Ice Nuclei, Rainwater Chemical Composition, and Static Cloud Seeding Effects in Israel

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ABSTRACT

This study analyzed the temporal and spatial variation of natural ice nuclei (IN), total suspended particles (TSP), and rainwater chemical composition (RCC) in Israel. This research is complementary to the statistical analyses of cloud seeding, which have shown significant positive seeding effects only in northern Israel, together with detrimental effects of desert dust.

It was observed that the concentration of continental components dissolved in rainwater increases by about an order of magnitude from northern to southern Israel. High values of IN, TSP, and continental origin matter dissolved in rainwater were coincident with geographic location and synoptic conditions that are indicative of the presence of desert dust. These conditions are coincident with the situations in which cloud seeding did not result in significant rain enhancement.

According to geochemical ratios, a major fraction of the dust found in the rainwater originated at the Sahara Desert. These findings support the hypothesis that desert dust has a detrimental effect on cloud seeding in Israel, due to its cloud condensation nuclei and/or ice nucleating activity.

1. Introduction

One of the outstanding problems of cloud physics is the nucleation during phase transitions in cloud forming processes. Aerosol particles play an essential role in reducing the activation energy of these phase transitions. Soluble particles serving as cloud condensation nuclei (CCN) determine the cloud droplet spectra at cloud base. Ice nuclei (IN) determine the initiation of the ice phase. If the CCN develop a wide cloud droplet spectra, collision and coalescence processes can create warm rain. When narrow spectra develop, only processes involving ice particles can create precipitation.

Experimental and operational artificial IN (AgI) seeding for rain enhancement has been conducted in Israel since 1949. The static cloud seeding is based on the hypothesis that the CCN develop a highly continental (narrow) droplet spectra, and the conversion of cloud water into precipitation is inhibited due to a lack of natural IN in the clouds (Gagin 1975). The basic statistical evaluation of the randomized experiments of cloud seeding has suggested a positive significant seeding effect of 12% in the north (Fig. 1a) and no overall seeding effect in the center and south (Rosenfeld and Farbstein 1992). These findings raised the question of whether or not natural aerosol properties determine the response of clouds to artificial IN seeding. Such a question is not easy to elucidate, but a study of natural aerosols including IN concentrations may shed some light on this problem.

Israel is located at the southeast corner of the Mediterranean Sea, in a transition zone between the Mediterranean climate in the north and the desert to the south. As a consequence, southwesterly (SW) winds (azimuth: 180°–240°) imply airmass trajectories over the Sahara, Sinài, or Negev Deserts. Northwesterly (NW) winds (azimuth: 280°–360°) imply airmass trajectories over the Mediterranean Sea. A case study showing the change in airmass trajectories with the succession of a cyclone is presented in Fig. 1b. The trajectories were computed with a constant-level model based on averaged winds calculated from rawisonde data.

High IN concentrations in air masses with trajectories over arid or semiarid land have already been reported by Schaefer (1954) and Isono et al. (1959). Mason (1971) considered clay minerals to be the most important sources of IN. Supporting evidence for this view was given by Kumai (1976), who found that about 70% of the nuclei in the centers of snow crystals in Japan, the United States, Greenland, and the South Pole were clay minerals. Clays minerals such as kaolinite and illite are a major components of Sahara Desert dust sampled in Israel (Ganor 1991; Levin et al. 1990) and Italy (Prodi and Fea 1979).

Kaolinite and illite have activation threshold temperatures of −9°C, while AgI is already activated at −4°C (Mason 1971). Thus, AgI seeding should accelerate the conversion of cloud droplets to precipitable

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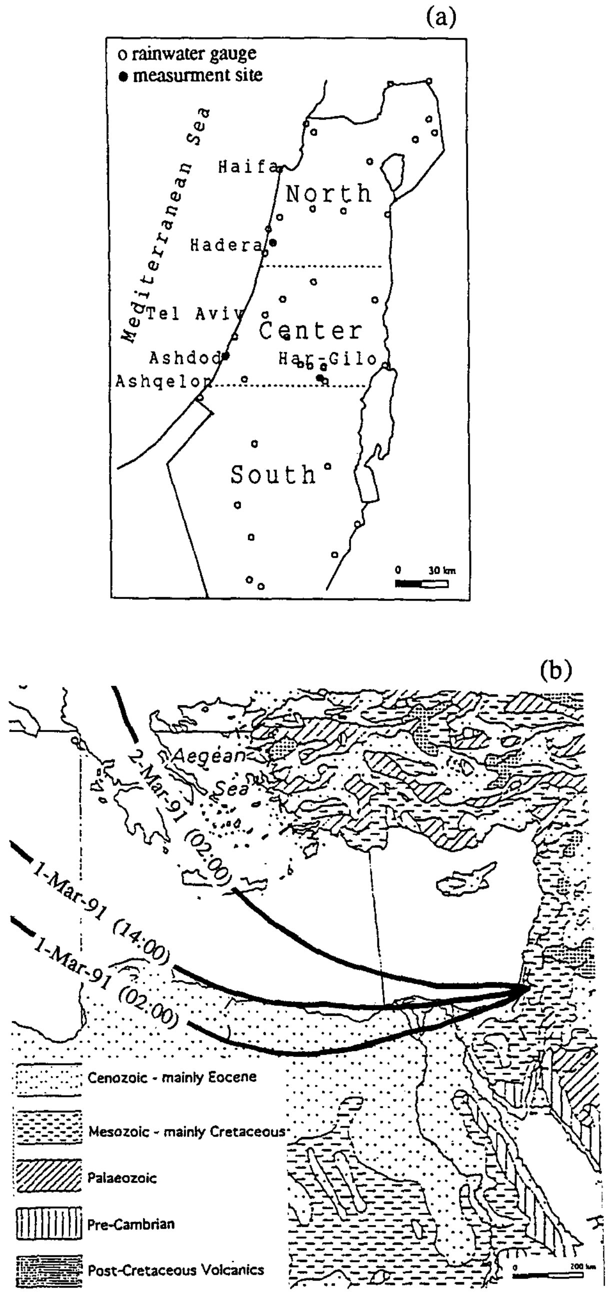

FIG. 1. (a) Map of Israel including the north, center, and south regions; rainwater sampling sites (circles); and TSP and IN sampling sites (dots). (b) Geological map of the Middle East and airmass trajectories to Israel (Tel Aviv) during the rainy period of 1–2 March 1990. (The local time of arrival is given in parentheses.)

ice particles. On the other hand, if AgI particles are artificially added to clouds containing high concentrations of natural IN, this may not be effective if cloud tops are colder than the natural IN activation temperature, and overseeding even may occur. Overseeding causes competition for the available vapor, and as a consequence the ice crystals are too small to have a sufficient terminal velocity for falling as precipitation. Another influence of desert dust on clouds may be by its soluble components. Levin (1990, 1994) reported that desert dust contains mixed particles of sulfate and mineral dust. These mixed particles, when serving as giant CCN (radius greater than 1 μm), may produce some large cloud droplets (diameter greater than 25 μm). Large cloud droplets may initiate warm rain processes (Johnson 1982) and ice multiplication (Hallet and Mossop 1974), and may invalidate the static cloud seeding hypothesis.

Rosenfeld and Farbstein (1992) investigated the influence of desert dust on seeding effects. They defined a daily "dust index," using the standard historical weather reports of dust or sand storms taken at 14 weather stations in central and southern Israel (Fig. 1a). A day was classified as a dust day if a dust or sand storm was reported at least once at any of the stations. They have shown evidence that in the experiment in northern Israel the rainfall was significantly increased by 26% on days for which dust was not reported. On the other hand, an insignificant decrease of 2% was indicated on dust days. Similar differential effects, with respect to desert dust, were observed in the operational seeding that started in the north in 1975 (Nirel and Rosenfeld 1995) and in the experiments in the center and the south of Israel. The present study investigates whether or not the crude dust index can serve as a reasonable proxy for total suspended particles (TSP), ice nuclei concentration, and rainwater chemical composition (RCC).

Dust interacts with rain cloud systems in different ways. Rain events typically start with a cold front associated with a cyclone passing to the north. The cold front typically is oriented from northeast to southwest and crosses the Israeli coast first in the north and then propagates southward. In the cold air mass behind the cold front, rain is formed by convective clouds that develop over the relatively warm sea. The leading edge of the cold-frontal rainband and the southern edge of the postfrontal rain clouds constitute the southern margins of the rain cloud systems (SMR). Rosenfeld and Nirel (1996) further assumed that due to flow pattern in the southeastern Mediterranean Sea the air masses to the north of the SMR will be relatively free of desert dust, as compared to the air mass at the SMR itself. They tested the influence of the SMR location on the seeding effect in northern Israel (combining experimental and operational data). Their results indicated a significant positive effect of 13% when the SMR was in the south and an insignificant decrease of 6% when the SMR was in the center or the north.

The present study investigated whether or not the SMR location really influenced the IN, TSP, and RCC values in the way suggested by the statistical analysis. Additionally, the changes of IN and TSP concentrations with the evolution of the synoptic and cloud systems were analyzed. Geochemical ratios were also used to investigate the origin of the dust that interacted with the rain. The ultimate goal of this study was to explain the statistical results of the static cloud seeding in Israel by related physical measurements.

2. Measurement methods

a. *Ice nuclei*

Ice nuclei samples were collected at Har-Gilo, Israel (7 km south of Jerusalem at 920 MSL), and at Ashdod, Israel (on the coast, 30 km to the south of Tel Aviv, at the same latitude as Har-Gilo). The IN were sampled in 3-h intervals during the rainy periods of winter 1990/91. In all, 73 samples were taken at Har-Gilo and 66 at Ashdod.

Each sample was obtained by pumping 100 L of air through a 0.05-μm-pore-size Millipore membrane filter (catalog No. vmwp04700). These filters were exposed in a static thermal diffusion chamber (Gagin and Aroyo 1969) to a temperature of $-15°C$ at water saturation. Calculated IN concentrations were corrected by blank counts from filters through which air was not pumped.

Extensive investigations (Vali 1985) have shown that hygroscopic particles serve as vapor sinks and, therefore, diminish the supersaturation production by the ice source in the chamber. As Ashdod is located on the coast, sea salt particles could cause an underestimate of the IN concentrations. Due to this effect, and because the Har-Gilo location is free from direct urban pollution and is close to the typical cloud-base level in Israel, Har-Gilo was taken to be the representative station.

In addition to the vapor depletion effect, IN measurements are influenced by many other factors such as the wall effect, flow rate, sampling volume, collection efficiency, and the filter type. Keeping all these parameters constant provides the possibility to measure the relative variations of IN with time, but not the absolute IN concentrations in the air.

b. *Total suspended particles*

The total suspended particles in the air were sampled by the two different instrument networks below.

1) High-volume samplers measured daily TSP at the Hadera, Israel, region (50 km north of Tel Aviv). One hundred fifty-four daily samples were obtained during the winters of 1981–90.

2) Automatic β absorption samplers measured TSP continuously at the Ashdod region. This method emphasizes the small particle range ($<10\ \mu$m), while the high-volume method also samples the larger particles. Three-hourly mean values of the TSP were calculated in synchronization with the 3-h IN measurements.

High cross correlations of TSP within each region were found: 0.81–0.89 at Ashdod and 0.83–0.98 at Hadera. No correlation with SO_2 was found. Therefore, it is concluded that most of the variability in TSP is associated with synoptic-scale conditions and not with local aerosol sources.

c. *Rainwater chemical composition*

The rainwater chemical composition (RCC) data were obtained by analyzing 554 daily rain samples taken all over Israel during the years 1982–92 (Fig. 1a). It should be mentioned that some stations in the south are to the south of the cloud seeding target area. The concentrations of Ca and Sr were obtained by inductively coupled plasma, and Na was determined by atomic absorption spectrometry (Herut 1993).

In order to analyze the marine and continental materials separately, Na and Ca were chosen as the indicators for the two origins. Assuming all Na dissolved in rainwater is marine in origin and that no chemical fractionation between solutes took place after seaspray injection into the atmosphere, the nonseaspray fraction of each ion is given by

$$(X)_{\rm nss} = (X)_{\rm tot} - \left(\frac{X}{\rm Na}\right)_{\rm sw} ({\rm Na})_r, \qquad (1)$$

where $(X)_{\rm nss}$ is the nonseaspray fraction of the ion X, $(X)_{\rm tot}$ is the total concentration of the ion X in the sample, $(X/{\rm Na})_{\rm sw}$ is the ratio of the ion X and Na in seawater, and $({\rm Na})_r$ is the Na concentration in the rain sample.

d. *Statistics*

By applying the nonparametric Wilcoxon test, the significance probability of the differences between the subgroups was inferred.

3. Results and discussion

a. *North–south gradients in RCC*

The RCC was strongly influenced by latitude, especially the continental origin elements. In the center of Israel the mean $({\rm Ca})_{\rm nss}$ concentration was double that in the north (Fig. 1a), and in the south it was 8 times higher than in the north (15 times in median values). In contrast, the mean Na concentration in the north was insignificantly higher than in the center. In the south the Na concentration was only 30% to 60% higher than in the north and center, respectively. An increase in the $({\rm Ca})_{\rm nss}/{\rm Na}$ ratio from 0.59 in the north to 1.4 in the center and to 3.3 in the south indicates an increase from north to south in the enrichment of the continental elements. These fundamental differences in continental aerosols interacting with rain may point out their possible signature on the differential seeding effects in northern and southern Israel.

b. Dust index

The 3-h measurements of IN and TSP were classified as dust periods if a dust or sand storm was reported in the standard meteorological observations at least once in central or southern Israel during the measurement time or up to 3 h before. If dust was not reported, the period was considered a no-dust period. The IN and TSP concentrations as a function of this dust index are presented in Fig. 2. The variation of the daily RCC as a function of geographic location, together with the daily dust index as defined by Rosenfeld and Farbstein (1992), is presented in Fig. 3.

It can be seen in both figures that for periods with reported dust all the measured components were higher than for periods without reported dust. On average, during periods with reported dust, the concentrations of TSP were higher by 50%–90% than during the no-dust periods. The Na and $(Ca)_{nss}$ were higher by 42% and 47%, respectively, and IN concentrations were more than doubled in the "dusty" periods. All values were significant at the 5% level. Although the dust index was defined according to stations in central and southern Israel, the most significant separation of RCC by the dust index was obtained in the north. On dusty days the Na concentration was higher by 65% (significant at the 0.5% level), and $(Ca)_{nss}$ concentration was more than double that of no-dust days (significant at the 0.01% level). In the center the differences were smaller but still significant at the 5% level. In the south the Na concentrations were the same in the two groups, and $(Ca)_{nss}$ concentrations were higher by 56% on dusty days, but not significant at the 5% level.

These findings show that the dust index has good skill in separating times with high IN and dust concentrations from times with relatively low IN and dust concentrations. This result supports the sensitivity of the statistically obtained seeding effect to the dust index as shown by Rosenfeld and Farbstein (1992).

c. Wind direction

The data were further stratified by wind direction at the 850-mb level as an indicator for the airmass trajectories. The 850-mb level was chosen as representative of the layer between the surface and upper-level winds

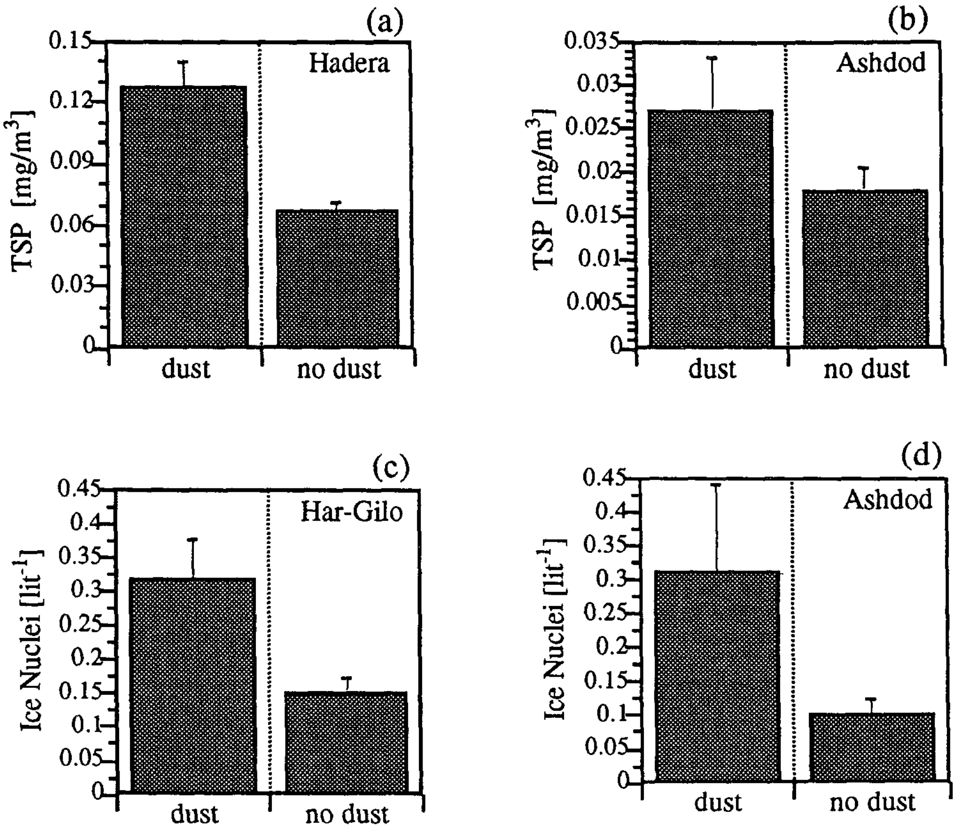

FIG. 2. Stratification by the dust index of the mean concentrations of (a) total suspended particles (TSP) at Hadera and (b) Ashdod, and (c) ice nuclei (IN) at Har-Gilo and (d) Ashdod. No dust—when dust was not observed during the measurement. Dust—when dust was observed during the measurement. The standard error is given as vertical T bars.

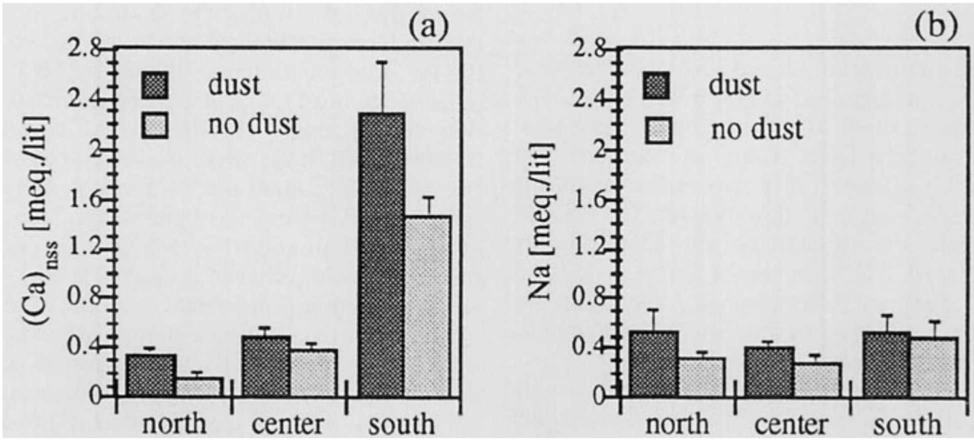

FIG. 3. As in Fig. 2 for the mean concentrations of (a) $(Ca)_{nss}$ and (b) Na in northern, central, and southern Israel.

in which Saharan dust is frequently transported, as noted by Prodi and Fea (1979).

From the wind rose presented in Fig. 4, it is clear that the maximum concentration of IN was obtained at a wind direction from 230°, in agreement with Gagin (1965). A comparison between the IN, TSP, Na, and $(Ca)_{nss}$ concentrations during SW and NW winds is presented in Table 1. It can be seen that the wind direction influenced IN and $(Ca)_{nss}$ in the most significant way. The IN concentrations at Har-Gilo were three times higher, and the $(Ca)_{nss}$ concentrations were higher by about 50% (double in median values) during SW winds than during NW winds. In contrast, the Na concentrations were not significantly influenced by wind direction. The TSP was also significantly higher during SW winds, but the differences were moderate, possibly due to the maritime component of TSP.

The way that $(Ca)_{nss}$ depends on wind direction and latitude suggests a continental aerosol source to the south and southwest of Israel—that is, desert dust. It is also evident that these aerosols serve as good ice nuclei.

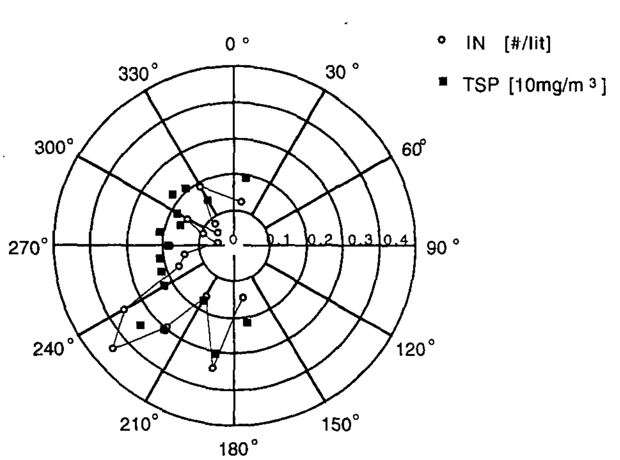

FIG. 4. A wind rose of the mean concentrations of total suspended particles at Ashdod and ice nuclei at Har-Gilo.

d. *Southern margins of the rain cloud system*

In typical rain systems over Israel, the southern margins of the rain cloud system (SMR) migrate from north to south with the progression of the rain system.

TABLE 1. Concentrations of IN, TSP, $(Ca)_{nss}$, and Na as a function of wind direction (WD) and the significance probability (sig.) of differences between the concentrations obtained during southwesterly winds and northwesterly winds.

	WD	*n*	Mean	Standard error	Median	Sig.
IN						
Har-Gilo	NW	21	0.11	0.017	0.08	0.1%
	SW	26	0.34	0.059	0.26	
IN						
Ashdod	NW	27	0.12	0.033	0.07	18%
	SW	15	0.31	0.16	0.10	
TSP						
Ashdod	NW	50	0.018	0.0006	0.017	0.4%
	SW	39	0.026	0.0023	0.021	
(Ca)nss						
North	NW	92	0.15	0.025	0.065	4.4%
	SW	66	0.23	0.033	0.11	
Center	NW	63	0.31	0.067	0.11	1.2%
	SW	34	0.44	0.093	0.28	
South	NW	49	1.34	0.171	0.97	16%
	SW	23	2.32	0.609	2.02	
Na						
North	NW	92	0.25	0.049	0.14	47%
	SW	66	0.29	0.057	0.15	
Center	NW	63	0.23	0.048	0.13	12%
	SW	34	0.37	0.123	0.15	
South	NW	49	0.47	0.070	0.32	64%
	SW	23	0.55	0.183	0.40	

For the 3-h interval of IN and TSP measurements, the SMR location was defined by regular synoptic observations. The daily $(Ca)_{nss}$ measurements were partitioned into two groups:

1) southern SMR—days with rain exceeding 2 mm in the south (Fig. 1a), and
2) central or northern SMR—days without rain exceeding 2 mm in the south.

Figure 5 presents the influence of the SMR location on the concentrations of IN, $(Ca)_{nss}$, and TSP. A clear decrease of IN and TSP with the migration of the SMR from northern to central Israel was observed. A further decrease in IN and $(Ca)_{nss}$ values was observed with the migration of the SMR to the southern part of Israel. The differences between all these values with respect to the SMR location were significant at the 5% level, except for the IN and TSP concentrations in Ashdod. Conversely, it was found that the Na concentration was insignificantly higher when the SMR location was in the southern part of Israel. This may explain the relatively small variability in the TSP concentration measured at Ashdod (Fig. 5c).

In summary, these findings lead to the conclusion that the SMR may serve as a floating boundary between an area relatively free of desert dust to the north of the SMR and the SMR area itself where high concentrations of IN and desert dust interact with the rain clouds.

e. The evolution of cloud systems with cyclone passage

The most frequent synoptic condition during rainy periods in Israel is an extratropical cyclone centered to the north of Israel. Different types of cloud systems develop with the eastward movement of the cyclone.

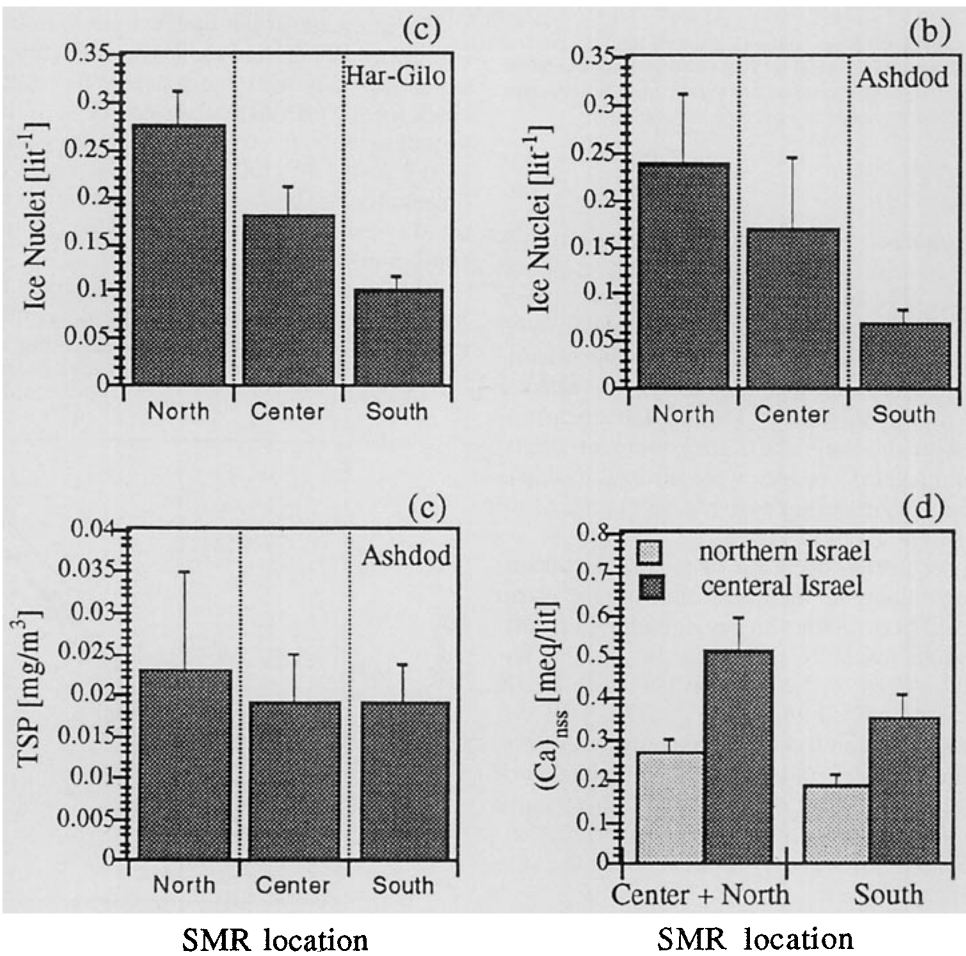

FIG. 5. Stratification by the southern margins of rain (SMR) of the mean concentrations of (a) ice nuclei at Har-Gilo and (b) Ashdod, (c) total suspended particles (TSP) at Ashdod, and (d) $(Ca)_{nss}$ and Na in central and northern Israel. South—rain spreading south of Ashqelon. Center—rain spreading north of Ashqelon and south of Hadera. North—rain spreading to Hadera and farther north. The standard error is given as vertical T bars.

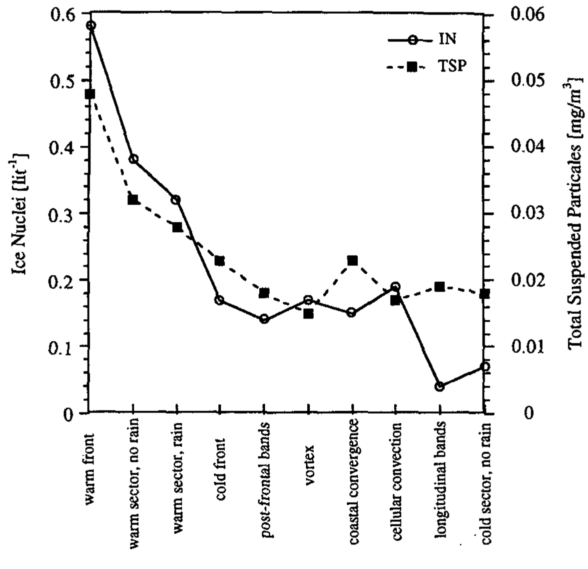

FIG. 6. The changes in mean concentrations of total suspended particles (at Ashdod) and ice nuclei (at Har-Gilo) with the evolution of cloud systems. The cloud systems are sorted from left to right according to a typical sequence in a cyclone lifetime.

The general sequence in a cyclone passage is from a warm front to a cold front, convection in the cold air mass, and then cold air without precipitation. These transitions of cloud systems are also associated with the veering of the wind and the SMR southward migration. The timescale on which cloud systems change is within the time resolution of IN and TSP measurements but not of the daily RCC measurements. Synoptic maps and satellite images were utilized to determine the cloud system during each measurement. More specific details are given in Fig. 6.

A large and constant decrease in mean IN concentration was indicated with the transition from the warm front to the cold front. After the passage of cold fronts, IN concentrations reached values close to 0.2 L^{-1}. Toward the final stages of the cyclone passage, the IN concentrations decreased to 0.1 L^{-1}. The TSP decreased moderately with the first stages of the cyclone passage, and at the final stages the TSP remained constant. This relatively small decrease in TSP, as compared to the IN, can be explained by the marine component of TSP, which was less influenced by the synoptic conditions (Table 1).

The analysis by cloud systems inherently combines many of the synoptic conditions that change in a timescale of a few hours. These results emphasize the higher variability of IN that is continental in origin, as compared to the TSP, which also combines a marine component. As the cloud systems are easy to classify by looking at satellite and radar images, they may help in the nowcasting of flow patterns and aerosol concentrations.

f. Dust origin

From the $(Sr/Ca)_{nss}$ ratio it is possible to identify the origin of the carbonate minerals dissolved in rainwater. The mean $(Sr/Ca)_{nss}$ ratio in eocene chalk exposed in North Africa is 1.2×10^{-3}. The $(Sr/Ca)_{nss}$ ratios of local upper cretaceous limestone and dolomite exposed in the Judea and Galilee mountains are 2.8×10^{-4} and 3.4×10^{-4}, respectively. The ratio in recent marine minerals (gypsum) is 2×10^{-3} (Herut 1993).

The $(Sr)_{nss}$ concentration as a function of the $(Ca)_{nss}$ concentration obtained in the Judea and Galilee mountains is presented in Fig. 7. The correlation coefficient between $(Sr)_{nss}$ and $(Ca)_{nss}$ concentrations was 0.88, and the linear regression equation was

$$(Sr)_{nss} = -6.29 \times 10^{-5} + 1.29 \times 10^{-3}(Ca)_{nss}. \quad (2)$$

The intercept of the linear regression line was small (-6.29×10^{-5}), and the slope was 1.29×10^{-3} [Eq. (2)], which is near the mean Sr/Ca ratio of eocene chalk from North Africa. As can be seen in Fig. 7, most samples with relatively low $(Ca)_{nss}$ concentrations $(<0.2 \text{ meq } L^{-1})$ fall between the ratios of chalk and limestone. For higher $(Ca)_{nss}$ concentrations, there is a trend of an increase in the $(Sr/Ca)_{nss}$ ratio toward the chalk ratio.

Higher $(Sr/Ca)_{nss}$ ratios than the local dolomite and limestone ratios were found regardless of the wind direction. These observations indicate that a major frac-

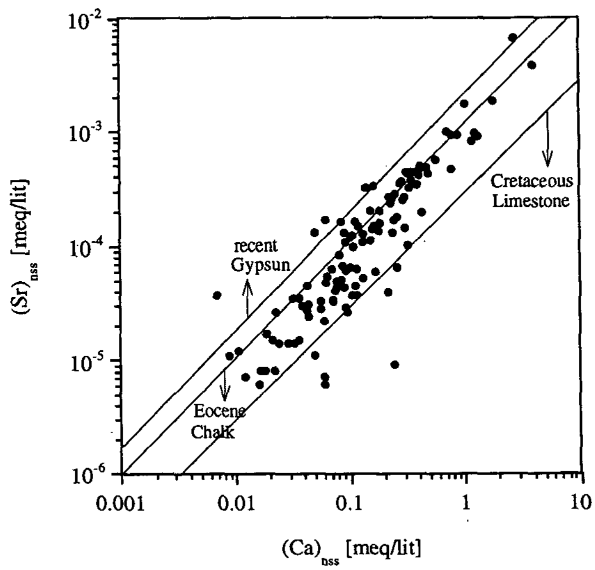

FIG. 7. The $(Sr)_{nss}$ concentration as a function of the $(Ca)_{nss}$ concentration in 117 rainwater samples from the Judea and Galilee mountains. The mean slope values for local cretaceous limestone, eocene chalk from North Africa, and recent gypsum are plotted.

tion of aerosols interacting with rain are from long-range transport origin. In cases with high $(Ca)_{nss}$ concentrations and during SW winds, the most probable origin is from the North African deserts. It should be mentioned that the coastal front mechanism is capable of injecting desert dust into the SMR also in purely westerly winds, which seemingly have no trajectory over North Africa (Rosenfeld and Nirel 1996).

4. Summary and conclusions

Analysis of combined measurements of IN, TSP, and RCC demonstrated the most important impact of synoptic conditions and geographical location on the concentrations of continental origin constituents. Geochemical ratios indicated that the origin of a major fraction of the carbonate minerals dissolved in rainwater is from chalk exposed in the Sahara Desert. In cases with high $(Ca)_{nss}$ concentrations, this origin is the most probable. These findings also reconfirmed that desert dust is a good source of ice nuclei (Gagin 1965; Perez et al. 1985; Isono et al. 1959).

The dust index defined by Rosenfeld and Farbstein (1992) proved to be useful in determining high concentrations of IN, TSP, and elements of marine and continental origin dissolved in rainwater. Therefore, it can be concluded that the dust index is a valid parameter for the stratification analysis of cloud seeding effect in Israel.

According to the static cloud seeding hypothesis, rain can be enhanced only if the natural concentration of ice nuclei is less than a certain optimum required for the most efficient conversion of cloud water into precipitation. The statistical analysis of static cloud seeding in Israel showed that low seeding effects were obtained 1) in southern Israel, 2) when dust was observed, and 3) near the SMR.

These temporal and spatial variations of the seeding effect can be related to variations in natural aerosol. The major points concerning the possible aerosol impact on differential seeding effects are presented below.

- There is both an enrichment and an absolute increase in the concentration of the continental elements interacting with rain, from northern toward southern Israel.
- Concentrations of $(Ca)_{nss}$ and IN were higher when dust was observed, during SW winds or close to the SMR, as compared to conditions in which dust was not observed, the winds wer NW or to the north of the SMR. This leads to the point below.
- The SMR may serve as a floating boundary between the following: (a) An area to the north of the SMR, which is relatively free of desert dust, and has high potential for static cloud seeding, and (b) the SMR area itself, where high concentrations of IN and desert dust interact with rain clouds, thus causing a relatively small potential for glaciogenic cloud seeding.

The findings presented here are consistent with the static cloud seeding hypothesis and with the statistical results showing a detrimental effect of desert dust on seeding effectiveness. In conclusion, clouds seeding with AgI should be most effective in the absence of desert dust, but still much research is needed to determine the exact role of the dust, as CCN and IN, on cloud microphysics in Israel.

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