EPSC Abstracts
Vol. 6, EPSC-DPS2011-352, 2011
EPSC-DPS Joint Meeting 2011

Uptake of ammonia gas by Jovian ices

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Abstract

The altitude profile of ammonia (NH_3) in Jupiter's atmosphere, as constrained by microwave spectra, is poorly understood. A global-scale NH_3 depletion mechanism appears to be operating between the 2- and 6-bar levels. Candidate depletion mechanisms include dynamics, condensation, and adsorption of NH_3 onto ices.

Laboratory measurements of NH_3 uptake provide valuable constraints for model calculations testing the NH_3 depletion hypothesis in Jupiter's atmosphere. We have performed experiments designed to measure the uptake coefficient of NH_3 by H_2O ice using a Knudsen cell apparatus at temperatures relevant to the troposphere of Jupiter. We have initiated model calculations to apply the experimental results to Jupiter's atmosphere and evaluate the key parameters and processes that influence the ammonia uptake and their relevance to Jupiter's atmosphere.

1. Introduction

The microwave spectrum of Jupiter is consistent with supersolar NH_3 deeper than 6 bar and subsolar NH_3 at pressures less than 2 bar [3]. Because the Galileo Probe Mass Spectrometer measured a deep NH_3 / H_2 mixing ratio of 6.4×10^4 [7]—or five times the protosolar ratio [4]—the microwave data show that there must be a global-scale depletion mechanism for NH_3 gas, operating between the 2- and 6-bar levels. Candidate depletion mechanisms include dynamics, condensation of species with higher nitrogen fractions than NH_4SH (including clathrates [6]), and adsorption of NH_3 onto NH_4SH or water ices. The goal of this study is to test whether uptake of NH_3 by water ice can explain the depletion of NH_3 in Jupiter's troposphere.

2. Experimental Apparatus

A Knudsen cell is a low-pressure, stirred-flow reactor with two chambers separated by a valve. The sample under study is placed in the bottom chamber, and a mixture of gas-phase reactants is introduced into the top chamber. The top chamber has ports for diagnostics and a small aperture leading to a differentially pumped mass spectrometer. The pressure in the top chamber is kept at low enough levels so that molecular flow applies. Thus, the residence time of gas-phase species in the top chamber is determined by the size of the escape aperture and any loss by heterogeneous reactions on the cell walls or the sample surface. The pressure in the top chamber can be altered when either the size of the escape orifice is changed, or the valve between the top and bottom chambers is opened to allow for interaction of the flowing gas with the ice sample. A change in the gas-phase reactant concentration can be observed by mass spectrometry or another spectrometric technique.

Helium gas containing traces (~1%) of NH_3 and Ar is introduced into the Knudsen cell under low-pressure, molecular-flow conditions (~10–20 mTorr). Under these conditions, the collision frequency of NH_3 with the water ice substrate can be calculated and is approximately the same as the collision frequency with the escape orifice. The composition of the gases exiting through the orifice is monitored continuously with a residual gas analyzer mass spectrometer (RGA).

Uptake of NH_3 on water ice appears as a depletion of gas phase NH_3 in the signal detected by the RGA. The temperature of the ice substrate can be cooled externally and, in the present study, was varied in the temperature range 170–195 K.

2. Results and Discussion

The raw RGA signal is first corrected for background contributions from residual gases in the RGA compartment and for mass interference, which occurs when multiple gases produce signal at the same mass-to-charge ratio (m/z). For example, at m/z = 17, there is signal originating from both water and ammonia (the OH^+ and NH_3^+ ions, respectively). Measured signal ratios for fragmentation are used to account for such mass interference contributions. Argon gas serves as a reference to help distinguish between changes in gas composition and NH_3 partial pressure due to adsorption on the ice sample. The NH_3 partial pressure in the Knudsen cell is the net result of the ammonia flow in and out of the reactor and the ammonia adsorbed on the walls and the ice.

The mass accommodation coefficient or sticking probability, α, is the probability that a collision of a gas molecule with a condensed phase boundary will result in uptake of the gas by the condensed phase. The measured uptake coefficient for solids, γ, is defined as the net number of molecules sticking to the surface divided by the number of collisions with the surface. Desorption or saturation of the surface limits the measured net uptake.

The uptake coefficient can be determined by relative measurements of the ammonia partial pressure in the cell at constant input flow in two ways: i) the diameter of the escape orifice is changed; and ii) the ice substrate is exposed to the NH_3 flow vs. being isolated from it [5]. We applied both approaches in the temperature range 170–195 K and determined an average accommodation coefficient (initial uptake) with an average value of $\sim 4 \times 10^{-3}$ and an uptake coefficient at equilibrium of $\sim 3 \times 10^{-4}$ for NH_3 uptake by H_2O ice. Analysis of recent measurements is currently in progress.

Using our results for the uptake coefficient and a simple atmospheric model assuming 1-μm spheres and cloud mass densities from the equilibrium cloud condensation model of Atreya and Romani [1], we find that for a significant depletion of ammonia to take place at least one thousand monolayers of coverage would be required. A depletion of approximately 90% is needed to be consistent with globally averaged NH_3 concentrations derived from microwave measurements [3].

Future efforts will investigate the effect of ammonia uptake by liquid water aerosols. Because of diffusion into the bulk and dissolution, the uptake of NH_3 by liquid water is anticipated to be at least three orders of magnitude larger than uptake by water ice.

Acknowledgements

This work is supported by the NASA Planetary Atmospheres Program under grant NNX09AB69G.

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