



**Venus: Halide Cloud Condensation and Volatile Element Inventories**

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that ion as an essential component of the geotropic-sensing system, it adds a new characteristic to our understanding of the gravity-sensing organelle and raises the possibility that calcium is associated with the translation of the gravity-induced movement of amyloplasts into a physiological gradient, leading eventually to geotropic growth.

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## Venus: Halide Cloud Condensation and Volatile Element Inventories

**Abstract.** Several recently suggested Venus cloud condensates, including aluminum chloride and halides, oxides, and sulfides of arsenic and antimony, are assessed for their thermodynamic and geochemical plausibility. Aluminum chloride can confidently be ruled out, and condensation of arsenic sulfides on the surface will cause arsenic compounds to be too rare to produce the observed clouds. Antimony may be sufficiently volatile, but the expected molecular form is gaseous antimony sulfide, not the chloride. Arsenic and antimony compounds in the atmosphere will be regulated at very low levels by sulfide precipitation, irrespective of the planetary inventory of arsenic and antimony. Thus arguments for a volatile-deficient origin for Venus based on depletion of water and mercury (relative to the earth) cannot be tested by a search for atmospheric arsenic or antimony.

Soviet spacecraft have analyzed cloud particles in the main Venus cloud layer by means of x-ray fluorescence (XRF) spectroscopy (1). On the basis of these analyses it is claimed that chlorine is present as a cloud constituent and sulfur is not detected. These results are in conflict with the weight of evidence from Earth-based studies (2) and from other spacecraft experiments (3), which strongly suggest that the dominant cloud constituent on Venus is H<sub>2</sub>SO<sub>4</sub> droplets. Satisfactory photochemical models for the production of an H<sub>2</sub>SO<sub>4</sub> aerosol from geochemically plausible primary gases, including COS, H<sub>2</sub>S, and SO<sub>2</sub>, are available (4). Direct evidence regarding the abundances of these species in the lower troposphere is lacking: copious production of COS and so on by reactions between sulfuric acid and the inlet system of the mass spectrometer on the Pioneer Venus large probe leads to masking of the atmospheric sulfur gases,

and abundances up to 100 ppm are possible for COS and H<sub>2</sub>S (5). No composition data are available for the bottom 22 km of the atmosphere, a region which contains not only the best-equilibrated gases, but also over 80 percent of the total atmospheric mass.

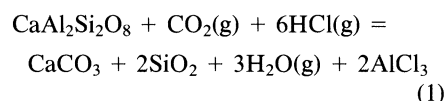
The source of a chlorine-bearing aerosol is less obvious. We have pointed out the high volatility of halides and sulfides of mercury, arsenic, and antimony (6) and have shown that the terrestrial crustal abundance of even the rarest of these elements, mercury, would suffice to produce substantial masses of halide cloud condensates on Venus: mercury is so volatile at the surface temperature of Venus that it would reside almost completely in the atmosphere.

More recently, in the context of a model for the formation of the planets in the presence of a steep gradient outward from the proto-sun, we have favored compositional models in which the vola-

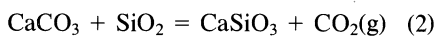
tile elements are severely depleted in the accreting Venus relative to the earth. Combined condensation-accretion models with a variety of recent estimates of the accretion sampling functions of the terrestrial planets (7) show a primordial water content on Venus from 10<sup>-1</sup> to as low as 10<sup>-4</sup> of the terrestrial value. The failure of the 1978 Pioneer Venus mission to detect even a trace of mercury in the lower atmosphere (5) strongly implies that Venus is deficient in mercury relative to the earth. The severe depletion of the most abundant terrestrial volatile, H<sub>2</sub>O, on Venus is well known and is attributed to either a lack of water in preplanetary solids at the orbit of Venus (8) or massive loss of oceans of H<sub>2</sub> after differentiation and outgassing of the planet (9). Donahue (10) found an enhancement of the D/H ratio on Venus by about a factor of 100 over the terrestrial value, requiring either loss of hydrogen from 100 times the present water inventory or accumulation of deuterium-rich material on Venus. Such a late loss mechanism could not deplete mercury while leaving vast amounts of the lighter and more volatile species N<sub>2</sub> and CO<sub>2</sub>. Thus the observed severe depletion of mercury is more convincing evidence for a volatile-poor high-temperature origin of Venus than is the depletion of water. Other moderately volatile elements, such as arsenic, antimony, bismuth, and germanium, are also potential indicators of the overall volatile content of Venus. In addition, such species, if present in the hot lower atmosphere, would condense at intermediate altitudes to form solid halides, sulfides, and oxide cloud particles.

In the past year, Krasnopolsky and Parshev (11) suggested Al<sub>2</sub>Cl<sub>6</sub> as the major cloud layer constituent and Barsukov *et al.* (12) suggested arsenic and antimony halides and oxides. Mole fractions of at least 10 ppm of condensable gases are required to provide the observed cloud density (13). Are these species plausible cloud constituents? Can useful limits on the abundances of the volatile elements arsenic and antimony be derived from atmospheric measurements?

It is simple to estimate the volatility of aluminum compounds at the mean surface conditions of Venus. Consider coexisting anorthite (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>), quartz (SiO<sub>2</sub>), and calcite (CaCO<sub>3</sub>) at 750 K and 95 bars in contact with the atmosphere



The equilibrium constant for this reaction at this temperature is  $10^{-26.1}$ . Assuming the spectroscopic HCl abundance in the clouds, about 1 ppm (14), and the most typical water vapor abundance figures for the lower atmosphere, about 100 ppm (15), an  $\text{AlCl}_3$  partial pressure of  $10^{-21.0}$  bar is calculated. With an extreme effort to bias the equilibrium in favor of  $\text{AlCl}_3$  production (raising HCl to 10 ppm and lowering  $\text{H}_2\text{O}$  to 10 ppm), we can force the  $\text{AlCl}_3$  pressure only as high as  $10^{-16.5}$  bar. This is still ten orders of magnitude too small to provide detectable amounts of condensate. Note that the presence of granitic rocks on Venus, with anorthite and quartz as common primary minerals, is expected both from consideration of the atmospheric composition (16, 17) and from surface passive gamma-ray spectroscopy (18). The surface of Venus lies, as accurately as can be determined, precisely on the calcite-quartz-wollastonite ( $\text{CaSiO}_3$ ) buffer



and calcite is therefore also a plausible surface mineral. Since weathering reactions at higher altitudes on Venus will preferentially tend to mobilize a fine calcium-rich dust, which can be transported readily by winds to the hot lowlands, this buffer may not be difficult to establish on Venus (19).

Thermodynamic treatment of arsenic and antimony volatilization can be carried out without a precise a priori knowledge of the minerals formed by these elements on the surface (20, 21). We will calculate the partial pressures of a number of arsenic and antimony gases at the surface as a function of the activities of arsenic and antimony: an activity of 1 means the pure element is present on the surface and  $10^{-4}$  means the pressure of the monatomic vapor of that element is  $10^{-4}$  times its abundance at saturation. We can then assess the stability of possible surface minerals containing these elements. Figure 1 presents the results for arsenic (22). For activities greater than about  $10^{-2}$  the dominant gas is  $\text{As}_4$ , with  $\text{As}_4\text{O}_6$  second. Based on recent thermochemical data of Johnson *et al.* (18), we calculate that liquid  $\text{As}_2\text{S}_3$  will precipitate if the elemental arsenic activity is greater than 0.13, thus ruling out higher arsenic activities. This, in turn, places firm upper limits on the pressures of arsenic-bearing gases, as indicated in Fig. 1.

The results of a similar calculation for antimony are given in Fig. 2. Precipitation of  $\text{Sb}_4\text{O}_6(\text{s})$  occurs for an elemental

antimony activity of 0.20. Thus for all possible antimony activities,  $\text{SbS}$  is the dominant gas. The best available data on  $\text{Sb}_2\text{S}_3$  (stibnite) from Johnson *et al.* (20) indicate that  $\text{Sb}_2\text{S}_3(\text{s})$  precipitates at an antimony activity of  $10^{-2.3}$ . Therefore we expect that stibnite precipitation on the Venus surface will regulate the antimony gas phase abundance.

We conclude that the mole fraction of all arsenic gases is below  $10^{-7}$ , probably making these species too rare to account for the clouds no matter what species condenses. We expect that the total mole fraction of antimony gases will be lower than for arsenic, but that the most stable

gas is  $\text{SbS}$ , not a halide. Partial pressures as high as  $10^{-4}$  bar are conceivable for  $\text{SbS}$  and cannot be ruled out.

Observational constraints on the abundance of arsenic in the Venus atmosphere even down to the level of 0.1 ppm are therefore not sufficient to test whether arsenic is, like water and mercury, depleted in Venus relative to the earth: the stability of arsenic sulfides is great enough to preclude a larger abundance of gaseous arsenic compounds irrespective of the crustal abundance of arsenic. The same may be true of antimony, since the mineral  $\text{Sb}_2\text{S}_3$  seems to have low enough volatility to hide antimony in the lithosphere. We have also briefly considered bismuth, and find that  $\text{Bi}_2\text{S}_3$  is so stable that the most abundant bismuth-bearing gas,  $\text{BiS}$ , should have a mole fraction below  $10^{-12}$ .

In any event, we call into serious question the geochemical plausibility of all the species so far suggested as sources of chlorine-bearing clouds. We suggest that either the chlorine compound is a species which has not been considered or the XRF data used to deduce the presence of chlorine are in error.

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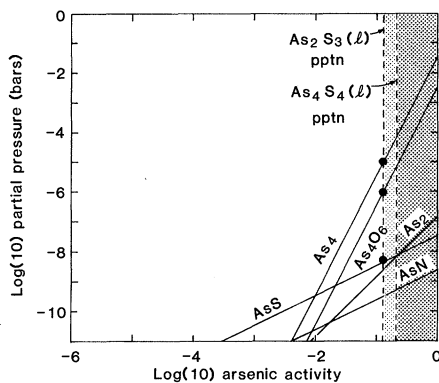


Fig. 1. Partial pressures of arsenic gases as a function of elemental arsenic activity at the Venus surface. Limitations of the arsenic activity imposed by precipitation of liquids of  $\text{As}_2\text{S}_3$  (orpiment) and  $\text{As}_4\text{S}_4$  (realgar) composition are indicated by vertical dashed lines. Heavy dots indicate the upper limits on the  $\text{As}_4$ ,  $\text{As}_4\text{O}_6$ , and  $\text{AsS}$  partial pressures. A condensable species with a partial pressure near  $10^{-4}$  bar is needed to provide the observed cloud density. The maximum total mole fraction of arsenic gases is  $\leq 0.1$  ppm.

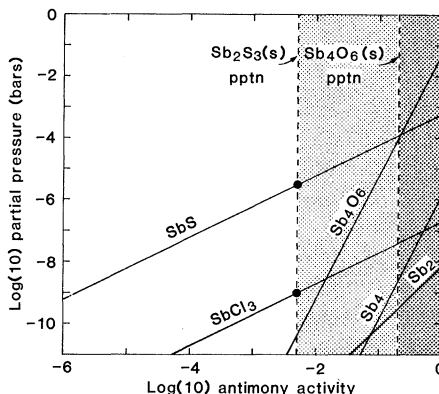


Fig. 2. Partial pressure of antimony gases as a function of elemental antimony activity. The  $\text{Sb}_4\text{O}_6$  (valentinite) and  $\text{Sb}_2\text{S}_3$  (stibnite) precipitation points are indicated by vertical dashed lines. Firm upper limits on the antimony gas pressures are indicated by the dots. The maximum total mole fraction of antimony gases is about 0.03 ppm. An  $\text{SbS}$  gas abundance of 0.3 ppm is needed to make clouds.

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21. The pressure, temperature, composition conditions assumed in these calculations are 95 bars total pressure, 750 K surface temperature,  $P_{O_2} = 10^{-22.5}$  bar,  $P_{HCl} = 10^{-4.0}$  bar,  $P_{S_2} = 10^{-4.0}$  bar, and  $P_{HF} = 10^{-5.2}$  bar.
22. Other As- and Sb-bearing gases included in the calculations but not abundant enough to be graphed are As, AsO, AsH<sub>3</sub>, AsF, AsF<sub>2</sub>, AsF<sub>3</sub>, AsF<sub>5</sub>, AsCl, AsCl<sub>2</sub>, AsCl<sub>3</sub>, Sb, SbO, SbH<sub>3</sub>, SbN, and SbF<sub>3</sub>. The solid and liquid arsenic oxides As<sub>2</sub>O<sub>4</sub>, As<sub>2</sub>O<sub>5</sub>, and As<sub>4</sub>O<sub>6</sub> are unstable and do not precipitate as pure condensates on the surface of Venus. Formation of more complex minerals, such as As-bearing apatites and complex Sb oxides, for which thermodynamic data are unavailable, may result in lower gas phase abundances for As and Sb. Thus our results are firm upper limits.
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cles, as well as on their size and shape. This can explain the ability of filter feeders to capture ultrafine particles such as bacteria, clays, and colloids that are much smaller than the minimum mesh size of the filtering appendages.

Filter feeding by small invertebrates occurs at low Re, where fluid flow is slow and laminar and all motion is dominated by viscous rather than inertial forces (2, 5). The Re for the filtering combs on the third and fourth appendages of *Daphnia magna* (Fig. 1, A to C) is in the range 0.4 to 2.0; for a single seta it is  $10^{-2}$  to  $10^{-3}$ ; and for a single setule it is  $10^{-3}$  to  $10^{-4}$  (6). The boundary layer, or the region of reduced flow around a single setule, extends approximately 10  $\mu$ m under these conditions, which is far beyond the next setule (7). Therefore, little or no flow occurs between setules, and the appendage functions as a solid wall, analogous to the hirsute wings of small insects (8). Observations of a scaled-up model of a *Daphnia* appendage, made with 110- $\mu$ m mesh plankton netting oscillated in glycerin, support this since no fluid was observed to pass through the mesh of the model (9).

A capture mechanism that may function under these conditions is electrostatic or ionic attraction. It operates over shorter distances in water than in air but nevertheless can be important in particle capture and retention, especially in slow laminar flow regimes (10). We expand and redefine this mechanism to include all surface chemical interactions between particle and filter, such as ionic or hydrophobic-hydrophilic interactions.

We tested the sieving model and the role of surface chemistry in filtering by feeding suspensions of polystyrene

## The Role of Surface Chemistry in Filter Feeding by Zooplankton

**Abstract.** Surface chemistry of both particles and animals is important in filter feeding at low Reynolds number. *Daphnia magna*, fed mixtures of three sizes of polystyrene particles, retained particles that were smaller than the mesh size of the animals (1.0 micrometer) at greater efficiencies than predicted by a sieving model. Retention efficiency of the smallest particles (0.5 micrometer) was increased when negative surface charge on the particles was neutralized, and retention was decreased when a nonionic surfactant was added to reduce wettability.

Filter feeding is the dominant process of primary consumption in freshwater and marine environments. It is often described as sieving, in which particles too large to pass through an animal's filtering mesh are captured. Capture efficiency of various sized particles is thought to be related to the array of mesh sizes on the filtering appendages (1); however, it appears that sieving is not the mechanism by which fine particles are captured by small filter feeders. Films of copepod feeding show little or no movement of water through the filtering meshes (2). Brittle stars are able to catch particles smaller than the spaces between their mucus-covered tube feet, and the surface charge on particles determines their affinity for retention (3). Our observations of *Daphnia* indicate that the majority of water movement is over, not through, the meshes of the filtering combs. Also, there is no evidence of mucus enhancement of filtering in *Daphnia* (4).

We report that particle capture in these animals occurs at low Reynolds number (Re), is not a sieving process,

and is strongly influenced by surface chemistry, such as surface charge and wettability of the particles. The findings suggest that differential or selective feeding on natural particles may occur on the basis of surface properties of the parti-

Table 1. Relative ingestion of three particle sizes of unmodified polystyrene by *Daphnia magna*. Particles were presented at equal densities by volume, with total densities similar to those found in nature ( $10^4$  cm<sup>-3</sup> for alga-sized particles and  $10^6$  to  $10^7$  cm<sup>-3</sup> for bacteria-sized particles). Data reported are proportions transformed with an arcsin-square root transformation, and are thus shown as degrees. Numbers in parentheses are percent feeding efficiencies expressed as the ratio of observed to expected proportions of the 0.5- $\mu$ m particles. Relative ingestion of all three particle sizes were compared by *t*-tests adjusted for multiple comparisons (12). Expected proportions, based on concentrations in feeding suspensions (11), were compared with the control test (treatment 1). Deionized (0.5- $\mu$ m particles) and surfactant treatments (treatments 2 and 3, respectively) were compared to the control treatment. In all treatments where *t* was significant, 0.5- $\mu$ m particles were ingested in a significantly lower proportion than expected. When 0.5- $\mu$ m particles were dropped from the analysis and proportions of the larger particles were recalculated, no statistically significant differences were observed in the relative ingestion of 1.1- and 5.7- $\mu$ m particles, indicating that both of the larger particles were collected in the same proportions at which they were present in suspension.

Particle size ( $\mu$ m)	Concentration in suspension (No./cm <sup>3</sup> )	Expected proportion (degrees)	Proportion in gut (degrees)		
			Treatment 1 (N = 9)	Treatment 2 (N = 9)	Treatment 3 (N = 9)
5.7	$10^4$	8.6	18.0*	16.8	20.4
1.1	$1.4 \times 10^6$	16.8	34.6†	39.6	43.4†
0.5	$1.5 \times 10^7$	71.0	49.2† (64)	45.4 (51)	39.1* (44)

\**P* < .05. †*P* < .005.