Collecting Supercooled Cloud Droplets as a Function of Droplet Size

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ABSTRACT

Supercooled cloud droplets were inertially impacted onto “cloud-sieves” at a mountaintop location. The large cross-sectional areas of the sieve meshes permitted grams of cloud water to be passively collected in minutes. Each sieve was constructed from specific diameter cylindrical strands and collected all cloud droplets larger than a critical size. Procedures are developed to produce liquid water content (LWC) and chemical composition values as a function of droplet-size interval.

The sieve LWC measurements were compared with simultaneous LWC measurements obtained from a standard cloud droplet spectrometer. The sieve and spectrometer values were consistent for droplets between approximately 4 and 13 μm in diameter. The sieves overestimated the water contents of larger and smaller droplets in low LWC clouds (<0.1 g m⁻³). In high LWC clouds, the sieve LWC values for all droplet sizes closely approximated the spectrometer values.

Sources of error were investigated. Rime “feathers” and frost grew on the larger sieves in low-LWC clouds, capturing droplets smaller than the sieves critical size. Frost growth on the smallest sieve overestimated LWC values of the smallest droplets. Procedures are suggested to overcome these limitations.

1. Introduction

The chemical composition of cloud droplets can vary as a function of droplet size, as shown by Seidl (1989). This knowledge is important because Borys et al. (1988) have shown that ice-crystal chemical composition is governed primarily by the composition of the accreted droplets, presumably the larger droplets. Further, Twomey et al. (1989) and Ogren and Charlson (1989) report that the size dependence of droplet composition may affect droplet gas solubilities and reaction chemistry. Therefore, size-segregated collections of cloud droplets are necessary for chemical analyses, a conclusion also reached by Fuzzi and Waldvogel (1990).

The multicylinder device used by Langmuir and associates was the first instrument, to our knowledge, developed to collect cloud droplets as a function of droplet size. The instrument consists of a stack of different diameter cylinders as described by Brun et al. (1955) and Howe (1981). The instrument is based on the principle of inertial separation, where the smaller cylinders collect all droplets and the larger cylinders collect only the larger droplets.

Hindman and Borys (1987) employed the multicylinder to collect droplets and determine their chemical composition at Mount Washington Observatory (MWO), New Hampshire. They found the multicylinder produced insufficient collections of the smallest droplets for thorough chemical analyses because of the small cross-sectional area of the smallest cylinder. Consequently, they redesigned the multicylinder to overcome this limitation and, in doing so, developed the “cloud sieve,” which is the subject of this paper. Hindman (1988) also has designed a “cloud sieve” for airborne use.

Ogren et al. (1985) and Noone et al. (1988a) have developed a counterflow virtual impactor (CVI), which inertially separates droplets larger than about 10 μm in diameter and then collects the droplet residues on a filter. Noone et al. (1988b) and Ogren et al. (1989) have used the CVI to determine the chemical composition of cloud droplets as a function of droplet size from collections in northwestern Washington state and in Sweden. Their preliminary results are similar to those obtained by Hindman and Borys (1987) at Storm Peak Laboratory (SPL) in northwestern Colorado using the “cloud sieves”; the larger droplets are more chemically concentrated than the smaller droplets. Consequently, the CVI and the sieves have produced similar results, even though the CVI is a droplet residue collector, while the sieves are bulk water collectors.
Using a cloud-water collector capable of sampling in two size-segregated fractions, Munger et al. (1989) found large differences between the chemical composition of the larger and smaller cloud droplets. Species associated with larger aerosol particles (e.g., Na⁺, Ca²⁺, and Mg²⁺) had higher concentrations in the larger droplets, while species associated with smaller secondary aerosol particles (e.g., SO₄²⁻, NO₃⁻, and NH₄⁺) had higher concentrations in the smaller droplets. Their device was based on the dependence of droplet impaction on the size of the collecting cylinder.

Finally, a cascade impactor was utilized by Levin et al. (1990) to collect both haze and cloud droplets from a mountaintop in Israel. The residue particles from these populations were chemically analyzed; the haze droplets were more concentrated (mass solute/mass solvent), while the cloud droplets were less concentrated, confirming the calculations of Seidl (1989). Further, the concentration decreased with increasing cloud-droplet size. This result is in contrast to that of Hindman and Borys (1987) and Ogren et al. (1989) and may have something to do with the influence of desert aerosol particles in Israel.

The dependence of cloud-droplet chemical composition as a function of droplet size is most likely due to a variety of size-dependent factors: chemical com-
Fig. 2. Cloud sieves for collecting cloud droplets as a function of droplet size. The cylinder diameter $d_c$ (mm), droplet cutoff diameter at 50% collection efficiency $d_{50}$ (μm), cross-sectional area of the cylinders $A$ (m²), length of the cylinders $L$ (m), Reynolds number $Re$ of the cylinder, and maximum ice deposit $M_{max}$ are given. The $d_{50}$ and $Re$ values are valid for average SPL wintertime conditions: $-10^\circ C$, 700 mb, and 9.5 m s⁻¹ airspeed.
position of the droplet-forming nuclei, differential adsorption of gases and particles, internal droplet chemical reactions, and, no doubt, others. Cloud-droplet composition results from the SPL sieve samples are reported by Carter (1991).

The purpose of this paper is to describe the design, operation, and analysis procedures of the cloud sieves. The calibration for the sieves is developed; they are shown to be most accurate for collecting the numerous small cloud droplets and less accurate for collecting the fewer large droplets.

2. Sieve design

a. Adaptation of the sieving principle to the atmosphere

Sieves are devices commonly used to sort particles into size bins. The principle of inertial impaction is employed to collect supercooled cloud droplets on the strands of specially constructed sieves rather than wedging particles in the mesh of the sieve. The supercooled droplets freeze upon impacting the mesh, producing an easily manageable ice deposit.

The "cloud-sieving" process is illustrated in Fig. 1. As seen in the figure, all droplets within the geometric projection of the cylinders with diameters greater than or equal to about 2 μm are collected on the strands of the first sieve. Droplets smaller than 2 μm flow around the cylinders and are not collected. Simultaneously, an adjacent sieve, not in series, collects all droplets with diameters greater than or equal to about 5 μm, droplets smaller than 5 μm pass through the mesh, and so on. The physical and chemical properties of droplets between 2 and 5 μm are found by subtracting the collections made on the larger sieve from the collections made on the smaller sieve. The details of the data reduction procedures are given in section 3.

A series of sieves is constructed as shown in Fig. 2. Each sieve has a different cylinder diameter; the smallest diameter is 0.152 mm and the largest is 20 mm. The sieves with the smaller diameter cylinders were woven into a "tennis racquet" form using strands of

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**FIG. 3.** Cloud-droplet diameter (μm) collected with a 50% efficiency as a function of airspeed (m s⁻¹) and sieve cylinder diameter (mm); conditions: -10°C, 700 mb.

**FIG. 4.** The collection efficiency of a cylindrical collector used to make a sieve (CS₀, CS₁, CS₂, CS₃, CS₄, CS₅, CS₆) as a function of cloud-droplet diameter at an airspeed of 9.5 m s⁻¹; conditions: -10°C, 700 mb.
polypropylene monofilament. The sieves with the larger diameter cylinders are constructed in a "jail-cell window" form; the "bars" are made from either solid, polypropylene, or Teflon-covered polypropylene cylinders. The values of cylinder diameter $d_c$, droplet diameter collected with 50% efficiency $d_{50}$, cross-sectional area of the sieve mesh $A$, length of the cylinders in the mesh $L$, Reynolds numbers Re of the cylinders, and maximum ice deposit $M_{\text{max}}$ for each sieve are given in Fig. 2. The values are for average wintertime cloud characteristics at SPL from Hindman (1987a,b); liquid water content (LWC) $= 0.19 \text{ g m}^{-3}$, median-volume diameter $d_v = 8.8 \text{ µm}$, and average meteorological conditions; temperature $T = -10 \degree \text{C}$, pressure $p = 700 \text{ mb}$, wind speed $U = 9.5 \text{ m s}^{-1}$.

The sieve $d_c$ and $A$ values shown in Fig. 2, designed for SPL conditions, will have to be adjusted if the sieves are to be used in different conditions than found at SPL. For example, average wind speeds at MWO are much higher than at SPL. Consequently, for the same LWC and $d_v$ values at the two locations, the $d_c$ values should be increased and the $A$ values decreased to collect the same mass of cloud water at MWO.

b. Cloud-droplet cutoff sizes

The $d_{50}$ values (cm) as a function of $d_c$ (cm) and $U$ (cm s$^{-1}$) were determined using the following relationship from Ranz and Wong (1952)

$$d_{50} = \Psi^{1/2}[\gamma \rho U/(18\eta d_c)]^{-1/2},$$  

(1)

where $\Psi^{1/2}$ is the square root of the Stokes number, a parameter related to the collection efficiency $E$ of the cylinder [$\Psi^{1/2} = 0.9$ for $E = 0.5$ from the Langmuir and Blodgett (1946) cylinder values in Ranz and Wong]); $\gamma$ is the slip correction factor that is nearly equal to unity for cloud droplets; $\rho$ is the density of the droplets (g cm$^{-3}$), again nearly unity; and $\eta$ is the dynamic viscosity of the air (g cm$^{-1}$ s$^{-1}$). The $d_{50}$ val-

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**Fig. 5.** The approximate cross-sectional area of the maximum ice deposit ($d_c^2/4$) to maintain an accurate collection efficiency, where $d_c$ is the diameter of the sieve cylinder. This sketch is based on the initial ice accretions onto cylinders as photographed by Prodi et al. (1986) and Lozowski et al. (1983b) and calculated by Lozowski et al. (1983a).

**Fig. 6.** Average cumulative cloud liquid water spectrum at SPL (7-8 January 1985, 31 cloud-gun samples) as reported by Hindman (1987b). The dashed line is a reasonable extrapolation of the data.

**Fig. 7.** Schematic (not to scale) of a portion of a cloud sieve constructed in the "tennis racquet" style. The area of the crossed cylinders is a small percentage of the total area of the sieve (for example, 0.05% in the case of CSo).
ues as a function of $d_c$ and $U$ from (1) are presented in Fig. 3. Also, shown in Fig. 3 are the $d_c$ values for the various cloud sieves used at SPL. The $d_{50}$ values shown in Fig. 2 were produced using values from Fig. 3.

The “sharpness” of the sieve droplet “cuts” are determined. The sharpness, for a given airspeed, is defined as the difference in the size of droplets collected at 90% efficiency from those collected with 10% efficiency. The ideal collector, as described by Marple and Willeke (1976), is one that collects all droplets equal to or larger than the cut size with 100% efficiency, and does not collect any droplets smaller than the cut size. The sharpness of the sieve droplet cuts is determined using a general form of (1), where the $E$ versus $\Psi^{1/2}$ data of Langmuir and Blodgett for a cylinder, as reported by Ranz and Wong (1952), are fit with the following polynomial:

$$E = -0.21388 + 0.79382\Psi^{1/2} + 0.26979(\Psi^{1/2})^2 - 0.43282(\Psi^{1/2})^3 + 0.1477(\Psi^{1/2})^4 - 0.016237(\Psi^{1/2})^5,$$  \hspace{1cm} (2)

where (2) is valid for values of $\Psi^{1/2}$ between 0.25 and 3.5; for values less than 0.25, $E$ is set equal to 0.001, and for values greater than 3.5, $E$ is set equal to 0.95. The correlation coefficient for (2) is 0.99956.

Using (1) and (2), values of $E$ were determined as a function of $d_c$ and $d$ (cloud-droplet diameter). The results are given in Fig. 4, where the sharpness of the size cuts are illustrated. For example, the sieve made of 0.152-mm cylinders ($C_0$) has the “sharpest” cut; the sieve collects 8-$\mu$m droplets with 95% efficiency and collects 0.8-$\mu$m droplets with 10% efficiency (the $d_{50}$ value is 2 $\mu$m). In contrast, $C_5$ has the least sharp cut; the sieve collects 70-$\mu$m droplets with 100% efficiency and collects 10-$\mu$m droplets with 10% efficiency (the $d_{50}$ value is 23 $\mu$m). It can be seen from Fig. 4 that $C_0$ and $C_1$ have efficiency curves close together, and $C_3$ and $C_4$ likewise. Therefore, a typical sieve set exposed at SPL consists of $C_0$, $C_1$, $C_4$, $C_5$, and $C_6$.

\[c. \text{Design of sieves for SPL}\]

The steps in the design of the sieves used at SPL are detailed here. This information should enable sieves to be constructed for use at other sites with cloud conditions different than those at SPL.

The sieves are designed to collect at least 2 g of cloud water in minutes. The 2-g value was established because it is about the smallest sample that enables a series of chemical analyses (pH, cations and anions, and elements). The sample period must be kept as short as possible so the ice deposit remains cylindrical and the cylinder diameter is not effectively increased. Long-period deposits take the shape of a lemniscate (see Fig. A1). Accordingly, the cross-sectional area of each sieve is constructed to collect at least 2 g of water from a typical cloud at SPL in an exposure period of a few minutes. The cloud-water collection on the sieve is in the form of either rime or clear ice because the sieves are exposed to supercooled clouds. To ensure that the ice deposit remains cylindrical, the thickness of the ice deposit must be less than $d_c/4$, as illustrated in Fig. 5. Thus, it was estimated from Fig. 5 that the cross-sectional area of the maximum ice deposit $A_{\text{max}}$ on a cylinder should be approximately $d_c^2/4$. Assuming the density of the ice deposit is nearly 1 g cm$^{-3}$, 2 g of cloud water occupies approximately 2 cm$^3$ of ice. The volume $V$ of the ice deposit is given by $V = LA_{\text{max}}$, where the length of the cylinder $L$ (cm) to collect 2 cm$^3$ of cloud water is given by

\[L = 8d_c^2,\]  \hspace{1cm} (3)

and the cross-sectional area of the sieve $A$ (m$^2$) is given by

\[A = d_cL \times 10^{-4}.\]  \hspace{1cm} (4)

Using (4), the values of $A$ for the sieves employed at SPL are calculated; the results are listed in Fig. 2.

The exposure period $T$ (min) required to collect the 2 g of cloud water by a sieve is determined from the cloud LWC (g m$^{-3}$) for droplets greater than or equal to the $d_{50}$ value of the sieve, the sieve $A$ (m$^2$) value, and the average wind speed $U$ (cm s$^{-1}$) for the period,

\[T = 3.33[\text{LWC}(d_{50})A]^{-1}.\]  \hspace{1cm} (5)

A typical LWC spectrum in clouds at SPL is given in Fig. 6. Using this spectra and (5), and assuming $U$ equals 950 cm s$^{-1}$, the exposure periods required for $C_0$, $C_2$, $C_4$, $C_5$, and $C_6$ to collect 2 g of water are 0.77, 0.85, 0.86, 1.78, and 130 min, respectively. These results indicate that short exposure periods are required with $C_0$, $C_2$, $C_4$, and $C_5$. A considerably longer period is required with $C_6$, because of the low LWC value for drops greater than 23 $\mu$m. If the $A$ value for $C_6$ is increased by a factor of 10, then the $T$ value would be reduced from 130 min to a reasonable 13 min (but, this would produce an unrealistically large sieve). Exposure periods of around 1 min in duration are beneficial because airspeed fluctuations should be minimized.

By minimizing airspeed fluctuations, the “smearing” of $d_{50}$ values for a particular sieve will be minimized. SPL is located on the windward side of a round-top peak, so it has the most even possible airflow; no eddies from upwind peaks occur because all upwind peaks are significantly lower in elevation. Mean wind speeds are $10 \pm 2$ m s$^{-1}$, causing $d_{50}$ values to vary from 1.8 to 2.2 $\mu$m for $C_0$ and 19 to 23 $\mu$m for $C_6$, which are insignificant fluctuations.

There are two physical constraints when designing the cross-sectional area of a sieve. First, the sieves that are worn “tennis racquet” style must have sufficiently
large openings so the area of the sieve with crossed cylinders is minimized; consequently, the area of uncrossed cylinders is maximized (see Fig. 7). In the case of CS₀, the area of the crossed cylinders is 0.05% of the total area; 99.95% of the sieve area is unobstructed. Second, the sieves constructed in the "jail-cell window" style should have, based on experimentation, at least two diameters between adjacent cylinders to minimize possible airflow restrictions (the two \( A \) values for CS₀ in Fig. 2 result from this constraint, the smaller \( A \) value corresponds to a cylinder spacing of at least two cylinder diameters).

3. Procedures

The sieves illustrated in Fig. 2 were simultaneously exposed to supercooled, cloudy, and frequently snowy airstreams approximately 4 m above the snow surface at SPL. [at this location, the sieves were not significantly affected by blowing snow because Mellors (1965) reports the mean drift density decreases two orders between 0.1 and 4 m above a snow surface]. Airspeed measurements were obtained to determine the average airspeed during an exposure period (wind-run measurements for the period would be more representative). During each period, one to two droplet spectra were obtained using the cloud gun (CG) described by Hindman (1987). During select exposure periods, LWC measurements were obtained using the roto-rod (RR) device described by Rogers et al. (1983). These measurements are used to calibrate the sieves in section 4.

At the end of the exposure period the sieves were quickly removed from the airstream and brought into a cold room. The room was kept nearly at ice saturation to minimize sublimation of the collected ice deposits. Each sieve was individually scraped clean of its ice deposit (occasionally large snowflakes and graupel wedged in the mesh of CS₀; the particles were removed from the deposit before the deposit was removed from the sieve). The ice samples from each sieve were individually sealed in plastic bags and weighed for LWC determinations. The samples were kept frozen until the time of the chemical analyses.

Liquid water contents (g m⁻³) were determined from the sieve ice collections using the following relationship:

\[
LWC (d \geq d_{50}) = M(UA\Delta t)^{-1},
\]

where \( M \) is the mass of ice collected on the sieve and \( U \) (m s⁻¹) is the average airspeed during the exposure period \( \Delta t \) (s). Values of \( d_{50} \) are determined from Fig. 3. The maximum values of \( M \) that can be collected while still keeping the sieve collection surface spherical are derived in appendix A and are listed in Fig. 2 as \( M_{\text{max}} \). If \( M > M_{\text{max}} \), a correction procedure to utilize \( M \) is also developed in appendix A.

The uncertainty in the LWC values from (6) can be estimated. Values of \( A \) and \( \Delta t \) are accurately measured. Values of \( M \) and \( U \) are uncertain. A small portion of the ice mass cannot be scraped from the sieves, while some of the ice mass outside the collection area is inadvertently removed and included in the sample. These effects tend to compensate. Nevertheless, the values of
Fig. 9. Simultaneous cloud sieve (CS<sub>c</sub> - CS<sub>s</sub>) and cloud-gun (CG) liquid water content spectra obtained at SPL on 8 February 1988: (a) 0342-0538 MST, (b) 0824-0941 MST, (c) 1223-1332 MST and (d) 1742-1802 MST. Notes: 1) The periods correspond to CS<sub>c</sub> exposures, the other sieves were exposed within the CS<sub>c</sub> exposure periods; 2) the dashed portions of the CG curves are based on extrapolation of the LWC spectra below the minimum-detectable LWC value (shown by the solid symbol in the droplet spectra; the minimum-detected LWC value is the value of a drop-size range containing the minimum-detectable droplet concentration that equals one per sample volume); and 3) the error bars in (a) indicate the uncertainties in the data.

$M$ can be overestimated/underestimated by, at most, ±20%. The fluctuations in values of $U$ are also ±20% (SPL average 10 ± 2 m s<sup>-1</sup>). Thus, the uncertainty in LWC values from (6) are ±20%.

Equation (6) produces integral water content values. To obtain the LWC for drops with diameters between $(d_{50})_1$ and $(d_{50})_2$, where $(d_{50})_1 < (d_{50})_2$, the LWC value for $(d_{50})_1$ is subtracted from the value for $(d_{50})_2$,

$$\text{LWC}[(d_{50})_1, (d_{50})_2] = \text{LWC}(d_{50})_1 - \text{LWC}(d_{50})_2.$$  

(7)

The trace constituents of the ice collections are determined through chemical analyses as described by Carter (1991). Each ice deposit from a sieve contains droplets equal to and greater than $d_{50}$ for that sieve. The trace constituents for drops within a given diameter interval are determined following the procedures developed in appendix B.

4. Calibration

Simultaneous LWC values from the RR device and from CS<sub>1</sub> are compared as one check of the design, operation, and analysis of the sieve (the CS<sub>1</sub> measurements were made at SPL in December 1986 and February and March 1988). The comparison is possible because the RR has a $d_{50}$ value of about 3 μm, which is close to that of the sieves. Further, the RR is an integrating device like the cloud sieves. The uncertainty with the RR values is ±5%, as claimed by Rodgers et al. (1983).

The CS<sub>1</sub> measurements are analyzed both with and without the correction procedure (appendix A). The results are shown in Figs. 8a,b. As expected, the uncorrected sieve data produce an average LWC value (and uncertainty of the mean) systematically greater than the value from the RR: 0.23 ± 0.035 versus 0.20 ± 0.026 g m<sup>-3</sup>. The corrected sieve data produce LWC values similar to RR values for low LWC values, but the sieve produces smaller LWC values than the RR for large LWC values. The small LWC values from the sieve generally correspond to small ice-mass collections, while the large LWC values from the sieve generally correspond to large ice-mass collections. The correction procedure reduces the large LWC values more than the small values, resulting in a smaller range in the CS.
LWC values. Nevertheless, the regression analyses between the CS$_1$ and RR water content values in both Fig. 8a,b are considered significant because of the high r values. Further, the average LWC values from the CS and RR agree within the expected uncertainty.

To evaluate the other sieves, simultaneous CG and CS samples were compared. The LWC spectra from the CG were compared with the LWC spectra from the sieves (the sieve data were corrected for overloading where applicable). Typical results are given in Figs. 9a–d. The CG LWC spectra in Fig. 9 are integrations of droplet spectra giving cumulative LWC spectra. The dashed portions of the CG curves are extrapolations of the LWC spectra below the minimum detected LWC value and represent the largest droplets expected in the cloud.

The CS spectra in Fig. 9 are determined from (6) and, therefore, are also cumulative LWC spectra. When the CS spectra do not monotonically increase with decreasing droplet size, sampling variations between the sieves apparently occurred. It can be seen in Fig. 9 that the best agreement between the CS and CG LWC values occurs for the smaller droplets and the poorest agreement occurs for the larger droplets.

To further evaluate the sieve performance, all of the simultaneous CS and CG LWC measurements are compared. The measurements were made at SPL December 1985, January 1986, December 1986, February and March 1988, and January 1989. The comparison is performed by plotting the CS water content values (CS$_0$–CS$_6$) as a function of d$_{50}$, as in Fig. 9. Then, the cumulative CG water content values corresponding to the d$_{50}$ values are superimposed on the figure. Finally, paired CS and CG water content values were plotted as scatter diagrams in Figs. 10a–g.

It should be noted in Fig. 10 that the number of samples are not the same for each sieve, indicating all seven sieves were not always operated simultaneously. This fact explains the different average LWC values between sieves. For example, 32 samples are made with CS$_2$ (Fig. 10c), while 16 samples are made with CS$_3$ (Fig. 10d); thus, CS$_2$ sampled 16 clouds that are not sampled with CS$_3$.

There is considerable scatter in Fig. 10. Nevertheless, there are sufficient data to reveal the following patterns: the average CS$_0$ LWC values (Fig. 10a) are 4.5 times greater than the CG values; the average CS$_1$, CS$_2$, and CS$_3$ LWC values (Figs. 10b–d) are 1.6–2.0 times greater than the CG values; and the average CS$_4$, CS$_5$, and CS$_6$ values (Figs. 10e–g) are 4.4–2449 times greater than the CG values.

The CS$_0$ LWC values in Fig. 10a are larger than the corresponding CG values, indicating the correction procedure in appendix A cannot account for all of the
Fig. 10. Simultaneous liquid water contents from the cloud sieves (LWC_{CS}) and cloud gun (LWC_{CG}) for the same d_{50} values: (a) CS_0, (b) CS_1, (c) CS_2, (d) CS_3, (e) CS_4, (f) CS_5, and (g) CS_6. On each individual figure, the following items are indicated: the number of simultaneous samples (n), the average LWC values from the samples, the linear correlation coefficient r for the least-squares fit to the data, and the average d_{50} values. The uncertainty in the data is illustrated in Fig. 10a.

difference. A portion of the elevated CS_0 LWC values appears to be vapor deposition; as shown in appendix C (see Table C1), vapor deposition to CS_0 is a large fraction (0.28) of the total deposition (liquid plus vapor).

The CS_4, CS_5, and CS_6 LWC values in Figs. 10e-g are larger than the corresponding CG values; a result consistent with Fig. 9. These cloud sieves significantly overestimate the amounts of liquid water contained in the large drops. However, as seen in Fig. 10 for all sieves, the larger the CG LWC values, the closer the agreement between the CS and CG liquid water contents. This result is consistent with the result reported by Hindman and Boros (1987) from the early sieve measurements.

5. Discussion

There are five possible mechanisms that may explain why the LWC values from sieves CS_5 and CS_6 are sig-
significantly larger than the LWC values from the CG: 1) the sieves are collecting large droplets missed by the CG due to the small sample volumes of the CG (cubic centimeters for the CG versus cubic meters for the sieves); 2) the sieves are collecting significant numbers of droplets with sizes smaller than \( d_{90} \) due to the smearing of the size cuts for the larger diameter sieves, as illustrated in Fig. 4; 3) the collection efficiency curves in Fig. 4, which are valid for smooth cylinders, are not accurate for the rough, lemniscate shape of the ice deposits; 4) a significant amount of frost is being deposited onto the sieves due to the ice-supersaturated conditions within the supercooled cloud; and 5) snow or ice crystals are being collected by the larger sieve cylinders.

Assuming the concentrations of large droplets were steady state, it is unlikely that the CG missed significant numbers of large droplets due to its small sample volumes. Inspection of Figs. 9a–d shows that the sieves are producing significantly larger LWC values than the CG for droplets around 15 \( \mu \)m in diameter. For droplets of these sizes and smaller, the CG produces reliable droplet size and concentration values, hence, reliable LWC values because adequate numbers of droplets are sampled. Further, due to the rapid decrease in minimum-detectable CG LWC values with increasing droplet size in Fig. 9, it is unlikely that droplets larger than about 30 \( \mu \)m in diameter are present.

The effect of a sieve collecting droplets with diam-
eters smaller than \(d_{50}\) was estimated. For example, in Fig. 9a, it can be seen that CS\(_2\) collects 0.006 g m\(^{-3}\) of liquid water with droplet diameters greater than or equal to 8 \(\mu\)m. However, the average CG data indicate that about 0.001 g m\(^{-3}\) of liquid water is present for droplets \(\geq 8 \mu\)m. The difference between the sieve and CG LWC values is 0.005 g m\(^{-3}\); how much of this liquid water could have come from collection of droplets with diameters less than 8 \(\mu\)m? The amount of water in droplets with diameters between 7 and 8 \(\mu\)m is 0.002 g m\(^{-3}\), as seen in the average CG data. The collection efficiency for the 5 m s\(^{-1}\) airspeed of CS\(_2\) for droplets with diameters greater than or equal to 7 \(\mu\)m is 0.38. Therefore, 0.002 g m\(^{-3}\) \(\times\) 0.38, or 0.00076 g m\(^{-3}\), could have come from droplets between 7 and 8 \(\mu\)m. This procedure is repeated for droplets between 6 and 7 \(\mu\)m, 5 and 6 \(\mu\)m, 4 and 5 \(\mu\)m, and 3 and 4 \(\mu\)m. The individual contributions are summed, and 0.0010 g m\(^{-3}\) of the 0.0030 g m\(^{-3}\) can be explained; about 33%. Consequently, only a small portion of the LWC difference between the CS and CG values can be explained by collection of droplets with diameters less than \(d_{50}\).

The \(d_{50}\) values for the larger sieves may be overestimated. This is most likely due to rough ice deposits causing turbulence on the upwind face of the large cylinders and, thus, causing collection of smaller droplets than indicated by present theory; a result investigated by Keith and Saunders (1988). They report that “feathers of rime” grow from a cylindrical surface when the rime density is low. The rime feathers have much higher collection efficiencies for smaller droplets than do the smooth cylinders of the sieves. This condition occurs with low droplet impact speeds and cold temperatures, according to Macklin (1962). The magnitude of this effect, they report, is as follows: at an impact speed of 3 m s\(^{-1}\), a rough 5-mm-diameter cylinder has an effective smooth diameter of 3.2 mm, and at 5 m s\(^{-1}\), the effective smooth diameter is 4.8 mm. Supporting this finding, Hindman (1986) reported that at wind speeds below 7 m s\(^{-1}\), the LWC values from a 6.25-mm cylinder are systematically high; perhaps this result indicates rime feather growth on the cylinder is increasing the sweep out of droplets over that of a smooth cylinder. Further, the sieve collections reported in Fig. 9 are made at wind speeds \(\leq 7\) m s\(^{-1}\). Thus, the results in Fig. 9 indicate that CS\(_3\) and CS\(_6\) may have been collecting a portion of the droplets that theoretically only should have been collected by smooth cylinders with diameters of, respectively, CS\(_3\) and CS\(_6\); that is, rough sieves 7.9 and 20 mm in diameter may have collected a portion of the droplets that only should have been collected by smooth cylinders 1.9 and 3.8 mm in diameter. This diameter discrepancy is greater than that reported by Keith and Saunders (1988); however, they did not incorporate the effects of frost growth and snow-crystal capture, which would further roughen the cylinder.

It is shown in appendix C that frost deposition onto the sieves for typical SPL cloud conditions is 4–7 times less than the liquid water deposition for CS\(_3\)–CS\(_6\). However, for CS\(_3\) and CS\(_6\), frost deposition can be equal to or greater than the LWC flux. This finding supports the previous conclusion that frosty surfaces will collect smaller droplets than indicated by \(d_{50}\) values.

Deshler and Vali (1985), Keith and Saunders (1989), and Baumgardner and Rodi (1989) report that most ice crystals bounce off a riming surface. Using Keith and Saunders (1989) bounce-off probabilities in appendix C, we show the mass flux due to snow crystals impacting the sieves to be on the order of \(4 \times 10^{-6}\) g cm\(^{-2}\) s\(^{-1}\). This value is generally an order of magnitude less than the liquid water flux, except for CS\(_3\) and CS\(_6\). Therefore, snow impaction is expected to interfere little with the collection of cloud water for the sieves smaller than CS\(_3\). The large discrepancy between the CS\(_3\) and CS\(_6\) LWC spectra and the CG spectra, therefore, appears to be primarily due to frost and impacted snow roughing up the cylinder surface of the sieves so they collect droplets significantly smaller than predicted by the \(d_{50}\) values.

The behavior of the LWC\(_{cs}\) and LWC\(_{cg}\) values in Fig. 10 (for low-LWC clouds, LWC\(_{cs}\) \(>\) LWC\(_{cg}\), and for high-LWC clouds, LWC\(_{cs}\) \(\approx\) LWC\(_{cg}\)) can now be explained. At low LWC, rime feathers appear to grow because of the more random and uneven droplet collisions at low droplet concentrations, but at high LWC the feathers do not grow due to large liquid water fluxes onto the surface; the riming surface is more nearly spherical, smoother, and hence closer to the \(d_{50}\) value predicted for a cylinder. Further, vapor deposition to the sieves is independent of LWC; at very low LWC the sieves are collecting as much frost as liquid water, but at high LWC the sieves are collecting primarily liquid water. Development of a theory of droplet collection in a turbulent boundary layer caused by feather growth and frosting is beyond the scope of this paper. However, Lozowski et al. (1983a) appear to have made progress with this problem.

The data collection and analysis procedures originally discussed in section 3 are modified here to incorporate the results of the sieve calibrations in section 4. The one to two cloud-gun measurements during a sieve exposure period should be replaced by continuous droplet-spectra measurements obtained with an electro-optical spectrometer that detects only droplets and not snow crystals [e.g., the CSASP of Knollenberg (1981)]. If this is not possible, then, numerous and regular cloud-gun measurements should be obtained throughout the sieve exposure period. These droplet measurements will complement the sieve measurements and provide a reference liquid water spectra during the sieve exposure period. Continuous droplet measurements are required because the continuous droplet measurements
by Rogers et al. (1983) and Politovich and Vali (1983) at Elk Mountain Observatory (EMO, which is 80 km north of SPL) demonstrate rapid LWC and droplet concentration variations in periods of minutes.

The sieves should not be exposed to wind speeds $< 7 \text{ m s}^{-1}$ to avoid rime feather growth and the increased relative growth rate of frost. Further, the larger sieves are best exposed to high-LWC, large-droplet clouds, and clouds that occur about 10% of the cloud events at SPL, according to Hindman and Phillip (1990). The larger the $d_c$ value, the more frost the sieve is expected to collect than cloud water, especially for low-LWC clouds.

6. Conclusions

The principle of inertial impaction was utilized to collect supercooled cloud droplets of different sizes from a mountaintop location. Meshes of monofilament cylinders called "cloud sieves" were exposed to the cloudy airstream. The unique feature of the sieves is their large cross-sectional area, which permits grams of cloud water to be collected in minutes from an unaccelerated air stream. Each sieve was constructed from a different diameter cylinder from 0.152 to 20 mm; the smaller cylinders collected all cloud droplets down to about 3 $\mu$m in diameter, and the larger cylinders collected the larger droplets.

The sieves were designed to collect droplets from supercooled clouds that envelope Storm Peak Laboratory (SPL) in the northern Colorado Rockies. The SPL sieves may have to be modified to collect droplets at different cloud-impacted locations. For example, a location with average wind speeds significantly higher than the typical 10 m s$^{-1}$ value at SPL will require the sieve cross-sectional areas to be reduced and cylinder diameters to be increased. Then, similar amounts of cloud water and similar droplets sizes are expected to be collected.

The sieve samples are used to produce cumulative liquid water content and chemical concentration measurements. Procedures are developed for differentiating these integral values to produce liquid water content and chemical composition values as a function of droplet-size interval.

The sieve liquid water content values were compared with LWC values obtained from the standard cloudgun droplet spectrometer to calibrate the sieves. It was found that the sieves produced water content values within a factor of 4 of values from the cloud-gun for droplets between about 4- and 13-$\mu$m diameters. But the sieves consistently overestimated the water contents of the larger and smaller droplets in clouds with low liquid water contents (less than about 0.1 g m$^{-3}$). In clouds with higher liquid water contents, the sieve and cloud-gun LWC values were more nearly consistent.

Sources of error were investigated. It was concluded that the droplet sizes collected with 50% efficiency for the larger sieves were overestimated using current inertial-impaction theory for smooth cylinders. This discrepancy is most likely due to "rime feathers" and frost growing on the windward face of the large cylinders in a low LWC cloud and thus causing collection of smaller droplets. Frost growth on the smallest sieve explains the overestimated LWC values of the smallest droplets. Growth of "rime feathers" is minimized by exposing the larger sieves at wind speeds $> 7 \text{ m s}^{-1}$, while exposing all sieves to water contents greater than 0.1 g m$^{-3}$ minimizes the effect due to frost growth.

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APPENDIX A

Correction for Sieve Overloading

To obtain an accurate collection from the cloud sieves, the width of the collected ice deposit must not exceed the diameter of the sieve cylinder and the depth of the deposit should be less than $d_c/4$ (see Fig. 6). If the deposit width exceeds the cylinder diameter, then the sieve collection area will increase. In this case, the sieve is "overloaded." In the overloaded condition, the cross-sectional area of the sieves is determined from the width of the ice deposit and not the diameter of the cylinder.

The width of the ice deposit is determined assuming the deposit forms a lemniscate as reported by Rogers et al. (1983) and illustrated in Fig. A1. They determined the following relationship between the width of the ice deposit $d_{\text{new}}$ and the length of the major axis of the lemniscate $A'$,

$$d_{\text{new}} = 0.56 A'.$$

Further, the area of the ice deposit (hatched area in Fig. A1) is given by

$$\text{area} = \left(\frac{(A')^2}{x}\right) - D,$$

where $x$ is a parameter found by Rogers et al. to be $2.6 \pm 0.04$ and $D$ is the area of the lemniscate occupied by the sieve cylinder (crosshatched area in Fig. A1). The angle the lemniscate forms at its apex was found by Rogers et al. to be 46°. Thus, the value of $D$ equals...
one-eighth of the cylinder area or $\pi d_c^2 / 32$. Substituting the value of $A'$ from (A1) and the value of $D$ into (A2) results in

$$\text{area} = \frac{(d_{\text{new}}/0.56)^2}{2.6} - \frac{\pi d_c^2}{32}. \quad (A3)$$

The volume of the ice deposit on a sieve, then, is given by

$$\text{volume} = \text{area}(L), \quad (A4)$$

where $L$ is the length of the cylinders that constitute the sieve.

The volume of the ice deposit on a sieve is unknown. However, the mass $M$ of ice deposit is measured. Therefore, the following relationship between $M$ (g) and volume (cm$^3$) is established assuming an ice density of 1 g cm$^{-3}$ [the ice-density relationship of Macklin (1962) was investigated but did not improve the results],

$$\text{volume} = M. \quad (A5)$$

Combining (A3), (A4), and (A5) and solving for $d_{\text{new}}$ (cm) produces the following expression:

$$d_{\text{new}} = 0.90297(M/L)^{1/2} + 0.282913d_c. \quad (A6)$$

The values of $L$ and $d_c$ from Fig. 2 are used with (A6) to develop the $d_{\text{new}}$ relationship for each sieve,

- $CS_0$ $d_{\text{new}} = 7.13 \times 10^{-3}M^{1/2} + 4.30 \times 10^{-3}$
- $CS_1$ $d_{\text{new}} = 1.27 \times 10^{-2}M^{1/2} + 1.02 \times 10^{-2}$
- $CS_2$ $d_{\text{new}} = 2.11 \times 10^{-2}M^{1/2} + 3.74 \times 10^{-2}$
- $CS_3$ $d_{\text{new}} = 3.14 \times 10^{-2}M^{1/2} + 5.39 \times 10^{-2}$
- $CS_4$ $d_{\text{new}} = 2.76 \times 10^{-2}M^{1/2} + 1.09 \times 10^{-1}$

The maximum mass $M_{\text{max}}$ that can be deposited on a sieve before the collection efficiency degrades is assumed to occur when $d_c$ equals $d_{\text{new}}$. Therefore, setting $d_{\text{new}}$ equal to $d_c$ and solving the above series of equations results in the values of $M_{\text{max}}$ listed in Fig. 2. It can be seen from those values that the sieves with the smallest cylinder diameters ($CS_0$, $CS_1$, and $CS_2$) are the most likely to overload.

When a sieve becomes overloaded, the $d_{50}$ size and the LWC value must be corrected. The $d_{50}$ value from (1) is the initial minimum droplet size collected by the sieve of diameter $d_c$. The final $d_{50}$ value collected by the overloaded sieve is found by substituting $d_{\text{new}}$ into (1). Essentially, the ice collection is "smeared" between droplets of the initial $d_{50}$ value and the final value. Therefore, the $d_{50}$ value is established by calculating the geometric-mean diameter of the initial and final $d_{50}$ values: $\left[\frac{d_{50}\text{initial}}{d_{50}\text{final}}\right]^{1/2}$.

The initial LWC value is obtained by substituting $M$ and $A$ values into (6). However, when the sieve becomes overloaded, the final $A$ value is larger than the initial $A$ value. Consequently, the corrected LWC value is found by multiplying the value obtained from (6) by $d_c(d_c/d_{\text{new}})^{-1/2}$.

### APPENDIX B

**Determination of Trace Constituents**

The sieve samples produce a series of integral LWC values and trace constituent values that must be differentiated to determine trace constituent concentration as a function of cloud-droplet size (more precisely, droplet-size interval). The LWC ($d \geq d_{50}$) values from (6) are used to determine the trace constituents as a function of droplet size. Let the LWC value from the sieve with the larger $d_{50}$ value be LWC$_2$ and the LWC value from the sieve with the smaller $d_{50}$ value be LWC$_1$. Also, let the corresponding aqueous trace constituent concentration measurements be $C_2$ and $C_1$ (mass solute/mass solvent), respectively. The problem is to find $C_{1,2}$, the trace constituents for droplets with diameters greater than or equal to $(d_{50})$, and less than $(d_{50})$. The value of $C_1$ is made up of $C_{1,2}$ and $C_2$. Further, the value of LWC$_1$ is made up of LWC$_{1,2}$ plus LWC$_2$. Therefore, by expressing concentrations as the amount of dissolved species per unit volume of cloudy air, a statement of mass conservation within the droplet distribution may be expressed as

$$\text{LWC}_1 C_1 = \text{LWC}_{1,2} C_{1,2} + \text{LWC}_2 C_2. \quad (B1)$$

Also, from (7),

$$\text{LWC}_{1,2} = \text{LWC}_1 - \text{LWC}_2. \quad (B2)$$
Now, substituting (B2) into (B1) and solving for \( C_{1,2} \) results in
\[
C_{1,2} = (\text{LWC}_1 C_1 - \text{LWC}_2 C_2)/(\text{LWC}_1 - \text{LWC}_2)^{-1}.
\]  
(B3)

Hence, if sieves CS\(_0\), CS\(_2\), CS\(_4\), CS\(_5\), and CS\(_6\) are used to sample the cloud, then four \( C_{1,2} \) values can be determined.

In a similar fashion, the mass of a trace constituent \( M_{1,2} \) in a given droplet-size interval can be determined as follows:
\[
M_{1,2} = \text{LWC}_1 C_1 - \text{LWC}_2 C_2,
\]  
(B4)

where \( M \) is expressed in mass of dissolved species per unit volume of cloudy air.

APPENDIX C

Fluxes of Liquid Water, Water Vapor, and Snow Crystals onto the Cloud Sieves

The fluxes (g cm\(^{-2}\) s\(^{-1}\)) of liquid water, water vapor, and snow crystals deposited onto the sieves for typical SPL cloud conditions were determined in the following manner.

The flux of liquid water onto the sieves, \( F_r \), is given by,
\[
F_r = \text{LWC} V \rho_m \left[ \sum D^3 n(D) E_c(D) dD \right],
\]  
(C1)

where \( \text{LWC} \) is the cloud liquid water content (g cm\(^{-3}\)), \( V \) is the air speed (cm s\(^{-1}\)), and \( \rho_m \) is the mass-weighted collision efficiency. alternately, (C1) can be written
\[
F_r = \rho \left[ \frac{\pi}{6} \right] \sum \left[ D^3 n(D) E_c(D) dD \right],
\]  
(C2)

where \( \rho \) is the density of water, \( D \) is the diameter of the cloud droplets, and \( E_c \) is the collection efficiency given by (2). The \( n(D) \) function was developed for typical SPL droplet spectra as described by Mitchell (1990) using the form of the Khrgian–Mazin cloud-droplet distribution,
\[
n(D) = AD^v \exp(-BD) dD,
\]  
(C3)

where \( v = 9, B = \Gamma(11) [\Gamma(10) D_{bar}]^{-1} = 10/D_{bar} \) and \( A = \text{LWC} B^{1/2} [(\pi/6) \rho \Gamma(13)]^{-1} = 3.987 \times 10^4 \text{ LWC}/(\rho D_{bar}^{1/2}) \). For typical conditions at SPL \((V = 7 \text{ m s}^{-1}, \text{LWC} = 0.1 \text{ g m}^{-3}, D_{bar} = 7.04 \text{ \mu m}, p = 0.65 \text{ atm}, T = -10^\circ \text{C})\), the values of \( F_r \) were calculated using (C2). The values are listed in Table C1.

The flux of water vapor onto the sieves (cylinders), \( F_d \), is given by Baker et al. (1987) as
\[
F_d = f_v D_g (\rho_w - \rho_i)/R,
\]  
(C4)

where \( f_v \) is the ventilation coefficient, \( D_g \) is the diffusion coefficient for water vapor, \( \rho_w \) and \( \rho_i \) are, respectively, the saturation vapor densities over water and ice surfaces, and \( R \) is the cylinder radius. The ambient vapor pressure is assumed to be at water saturation because the sieves are exposed only when cloud droplets are present. For \( \text{Sc} = 1/3 \text{Re}^{1/2} > 1.4 \) (Sc and Re are defined in the following text), as pertains to the sieves, \( f_v \) for a cylinder may be approximated from \( f_v \) for spheres as
\[
f_v = 0.78 + 0.308 (\text{Sc}^{1/3} \text{Re}^{1/2}),
\]  
(C5)

following Beard and Pruppacher (1971), where \( \text{Sc} = \eta/D_g (\eta = \text{kinematic viscosity of air}) \) and \( \text{Re} = (V 2R)/\eta \). Values of \( F_d \) were calculated using (C4), and the values are listed in Table C1.

The flux of snow crystals onto the sieves (cylinders), \( F_i \), is given by
\[
F_i = \text{IWC} V \rho_s P_s,
\]  
(C6)

where \( \text{IWC} \) is the water content of the cloud ice (g cm\(^{-3}\)} \{i.e., \( \text{IWC} = \sum [m(D) n(D) dD] \} and \( P_s \) is the collision efficiency multiplied by the sticking probability. Using laboratory measurements of \( E_c \) and

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<thead>
<tr>
<th>Table C1. Characteristic fluxes onto cloud sieves at SPL for cloud liquid water ((F_r)), water vapor ((F_d)), and snow crystals ((F_i)).</th>
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</thead>
<tbody>
<tr>
<td>Cylinder diameter (mm)</td>
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<tr>
<td>Flux (g cm(^{-2}) s(^{-1}))</td>
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<tr>
<td>( F_r )</td>
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<td>( F_d )</td>
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<tr>
<td>( F_i ) (exponential spectra)</td>
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<tr>
<td>( F_i ) (subexponential spectra)</td>
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<tr>
<td>( F_i = F_d + F_r )</td>
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<tr>
<td>( F_i = F_r + F_d )</td>
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</table>

Assumed conditions: wind speed = 7 m s\(^{-1}\), LWC = 0.1 g m\(^{-3}\), IWC = 0.1 g m\(^{-3}\), T = -10°C, p = 0.65 atm.
\( P_\text{s} \), Keith and Saunders (1989) determined an event probability, \( EP \), (probability a crystal will rebound after a collision) as a function of \( D \), \( V \), and cylinder diameter. Therefore, the \( E_\text{s}P_\text{s} \) term in (C6) is rewritten as \( E_\text{s} = 1 - EP \). In the IWC expression, the \( m(D) \) term is approximated by \( aD^b \) and the \( n(D) \) term is approximated by \( N_0D^e \exp(-\lambda D) \). Now (C6) can be written as

\[
F_i = V \propto N_0 \sum [D^{(\lambda+e)} \exp(-\lambda D)E_i(D)dD].
\]

(C7)

It was assumed that \( \lambda \) for exponential spectra was 10 cm\(^{-1} \) and 12.3 cm\(^{-1} \) for superexponential spectra [equivalent exponential slope = 10 cm\(^{-1} \) as described by Mitchell (1991)]. Measured values for \( \lambda \) near cloud base, where SPL typically resides, have been found to be generally around 10 cm\(^{-1} \) (Houze et al. 1979; Mitchell 1988; Braham 1990). Values of \( N_0 \) were determined letting IWC = 0.1 g m\(^{-3} \) (from measurements at SPL by Borys et al. 1988), \( \beta = 2, \alpha = 0.0022 \) cgs (Mitchell et al. 1990), and \( v = 1 \) (subexponential spectra) and \( v = 0 \) (exponential spectra) as described by Mitchell (1991). Values of \( F_i \) were determined from (C7) using the exponential and subexponential spectra for cylinders 3 and 5 mm in diameter (diameters used by Keith and Saunders) and the resulting values are listed in Table C1. Mellor (1965) reports mass fluxes for blowing snow much larger than our mass-flux measurements at SPL. Consequently, the IWC and snow-crystal spectra values at SPL are not contaminated significantly by blowing snow.

It can be seen in Table C1 that the amount of water vapor and snow crystals deposited onto the sieves is a small fraction of the amount of cloud liquid water typically deposited for \( CS_1 \) through \( CS_4 \). But the amount of water vapor and snow crystals deposited becomes significant for \( CS_5 \), \( CS_6 \), and dominates for \( CS_7 \).


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