A New OH Dilemma?

—— The Gap Between Model and Aura MLS Observations in Mesospheric OH

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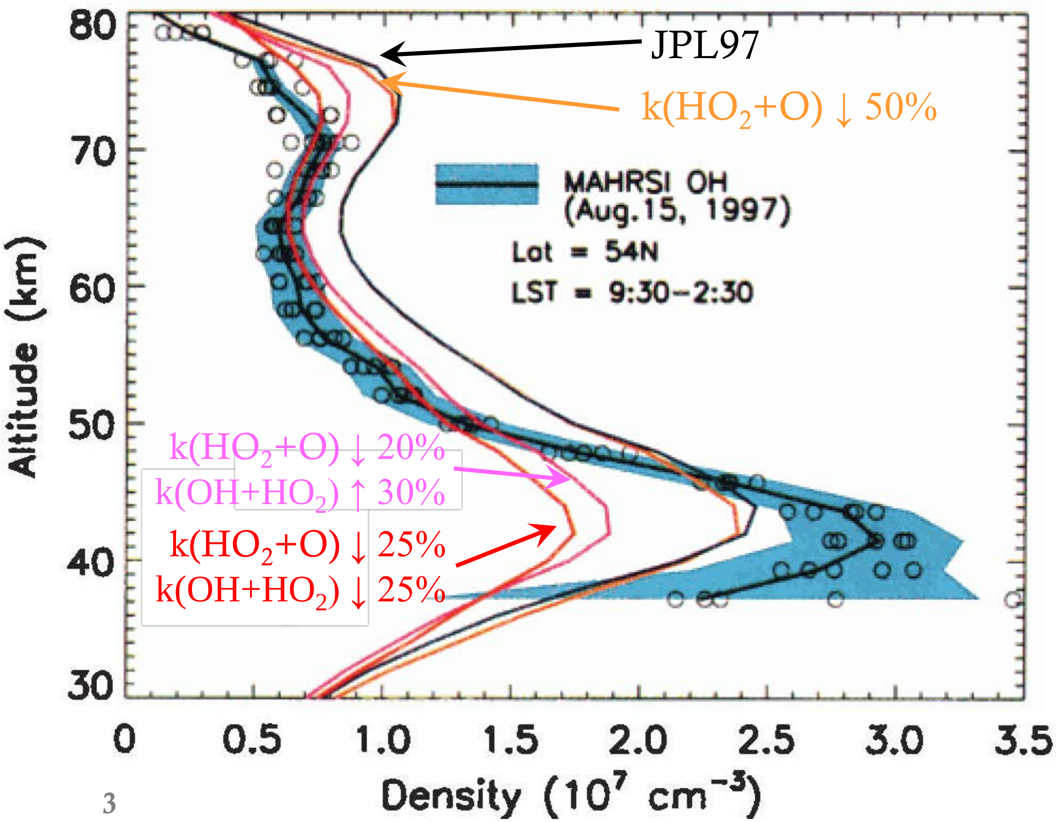
History of "OH Dilemma"

- Middle atmospheric OH are mainly produced through UV photolysis. The HO_x [$OH+HO_2$] catalytic reaction cycles plays an important role controlling O_3 loss.

[Conway et al., 2000]

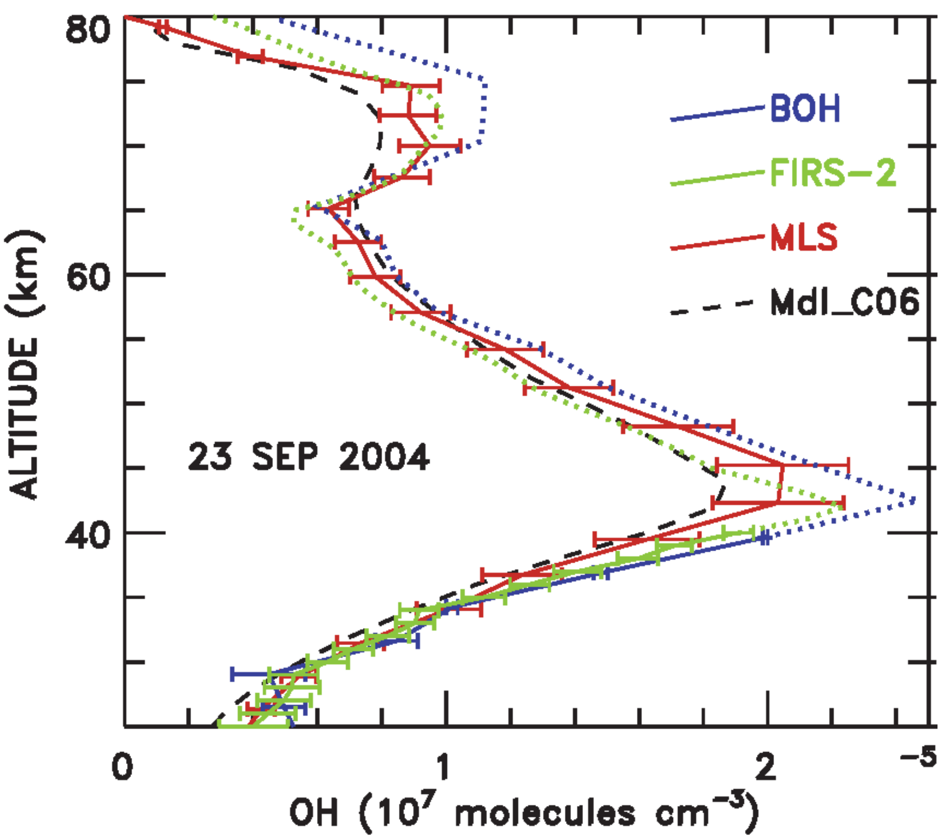
- MAHRSI, the first space-borne OH measurements in the middle atmosphere and model predictions show puzzling discrepancies.
- Model seemed to under-predict OH in the mesosphere and over-predict OH in the stratosphere.
- The shape of the observed OH profile could not be explained by standard chemistry.
- ➔ **"OH Dilemma"**



History of "OH Dilemma"

- Aura MLS observations (v2.2) show significantly higher mesospheric OH than MAHRSI.
- MLS OH profiles matched model calculations with reasonably adjusted chemistry.
- ➔ It was concluded that **there was no such OH dilemma.**

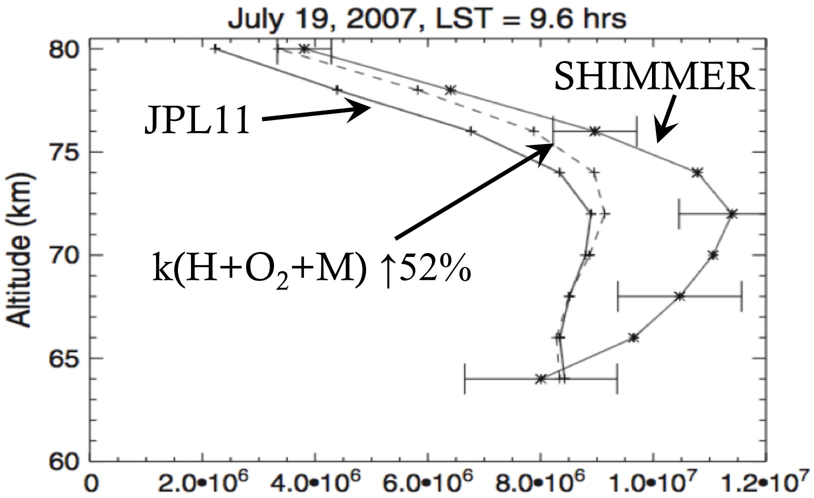
[Pickett et al., 2008]



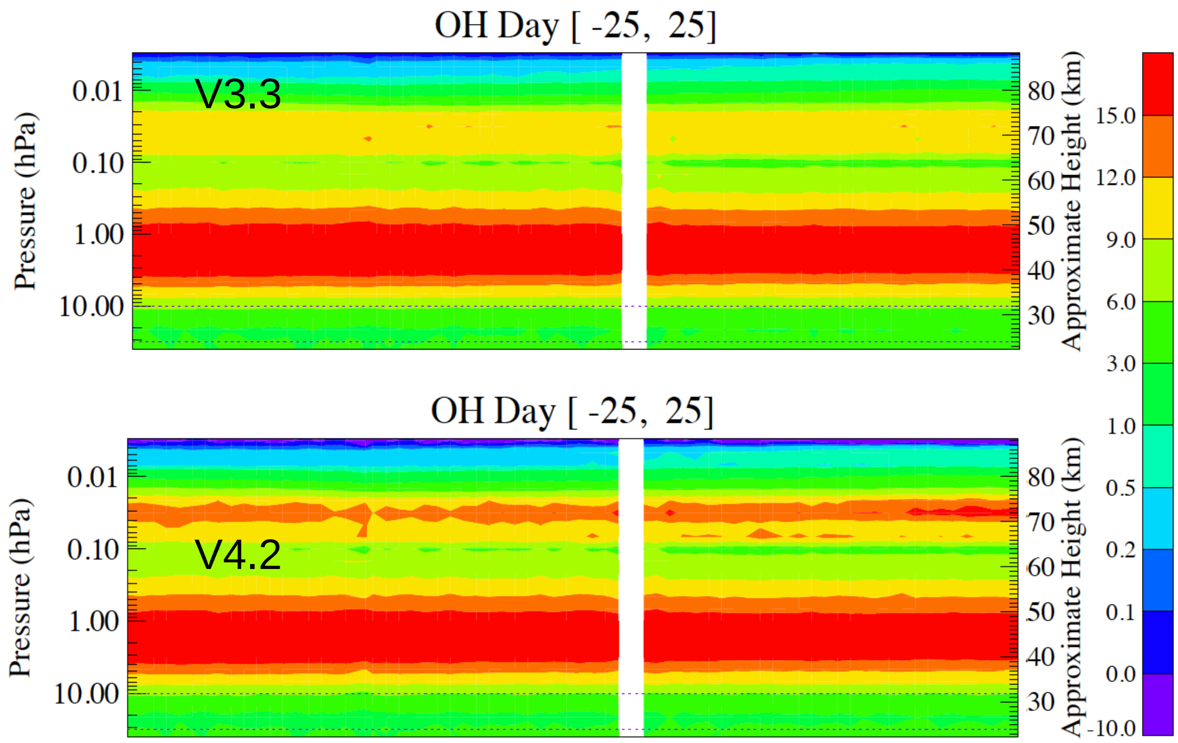
History of "OH Dilemma"

- SHIMMER mesospheric OH data were compared with model.
- Above 75 km, SHIMMER OH levels are higher than model in the morning and lower than model in the evening.
- Using a higher rate coefficient for $H + O_2 + M \rightarrow HO_2 + M$ improves the agreement, but definitely not enough.
- In particular, SHIMMER daytime OH in the entire mesosphere is higher than model. ➔ **Opposite from the original "OH dilemma"**

[Siskind et al., 2013]



The Updated Aura MLS OH Data

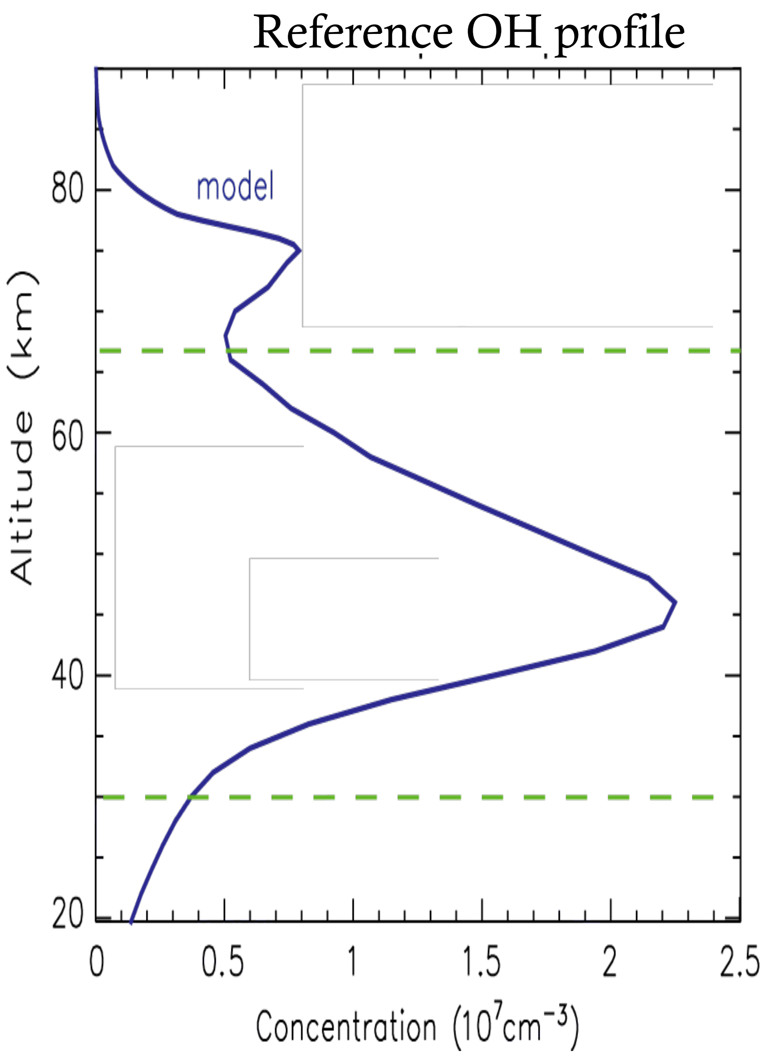


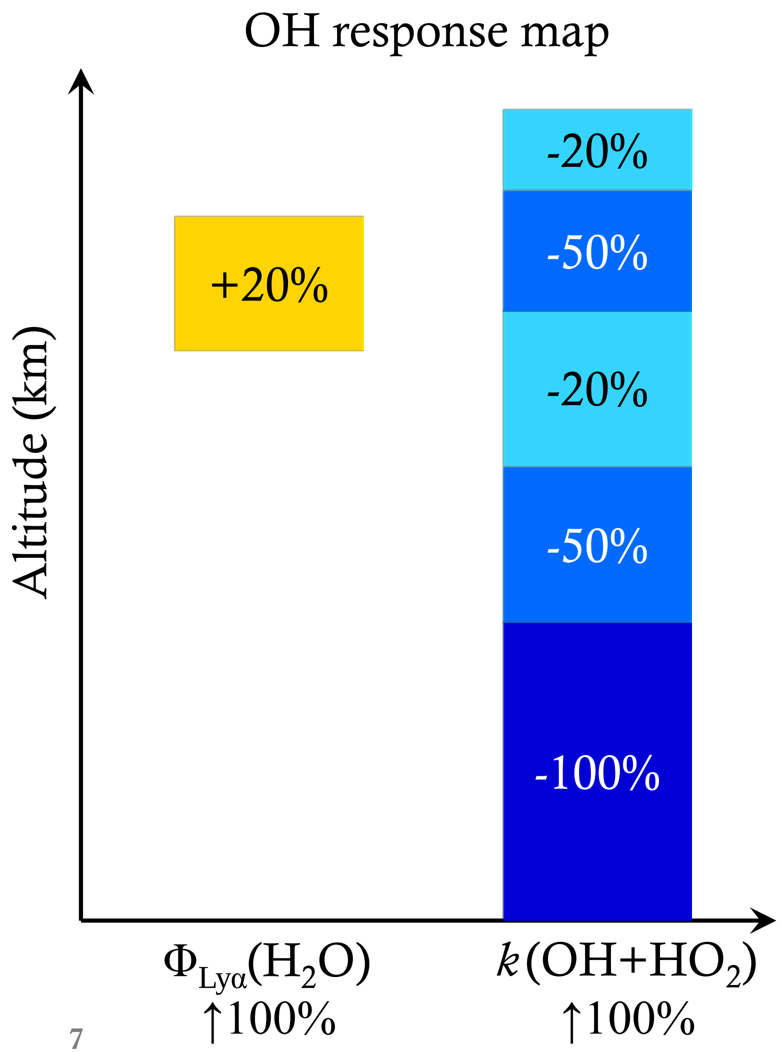
Zonal mean OH timeseries during June 2005 (v3.3 vs v4.2)

- The new MLS HO_2 offline product with extended vertical coverage allows for investigations of HO_2 as well as OH [Millán et al., 2014].

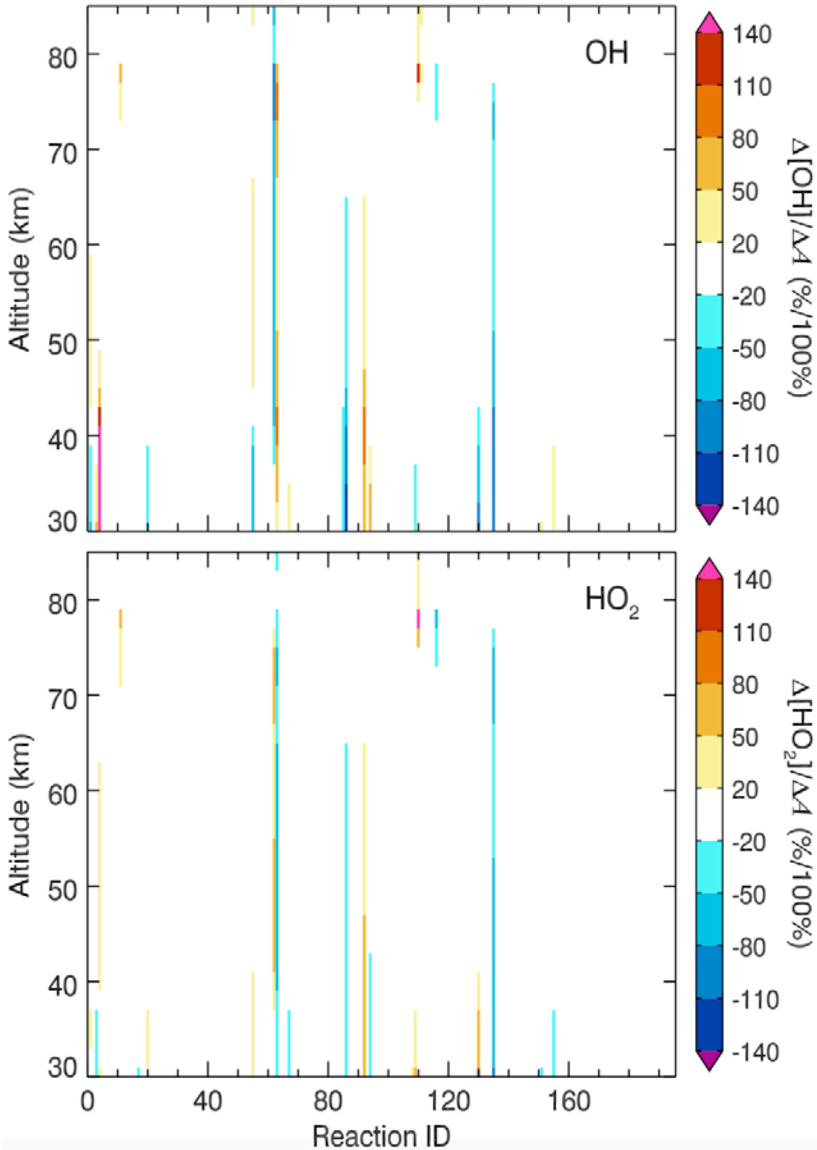
- The most recent MLS v4.2 OH data show significantly higher mesospheric OH than previous versions.
- The differences are the largest in high-OH season/latitudes.
- The change from v3.3 to v4.2 is mainly due to fixing an overly tight a priori constraint in the retrieval process [Livesey et al., 2015].

Kinetic rate perturbations





Model --- Impacts of Chemical Kinetics

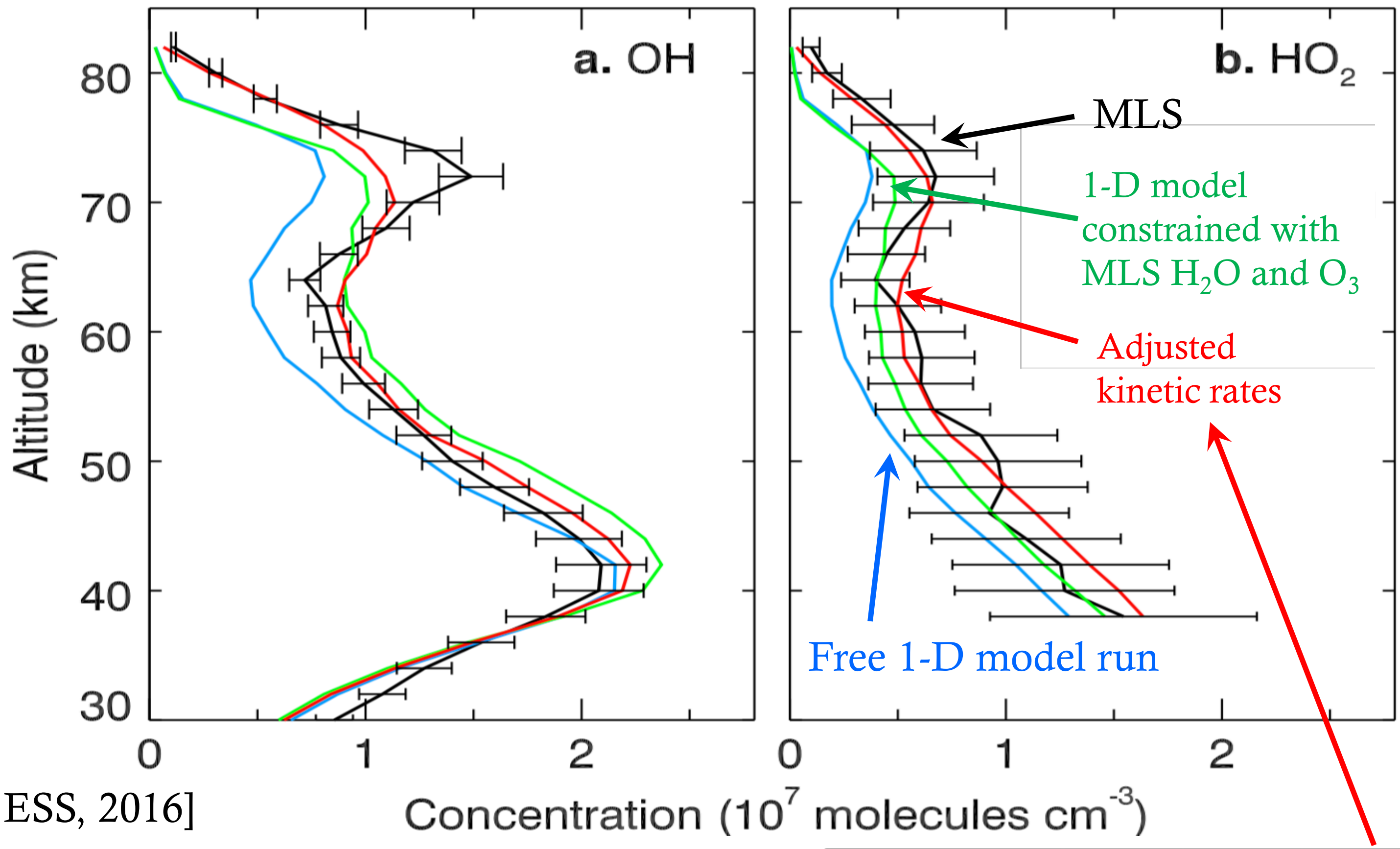


Some important reactions:

ID#	Reactions
4	$O_3 + h\nu \rightarrow O_2(^1\Delta) + O(^1D)$
11	$H_2O + h\nu \rightarrow H + OH$
62	$O + OH \rightarrow O_2 + H$
63	$O + HO_2 \rightarrow O_2 + OH$
86	$O(^1D) + N_2 \rightarrow O + N_2$
92	$H_2O + O(^1D) \rightarrow 2OH$
110	$H + O_2 + M \rightarrow HO_2 + M$
116	$H + HO_2 \rightarrow H_2 + O_2$
130	$OH + O_3 \rightarrow HO_2 + O_2$
135	$OH + HO_2 \rightarrow H_2O + O_2$

- We use the Caltech/JPL 1-D photochemical model.
- 66 levels from the surface to 130 km.
- 34 photolytic reactions and 142 bi-/ter-molecular reactions [*Li et al.*, 2017].
- We used a Bayesian optimal estimation to retrieve reaction rate coefficients for better agreements with observations.
- 4 reactions that have large impacts on OH and HO_2 are identified.

Model --- Impacts of Chemical Kinetics



[Li et al., ESS, 2016]

Conjecture:

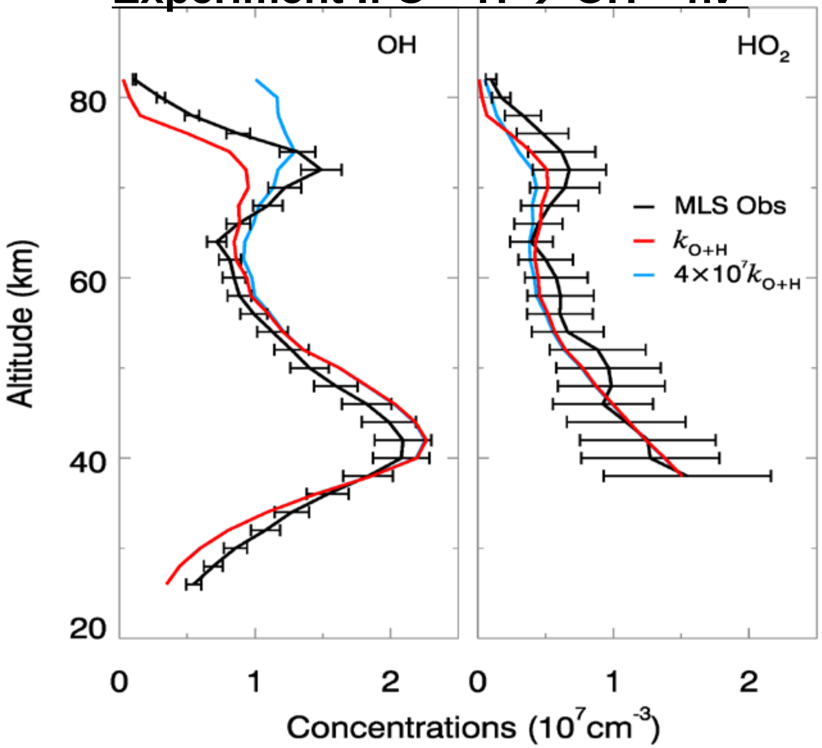
$H + O_2 + M \rightarrow HO_2 + M$ is not enough

$H + O_2 \rightarrow HO_2 + h\nu$ may be important in Earth's mesosphere

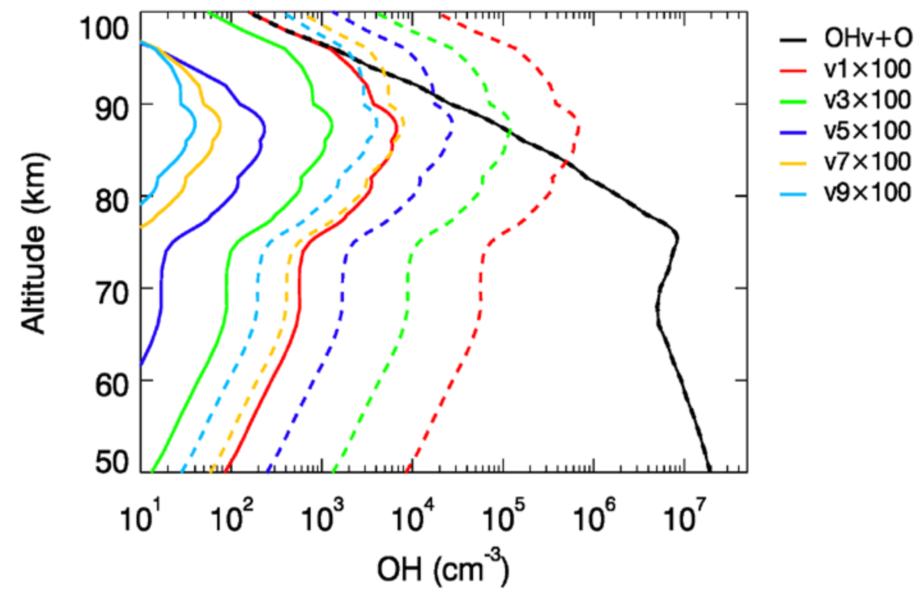
#110	$H + O_2 + M \rightarrow HO_2 + M$	+310%
#4	O_2 absorption at Lyman α	-33%
#62	$O + OH \rightarrow O_2 + H$	+12%
#135	$OH + HO_2 \rightarrow H_2O + O_2$	-10%

Further Explorations

Experiment I: O + H → OH + hν



Experiment II: Excited State OH Reaction



- O + H is an important pathway to produce OH in the interstellar medium. Due to the steep vertical gradient of H, introducing this reaction causes higher mesospheric OH without changing stratospheric OH.

- However, the reaction rate coefficient is too low to cause significant impacts.

- $OH^*_{(v>=5)} + O(^3P) \rightarrow OH_{(lower\ v)} + O(^1D)$

  $O(^1D) + H_2O \rightarrow 2OH$

- This new reaction involving O and excited sate OH was recently proposed [Kalogerakis et al., 2016].

- The abundance of $OH^*_{(v>=5)}$ is much smaller than OH, the impact of this reaction is found to be negligible.

Summary

- The 20-year-old "OH dilemma" remains, although in a different way.
- The new v4.2 MLS data show higher mesospheric OH than model prediction.
- Adjustments of kinetic rates do not resolve this model-obs discrepancy.
- Chemical reactions not included in standard model may be required.

Take-Home Message

Considering the long-standing ozone deficit problem above 45 km, it is important to continue investigating the gaps between observations and model in upper stratospheric/lower mesospheric OH.