot wire LWC/TWC probe: Principles of operation and performance characteristics. *J. Atmos. Ocean. Tech.*, 15, 1496-1511.
- Meyers, P. M., P. J. DeMott, and W. R. Cotton, 1992: New primary ice-nucleation parameterizations in an explicit cloud model. *J. Appl. Meteor.*, 31, 708-721.
- Lawson P.R., Baker B.A., , and Schmitt C.G., 2001: An overview of microphysical properties of Arctic clouds observed in May and July 1998 during FIRE.ACE. *J. Geophys. Res.*, 106, 14989-15014.
- Mossop, S. C., 1985: The origin and concentration of ice crystals in clouds. *Bull. Amer. Meteor. Soc.*, 66, 264-273.
- Mossop, S. C., and A. Ono, 1969: Measurements of ice crystal concentration in clouds. *J. Atmos. Sci.*, 26, 130-137.
- Mossop, S. C., 1970: Concentration of ice crystals. *Bull. Amer. Meteor. Soc.*, 51, 474-479.
- Rogers, D. C., and G. Vali, 1987: Ice crystal production by mountain surfaces. *J. Clim. And Appl. Meteor.*, 26, 1152-1168.
- Szyrmer, W., and I. Zawadzki, 1997: Biogenic and anthropogenic sources of ice forming nuclei: A review. *Bull. Amer. Meteor. Soc.*, 78, 209-228.

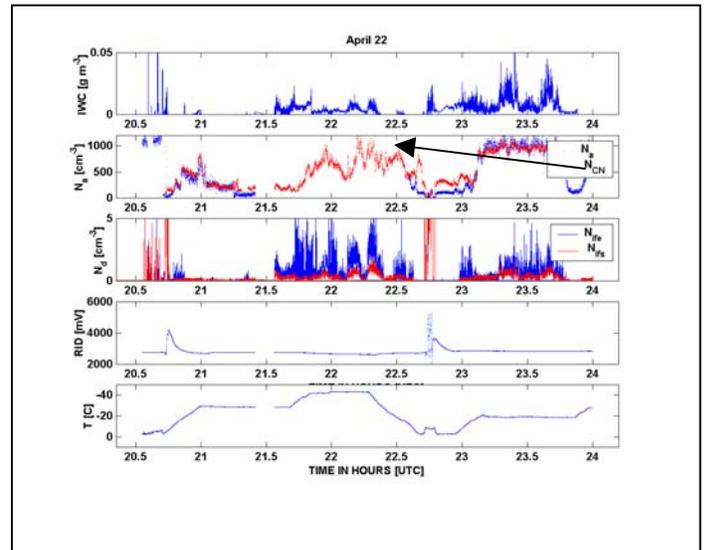


Fig. 1: Time series for April 22 case

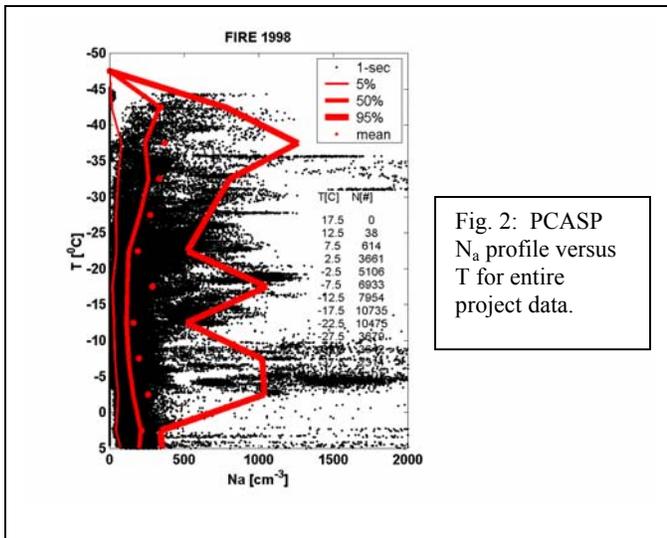


Fig. 2: PCASP  $N_a$  profile versus T for entire project data.

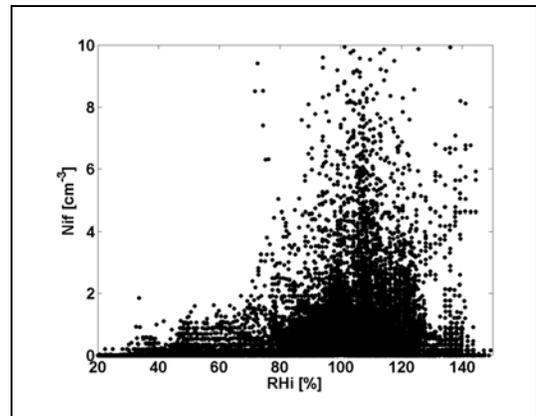


Fig. 5:  $N_{ife}$  versus RHi for all cases.

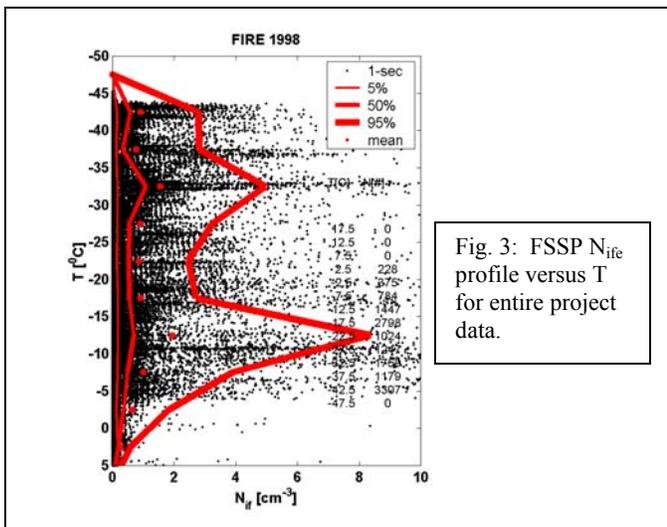


Fig. 3: FSSP  $N_{ife}$  profile versus T for entire project data.

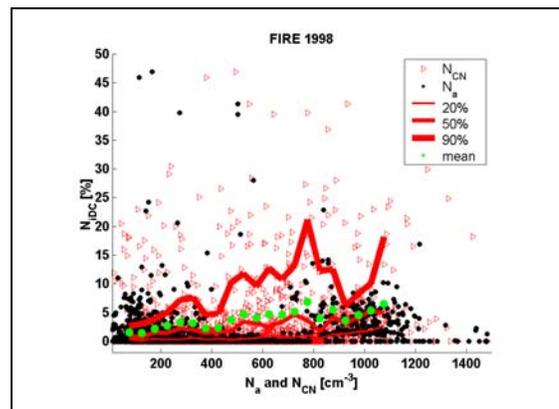
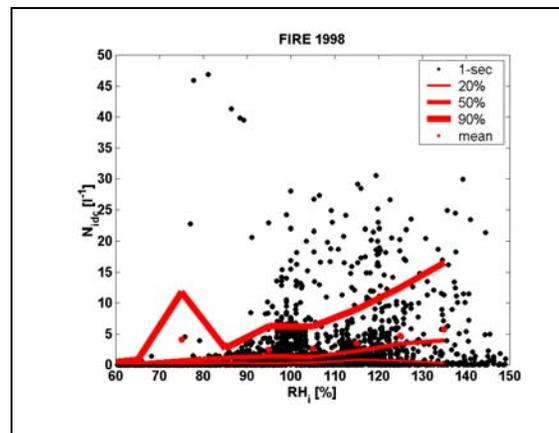


Fig. 6:  $N_{IDC}$  versus RHi (a), and versus  $N_a$  overlaid on  $N_{CN}$  (b).

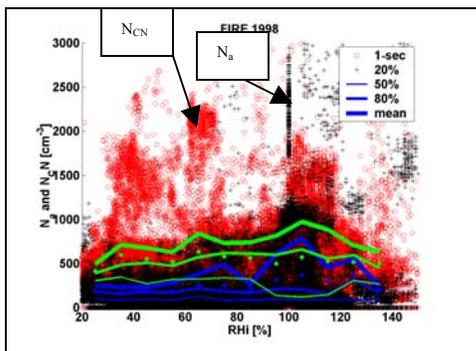


Fig. 4:  $N_a$  and  $N_{CN}$  versus RHi for entire FIRE.ACE data set. The green lines (blue lines) are for  $N_{CN}$  ( $N_a$ ) values.