Molecular hydrogen as a mesospheric hydrogen reservoir

Laura K. Meredith[1], Eric A. Ray[2], R. Alan Plumb[1], Fred L. Moore [2]

[1]Department of Earth, Atmospheric and Planetary Science; Massachusetts Institute of Technology; Building 54-1320; Cambridge, MA 02139

[2]National Oceans and Atmospheres Administration; 325 Broadway, Boulder, CO 80305

predawn@mit.edu

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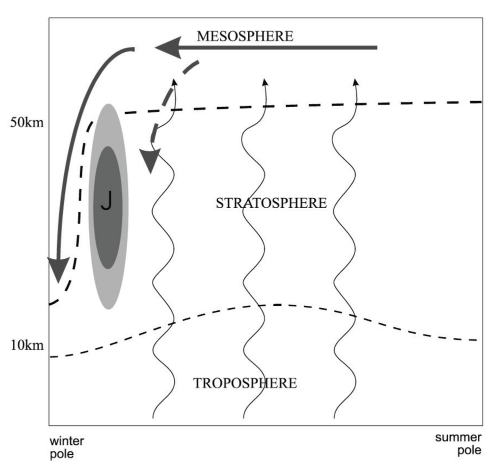
Introduction

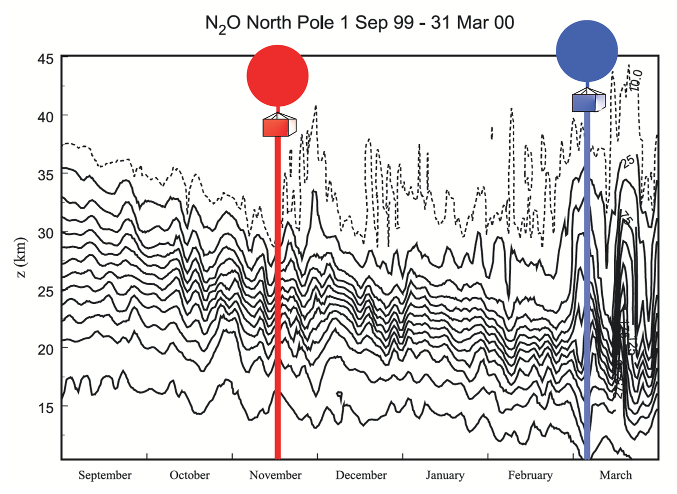
A rare glimpse into the chemical and dynamical evolution of the Arctic polar vortex is provided by a suite of *in situ* balloonborne measurements. A set of mesospheric tracers observed in the late vortex above 25 km validate theoretical mesospheric chemical profiles, which is especially valuable for mesospheric H_2. Early vortex mesospheric profiles are constructed to explain mixing in tracer-tracer space, and they will constrain estimates of the amount of mesospheric air that descended to stratospheric altitudes by vortex end.

Observations of descent

Dynamics: The polar vortex is characterized by summer-to-winter pole mesospheric circulation and rapid, isolated descent within the vortex.

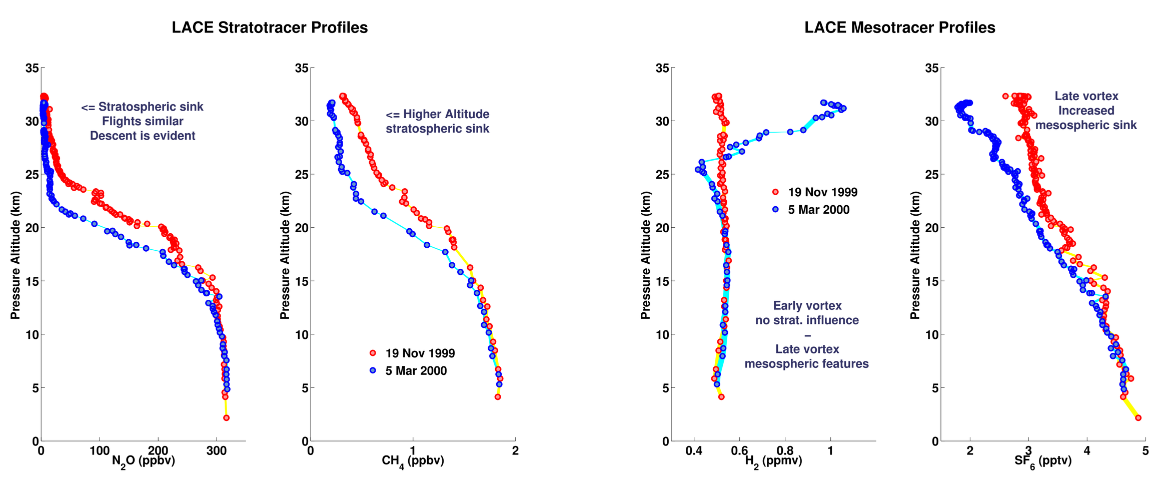


Descent of mesospheric air [5]



19 Nov 1999 (red) to 5 Mar 2000 (blue)

Chemistry: The distribution of chemical species with sufficiently slow production and loss processes will be controlled by transport.

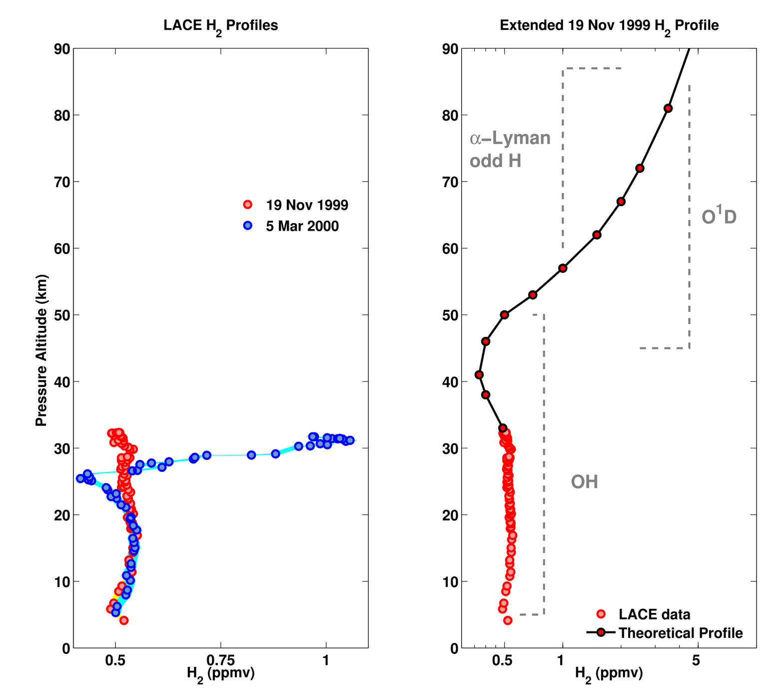


Measurements: Balloonborne Lightweight Airborne Chromatograph Experiment (LACE) measurements made from Kiruna, Sweden (68°N) [4]. Profiles were measured up to 35 km in altitude on 19 November 1999 and 5 March 2000 as part of the SAGE III Ozone Loss and Validation Experiment (SOLVE). The March, 2000 LACE measurements of descended air parcels may be the first direct measurements of the mesospheric H_2 profile.

What were early vortex mesotracer profiles?

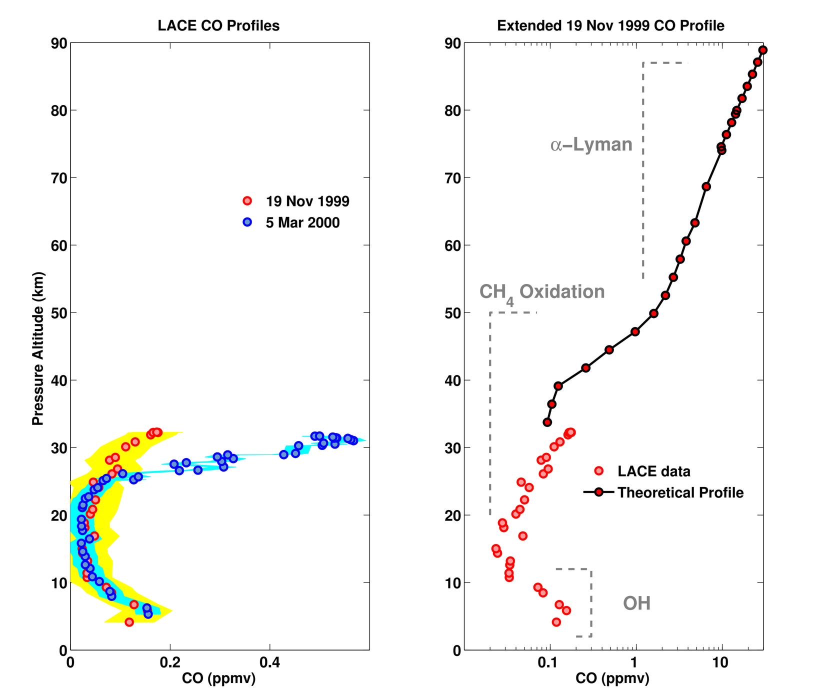
Three measured LACE species can be used as *mesotracers*: H_2, CO, and SF_6. All are relatively stable in the stratosphere, but have marked mesospheric sinks or sources that imprint upon the descending vortex air.

H_2



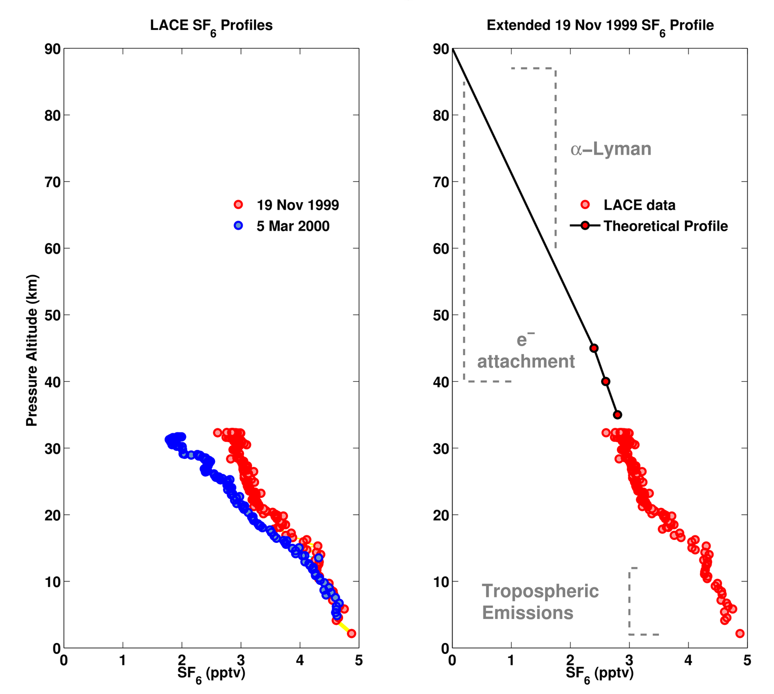
H_2 mixing ratios increase into the mesosphere where H_2O is photodissociated by α-Lyman radiation and odd hydrogen recombines efficiently. A minimum near the mesopause is predicted where H_2 oxidation dominates [3].

CO



CO is produced by the oxidation of stratospheric methane and by the photodissociation of carbon dioxide in the mesosphere. The influence of tropospheric OH on surface emissions is evident [1].

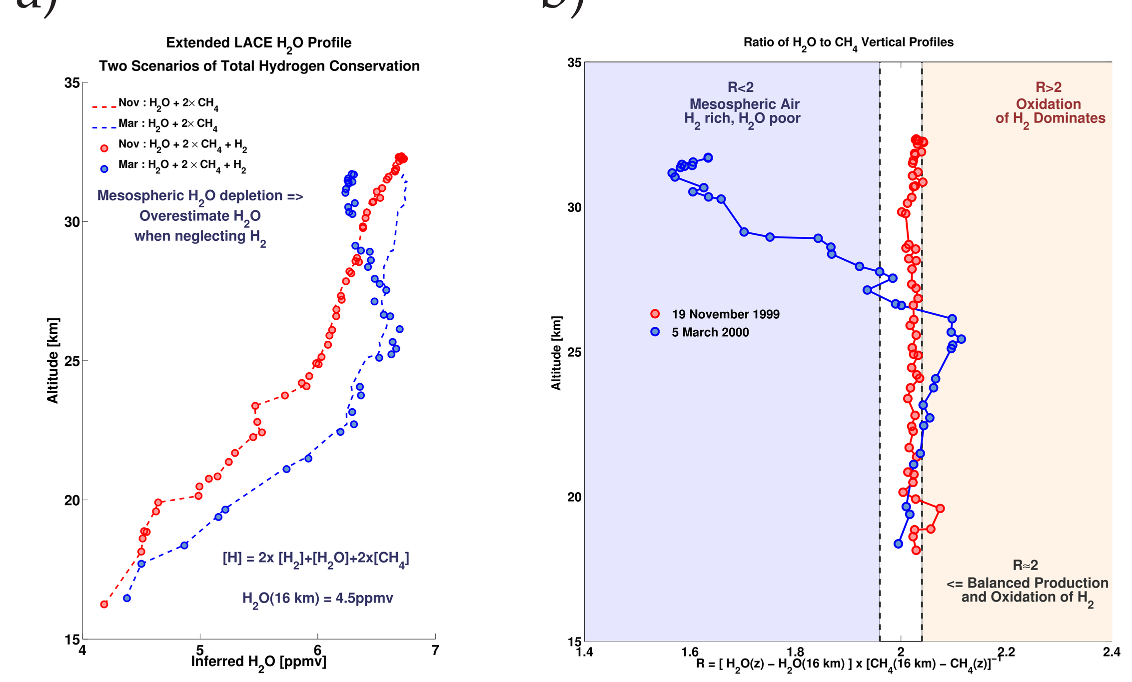
SF_6



SF_6 is depleted in the mesosphere by α-Lyman radiation and electron attachment. It is emitted anthropogenically from the surface [2].

Hydrogen chemistry

a) b)

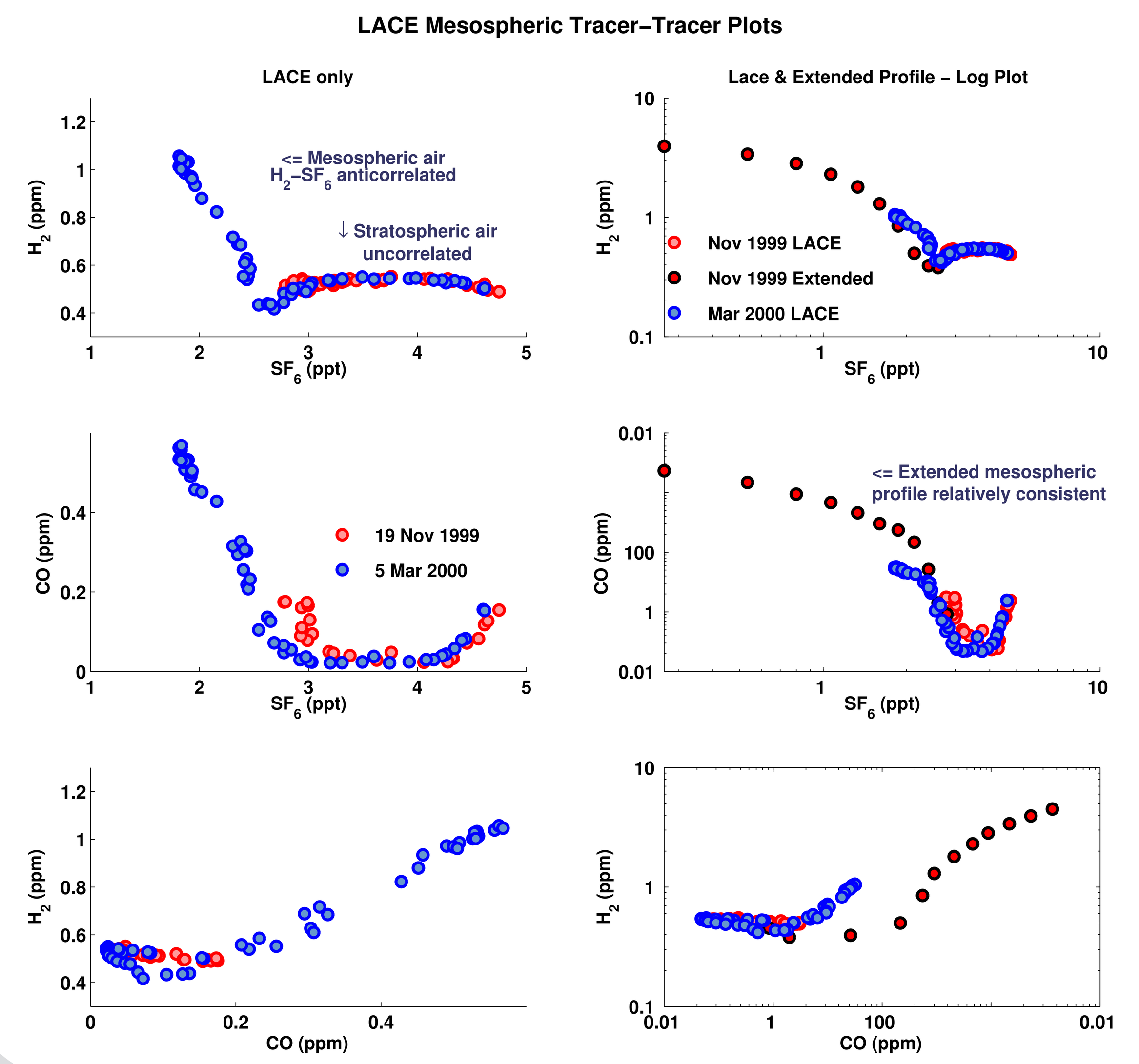


We infer the H_2O profile by assuming total (even) hydrogen conservation in **Figure a**, which highlights 1) the overestimation of H_2O when neglecting the mesospheric H_2 reservoir and 2) dehydrated mesospheric air enters the vortex.

The H_2O/CH_4 gradient in **Figure b** indicates different regions of hydrogen chemistry. The early vortex exhibits balanced oxidation and production of H_2 (R=2). Departures in the late vortex arise from increased oxidation near the mesopause (R>2) and from H_2 production by H_2O photolysis (R<2) above 27 km.

Tracer-tracer verification

Extended mesospheric profiles are consistent with LACE tracer-tracer relationships.



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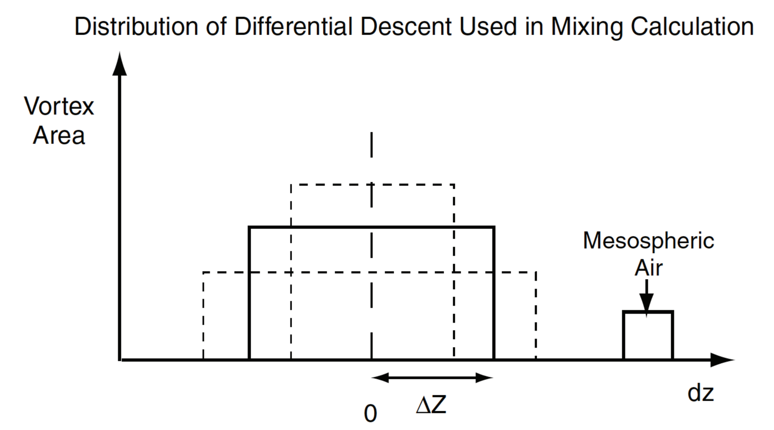
Next step - model concept

A model of differential descent and mixing in the polar vortex, constrained by stratospheric tracer-tracer relationships and SF_6 as a mesotracer, has previously calculated 2-4% mesospheric air in the late vortex [6].



We plan next to update the model with extended initial profiles of the mesotracers H_2 and CO, which provide additional constraints at different heights in the mesosphere. The result should be a more robust estimate of the amount and characteristics of mesospheric descent within the polar vortex.

Conclusions

LACE measurements of the early and late 1999-2000 polar vortex offer an unparalleled set of stratospheric and mesospheric tracer measurements. Over the evolution of the vortex, we observe three mesotracers, SF_6, CO, and H_2 descend to stratospheric heights. LACE profiles and tracer-tracer relationships validate current understanding of mesospheric chemistry. Theoretical profiles were constructed and tested, and will be used to constrain a model of differential descent and mixing to determine the mesospheric contribution to the polar vortex.

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