Observations of Hydration and Dehydration in the Winter 2000 Arctic Stratosphere

R. L. Herman\textsuperscript{1}, K. Drdla\textsuperscript{2}, J. R. Spackman\textsuperscript{3}, D. F. Hurst\textsuperscript{4,5}, C. R. Webster\textsuperscript{1}, J. W. Elkins\textsuperscript{4}, E. M. Weinstock\textsuperscript{6}, B. W. Gandrud\textsuperscript{7}, G. C. Toon\textsuperscript{1}, M. R. Schoeberl\textsuperscript{8}, H. Jost\textsuperscript{2}, E. L. Atlas\textsuperscript{7}, P. J. Popp\textsuperscript{5,9}, and T. P. Bui\textsuperscript{2}

\textsuperscript{1}Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA.
\textsuperscript{2}NASA Ames Research Center, Moffett Field, CA.
\textsuperscript{3}Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA.
\textsuperscript{4}Climate Monitoring and Diagnostics Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO.
\textsuperscript{5}Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO.
\textsuperscript{6}Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA.
\textsuperscript{7}National Center for Atmospheric Research, Boulder, CO.
\textsuperscript{8}NASA Goddard Space Flight Center, Code 916, Greenbelt, MD.
\textsuperscript{9}Aeronomy Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO.

ICMA conference
July, 2001
Innsbruck, Austria
Introduction

The 1999-2000 winter was characterized by unusually cold conditions in the Arctic lower stratosphere, including a larger area exposed to temperatures cold enough to nucleate nitric acid trihydrate (NAT) than ever measured before in the Arctic [e.g., Manney and Sabutis, 2000]. If the polar vortex gets colder over the next several decades, this winter may typify Arctic winters of the future. Solid PSCs that form below 195 K are ubiquitous in the Arctic winter stratosphere [Fahey et al., 2001] and are predominantly NAT [Voigt et al., 2000]. Ice PSCs are stable below the frost point (~188 K), so Arctic dehydration has been observed mainly in the coldest winters. That raises the question: did the widespread presence of polar stratospheric clouds (PSCs) in 1999-2000 cause irreversible removal or redistribution of stratospheric water?

We examine here total hydrogen \( H = H_2O + 2CH_4 + H_2 \) from the NASA ER-2 high-altitude aircraft during the SAGE III Ozone Loss and Validation Experiment (SOLVE). The partitioning of hydrogen between these species is changed by photochemical oxidation of CH\(_4\) into H\(_2\)O and H\(_2\), oxidation of H\(_2\) into H\(_2\)O, and HO\(_x\) chemistry that includes both sources and sinks of H\(_2\) [e.g., Le Texier et al., 1988; Dessler et al., 1994; Hurst et al., 1999]. Since these are the three dominant hydrogen-bearing species in the stratosphere, \( H \) should be conserved in air in which the seasonal cycle of water has been averaged out. In this case, \( H \) is changed only by long-term trends in CH\(_4\) or H\(_2\)O, dehydration, or hydration.
### Total Hydrogen Budget

<table>
<thead>
<tr>
<th></th>
<th>( \text{H}_2\text{O} + 2\text{CH}_4 )</th>
<th>( \text{H}_2\text{O} + 2\text{CH}_4 + \text{H}_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>JLH</td>
<td>Harvard</td>
</tr>
<tr>
<td>Extravortex (E)</td>
<td>7.32±0.13</td>
<td>7.07±0.18</td>
</tr>
<tr>
<td>Mean Vortex (V)</td>
<td>7.38±0.11</td>
<td>7.11±0.15</td>
</tr>
<tr>
<td>Vortex, deploy 1</td>
<td>7.41±0.11</td>
<td>7.09±0.11</td>
</tr>
<tr>
<td>Vortex, deploy 2</td>
<td>7.36±0.11</td>
<td>7.13±0.11</td>
</tr>
<tr>
<td>Difference, E-V</td>
<td>-0.06±0.17</td>
<td>-0.04±0.23</td>
</tr>
</tbody>
</table>

Was there a net loss of total hydrogen from the Arctic stratospheric polar vortex during the 1999-2000 winter? Shown above are mean stratospheric data filtered by \( \text{CH}_4 < 1.45 \text{ ppmv} \) and \( \theta > 400 \text{ K} \) to eliminate the seasonal cycle of water. The difference between mean extravortex and vortex values show insignificant changes in total hydrogen. Extravortex \( \text{H} \) is greater than vortex \( \text{H} \) solely due to low \( \text{H}_2 \) in the Arctic vortex. The largest difference between the measurements is due to a bias between the JPL Laser Hygrometer (JLH) [May, 1998] and the Harvard Lyman-\( \alpha \) Hygrometer [Weinstock et al., 1994]. \( \text{CH}_4 \) data are from the Aircraft Laser Infrared Absorption Spectrometer (ALIAS) [Webster et al., 1994], and \( \text{H}_2 \) data are from the Airborne Chromatograph for Atmospheric Trace Species (ACATS-IV) [Elkins et al., 1996]. SOLVE deployment #1 is Jan. 14 through Feb. 3, 2000, and deployment #2 is Feb. 26 through Mar. 16, 2000.
Dehydration / Rehydration Episodes

Shown above are the largest excursions from local background values of H\textsubscript{2}O + 2CH\textsubscript{4} measured during SOLVE with JH H\textsubscript{2}O and ALIAS CH\textsubscript{4}. Above 430 K, ∆(H\textsubscript{2}O + 2CH\textsubscript{4}) < 0 due to dehydration. Below 430 K, ∆(H\textsubscript{2}O + 2CH\textsubscript{4}) > 0 due to hydration, with one notable exception of dehydration on March 7, 2000.
This figure shows the sum $\text{H}_2\text{O} + 2\text{CH}_4 (\text{JLH H}_2\text{O}, \text{ALIAS CH}_4)$ during the ER-2 flight of Jan. 27, 2000. $\text{H}_2\text{O} + 2\text{CH}_4$ was low for more than 430 km along the ER-2 flight path, indicating up to 0.63 ppm dehydration. Negligible $\text{H}_2\text{O}$ was sequestered in the condensed phase during this flight segment. The dehydrated air parcels were located at potential temperatures ranging from 46.5 to 46.8 K, pressures from 47.5 to 55.1 hPa, and altitudes of 20.4 to 20.9 km. The dehydrated air parcels were located at potential temperatures ranging from 46.5 to 46.8 K, pressures from 47.5 to 55.1 hPa, and altitudes of 20.4 to 20.9 km.
As shown above, a cluster of three-week diabatic back-trajectories from the dehydrated air parcel (48400 sec UT on Jan. 27, 2000) passed through the cold pool of the Arctic vortex in early Jan., as shown above, a cluster of three-week diabatic back-trajectories depleted the air parcel of water vapor. Growth of ice PSCs. Sedimentation of these particles irreversibly depleted the air parcel of water vapor. The temperature was below the ice frost point, allowing freezing and growth of ice PSCs. For several days, the temperature experienced temperatures as low as 183 K.
In the Arctic vortex, the sum $\text{H}_2\text{O} + 2\text{CH}_4$ decreases at lower CH$_4$ mixing ratios (higher altitude). JLH H$_2$O and ALIAS CH$_4$ are shown here, but the trend is also seen with data from the Harvard Lyman–α Hygrometer.
Outside the Arctic vortex, in the extravortex and vortex edge regions, the sum \( \text{H}_2\text{O} + 2\text{CH}_4 \) does not change as systematically as it does inside the vortex. JLH \( \text{H}_2\text{O} \) and ALIAS \( \text{CH}_4 \) are shown here, but the trend is also seen with data from the Harvard Lyman–\( \alpha \) Hygrometer.
Modeling

To further explore the presence of ice PSCs in the winter Arctic polar vortex, we utilized the Integrated Microphysics and Aerosol Chemistry on Trajectories model (IMPACT) [Drdla, 1996]. This model follows diabatic trajectories of more than 2000 air parcels within the 1999-2000 Arctic polar vortex using UKMO temperatures [Schoeberl et al., 1993; Schoeberl et al., 2000]. A full particle microphysics code allows condensation, freezing, sublimation, and sedimentation of PSCs [Drdla et al., 2001].
Results for HetFrzB, f=0.02% Scenario
Dehydration on February 1st

- Average of points inside vortex (maximum = 4.5)
- Average of all points (maximum = 2.87, minimum = -1.45)
- Points outside vortex
- Points in vortex edge region
- Points inside vortex
The figure to the left shows model dehydration (%) of each individual diabatic trajectory, plotted against the value of the trajectory’s potential temperature on February 1, 2000. The "HetFrzB" scenario of the IMPACT model assumes that NAT PSCs form by heterogeneous freezing. Shown is the case where 0.02% of particles are assumed to contain heterogeneous nuclei. The "HetFrzB" scenario of the IMPACT model assumes that NAT particles form by heterogeneous freezing.  Shown is the case where 0.02% of particles are assumed to contain heterogeneous nuclei.
Conclusions

- During the 1999-2000 winter, mean total hydrogen in the Arctic lower stratospheric vortex was not significantly different from the mean value outside the vortex. This implies negligible net loss of water from the Arctic stratosphere (400 – 470 K).

- Isolated episodes of dehydration and hydration were intercepted by the ER-2 aircraft on several flights. In particular, on the flight of Jan. 27, 2000, vortex air was dehydrated by as much as 0.63 ppmv.

- Diabatic back-trajectory calculations suggest this air parcel passed through the cold pool and experienced a dehydration event during Jan. 9 - 12, 2000.

- Total hydrogen decreased significantly with height throughout the lower stratospheric vortex, indicating a weak but widespread redistribution of water: water is low at altitudes corresponding to CH$_4$ < 1.0 ppmv ($\theta > 445$ on Feb. 1, 2000), and water is high at lower altitudes. A downward flux of water within the Arctic stratospheric vortex due to sedimentation and evaporation of PSCs is thus inferred.
Acknowledgements

We thank R. D. May for development of the JPL Laser Hygrometer; G. J. Flesch, D. C. Scott, and K. Modarress for field assistance; S.C. Wofsy for providing age of air data; P. Romashkin for processing H$_2$ data; and the ER-2 pilots, crew, and SOLVE logistical support staff. Support for this research and the SOLVE campaign was provided by the National Aeronautics and Space Administration (NASA) Upper Atmosphere Research Program (UARP). Part of the research described in this paper was carried out by the Jet Propulsion Laboratory, California Institute of Technology, under a contract with NASA.

References

Drdla, K., Applications of a Model of Polar Stratospheric Clouds and Heterogeneous Chemistry, University of California at Los Angeles, 1996.
Drdla, K., et al., Microphysical Modelling of the 1999-2000 Arctic winter: Polar stratospheric clouds, denitrification, and dehydration, subm. to JGR.