Comments on "Rocket Effluent: Its Ice Nucleation Activity and Related Properties"

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Parungo and Allee (1978) reported ice nucleus (IN) measurements made from an aircraft in the stabilized ground clouds (SGC) from Titan III launches at Kennedy Space Center, Florida. Particles from the SGC were collected by drawing air, which was grab-sampled and stored in a bag, through filters. The filters then were processed in the laboratory after the flights to determine the number of particles which could serve as IN. They concluded from the measurements that the SGC contained IN.

The filters obtained in the Titan SGC were cut in half, one-half being processed by Parungo and Allee (NOAA) and the other half at the State University of New York at Albany (SUNYA). The results from both of the laboratories are summarized in a report by Hindman *et al.* (1978). The purpose of this communication is to present the results of the SUNYA analysis of the filters and our reanalysis of the data from NOAA processing of the filters. From these results we conclude that the statement by Parungo and Allee (1978) that the filter devices detected IN in the SGC is not justified.

The processing of the filters in the SUNYA diffusion chamber followed standard preparation and processing techniques described by Zamurs *et al.* (1977). All filters were processed at −20°C and at 100% relative humidity with respect to water. These conditions, typical for IN measurements, are the best for the SUNYA filter method.

The processing of the filters in the NOAA diffusion chamber followed standard preparation and processing techniques described by Langer and Rogers (1975). All filters were processed at −20°C and at 101% relative humidity with respect to water.

Results of the SUNYA and NOAA processing of the filters for the 20 August and 5 September 1977 flights are given in Tables 1 and 2, respectively. It can be seen from the tables that, early in both flights, two pairs of filters were obtained from each bag; later in the flights only one pair was obtained per bag.

The IN concentrations obtained from the first pair of filters from each bag are plotted in Fig. 1. It can be seen from the figure that the IN concentrations from the SUNYA and NOAA processes are in reasonable agreement. The IN concentrations increased above background for a short period following both launches. Thereafter, the concentrations returned to background values. It can be seen in Fig. 1 that the filters exposed to the larger flow (larger volumes of air sampled) produced the lower IN concentrations and *vice versa*. This phenomenon is known as the "volume effect."

This effect has been studied theoretically by Lala and Jiusto (1972) and has been shown to depend not only on the number of IN in the sample but on the number of cloud condensation nuclei (CCN) as well. They show that measured IN concentrations for different volumes do not follow a simple proportion but rather exhibit a logarithmic behavior described by a function of the form

$$C = C_0 V^{-\alpha}, \qquad (1)$$

where C is the concentration of the sample volume V, C_0 is a scale factor and α is the slope. The sample volume effect occurs because the vapor supply available to a filter during processing is limited by diffusive transport. Increasing the sample volume leads to higher concentrations of particles on the filter surface competing for the available vapor. The competition results in a slightly lower humidity and consequently diminished crystal counts.

Sample volumes for both flights ranged from 0.1 to 40 ℓ. The volumes were varied to minimize the competition effect from CCN which were present in the SGC at high concentrations as reported by Hindman *et al.* (1978). Small volumes were taken early when the SGC were most dense and large volumes were taken late when the clouds became diffused.

The procedure at SUNYA for reducing filter data collected at different volumes (for both ambient and SGC samples) is to determine the best fit of (1) to the data and calculate C at some reference volume V. Ten liters were chosen for V because it is in the middle of the range of volumes sampled. For samples obtained at more than two volumes, the C_0 and α were determined by fitting (1) to the data by a

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least-squares procedure. For samples obtained at two volumes, C_0 and α were determined directly. The concentration C at $V = 10\ \ell$ then was calculated from (1). The values of C are plotted as a function of the age of the SGC in Figs. 2a and 2b.

It is our understanding from Parungo and Allee (1978) that the procedure to account for the effect of the sample volume on IN measurements was as follows. The data used were collected from the first pair of filters exposed to the air captured in the bags. The data from the second pair of filters exposed were not used because the particle population in the bag was observed to decrease in number due to coagulation and wall losses. The IN concentrations from a pair of samples listed in Tables 1 and 2 (representing two sample volumes) were plotted linearly against the sample volume. The IN concentrations corresponding to 1 and 10 ℓ samples were extracted from the plots. These IN concentrations are plotted in Figs. 3a and 3b.

The IN concentrations corrected for the sample-volume effect, following the SUNYA procedure (Figs. 2a and 2b), do not contain the large increases in concentrations shortly after launch which appear in the uncorrected data (Figs. 1a–1d). The SUNYA data set in Fig. 2b is one exception. The SUNYA data indicate SGC IN concentrations increased above background concentrations shortly after launch and the increased IN values returned to background concentrations after ~2 h. However, the NOAA data in Fig. 3b do not support this feature in the SUNYA data. Furthermore, the NOAA data (Fig. 3a) indicate SGC IN concentrations were above background concentrations between 1540 and 1620 GMT; the SUNYA data (Fig. 2a) do not support this feature in the NOAA data. Consequently, the data in Figs. 2a and 2b are interpreted to indicate the filter technique could not detect IN concentrations in the SGC greater than background concentrations.

The NOAA IN concentrations corrected for the sample-volume effect, using the NOAA procedure, are shown in Figs. 3a and 3b. It can be seen from these results that the concentrations of IN measured in the SGC were not substantially greater than background IN concentrations; the large increases

TABLE 1. Filter data from the 20 August 1977 Titan launch.

Time[2] (GMT)	Sample volume $V_a(\ell)$[1]	Concentration (ℓ^{-1}) C_a SUNYA	C_a NOAA	C_b SUNYA	C_b NOAA
1316	29.2	0.82	3.8	8.2	4.1
1346	29.8	0.94	1.5	33.0	6.7
1440	1.26	15.8	60	190.0	535
1440	12.6	2.22	6.3	9.49	47
1449	1.24	25.8	48	258.0	258
1449	12.4	3.55	2.6	—	3.2
1502	1.24	16.1	45	258.0	613
1502	12.4	3.55	5.2	226.0	52
1516	14.6	—[3]	7.1	8.22	33
1516	30.3	2.64	1.6	1.32	28
1531	14.8	8.60	3.0	29.6	19
1531	29.8	2.02	2.0	13.4	23
1546	22.8	1.41	11	12.3	109
1600	22.3	0.717	14	26.9	157
1607	22.3	1.61	5.7	16.1	77
1626	29.2	1.64	2.9	6.84	40
1656	29.7	1.21	1.5	9.41	5.4
1724	29.0	1.10	1.7	9.63	11
1745	29.7	1.35	1.3	10.8	4.0
1751	30.3	0.924	3.3	6.60	38

1. Sample volume for filter b is 0.1 V_a; flow rate $a = 10\ \ell\ \text{min}^{-1}$, $b = 1\ \ell\ \text{min}^{-1}$.
2. Underlined entries are clear air samples away from the cloud.
3. Blanks indicate missing data. These filters were used for other analyses.

TABLE 2. As in Table 1 for the 5 September 1977 Titan launch.

Time (GMT)	Sample volume $V_a(\ell)$	Concentration (ℓ^{-1}) C_a SUNYA	C_a NOAA	C_b SUNYA	C_b NOAA
1214	31.0	0.258	0.77	1.29	6.4
1227	31.0	0.258	0.77	3.87	9.0
1306	1.33	27.0	15.0	150.0	175.0
1306	13.3	2.1	2.4	12.0	8.2
1319	1.31	6.12	9.0	122.0	123
1319	13.1	1.53	0.91	15.3	9.1
1332	5.23	3.06	8.3	22.9	98
1332	20.9	1.34	1.8	9.56	18
1345	13.1	1.22	1.5	12.2	18
1345	26.1	0.306	0.61	10.7	6.1
1400	13.2	1.52	0.91	9.12	9.1
1400	26.8	9.12	0.75	6.08	9.1
1413	31.3	—	1.5	0.511	10
1422	31.4	—	1.0	—	8.9
1436	31.4	0.891	0.76	10.2	6.4
1452	31.3	0.766	0.89	6.39	7.6
1505	32.7	1.10	0.85	8.57	13
1512	31.3	0.894	0.89	5.11	6.4
1535	40.5	0.888	0.39	3.95	12
1542	38.9	0.720	0.31	1.03	3.1
1550	39.1	0.511	0.41	3.07	4.1
1600	39.9	0.100	0.6	4.01	10.0
1610	39.8	0.803	0.7	4.02	6.0
1621	39.8	0.703	0.6	5.02	8.0
1642	39.1	0.409	0.3	5.11	5.0
1655	39.8	0.100	0.21	3.01	4.1
1703	39.8	0.502	2.1	4.02	4.0
1712	39.7	0.202	0.3	5.04	2.0
1729	39.8	0.502	0.1	3.01	3.0
1745	39.2	0.613	0.4	4.09	3.1
1753	40.2	0.299	0.9	3.99	5.0
1801	40.6	0.296	0.8	3.94	2.2

See Table 1 for notes.

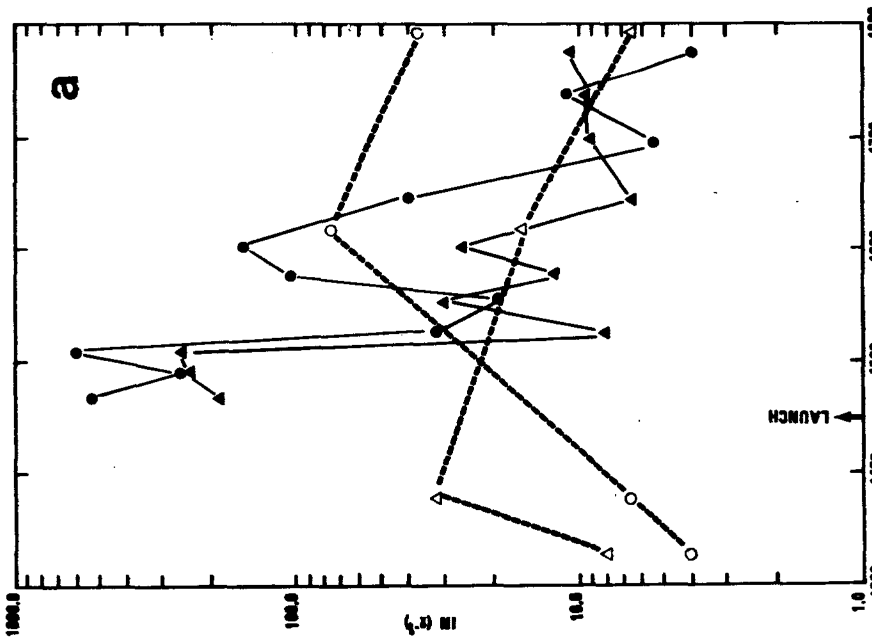


FIG. 1a. Ice nucleus concentrations (ℓ^{-1}) measured in the SGC (solid symbols) and in nearby air (open symbols). These data have not been corrected for the effect of the sample volume. Circles and triangles indicate data from NOAA and SUNYA filter processing, respectively. Samples pumped at 1 ℓ min^{-1}, 20 August 1977.

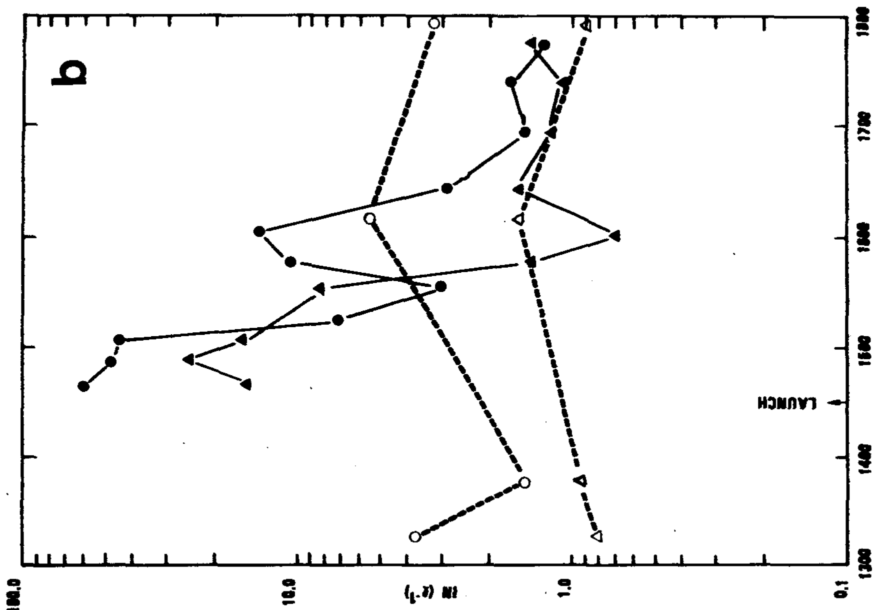


FIG. 1b. As in Fig. 1a except at 10 ℓ min^{-1}.

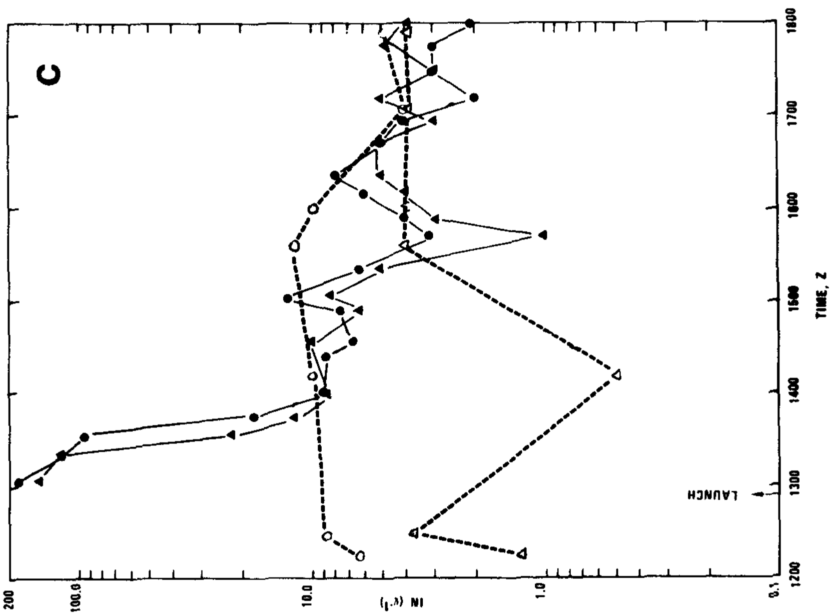


FIG. 1c. As in Fig. 1a except at 1 ℓ min⁻¹, 5 September 1977.

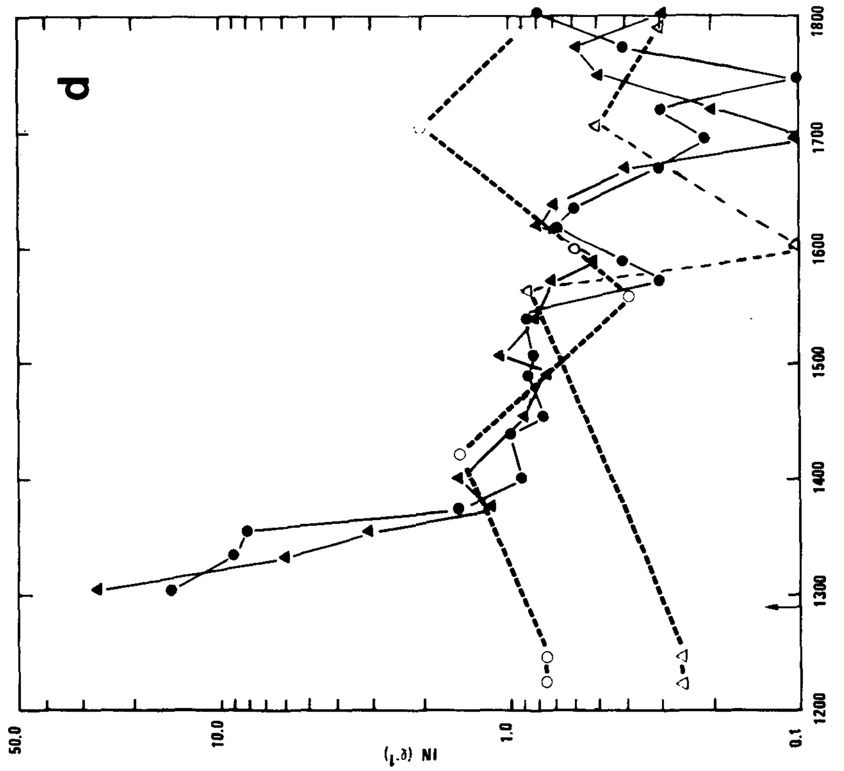
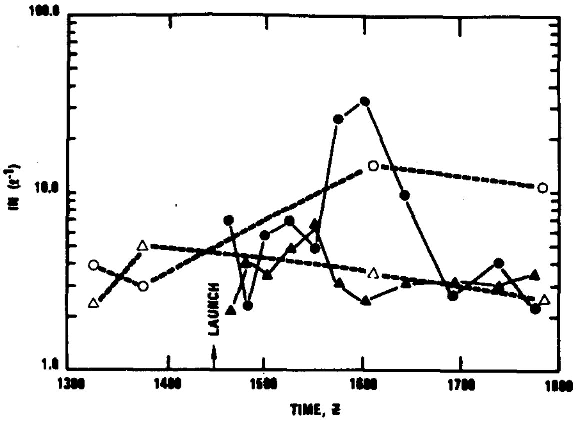
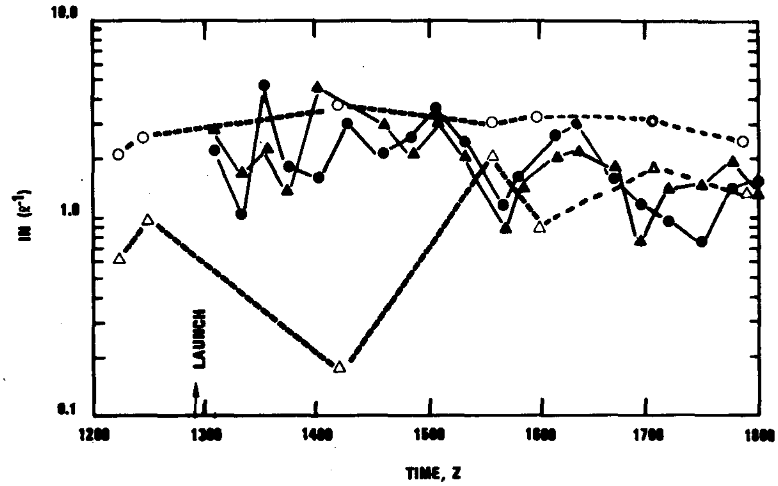


FIG. 1d. As in Fig. 1a except at 10 ℓ min⁻¹, 5 September 1977.



(a) 20 August 1977.



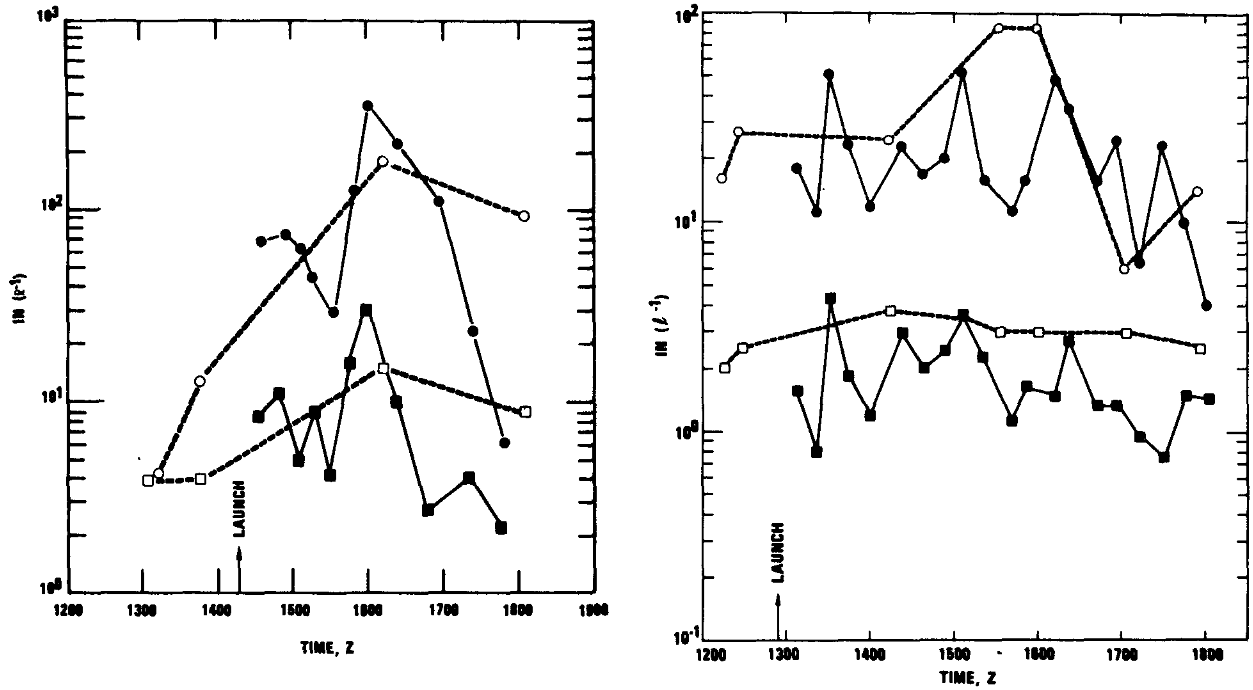
(b) 5 September 1977.

FIG. 2. Ice nucleus concentrations (IN, ℓ^{-1}) corrected for the volume effect following the SUNYA procedure. The measurements from the SGC and nearby ambient air are given by the solid and open symbols, respectively. The circles are data from the NOAA filters and the triangles are data from the SUNYA filters.

in IN concentrations immediately following launch which appear in the uncorrected data (Fig. 1) do not appear in Fig. 3. It is believed, however, that the high background IN concentrations measured at 1607 and 1751 GMT 20 August 1977 (Figs. 2a and 3a) are the result of contamination because other background IN measurements seldom show such high concentrations. Consequently, these data indicate the SGC on 20 August 1977 contained IN concentrations greater than the pre-launch atmosphere. However, the data from 5 September 1977 (Fig. 3b) indicate the filter technique could not detect IN concentrations in the SGC greater than either the pre-launch atmosphere or the background air near the SGC. This result contradicts that of Parungo and Allee (1978); they did not correct their 5 September 1977 data for the volume effect.

The lack of significant IN increases following launch in the data corrected for the volume effect (Figs. 2a, 2b and 3b) are interpreted to mean the large increases in IN in the SGC shown in Fig. 1 are due primarily to decreases in sampling volumes shortly after launch. The large increases in IN are not due to detection of IN with filters.

A comparison of the clear air IN concentrations with SGC values in Fig. 2 shows they differ at most by a factor of 2. This difference should not be interpreted to mean there are no significant concentrations of IN in the SGC. Other factors probably have influenced these results. First, the



(a) 20 August 1977. (b) 5 September 1977.

FIG. 3. As in Fig. 2 except that the volume effect is corrected for following the NOAA procedure. The circles and squares are data from the NOAA filters standardized to 1 and 10 ℓ, respectively.

presence of the high concentrations of CCN probably suppressed the peak relative humidity during processing by an unknown amount through competition for the available water vapor. This reduction in the humidity could cause a significant reduction in the activation of IN because of their sensitivity to humidity. Second, filter sampling concentrates the aerosol at the filter surface. With the high particulate concentrations in the SGC, it is possible that either IN were buried by other particles or they may have been inhibited from acting through some chemical poisoning. Until these complicating factors are clarified by laboratory work it will not be possible to reach definitive conclusions on the concentration of IN in the SGC relative to clear air concentrations using the filter technique.

Summarizing, filter samples for the detection of IN were obtained from the SGC resulting from the two Voyager launches. Processing of the filter samples by two laboratories (SUNYA and NOAA) produced IN concentrations (not corrected for the sample volume effect) that are consistent within the accuracy of the filter method. The concentrations corrected following the SUNYA procedure indicate the filter technique could not detect IN concentrations in either of the SGC greater than background concentrations. The concentrations corrected following the NOAA procedure indicate the SGC on 20 August 1977 contained IN concentrations greater than in the pre-launch atmosphere. In contrast, the concentrations corrected following the NOAA procedure indicate the SGC on 5 September 1977 did not contain IN concentrations greater than background values. There is reason to believe, however, that the chemical composition of the SGC and the high CCN concentrations may have substantially reduced the activity of IN in the samples. Nevertheless, we conclude, on the basis of the evidence presented here, that the filter devices were unable to detect IN in the SGC. We believe that IN exist in these clouds, based on laboratory data (Parungo and Allee, 1978; Hindman *et al.*, 1979) and IN measurements made in exhaust clouds from solid rocket motor firings (Hindman *et al.*, 1978).

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