The Microphysics of the Clouds of Venus: Results of the Pioneer Venus Particle Size Spectrometer Experiment

R. G. KNOLLENBERG

Particle Measuring Systems, Inc., Boulder, Colorado 80301

D. M. HUNTEN

University of Arizona, Tucson, Arizona 85724

The results of the particle size spectrometer experiment on the Pioneer Venus sounder probe are presented. The vertical cloud structure is found to consist of three primary cloud regions of approximately 20 km total thickness suspended within an ubiquitous aerosol haze which extends more than 10 km above and below it. The three cloud regions are separated by sharp transition regions where both particle chemistry and microphysics exhibit change. The size distributions are multimodal in all cloud regions. Three size modes are observed in the middle and lower cloud region which are composed of aerosol, H_2SO_4 droplets, and crystals. The crystals likely could be either sulphates or chlorides. We provide interpretations of the sources, growth characteristics, and fate of the particle species through a partitioning analysis of the LCPS size distribution data.

1. INTRODUCTION

The cloud particle size spectrometer (hereafter LCPS) was a primary experiment aboard the sounder probe during the Pioneer Venus (PV) mission. This single particle device individually sized particles traversing its illuminated field during descent from 66 km to the planet's surface. Covering a size range of 0.5-500 μ m, it detailed the cloud microstructure from medium-sized aerosol nuclei to small precipitation elements. The relatively higher number density of small aerosol particles observed proved sufficient in population to realize the LCPS's maximum vertical resolution of 100 m in the denser cloud regions. Preliminary results of this experiment have been reported by *Knollenberg and Hunten* [1979a, b].

The LCPS data and the nephelometer data on Venera 9 [Marov et al., 1979] and PV [Ragent and Blamont, 1979; Blamont and Ragent, 1979] all show a highly structured planetary cloud system. The main cloud system shows planetary transition levels at 56 and 50 km, where particle populations are grossly depleted, dividing the cloud system into three distinct regions, which we have named the upper, middle, and lower cloud regions. Each region has different microphysical and probably compositional properties. Above the cloud tops, the cloud photopolarimeter/imager experiment has defined an upper region of haze extending up to 90 km [Travis et al., 1979]. In addition, an extensive thin haze, detectable only by the LCPS, was found below the base of the cloud system (46 km) extending down to 32 km. Table 1 summarizes the general cloud system properties.

The cloud system microphysical properties from 66 km to the surface are best defined by the LCPS. Even though this comprehensive data set exists only at the sounder probe entry site, the low apparent volatility of the particles, together with the rapid circulation at cloud levels, could create sufficient microphysical homogeneity to lead one to conclude that the results are characteristic of the clouds most everywhere on the planet, i.e., we might be dealing with a planetary cloud system microphysically as well as morphologically (revealed by Venera and PV nephelometers).

Clearly, the most remarkable result of the LCPS measure-

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ments was the unexpected multimodal character of the size distributions at essentially all cloud levels. While multimodal distributions are quite common for higher moments, and mass distributions in particular, it is unusual to observe multimodal size distributions. At a minimum it is usually necessary to invoke differing growth conditions or compositional changes to achieve distinct multimodality. In fact, coexistence of two modes having the same composition requires peculiar equilibrium conditions (generally supersaturated). The multimodal observations at the upper cloud levels stand in stark contrast to the populations anticipated from earth-based work, which had predicted only a single very narrow distribution of H_2SO_4 with a modal radius of 1.05 μ m and a standard deviation of 0.26 μ m [Hansen and Hovenier, 1974].

This paper is a comprehensive summary of all of the LCPS data with more detailed analysis and expanded interpretation. In addition to the presentation of the entire data set and its scientific interpretation, we have provided a description of the operational limits of the LCPS performance which clarifies the statistical nature of the various populations, places limits on the number density of large particles possible where none was measured, and bounds the unseen aerosol below the lower limit of minimum detectable size (~0.5 μ m). A detailed instrument description of the LCPS is found in Knollenberg and Gilland [1980]; the reader is referred to that article for design, construction, and operational details. Here we only treat those aspects of the experiment that affect data interpretation, which immediately follows. The third section provides an overview of the vertical cloud structure as determined by the LCPS. The fourth section details the size distributions. The fifth section provides arguments favoring the existence of crystalline particles. Section 6 details the cloud structure in terms of modal partitions. Conceptual cloud particle composition and life cycles are presented in section 7, followed by our general conclusions.

2. LCPS SIZING, SAMPLING, AND PERFORMANCE CHARACTERISTICS

The LCPS was conceived as an in situ sampling instrument. An in situ sampling instrument has the advantage of allowing

Region	Altitude, km	Temperature, °K	Optical Depth $ au$	Average Number Density N, cm ^{-3*}	Mean Diameter, µm	Composition [†]
Upper haze‡	70–90	225°-190°	0.2-1.0	500	0.4	H_2SO_4 + contaminants
Upper cloud	56.5–70	286°-225°	6.0-8.0	(1) 1500 (2) 50	Bimodal 0.4 and 2.0	H_2SO_4 + contaminants
Middle cloud	50.5–56.5	345°–286°	8–10	(1) 300 (2) 50 (3) 10	Trimodal 0.3, 2.5, 7.0	H_2SO_4 + crystals
Lower cloud	47.5–50.5	367°–345°	6–12	(1) 1200 (2) 50 (3) 50	Trimodal 0.4, 2.0, 8.0	H ₂ SO ₄ + crystals
Lower haze¶	31-47.5	482°-367°	0.1-0.2	(2) 20	0.2	H_2SO_4 + contaminants
Precloud layers¶	46 and 47.5	378° and 367°	0.05 and 0.1	50 and 150	Bimodal 0.3 and 2.0	H_2SO_4 + contaminants

TABLE	1.	Summary	of	Venus	Cloud	Рго	perties
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*Mode 1 number density as determined by log normal fit to data in section 4; modes designated by parentheses.

†See sections 5, 6, and 7 for interpretations of composition

‡From Travis et al. [1979] and Kawabata et al. [this issue].

Observed_only at Sounder site.

a size-dependent sample volume (increasing with size) that is generally necessary to provide adequate samples of the less frequent larger particles. The LCPS design was modeled after devices used on aircraft by terrestrial cloud physicists [Knollenberg, 1976a]. Considering that earth clouds typically have concentrations of hundreds per cubic centimeter of 10 μ m size cloud droplets and a few per liter of millimeter size precipitation elements, a dynamic range of 2 orders of magnitude in size must necessarily be accompanied by 4-5 orders of magnitude growth in sample volume to provide equal count statistics. Cloud droplet spectra typically develop around a central mode because of competitive diffusional growth (see cloud physics texts, e.g., Pruppacher and Platt [1978]). In terrestrial clouds 10-20 μ m is the modal (highest frequency or most probable) diameter found in droplet growth regions. Values of $2 \,\mu m$ were expected for the cloud tops of Venus [Hansen and Hovenier, 1974]. Because the size mode shifts over some considerable range during growth and decay of a cloud while the number density remains relatively constant, there is generally no justification for anything but a fixed sample volume covering the cloud droplet spectrum. On the other hand, the growth of larger particles and, in particular, precipitation elements, generally requires increasing sample volumes by appropriately designed instruments. Since fixed sample volumes are characteristic of devices employing light scattering while

TABLE 2. LCPS Size Range Calibration

Size Channel				
Number	Range 1	Range 2	Range 3	Range 4
	Na	minal Range,	μm	
	0.5-5.0	5-50	20-200	50-500
	Actual	Calibrated Ra	nge, µm	
	0.6-5.0	5-53	16-181	35-490
Ca	librated Size	Intervals in M	licrons Diame	ler
1	1.6-2.1	5–7	16-26	35-66
2	2.1-2.8	7–9	26-36	66-106
3	2.8-3.9	9-12	36-48	106-147
4	3.9-5.0	10-16	48-62	147188
5	0.5-1.1	16-21	62–78	188-230
6	1.1-2.1	20-26	78–96	230-280
7	2.1-3.6	26-32	96-116	280332
8	3.6-5.0	32-39	116-137	332-385
9		39-46	137-159	385-435
10		46-53	159181	435490

size-dependent sample volumes are products of those employing imaging methods, both types of instruments are used in terrestrial cloud physics. Forward scattering particle size spectrometers covering 2-30 μ m and 3-45 μ m are most widely used for cloud droplet work, while imaging spectrometers utilizing size ranges of 20-300, 300-4500, 25-800, and 200-6000 μ m cover the region of cloud growth to precipitation sizes.

Thus the LCPS, being modeled after terrestrial instrument counterparts, similarly uses both light scattering and imaging approaches to cover the total dynamic range. The primary difference is that the cloud droplet mode on Venus was known to be somewhat smaller, and thus the light scattering range was set up for $0.5-5 \mu m$, with the imaging size range likewise extending to smaller sizes and covering $5-500 \mu m$. Because of the greater contribution of larger particles to important higher distribution moments, for example, D^2 (optical cross section) and D^3 (mass), imaging was considered the primary measuring technique and light scattering secondary, even though the results would hardly support either as secondary.

In the primary imaging measurement, the shadows of particles are imaged onto three linear photodiode arrays, using a tri-split laser beam and three tailored optical trains. The shadow image is sized by determining the number of array elements occulted during transit. This type of instrument is known as an optical array spectrometer (OAS). (This optical array spectrometer is of the one-dimensional type since only the dimension colinear with the array is measured. Two dimensional variations which can provide fully two-dimensional images are now more widely used. The latter was not a feasible alternative for PV because of telemetry bandwidth limits, although particle shape would have been extremely useful. To supplement that need, bulk particle aspect ratio could be measured in the 50–500 μ m range, using comparisons of shadow transit time with size. Unfortunately, no particles this large were observed.) The arrays each have 12 elements which produce 10 size classes covering size ranges of 5-50, 20-200, and 50-500 μ m (hereafter ranges 2, 3, and 4), using three appropriate magnifications. All three ranges are multiplexed and switched every $\frac{1}{4}$ s with one half the sample time on range 2 and one fourth on each of ranges 3 and 4. The data readout is also time shared, producing a complete sample of all three ranges every 8 s.

The light scattering technique employs the same beam as

range 2 but with a displaced object plane to establish a separate sample volume (a single objective window and prism are common to all four size ranges). Particles viewed by the light scattering technique are sized into four classes by a pulse height analyzer. The class limits are defined by a voltage divider which has high sensitivity and low sensitivity modes of operation and is alternately switched every 4 s. This produces two sets of four classes, hereafter denoted range 1.

A summary of the size calibration and sampling characteristics of each range is given in Table 2 with curves of the sample area (sample volume is a product of the sample area and descent velocity) for all size classes in each range in Figure 1. The shape of the curves for ranges 2, 3, and 4 results from a depth-of-field (sample area length) increasing as D^2 , while the sample area width decreases linearly with D. The depth-of-field also truncates because of mechanical aperture limits on ranges 3 and 4. For a more detailed discussion of sampling characteristics, see Knollenberg and Gilland [1980].

The optical array spectrometer technique is essentially insensitive to particle refractive index and measures the dimension projected colinear to the array, thus reducing systematic sizing errors when nonspherical shapes are involved. However, light scattering techniques must employ strong forward scattering to avoid systematic sizing errors when particles of complex morphology or varying index (whether real or imaginary) are involved. The theoretical response results in a po-



Fig. 1. LCPS sample area. The sample area for range 1 is a constant equal to 0.25 mm^2 . The OAS ranges 2, 3, and 4 all show a strong size dependency which results from a depth-of-field increasing as the square of the particle diameter.



Fig. 2. Theoretical Mie scattering for $2^{\circ}-11^{\circ}$ absorbing particles with 1.4 real index. Instruments using strong forward scatter have the least total sensitivity to particle shape and composition. However, the computed scattering response is ambiguous in the case of transparent spheres in certain size ranges owing to chance constructive or destructive interference of refracted and diffracted light. When the refractive index can be bounded reasonably well, a response relationship can be optimized and ambiguous regions avoided. This is the case of mode 2 with regard to the LCPS data. In this figure we show the further result of small amounts of absorption which tend to reduce the error. At large imaginary indices, this smoothing results in an ideal response relationship.

tentially ambiguous size calibration for transparent spheres as shown in Knollenberg and Gilland [1980, Figure 4]. For a narrow range of refractive indices from 1.35 to 1.45 (which covers the full range of H_2SO_4 concentrations compatible with the Venusian clouds), the oscillating character of the response only limits absolute accuracy to approximately $\pm 0.3 \ \mu m$, which is less than the resolution set by the size class limits in Table 2. Figure 2 illustrates the further reduction in ambiguity when small amounts of absorption are present. A second instrumental effect is the finite signal-to-noise ratio of the lowlevel light scattering pulses which generate approximately a full size class spread for even monodispersed particles. However, the systematic errors in sizing over a large sample are small except when the mode is narrow and in an ambiguous region (for instance, from 0.8 to 1.6 μ m in Figure 2). Furthermore, essentially errorless computations of certain particle optical properties (e.g., extinction coefficient) can be made at all visible wavelengths regardless of the theoretical response oscillations (G. W. Grams and R. G. Knollenberg, manuscript in preparation).

The LCPS instrument was turned on and functioning at an altitude of 66 km. The first data readout occurred at 1849:18 ground receipt time (GRT), which was only partially recoverable and included bit errors and memory turn-on background counts. However, the smallest sizes stored in range 1 appeared reasonable in light of continuing data and were retained. From 1849:23 and every 8 s thereafter, all data were recovered until sounder probe impact. The lack of any probe/telemetry bit errors is easily confirmed by the fact that range 4 had no

			TABLE	3. Total	Numbers	of Counte	d Particles	s in Eacl	h Size (Channel	Measu	red in E	ach Hei	ight Re	gion				
																	Numb That C Witl	er of Size S an Be Supj 1 90% Statis	pectra borted ttics
				Rang	e l							Range	0			Range 3	Mode I	Mode 2	Mode 3
Size channel	_	2	س	4	<u>ہ</u>	0	1	∞	-	5	س	4	0	1	∞				
Upper cloud region	499	1116	664	36	14494	5252	3350	284	0	0	1	0	0	0	0	0	95	30	0
Middle cloud region	123	557	1115	8	2352	849	2070	220	18	39	37	11		1	0	0	20	22	ę
Lower cloud region	102	320	499	181	3220	895	1025	255	0	76	85 4	17 19	/ 16	4	2	Ē	22	15	7
Precloud layers	34	69	55	0	244	165	28	-	0	0	0	0	0	•	0	0	7	7	0
Lower thin haze	7	0	0	0	311	14	0	0	0	0	0	0	0	•	0	0	Ē	0	0
Total	760	2062	2333	313	20621	7473	6473	760	0	0	0	0	•	0	0	0	142	62	Ś
Channels 9 and 10 of r *A 90% statistical confi	ange 2, 2-16 dence requi	0 of rang	ce 3, and a oximately	ll channe 100 parti	ls of range cles since t	4 were ze: he uncerta	ro. intvis≊	, r											

counts observed in any of its 10 size classes; size ranges 2, 3, and 4 share the same processing electronic circuitry. This also discounts any noise sources or stray optical effects as generating potential false counts.

The LCPS data from each 8-s period represent (approximately) a sampled volume of 1-5 cm³ (varying directly with descent velocity) for particles in range 1 and 1500-3400 cm³ for the particles of 200 µm diameter in range 4 that had the maximum sample volume. The total volume sampled during descent for this 200 µm size was 2.4 m³. The total number of particles measured in each size class is shown in Table 3. No particles were measured with diameters greater than 35 μ m. Figure 3 shows the maximum number density that could exist undetected above 35 µm with varying levels of statistical confidence. Because of the finite limits in LCPS sample volume and size sensitivity, functional fits to the raw data are required. Data extensions to these regions of undetected particles, however, must be consistent with the sample probability depicted in Figure 3.

3. VERTICAL STRUCTURE OF THE CLOUDS OF VENUS: AN OVERVIEW

The vertical structure of the Venus cloud system observed by the LCPS was first reported in Knollenberg and Hunten [1979]. At that time, data gaps existed above 62.5 km and from 56.2 to 58 km. The complete data set is presented in Figure 4 in the same form as in Figure 1 of that earlier work, with all adjustments deemed appropriate. The overall form and signatures of the number density, extinction coefficient, and mass loading are largely unchanged. The greatest change was in the magnitude of the mass loading and the extinction coefficient, which are rather sensitive even to the small adjustments in size class widths made in the interim. The three cloud regions and lower haze defined in Knollenberg and Hunten [1979a] are even more evident, particularly that region between the upper and middle cloud regions which was obscured because of missing data. The integrations of extinction coefficient and mass loading are still based on the spherical particle assumption. An alternate interpretation is given in section 5 and is



Fig. 3. Maximum number density for undetectable particles larger than 35 µm. Values in parenthesis indicate the percentage confidence that no particles exist for the indicated size and number density.



Fig. 4. Vertical structure of Venus cloud system. The above data represent direct computations from the LCPS data. No allowance has been made for particles smaller than the 0.6 μ m lower limit of size sensitivity or above the largest sizes actually measured. A density of 2 g cm⁻³ was assumed for mass loading computations. The thickness of T_{um} is about 1 km while T_{ml} is several hundred meters thick.

based on the assumption that the large particles are asymmetric crystals.

The upper cloud region is populated by a mixture of aerosol and H₂SO₄ droplets creating a bimodal size distribution consisting of what we have called mode 1 and mode 2 particles [Knollenberg and Hunten, 1979b]. The average size distribution for this region is shown in Figure 5. We identified H_2SO_4 as the larger mode 2 particles by matching the narrow size distribution properties of mode 2 in the middle cloud region (where mode 2 is clearly separated from mode 1) with previous earth-based data. Mode 2 could then be traced back up into the upper cloud region, where the two populations tend to merge. Separating the two modes in regions where they were completely merged required making the assumption that the spectral variance was the same as in adjacent regions where the modes separate. The refractive index (1.44) computed by Ragent and Blamont [1979] is also consistent with 90% H₂SO₄. There are distinct altitudes in the upper cloud region where the bimodality is unmistakable; however, there are sufficient numbers of the smaller aerosol everywhere to mask the narrow size distribution properties of the mode 2 H_2SO_4 , especially if observed from earth.

The identity of the aerosol particles cannot be specified absolutely, although in section 7 certain constraints on the identity are offered. Certainly the LCPS only sees the large tail of this underlying aerosol population; the limitations imposed on the 0.6 μ m cutoff are dealt with in section 4. The upper cloud region is shown to have maximum number density, extinction coefficient, and mass loading in the vicinity of 60 km, decreasing above and more sharply below. A transition region is reached at 56.5 km between the middle and upper cloud regions where there is a sharp decrease in particle activity observed on the nephelometers as well [Blamont and Ragent, 1979]. Within this transition region, particle properties change from those characteristic of the upper cloud region to those characteristic of the middle cloud region. This transi-



Fig. 5. Average upper cloud region size distribution. The bimodality in this average size spectrum is not apparent; however, at certain altitudes it is obvious which has enabled us to separate modes 1 and 2 in all altitudes.



Fig. 6. Average middle cloud region size distribution. Trimodality is apparent in this average size distribution. The mass distribution was computed, using a spherical particle assumption and a density of 2 g cm⁻³.

tion region is approximately 1 km in thickness and is designated T_{um} .

The middle cloud region is characterized by three observed size distribution changes: (1) The mode 1 aerosol population decreases in number density by nearly an order of magnitude; (2) increased growth in the mode 2 aerosol provides clear separation between mode 1 and mode 2; and (3) larger particles appear, resulting in a trimodal size distribution. The average size distribution for the middle cloud region is shown in Figure 6.

The middle cloud region shows that the total number density gradually increases with decreasing altitude, while the extinction coefficient and mass loading remain essentially constant until reaching the 50.5-km region where there is a greatly reduced particle activity, defining a second transition region between the middle and lower cloud regions, designated $T_{\rm ml}$. The extinction coefficient and mass loading are higher owing to the presence of mode 3 particles, while the total number density is lower when compared to the upper cloud region.

Below $T_{\rm mb}$ the numbers of mode 1 and 3 particles both increase by roughly a factor of 5, while mode 2 is largely unchanged. However, the trimodal character of the average size distribution shown in Figure 7 is still very much evident. The lower cloud region contains roughly the same particle number densities as the upper cloud region but much higher extinction coefficients and mass loadings owing to the great increase in large mode 3 particles.

Below the lower boundary of the lower cloud region there is a gradual decrease in aerosol particles down to 31 km, below which essentially no particles were observed. Imbedded within this aerosol population (shown in Figure 8) are two sharp cloud filaments at 46 and 47.5 km, previously designated as precloud layers. The size distributions for these two layers are shown for the first time in Figures 9a and 9b. It is immediately evident that these distributions are also bimodal. Those in the upper precloud layer resemble modes 1 and 2 in the lower cloud region, while those in the lower precloud layer closely approximate modes 1 and 2 of the middle cloud region.

The lower thin haze extends from 46 to 31 km. The lack of any discernible particle activity below 31 km is rather remarkable, although in the last few kilometers nearer the surface the confidence in zero particle activity must be acknowledged as less than 100%, owing to the presence of noise bursts which, though easily discountable as spurious, could mask some real particle events. However, the existence of particles larger than $2 \mu m$ is denied in spite of the noise, and we favor the idea that the atmosphere is remarkably clear of particles larger than 0.6 μm all the way to the surface.

4. SIZE DISTRIBUTIONS

As we previously emphasized, the multimodal size distributions represent a most unique feature of the Venus cloud system. One might ask whether or not this is a singular chance happening at the sounder probe entry site. The answer is apparently no. Clearly, the haze observed by the PV cloud photopolarimeter (OCPP) as reported by Travis et al. [1979] is widely mixed into the upper cloud region, resulting in a bimodal distribution similar to that observed by the LCPS at cloud top. Also, the most recent analysis of the Venera 9 nephelometer results reported by Marov et al. [1979] is only consistent with multimodal size distributions at several altitudes. In fact, they proposed that it is necessary to invoke multimodality just below T_{um} to explain the multiangle nephelometer scattering signals. (It should be emphasized that Marov defines two transition regions (I' and II') at exactly the same locations as our $T_{\mu m}$ and T_{ml} .) It is also noteworthy that in the upper cloud region their size distribution is broader than either our mode 2 H₂SO₄ distribution or that inferred by Hansen and Hovenier [1974]. It is doubtful that Marov's data and analyses are sufficient to separate modes 1 and 2. It is more likely that their results are only sensitive to the addition of mode 3 which is first observed below T_{um} by the LCPS. There are a number of other properties deduced from the Venera 9 nephelometer data that are similar to our PV results and are discussed in Knollenberg et al. [1980]. Suffice to say





Fig. 7. Average lower cloud region size distribution. The trimodality observed in Figure 6 is not nearly so apparent here, although it is still evident at several altitudes. The mass distribution was computed, using a spherical particle assumption and a density of 2 g $\rm cm^{-3}$.

Fig. 8. Average lower haze region size distribution. There is essentially no evidence for bimodality in the lower haze region.



Fig. 9a. Upper precloud layer size distribution.

there exists considerable evidence to suggest multimodality is the rule rather than the exception on Venus. The LCPS data provide the first high-resolution detailed look at these unusual particle populations.

We have found the separation of the three modes relatively uncomplicated at most altitudes. Mode 3 resides almost entirely in range 2 sizes, while modes 1 and 2 are restricted to range 1. Within range 1, modes 1 and 2 are nearly completely resolved in the raw LCPS size distribution data at all altitudes in the middle cloud region as well as at discrete altitudes scattered throughout the cloud system. At those altitudes where separation between modes 1 and 2 could not be accomplished by inspection, mode 2 was first extracted by assuming its spectral variance was identical to that observed at the closest altitude where modes 1 and 2 were already resolved. The extreme narrowness of mode 2 made what might seem to be an extremely difficult task relatively simple. In fact, had we not lost the resolution between 1 and 2 μ m, because of inherent instrumental ambiguity (see Figure 2), modes 1 and 2 would have appeared strongly bimodal at almost all altitudes. Even so,





Fig. 9. The upper precloud layer size distribution fits closely with that shown in Figure 7 for modes 1 and 2, and the lower precloud haze distribution closely resembles that in the middle cloud region, Figure 6.

		Log Normal Distribution		Gaussian Distribution			Log Normal Distribution			
		M	ode 1		Mo	de 2		Mo	ode 3	
	Altitude Range	NT, ncm ⁻³	σ _g	$\overline{d}_{g}, \mu \mathrm{m}$	$\overline{N_{T}, \text{ncm}^{-3}}$	σ, μm	<i>д</i> , µm	N_T , ncm ⁻³	σg	d̄ _g , μm
Upper cloud	66.10-65.20	181	2.16	0.35	9	0.50	1.33			
	65.20-63.80	211	2.16	0.35	22	0.68	2.38			
	63.80-62.50	336	2.16	0.35	41	0.68	2.20			
	62.50-61.25	521	2.16	0.35	53	0.80	2.20			
	61.25-60.20	544	2.16	0.35	64	0.69	2.32			
	60.20-59.00	608	2.16	0.35	71	0.84	2.28			
	59.00-58.15	348	2.16	0.35	63	0.91	2.26			
	58.15-57.10	197	2.16	0.35	58	0.87	2.12			
Tum	57.10-56.20	54	2.16	0.35	29	0.82	2.38			
Middle cloud	56.20-55.40	69	2.16	0.35	30	0.74	2.54	17	1.25	7.40
	55.40-54.80	69	1.9	0.30	31	0.44	2.83	30	1.25	7.40
	54.80-54.30	69	1.9	0.30	30	0.49	2.73	37	1.25	7.40
	54.30-53.90	86	1.9	0.30	32	0.48	2.80	32	1.25	7.40
	53.90-53.25	72	1.9	0.30	38	0.55	2.70	50	1.26	7.28
	53.25-52.80	99	1.9	0.30	37	0.55	2.80	38	1.26	7.28
	52.80-51.30	112	1.9	0.30	44	0.65	2.73	35	1.26	7.28
	51.30-52.70	122	1.9	0.30	54	0.59	2.73	48	1.26	7.28
	52.70-51.10	110	1.9	0.30	56	0.63	2.70	40	1.27	7.41
	51.10-50.60	121	1.9	0.30	52	0.68	2.70	39	1.27	7.41
	50.60-50.10	93	1.9	0.30	46	0.63	2.45	14	1.27	7.41
T_{ml}	50.10-49.70	144	1.80	0.35	14	0.60	1.70	15	1.27	7.41
Lower cloud	49.70-49.25	528	1.80	0.4	57	0.75	2.58	126	1.29	7.20
	49.25-48.75	563	1.80	0.4	71	0.60	2.73	177	1.29	7.20
	48.75-48.30	404	1.80	0.4	75	0.55	2.60	72	1.29	7.20
	48.30-47.75	233	1.80	0.4	59	0.66	2.14	33	1.29	7.20
Upper precloud	47.75-47.50	474	2.02	0.35	111	1.25	2.10			
Lower haze	47.50-46.45	128	1.70	0.28						
Lower precloud	46.45-46.30	179	1.80	0.30	34	0.85	2.90			
Lower haze	46.30-43.25	218	1.57	0.25						
	43.25-38.70	41	1.57	0.25						
	38.70-31.00	46	1.57	0.25						

TABLE 4. Mode 1, 2, and 3 Size Distribution Parameters for LCPS Data

still further separation of modes 1 and 2 results if one removes the spectral broadening introduced by instrumental effects, as shown in *Knollenberg and Hunten* [1979b].

Before one can begin to compare variations in size spectral properties within resolved modes, it is desirable to ensure that the populations are in fact on firm statistical ground. This, of course, depends upon which parameters one desires to examine. However, one must be aware of the following fact. These size distributions generally provide the highest raw counts per size class at the smallest sizes. This means that we can reduce the statistical noise in the size distributions for modes 1 and 2 much more readily than for mode 3. Table 3 shows the total raw counts per size class in each of the regions. Also shown is the number of individual size distributions that can be justifiably separated if 90% confidence is desired in the statistical stability of each population. Modes 1 and 2 are sampled in sufficient number to give individual distributions at each 8-s readout period throughout the cloud regions at vertical resolutions ranging from 100 to 400 m. Mode 3 is sampled in sufficient number to give only two distributions in the middle cloud region and three in the lower cloud region. Each precloud layer only can be resolved as a single distribution, and up to three individual spectra can be resolved in the lower haze. Because there are more than 200 individual spectra, only selected spectra can be presented here. We have selected analytical functions as an appropriate method of further compressing and presenting data at 31 altitude intervals from 66 to 32 km for all three modes (which are presented in Table 4). Several adjustments to the analytical functions have been made since those reported in Knollenberg and Hunten [1979b]. There, the narrow mode 2 H_2SO_4 was fit with Gaussian, the mode 1 aerosol with power-law, and the mode 3 with log normal size distribution functions. In this paper, log normal fits were derived for both modes 1 and 3.

The log normal distribution function is typically used to describe aerosol populations and was selected for mode 1 to limit the distribution at some reasonable size below the 0.6-µm lower limit of the LCPS and provide an estimate of the total population. The choices of possible distribution parameters were constrained by using coagulation rate computations and assumptions about particle lifetime. These are of course only first-order refinements with an admitted large uncertainty; however, it would be far worse to simply omit treatment of this subpopulation. We proceeded by first fitting the LCPS data in range 1 (after extraction of mode 2) with log normal distributions of varying total number densities (N_T) , geometric means (\vec{D}_{e}) , and standard deviations (σ_{e}) as shown in Figure 10 for the upper cloud region. The LCPS data really provide only a single size class that is entirely mode 1. The amount of mode 1 within size class 2 is taken as the residual after extraction of mode 2. Fits to these size classes are shown in Figure 10 for modal diameters (D_{ml}) ranging from 0.03 to 0.5 and a range of geometric means (\bar{D}_s) from 0.07 to 0.81 µm. The total number density for mode 1 is also shown for each value of $D_{\rm ml}$. Assuming only thermal (Brownian) coagulation as active, the distribution was followed in N_T , \vec{D}_{e} , and σ_s over time periods of 4, 2, and 0.5 months, the assumed residence times of mode 1 particles in the upper, middle, and lower cloud haze regions. The results indicate modal diameters between 0.1 and 0.2 μ m for all regions within time periods of 1-2 weeks, in all cases,

4).



Figures 11 and 12 illustrate fits of the analytical functions to the LCPS at 54.2 and 49.0 km. Use of the analytical functions duplicates the extinction coefficients and number densities in the range of LCPS observations to within 10% and total mass to within 15%. However, the log normal distribution cannot be made to fit at sizes larger than 20 μ m, which accounts for the mass error. The extension of the size distributions to sizes larger than 20 µm and especially to sizes where individual counts were not recorded (>35 μ m) must be made with a power-law fit if reasonable accuracy is desired.

The mode 2 modal diameter increases slightly when proceeding from higher to lower levels. It provides important clues to growth processes when compared to the number density, which we will defer to section 6. However, the following trends are observed in the full data set (Table 4 only provides representative spectra). In the upper cloud region, very little change in D_{m2} is observed until the instruments descend through T_{um} where D_{m2} increases from 2.1 to 2.7 μ m and fluctuates between 2.7 and 2.9 µm for several kilometers until gradually decreasing to below 2 μ m at T_{ml} . Below T_{ml} it again increases to about 2.2 μ m before decreasing to about 1.5 μ m at the lower cloud base and 2.1 μ m in the upper precloud layer. A somewhat larger value of $D_{m2} = 2.9 \ \mu m$ is computed for the lower precloud layer.

5. EVIDENCE FOR CRYSTALLINE PARTICLES

It has previously been shown by Blamont and Ragent [1979] and Marov et al. [1979] that the refractive index lowers as one moves from T_{um} down through the middle and lower cloud regions. Both workers find that an unreasonably low value of n \approx 1.33 is found in the most dense region around 49–52 km. Values of 1.42-1.46 are required if H₂SO₄ exists in any concentrations. Blamont's computations assume conservative scattering by spherical particles consistent with H₂SO₄ droplets. The index is computed from the LCPS size distributions, using estimates of indices to generate the observed backscatter. Our own computations confirm the lower index for conservative scattering; however, reasonable indices (\sim 1.40) can be achieved with small amounts of absorption (imaginary indices of 2×10^{-3} to 3×10^{-3}) as shown in Figure 13. Marov suggested similar fits with imaginary index values of 5×10^{-3} . In general, as can be seen from Figure 13, the backscatter per unit cross section would decrease linearly with size, and the higher value of imaginary index found by Marov is consistent with his smaller mean size. However, while these imaginary indices are not large, they reduce the single scattering albedo sufficiently to affect markedly net solar flux profiles. Since Tomasko et al. [1979] find no noticeable absorption in the middle and lower cloud region, this solution to the index discrepancy is shaky.

Another possible solution to the problem requires the introduction of nonspherical particles. Such particles can produce more or less forward scattering than spheres of equal index, depending on the choice of crystalline shape or habit. The differences are greatest when particles of high aspect ratio are involved. Nephelometry would in this case indicate lower indices than for spheres. It turns out that the LCPS data support the nonspherical particle hypothesis for the large mode 3 particles. While the circuitry specifically designed to measure aspect ratio was limited to range 4, wherein no particles whatever were measured, a comparison of the data in the first



NŢ

n (D/Ďg)

10

using log normal functions. In deriving these log normal functions we ran a series of coagulation computations which not only verified the choice of the distributions function but limited N_T and bounded \bar{D} . The approximate time required for N_T to decrease to values associated with each distribution is indicated. Because of the added coagulation constraint, our N_T estimates for mode 1 are lower in all regions than first estimated. In spite of the large increase in total number of particles within mode 1 with variations in D_{ml} from 0.1 to 0.5, the effect on the extinction coefficient and mass loading is quite small.

with total number densities between 200 and 1300 cm⁻³. The results are not very sensitive to the initial input distribution because coagulation is very rapid at high number densities. The best fits determined for all cloud regions are given by the distribution parameters shown in Table 4. There is little impact to either extinction cross section (and related optical properties) or mass from the choice in distribution parameters. The extinction coefficient varies only 20% at 600 nm and 40% at 300 nm for the extremes in the distributions shown in Figure 10.

In the upper cloud region a value of $\bar{D}_g = 0.35 \ \mu m$ was adopted as shown in Figure 10. Kawabata et al. [1980] find that an effective radius (diameter) of $0.24 \pm .07 \,\mu\text{m}$ (0.48 ± .14 μ m) with a standard deviation of 0.08 (0.16) and a refractive index of $1.45 \pm .05$ fits the polarizations in the regions poleward of 50°N. Our smaller value of D_e is consistent with the OCPP values in that \bar{D}_{e} is about 0.1 μ m less than the effective diameter for the polarization analyses. In the middle cloud regions we found a lower value of $\bar{D}_s = 0.3 \,\mu\text{m}$, which is consistent with its lower number density here. In the lower cloud region, a higher value of $\bar{D}_g = 0.4 \ \mu m$ was obtained. In the precloud layers, values of $\bar{D}_{g} = 0.35$ and $\bar{D}_{g} = 0.3 \ \mu m$ were adopted for the upper and lower layers, respectively (as previously indicated, these distributions tend to mimic those in the lower and middle cloud regions, and \bar{D}_{g} was assumed to be the same while σ_s selected to provide best fit). The lowest



Fig. 11. Size distributions for modes 1, 2, and 3 at 54.2 km. Note the poor fit at the largest sizes in mode 3.

channel of range 3 with range 2 indicates that nonspherical high-aspect ratio particles are required to explain the measurements. The problem here can be stated quite easily. In range 2 we observed approximately 72 particles larger than 16 μ m, between 56 and 48 km ranging in size from 16 to 36 μ m, while in range 3 only three particles were found in the first

size class (which is 16-35 μ m). Because only three particles were observed, the data are obviously of no value as a statistical particle population, and no attempt has been made to use these data thus far. However, there should have been a greater number of counts in range 3 than for range 2 in this common size overlap region, since range 3 has more sample area than



Fig. 12. Size distributions for modes 1, 2, and 3 at 49.0 km. Note the poor fit at the largest sizes in mode 3.



Fig. 13. Sensitivity of nephelometer backscatter to real and imaginary index variations. As shown, one could account for most of the index discrepancy $(1.44 - 1.33 \approx 0.1)$ by adding absorbing material with a range of imaginary indices of 10^{-3} to 5×10^{-3} for mode 3 particles (larger than 5 μ). However, at a 10- μ m size, a value of $n_{im} = 10^{-3}$ corresponds to a single scattering albedo of 0.98 which is too low to be compatible with Tomasko's [1979] results.

range 2 even after adjusting for the factor of two difference in duty cycles. One has to be very careful, because there is a tendency for a particle to be slightly oversized at the ends of the depth of field, and some of the largest particles in range 2 actually may be 10–20% smaller than was indicated. However, even if we compare sizes larger than 21 μ m in range 2, we find 30 particles sampled. Since range 3 samples for one half the time of range 2, the actual ratio of raw counts of range 2 to range 3 in the overlap region is $30/3 \times 2$, or 5. In fact, from sample area considerations, the ratio should be 50% more in range 3 than in range 2. We are therefore left with the result that these ranges respond differently to the same particles. The possibility that this finding could have resulted simply from unlucky statistical count fluctuations is much less than 1%.

The question of response of optical array spectrometer instruments to nonspherical particles has received considerable attention in terrestrial instrument counterparts, and the problem just outlined has been treated by *Knollenberg* [1975, 1976b]. Discrepancies are commonly observed when paired instruments with different resolutions sample snow crystals; there is generally acceptable to good agreement when only liquid drops are encountered. Asymmetrical particles generally result in sizing roundoff errors toward smaller size when the resolution is coarse. In the LCPS the resolution in range 2 and range 3 is 5 and 20 μ m, respectively, although some nonlinearity is noted in Table 2 at the smaller sizes.

The behavior of the LCPS to spherical and nonspherical particles of high-aspect ratio is illustrated in Figure 14a. For an individual element to register a shadowing event, 62.5% of its area must be covered by a particle's dark shadow. Thus a spherical particle of 16 μ m, if properly registered, should begin to be accepted in the first class of range 3 as illustrated. Particles larger than 22 μ m should be 100% accepted in range 3. The nonspherical particle case is shown in Figure 14b. Now the problem is quite different. Given the image of the particle as oriented, a size six (six elements occulted) is registered on range 2, while no element on range 3 is occulted at the 62.5% level. It is clear from symmetry arguments that no orientation of the image will result in an element occultation in range 3,

although permutations of the orientation in range 2 result in a variety of smaller sizes.

Clearly, the interpretation of the LCPS mode 3 data is much more complicated for such particles. However, it is necessary to invoke such asymmetry to duplicate the observed range 2/range 3 particle ratio. In fact, assuming asymmetry of the type illustrated (basically column, needle, or thin-plate crystals), the minimum aspect ratio to achieve the Pioneer Venus results is 3:1. The computed mode 3 distribution for spheres and columnar crystals of 3:1 aspect ratio using the observed range 2/range 3 data is shown in Figure 15. In this treatment, no preferential orientation (owing to aerodynamic effects) has been assumed. While the lack of preferential orientation cannot be ascertained, it would seem highly unlikely at these sizes.

The primary difference between the two distributions is the reduction in number of small particles while increasing the number of larger ones in the crystal particle distribution and the fact that the particles are distributed according to length. The total cross section reduces by a factor of 1.5, while the mass reduces by as much as a factor of 2. A number of crystalline forms were modeled theoretically to determine response effects. The plate or column forms provide response functions generating the required anomaly; however, certain open structures (much like dendritic snow crystals) could as well. Since some of these forms could result in underestimates while others overestimates of cross section and mass, we have chosen to continue use of the spherical assumption for such computations. The reader is referred to Knollenberg et al. [1980] for additional insight and constraints on possible effects of other crystalline forms. The identity of the mode 3 crystals is of course a primary scientific question, and suggested compositions are given in section 7. The refractive index of mode 3 is likewise difficult to estimate. In fact, a wide range of indices (m = 1.3-2.0) could be acceptable with certain crystalline forms.

6. MODAL PARTITIONS

One interpretation of the multimodal size distributions is that they represent particles of different phase, composition,



RANGE 3 MINIMUM DETECTABLE SIZE = 18μ m FOR SPHERES

Fig. 14a. LCPS range 3 versus range 2 response to spheres. Twelve element arrays are used on ranges 2 and 3 with the element spacing shown. Examining the above, it is easy to see that at all sizes close to the minimum detectable size, the probability of detection is a function of how well the image aligns with a given element.

or growth history. Our treatment of the data has been to partition the particle number density, extinction coefficient, and mass loading between the three size modes as in Figures 16, 17, and 18. The most important partition that relates to origin or growth history is the number density. The extinction coefficient is generally related to optical properties at visible wavelengths. The mass loading can be used to determine vapor abundances required for equilibrium at various altitudes and relates to mass and heat fluxes. To provide a reasonable profile of the mode 3 particles in these partitions, the shape of the mode 3 size distribution was assumed to be invariant, but the number density was allowed to vary according to the observed mode 3 number density at each altitude. This provides a smooth profile without the noisy effects created when the isolated large particles appear (which particularly dominate the mass) and an extinction coefficient profile which has improved signature correlation with the nephelometer (which samples a particle ensemble). A discussion of the vertical structure of the three partitions and the implications of observed changes in microphysical parameters follows.

a. Number Densities

The mode 1 particles have the highest number densities (exceeding 850 cm⁻³) in the upper cloud region in the vicinity of 61 km. Above 60 km the number densities decrease somewhat faster than the gas. A ratio of scale heights of the gas to mode 1 particles, Hg/Hp, of $2 \pm \frac{1}{2}$ is appropriate. Below 59 km the mode 1 particles decrease even more rapidly, reaching a minimum value at T_{um} . Below T_{um} and into the middle cloud region, the mode 1 particles gradually increase from this minimum value. In this region the particles and gas appear well mixed since Hg/Hp ≈ 1 . Below T_{m1} the mode 1 particles increase in number density quite rapidly, reaching number density quite rapidly.

sities of 600 cm⁻³ at 49 km. Aside from the small peaks coincident with the precloud layers, the mode 1 particles fall off to around 50 cm⁻³ at 44 km and hold remarkably constant until 31 km, where all particle activity ceases.

The mode 2 particles do not appear separated from the mode 1 particles in the upper cloud region because of the limited LCPS resolution. However, as previously described in section 4, the modes are separable. In general, the mode 2 particles follow the mode 1 particles, although they do not deplete as rapidly above or below the region of maximum number density, which also occurs at 60 km and reaches 76 cm⁻³. Below $T_{\mu m}$ the signatures of the number density of mode 2 track mode 1 remarkably closely until T_{ml} . At T_{ml} there is a rapid loss of mode 2 particles, while only a slight lowering in mode 1 occurs. In the lower cloud region the mode 2 number density increases and appears as a continuation of the profile observed in the middle cloud region. The peak in the upper precloud region at 47.5 km appears to fit this general profile also and is the highest mode 2 number density observed anywhere. Except for the lower precloud layer at 46 km, mode 2 is no longer observed.

The third mode is first observed just above T_{um} and develops to number densities of 10 cm⁻³ from 55 to 51 km. These particles largely disappear for 0.5 km at T_{ml} but increase rapidly in the lower cloud region to concentrations of 50 cm⁻³ or a five-fold increase over the middle cloud region number density. There is a lot of scatter in the raw data in the middle cloud region which has been largely removed by the smoothing technique. The statistics are better in the lower cloud region, and the structure shown is in all likelihood real; it correlates closely with the nephelometer [Ragent and Blamont, 1979].

Rapid changes in number density generally reflect proximity to sources or sinks. We have concluded that the source of both modes 1 and 2 is located near 60 km since both modes exhibit maximum concentrations in that region. Since this is also the region of maximum solar deposition [Tomasko et al., 1979] in the UV, one is led to believe that both modes are photochemically produced. The mode 2 particles are certainly



CONCLUSION: ASPECT RATIOS 2:1 ARE REQUIRED TO ALLOW 30µm PARTICLES TO BE MEASURED ON RANGE 2 AND NOT SIZE CLASS 2 OF RANGE 3,

Fig. 14b. LCPS range 3 versus range 2 response for crystals in preferred orientation to give maximum size. Here we see that shadowgraph images of large asymmetric particles can occult many elements of range 2 while not registering on range 3 (approximately 62.5% of an element's active area must be shadowed to register).

 H_2SO_4 , and, in fact, a bimodal spectrum in the region can be entirely explained in our growth modeling by growth of H_2SO_4 on giant nuclei. During any condensation growth process, whether homomolecular or heteromolecular, there exists a critical size (generally submicron) at which a droplet has maximum vapor pressure. This barrier results from two competing effects: vapor pressure lowering owing to solute and vapor pressure elevation owing to droplet surface curvature (Kelvin effect; see standard cloud physics texts, e.g., *Byers* [1965, p. 35]). The mode 2 narrow H_2SO_4 droplet spectrum results from nuclei which have become activated passing over the free energy barrier, while the mode 1 aerosols are trying to climb the free energy barrier. We will treat this problem further in section 7.

In the middle cloud region the number densities of modes 1 and 2 are nearly the same from 56 to 51 km. The fact that modes 1 and 2 show Hg/Hp \approx 1 is indicative of this region's being well mixed. Mode 3 shows hints of the same mixing process, although that data set is not very convincing because of weak count statistics.

In the lower cloud region the numbers of both mode 1 and 3 particles increase by factors of 3-5, while mode 2 appears as an extension of the profile observed in the middle cloud region. We are thus left to conclude, for whatever reason, that mode 1 and 3 particles correlate and that their coexistence must be interrelated somehow to their growth history. It thus seems unlikely that the mode 1 particles can be simply unactivated H₂SO₄ droplets at all altitudes. It is also unlikely that these three particle sets can all be H_2SO_4 since they are all in competition for the same parent vapors. There is also no simple explanation for why the mode 1 particles should largely disappear when descending through T_{um} and reappear just as mode 3 does, unless below T_{um} modes 1 and 3 are growth related. The mode 2 particles tend to increase steadily from 50 to 48.5 km, with more than a doubling of number density from 50 to 100 cm⁻³ at the same time that mode 1 particles decrease by a factor of 2. One might argue that this is the effect of H_2SO_4 competition, as in the middle cloud region; however, here the signature correlation is wrong. The fact remains that modes 1 and 3, and not modes 1 and 2, are coupled here.

The precloud layers tend to be dominated by mode 1 and 2 particles, again with fairly strong bimodality. The precloud layer at 46 km is no more than 100 m thick, and the upper layer at 47.5 km is less than 200 m thick. The sharpness of these thin layers is consistent with fairly rapid condensation with relative ambient stability. (Seiff et al. [1979] find this re-



Fig. 15. Comparison of mode 3 size distribution for spheres and high aspect ratio crystals for lower cloud region. The crystal size distribution is for 3:1 aspect ratio columnar crystals (length is plotted).



Fig. 16. Modal partitioning of number density. Mode 1 is shown for both the sizes measured by the LCPS and the total expected from modeling efforts. The greatest difference is in the lower haze where we also are least confident.

gion to be stable.) There is general support for the belief that both of these layers result from direct condensation rather than particle formation by chemical reaction. However, the upper precloud layer tends to follow almost exactly the extension of mode 1 and mode 2 number densities observed in the lower cloud region. We thus conclude that it is probably not a precloud layer but simply a detached region of lower cloud with respect to modes 1 and 2. This argument certainly cannot be applied to the lower precloud layer, though. Here, the particle properties are only consistent with new condensate. It is difficult to imagine either a chemical particle generation that could be constrained to such a thin layer or dynamical processes that could clearly shear off such a thin cloud layer.

b. Extinction Coefficients

The modal partitioning of the extinction coefficient shown in Figure 17 is for 600 nm wavelength, but the results have general applicability within the visible spectrum. The accumulative optical depth is listed in Table 5 for each cloud region according to the modes and the mode 1 aerosol distribution model choices. The narrow mode 2 particles dominate the upper cloud region optical depth; however, the role of UV absorption may be more consistent with large absorption cross sections in mode 1 at shorter wavelengths [Knollenberg et al., this issue].

It is immediately evident from Figure 17 that the mode 3 particles dominate the cross section whenever present in the middle and lower cloud regions. In the middle cloud region, mode 3 is 59-70% of the total cross section, while in the lower cloud region it is 80% of the total. A comparison with the nephelometer data of *Blamont and Ragent* [1979] shows that mode 3 correlates best with the nephelometer backscattering profile. It is indeed the dominant particle optically in the middle and lower regions. In the precloud regions, mode 2 is responsible for the cloud optical properties at visible wavelengths. Of course, in the lower haze, only small amounts of mode 1 aerosol particles exist.

c. Mass Loadings

The modal partitioning of mass shown in Figure 18 illustrates the dominance of mode 3 in the overall mass loading. In



Fig. 17. Modal partitioning of extinction coefficient. This partitioning reveals the strong influence of mode 3 to the total extinction coefficient in the middle and lower cloud region (see Figure 4). It is clear that mode 3 dominates the observed nephelometer backscatter as well (compare with *Blamont and Ragent* [1979, Figure 1]). Below 44 km only mode 1 is present, and the profile follows the number density profile of Figure 16 (computation used n = 1.45).

fact, the computed total mass in mode 3 integrated over the entire vertical cloud depth is 5 times that in modes 1 and 2 combined, as shown in Table 5. We previously showed [Knollenberg and Hunten, 1976b] that mode 3 particles could potentially provide a considerable mass flux from which latent heats amounting to one half those of the solar net radiation could be computed.

The mode 1 particles clearly have very little mass; thus their impact on the parent vapor inventory is nearly negligible. The mode 2 particles in the upper cloud region average 4–5 times the mass of the mode 1 particles and in that region must be considered important, requiring some 10–20 ppm in parent vapor support. The total columnar mass in each of the three modes is 1.5×10^{-4} g cm⁻², 18.7×10^{-4} g cm⁻², and 1.6×10^{-2} g cm⁻². The mass in the lower haze is clearly negligible.

7. CLOUD PARTICLE IDENTITIES AND CONCEPTUAL LIFE CYCLES

We really can readily identify only two types of condensable gases in any quantity from which to build particles: water vapor and sulfur gases. A few parts per million of hydrochloric acid also can be added based on measurements taken from earth observations [Hunten, 1971]. But direct condensation of HCl is impossible at cloud temperatures. With regard to the sulfur gases, SO₂ is easily identified and the most abundant. However, it must be converted to SO₃ before it can truly be considered condensable at cloud temperatures and pressures. While the direct condensation of SO₃ is not precluded, the reaction of SO₃ with H₂O is so rapid that SO₃ would be below detection limits of measuring instruments, if indeed they could be designed to measure it. The H₂SO₄ produced by the reaction is rapidly absorbed in droplets. Therefore, detection of gaseous SO₃ and H₂SO₄ is unlikely. Consequently, we find it necessary to use SO₂ as a primary

'condensable' species. Possible crystalline solids for mode 3 are discussed below.

Figure 19 shows vertical profiles of SO₂ and H₂O vapor density with H₂SO₄-H₂O equilibria values within the clouds; also shown are reference values for CO₂. The H₂SO₄-H₂O vapor densities are computed from vapor pressure data (shown in Figure 20) which follow the work of Gmitro and Vermuelen [1964]. The SO₂ profile was constructed from available data assembled from PV and Venera gas chromatography, mass spectrometry, and the PV ultraviolet spectrometer (OUVS). The H₂O profiles include data from Venera 12 [Moroz et al., 1980] and our adopted profile based on Moroz's data, reguirements for equilibria within the clouds, and earth-based measurements near cloud top (65 km). Our adopted profile was generated assuming the same columnar H₂O abundance as above 40 km but with its maximum at the base of the lower cloud region. In reference to our adopted H₂O profile, it is apparent that there would be a tendency for the H₂SO₄ concentration to increase slightly with increasing altitude; however, a value of 85 \pm 5% would cover all altitudes. The H₂O and SO₂ have similar abundances just below cloud base ($\simeq 10^2 - 10^3$ mg m^{-3}) and drop off rapidly with increasing altitude. The abundances of H₂O and SO₂ are sufficient to account for all of the mass in the cloud particles and are more than 1 order of magnitude greater than the sum of modes 1 and 2. There is sufficient sulfur to support mode 3 as a crystalline sulfate.

Another possibility is that mode 3 is a chloride, although not HCl. The primary issue is whether Yu. A. Surkov's (private communication, 1979) chlorine-to-sulfur ratio of 10 can be totally accepted. (The basic problem in Surkov's measurements is that the sampler was opened for collection at about 275°K and closed at about 370°K. Obviously, the only particulate remaining must be one involatile at the higher temperature. All other possibilities would have evaporated!) This happens to be about the right ratio of mode 3 to the sum of modes 1 and 2. Surkov estimates 2 mg m⁻³ of chlorine, and



Fig. 18. Modal partitioning of mass loading. The partitioning of mass loading also reflects the importance of the mode 3 particles. A density of 2 g cm⁻³ was used for all particles, and mode 3 was assumed to be spherical.

		Optic	al Depth		Columnar Mass, g cm ⁻²					
	Mode 1	Mode 2	Mode 3	Total	Mode 1	Mode 2	Mode 3	Total		
Upper cloud region	2.15	5.12	0	7.27	1.1 × 10 ⁻⁴	9.5 × 10 ⁻⁴	0	10.6 × 10 ⁻⁴		
Middle cloud region	0.34	3.53	6.2	11.07	0.2×10^{-4}	4.7 × 10 ^{−4}	0.004	0.0045		
Lower cloud region	0.48	1.82	9.9	12.20	0.2×10^{-4}	3.8 × 10 ^{−4}	0.006	0.0064		
Precloud layers	0.06	0.24	0	0.30	<10 ⁻⁵	0.6 × 10 ^{−4}	0	0.6×10^{-4}		
Lower haze	0.20	0.05	0	0.25	<10 ⁻⁵	0.1 × 10 ⁻⁴	0	0.1 × 10 ⁻⁴		
Totals	3.23	9.76	16.1	31.09	1.5 × 10 ⁻⁴	18.7 × 10 ⁻⁴	0.01	0.012		

TABLE 5. Summary of Optical Depths and Columnar Mass According to LCPS Size Modes

Approximately 3.5 optical depths are indicated above the first region of LCPS measurements by the LSFR [see Knollenberg et al. this issue].

we can infer 0.2 mg m⁻³ of S, which would amount to about 0.8 mg m⁻³ of H₂SO₄. This is within a factor of 2 of the mode 2 average mass loading in the cloud system. However, the LNMS did not detect significant HCl at any altitude, although its inlet was blocked by what appears to be H₂SO₄. It is not at

all clear whether certain chlorides in the vapor form would be detectable by any of these instruments.

The mode 2 particles at all altitudes are surely sulfuric acid. This identification is based on the known particle properties of the Venus cloud tops which have been associated with



Fig. 19. Vertical structure of parent vapor inventory. Data presented here are from a variety of sources. Measured H_2O : PV Gas Chromatograph (LGC) after *Oyama et al.* [1979]. Venera 12 water vapor spectrophotometer (Venera 12) after *Moroz et al.* [1980]. Earth-based measurements after *Hunten* [1971]. Measured SO₂: PV ultraviolet spectrometer (OUVS) after *Esposito et al.* [1980]. PV neutral mass spectrometer (LNMS) after *Hoffman et al.* [this issue]. PV gas chromatograph (LGC) after *Oyama et al.* [1979]. Temperature: PV atmospheric structure experiment after *Seiff et al.* [1979]. H_2SO_4 and H_2O profile used the earth-based measurements and Venera 12 data. It is consistent with $85 \pm 5\%$ H_2SO_4 at all cloud levels.



Fig. 20. Vapor pressures of H₂O, SO₃, and H₂SO₄ (by weight percent H₂SO₄). Data source: *Gmitro and Vermuelen* [1964].

 H_2SO_4 from earth-based measurements [Young, 1973; Pollack, 1974; G. Sill, personal communication, 1973]. The peculiar microphysical properties (narrow size distribution of 2- μ m diameter particles of 1.44 refractive index) cited by Hansen and Hovenier [1974] are easily identified with mode 2 [Knollenberg and Hunten, 1979b; Ragent and Blamont, 1979]. As shown in section 6, the mode 2 particles were easily traced throughout the entire cloud system down to altitudes of 46 km where H_2SO_4 evaporates or decomposes. However, decomposition to SO₃ and H_2O is not nearly as rapid as one might think. Our own estimate from equilibria data shown in Figure 20 is that at 45 km, H_2SO_4 is only 10% dissociated. (Use 99% H_2SO_4 and compare H_2SO_4 and SO₃ vapor pressures.)

We are also of the opinion that the bulk of the mass in mode 1 is sulfuric acid or sulfates. In the upper cloud region it is probably mostly sulfuric acid. This is clear from the behavior of the number density profile in Figure 16 which shows the modes tracking at most altitudes, indicative of growth from the same parent vapor phase. We might argue that they are both being generated in this region simply because of the UV-rich environment and could be different condensates of photolytically produced vapors. However, it still would be unlikely for the number density signatures to mimic each other.

Various workers have suggested that at least mode 1 is a UV absorber [Tomasko et al., 1979; Pollack et al., 1979]. A particulate absorber seems necessary in spite of the absorption contribution from gaseous SO₂ which fails to absorb sufficiently longward of 3500 Å. We would also support contaminants in mode 2. In fact, we propose that mode 1 and mode 2 probably originally had similar amounts (masses, not fractions) of contaminants. After all, if we believe that mode 1 and mode 2 simply differ in that mode 2 is activated, there is little reason to suspect that in their early life (submicron sizes) mode 1 and mode 2 were distinguishable. This argument also can be extended to imply that mode 2 would have higher single scattering albedoes because of dilution and lower ab-

sorption optical depth in the UV since mode 1 greatly outnumbers mode 2. Because of coagulation and related processes, mode 2 clearly ends up with more contamination in due time, but its concentration of contaminants by weight should be less.

In the middle cloud region the H_2SO_4 vapor is less supersaturated than at the higher, colder levels. This is consistent with the H_2SO_4 vapor production source being a few kilometers away and the higher temperatures supportive of more rapid equilibria. Modes 1 and 2 see more growth competition, and mode 2 grows at the expense of mode 1. Thus, mode 1 may shrink back to essentially dry (no H_2SO_4) nulcei cores.

In the lower cloud region, mode 1 may well be a mixture of particles, residue cores from the evaporation of both modes 2 and 3, or even some other chemical species. There is some evidence that all of their mass is not H_2SO_4 . One simply has to observe the fact that mode 2 decreases in size while mode 1 increases (in number, if not size). We do not have any good evidence about what the primary constitutents of mode 1 are, but there is bound to be a lot of nondescript polyparticle aerosol in the cloud system since there are no precipitation mechanisms to carry material to the surface.

From the similarity in mode 1 size distributions the lower precloud layer resembles the middle cloud region and the upper precloud layer resembles the lower cloud region; we therefore infer that they have the same compositions. We have already suggested that the upper precloud layer may be a detached section of the lower cloud region. In the lower precloud layer it is surely a recondensate which, because of the still higher temperatures, causes mode 1 to be at a growthcompetitive disadvantage with respect to mode 2.

The crystals which we can now identify as mode 3 particles appear at T_{um} and increase in number density by a factor of 5 in the lower cloud region while not showing any great growth surge. One explanation for this large a change is that there may have been two production sources for mode 3, yet other explanations can be offered as well. First of all, it is clear that the mode 3 extinction coefficient signature in Figure 17 correlates better than any combination of modes 1 and 2 with the nephelometer backscatter signal [Ragent and Blamont, 1979, Figure 1], in the middle and lower cloud regions. Since similar nephelometer profiles exist on all probes, we infer that mode 3 exists at all sites; that is, mode 3 also is planetary in its distribution. If we keep in mind that mode 3 particles exist in regions of strong wind shear, and have comparatively low terminal velocities, it is certainly doubtful that the measured mode 3 particles in the middle and lower cloud regions were created at the same place or time. The general increase in number density during descent through the cloud layers is consistent with a source in the upper part of the middle cloud region, sedimentation, and accumulation through slowed terminal velocities in the more dense lower atmosphere; but the extreme wind shear implies a tremendous lateral stretching of any given trajectory. A useful analogy exists in the case of terrestrial cirrus where ice crystals form aloft and survive kilometers of vertical fall, producing similar (but less extreme) profiles. If you sample a vertical path, you intersect crystal trails from various generating regions [Heymsfield and Knollenberg. 1972]. Another important aspect of that analogy might also apply here (that is, that cirrus crystals are energetically difficult to nucleate heterogeneously directly from the vapor phase). They invariably first condense as liquids and then freeze. Thus in terrestrial cirrus clouds there is a transient liquid water phase at cloud top, referred to as the generating region by *Heymsfield and Knollenberg* [1972]. It is also a requirement for contrail glaciation [*Knollenberg*, 1972]. A further result is that this transient liquid phase grows rapidly and may continue to grow while falling out until reaching a lower level where it is no longer in a supersaturated region.

We might carry this analogy still one step further. The heads, or generating cells, in cirrus are typically at the base of a shear zone often being 'kicked up' by Kelvin-Helmholtz waves. While we cannot say whether the critical critera are satisfied at T_{um} (Ri > 0.125), this does appear to be a dynamical boundary, and it coincides with the first indication of mode 3 particles.

Since water is the most abundant condensable, it would be advantageous to develop the mode 3 mass in the form of hydrates. There are a number of possible chlorides and sulfates with their hydrates from which to build crystalline particles. There are problems with all of the compounds suggested, however. Generally, the hydrates are too volatile and the anhydrides are too involatile. Yu. A. Surkov (personal communication, 1979) found approximately 2 mg m⁻³ of chlorine in his samples, which averaged over 63 to 46 km interval and included all of the regions for mode 3 particles. This is about a factor of 10 less than the integrated mass of mode 3 for the same altitude column as measured by the LCPS, even if a large aspect ratio is assumed. The additional mass may be partly made up by the cation and possibly water of hydration. Also, the sounder probe may have penetrated a region where the lower cloud is denser than the planetary average, as suggested by the nephelometer data [Blamont and Ragent, 1979].

Barsukov et al. [1979] have suggested HCl, whose low refractive index is attractive. But the vapor pressure would give a mixing ratio of several percent, 2-3 orders of magnitude higher than can be accepted. Krasnopolsky and Parshev [1979] favor aluminum chloride. Here the problem is the stability of Al_2O_3 , which would surely retain the aluminum in the crust. The thermodynamics are more favorable for the generation of FeCl₃, and it has a suitable volatility. In the cloud region it could become hydrated. Although FeCl₃ absorbs in the visible, preliminary studies by G. Sill (personal communication, 1979) suggest that its absorption would not be noticeable on Venus. Mercury chlorides can be ruled out since Surkov's experiment was specifically designed to detect mercury and found none. Iron was not easily detectable owing to the ⁵⁴Fe source. Detection of manganese, titanium, and aluminum was also masked by surrounding spacecraft structures of similar materials, which is possibly why Al₂Cl₃ was suggested.

One of the other problems of the unhydrated $FeCl_3$ is the low saturation vapor pressure. Abundances of a part per million are hard to come by. The hydrate $FeCl_3 \cdot 6H_2O$ has about the right vapor pressure, but its 310°K melting point is too low. (Note that the primary volatile in equilibria with FeCl₃ · 6H₂O is H₂O although some HCl can evolve if its vapor abundance is low.) Whatever mode 3 is, its melting point must be above 355°K for crystals to exist at cloud base for the lower cloud region. Sufficient vapor density also must be capable of existing at cloud base temperatures to account for the average, if not highest, mass loading of the above cloud mass. We are looking for at least tens of parts per million, and FeCl₃ has a saturation mixing ratio at 355°K of <1 ppm while FeCl₃ · 6H₂O requires 1000 ppm of H₂O. One possibility is that hydrated FeCl₃ exists only in the middle cloud region and begins losing its water near T_{m1} , leaving excess H₂O and



Fig. 21. Conceptual Venus cloud particle cycles.

FeCl₃. The H₂O is partially picked up by the H₂SO₄. We are not at all sure that the FeCl₃ or its hydrate will turn out to be the mode 3 particle identity; suffice to say at this time it is a possibility along with AlCl₃ and even NH₄Cl. Again, we wish to emphasize the critical chlorine issue. If chlorine turns out to be a measurement artifact, then sulfates are the preferred choice. Compounds with both sulfur and chlorine may also exist.

The restriction of sulfuric acid to mode 2 greatly simplifies its life cycle and in many ways provides overall simplifications to potential cloud models. The determination that mode 3 could not be spherical and thus not H_2SO_4 has been only recently positively verified by laboratory studies with the LCPS flight spare. The lack of time has restricted extensive use of crystals in our conceptual models. Very little detailed analysis can be accomplished until the mode 3 particles are positively identified. However, particles of different phase behave differently, and this is sufficient to constrain their potential life cycle. The LCPS data provide the most important inputs to microphysical modeling. It is only necessary to obtain abundances for the parent vapors in equilibrium to develop a rudimentary one-dimensional model. Initial results of applying existing H₂SO₄ models have been reported by Toon et al. [1979] and Whitten et al. [1979]. While we have not yet attempted numerical modeling of these results, we have developed a conceptual model from which numerical techniques can be applied to modes 2 and 3. Interpretation of the LCPS data has given us the basic ideas. We still need detailed knowledge of the parent vapor inventory within the clouds for all modes; however, that also has been constrained by LCPS results.

Figure 21 is a qualitative depiction of the particle life cycles; the H₂SO₄ cycle draws to some extent on the work of *Young* [1979], *Toon et al.* [1979], and *Whitten et al.* [1979]. The H₂SO₄ cycle is more easily explained and indeed is the only one that we can discuss knowledgeably. Essentially, all sulfuric acid vapor is formed through homogeneous reactions of SO₃ and H₂O at altitudes of 60 km or higher, where SO₂ is able to photooxidize. The SO₂ arises from a thermochemical source deep within the atmosphere below the cloud layer and mixes vertically through eddy processes. At altitudes above 60 km, sufficient H₂SO₄ is produced to support growth of mode 1 and mode 2, and numbers of mode 1 aerosol pass through the free energy barrier to become H_sSO₄ droplets. Moving downward through T_{um} the available H₂SO₄ vapor excess is gradu-

Property	Earth	Venus
Percent coverage	40	100
Average optical depth	5–7	25-40
Maximum optical depth	300-400	40
Composition	solid and liquid H_2O	H ₂ SO ₄ droplets, crystals, plus contaminants
Number density	100–1000 cm ⁻³ (liquid)	50-300 cm ³ (liquid)
-	$0.1-50 \text{ cm}^{-3}$ (ice)	$10-50 \text{ cm}^{-3}$ (crystals)
Average mass loading (Mass Density)	$0.3-0.5 \mathrm{g m}^{-3}$	$0.01-0.02 \text{ g m}^{-3}$
Maximum mass loading (Mass Density)	10–20 g m ⁻³	0.1–0.2 g m ⁻³
Distribution function	normal-log normal bimodal (ice)	Multimodal
Typical background aerosol at cloud base 0.5 microns	l cm ⁻³	$100-200 \text{ cm}^{-3}$
Condensation process	homomolecular	heteromolecular
Average precipitable mass	0.03–0.05 mm	0.1–0.2 mm
Mean scattering size (diameter) (In the visible)	10 µm	2–4 µm
Mean mass size (diameter)	30 µm	10 µm
Dominant optical cloud form	stratiform	stratiform
Dominant mass cloud form	cumulus	stratiform
Potential latent instability	high	low
Temporal variability	high	slight
Dominant cloud atmosphere heat exchange process	latent heat	radiation

TABLE 6. Comparative Venus and Earth Cloud System Properties

ally depleted, and the source becomes effectively removed. Now the activated mode 2 H₂SO₄ droplets have a competitive growth advantage over the smaller mode 1 aerosols, because of the Kelvin effect, robbing them of both H₂O and H₂SO₄ until they are rather desiccated.

Growth of hydrated H₂SO₄ droplets involves heteromolecular condensation, and we want to emphasize the importance of both H₂O and H₂SO₄ to the growth problem. It has been argued that H₂SO₄ droplets resist evaporation since, if they lose water, they simply increase their H₂SO₄ concentration and reduce their H₂O vapor pressure to accommodate the new equilibria. However, in so doing, their H₂SO₄ vapor pressure is increased (dramatically at higher H₂SO₄ concentrations; see Figure 20). We would therefore expect the droplets to lose H_2SO_4 if initially at equilibrium. The growth and decay of a droplet having binary components follow a pingpong exchange of alternate components if near equilibria for both; we simply tend to follow the less volatile specie (H₂SO₄ in this case) since it governs all rates. The computed growth times for mode 2 H₂SO₄ droplets cover a range of 4-5 orders of magnitude from 70-45 km. This diffusional growth process dominates with coalescence and Brownian coagulation negligible.

The most rapid growth of the H2SO4 droplets appears to occur within T_{um} where the modal diameter increases from 2.2 to 2.7 μ m in a kilometer. Computations show that a change from 2.2 to 2.5 μ m would account for the loss in mode 1 mass transferred to mode 2. This transfer occurs in less than 1 week, assuming vapor equilibrium with respect to mode 2. For the droplet to grow to 2.7 μ m, some growth from the vapor excess is also required. The narrowness of the mode 2 H₂SO₄ droplet size distribution below T_{um} is surprising. However, in a slow competitive growth situation, such narrow spectra in fact are generated in model calculations.

The LCPS observed no evaporation of the H₂SO₄ droplets below T_{um} . This fact requires that either greater H₂SO₄ be produced to maintain equilibrium or the concentration shift in favor of H₂O. And, of course, both may well happen and in our model do, with the H_2SO_4 decreasing from 90% concentration at 68 km to 82% above the lower cloud region before starting to lose water again at 48 km and to shrink in size. Below 49 km, the H₂SO₄ can be no longer supported and largely evaporates. The H₂SO₄ vapor only partially decomposes, leaving sufficient vapor density to provide sources for new condensation in the lower precloud layer.

At this point in our discussion we need to clarify another point. The lower cloud layer has been described as layered by Knollenberg and Hunten [1979a] and Ragent and Blamont [1979]. We have shown that mode 3 is responsible for most of this observed structure at least at the sounder site. The H₂SO₄ particles' optical depth increases smoothly from the upper cloud layer down to where it evaporates near 48 km except for two transition regions. There is also little change in size. The maximum optical cross section for H₂SO₄ particles is near 49 km and accounts for much less than one half of the total at this altitude. The structure observed by the nephelometer at all entry sites below 56 km is also probably primarily due to variations in mode 3 and not in mode 1 or 2.

The proposed mode 3 life cycle starts with its vapors reaching a liquid condensation level near T_{um} condensing as a transient supercooled liquid phase which rapidly nucleates to form crystals. The crystals grow rapidly in the supersaturated environment, growing to maximum size in the upper part of the middle cloud region and sedimenting to lower levels. At $T_{\rm mi}$ there appears to be a chemical or dynamical boundary. All particles appear to evaporate near the bottom of the middle cloud region only to reappear anew upon entering the lower cloud region. Mode 3 increases in condensed mass most prominently of all (three- to five-fold). We believe that T_{ml} must be both a chemical and a dynamical boundary:chemical in the sense that greater vapor abundances must exist below $T_{\rm ml}$ than can be accounted for from exchange processes above and dynamical in the sense that the middle and lower cloud regions were formed at different times, and the observed pro-

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file thus reflects horizontal transport from regions different in vapor abundance and particulate mass.

The crystals largely decompose near 49 km very close to where we first lose mode 2. One suspects at this point that modes 2 and 3 were somehow feeding upon at least one of the same vapor components. The lone possibility here is H_2O . Perhaps a hydrate can yet be found that competes with H_2SO_4 for water.

8. CONCLUSIONS

It is useful to look at the LCPS results in the light of our own knowledge of terrestrial clouds. Table 6 gives a summary of cloud parameters, providing a useful first comparison of clouds on sister planets. There are striking similarities as well as differences in the listed properties. Both cloud systems have both solid and liquid particles with similar number densities. The apparent large differences in mass loading, potential latent instability, and temporal variability are believed to be the result of the higher volatility of earth water-ice clouds.

The LCPS experiment has provided the most extensive microphysical 'cloud truth' in the Venus cloud system to date. Of greatest importance is the clear evidence for the existence of two if not three separate particle constituents. The verticle distribution of these constituents generates the observed layered cloud structure evident planetwide in the cloud morphology. Planetary transition regions have been defined at two altitudes coinciding with changes in microphysical properties and perhaps particle chemistry. The following list of summary findings highlights our results. Further treatment of the LCPS data taking into account all PV cloud measurements is given in *Knollenberg et al.* [1980].

1. The bimodal and trimodal size distributions observed by the LCPS indicate two or three different particle constituents. Mode 1 in all cloud regions is a background aerosol population with high number density but little optical depth except in the UV. Free sulfur, extra-Venusian materials, and pure H_2SO_4 are candidates. The mode 1 aerosols seem to be rich in sulfur compounds, but are likely polyparticle collections of various chemistry. Mode 2 is sulfuric acid. It is highly concentrated in all cloud regions. Mode 3 in the middle and lower cloud regions is surely made up of crystalline, high aspect ratio particles. Crystal hydrates may be preferred in certain regions.

This interpretation of the three size modes appears necessary in spite of the fact that the three modes disappear at similar altitudes and one is tempted to simply identify all of them with H_2SO_4 . However, this paradox will simply have to remain for the present, since H_2SO_4 could only exist as liquid at altitudes where mode 3 is present and mode 3 is obviously not a liquid but a solid.

2. The background mode 1 aerosol is abundant in most cloud regions and at the cloud tops and appears to be more abundant than at prior times of observation from earth. Mode 1 aerosols appear to be desiccation products (cores) of cloud particle growth and decay. They are probably not pure H_2SO_4 in the middle and lower cloud regions.

3. The H_2SO_4 droplets are probably several months old in the upper cloud region unless vertical exchange is faster than suspected and at least days in the lower cloud region (inferred from growth rate calculations). All H_2SO_4 droplets are likely to be contaminated through scavenging aerosol over their long lifetime. In the upper cloud region, H_2SO_4 vapor may be highly supersaturated (even though droplet growth rates are extremely slow), permitting nucleation of pure H_2SO_4 embryo and contributing to nuclei production.

4. All cloud particles and most of the aerosol mass are volatile at temperatures >200°C.

5. Cloud dynamics are most important in transporting particles between atmosphere levels of high and low volatility. In the horizontal direction, the droplets go along for the ride and primarily result in variations in number density.

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