Diurnal variation of O_3 during the Polar day above Spitsbergen

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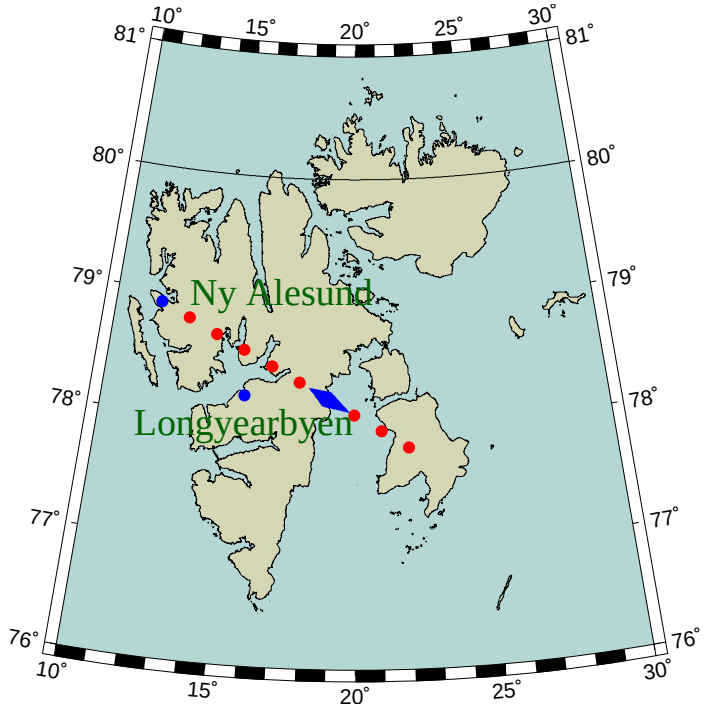
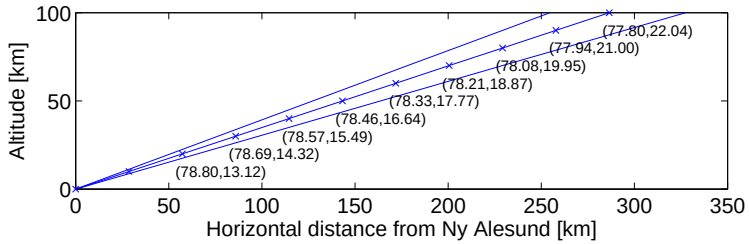
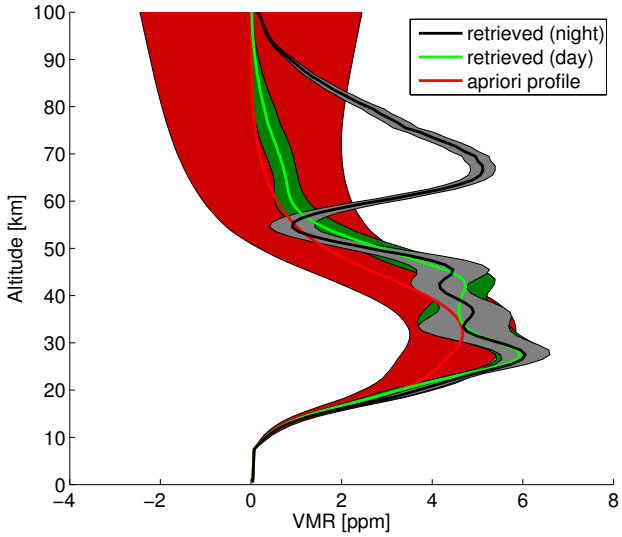
Measurement geometry



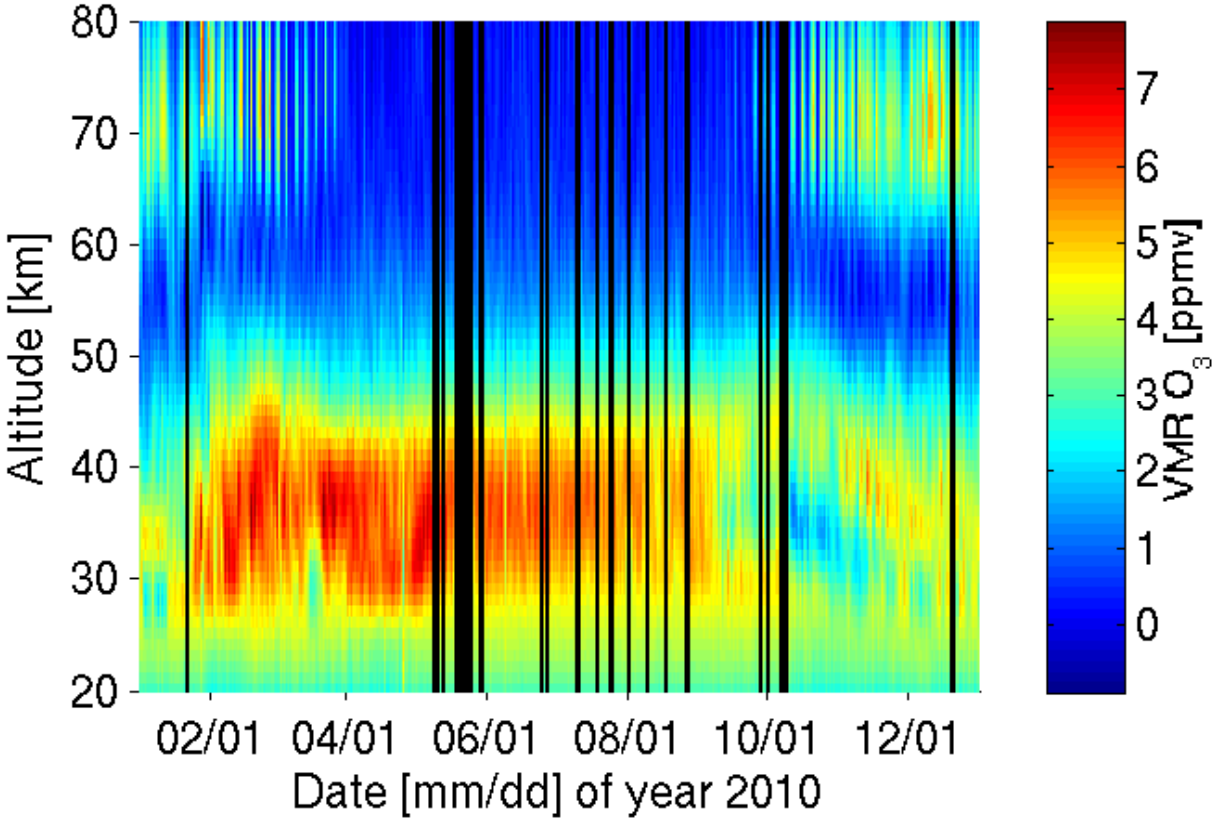


- Geoposition 78.9N, 11.9E, Altitude 10m
- Elev. 20°, Azi. 113°

- O_3 emission, 142.175 GHz
- Res. 60 kHz, Bandwith 900MHz
- Integration time, 1 hour
- $T_{\mathrm{REC}} \approx 1200K$



Observation



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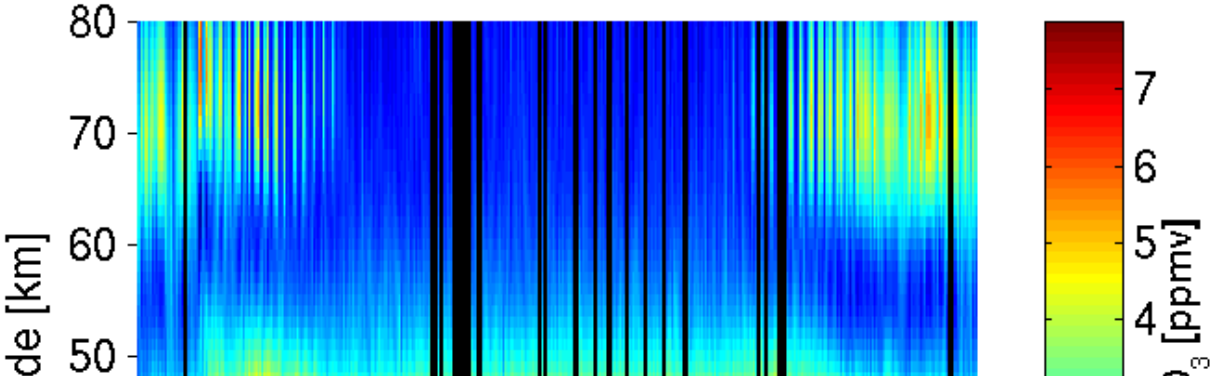

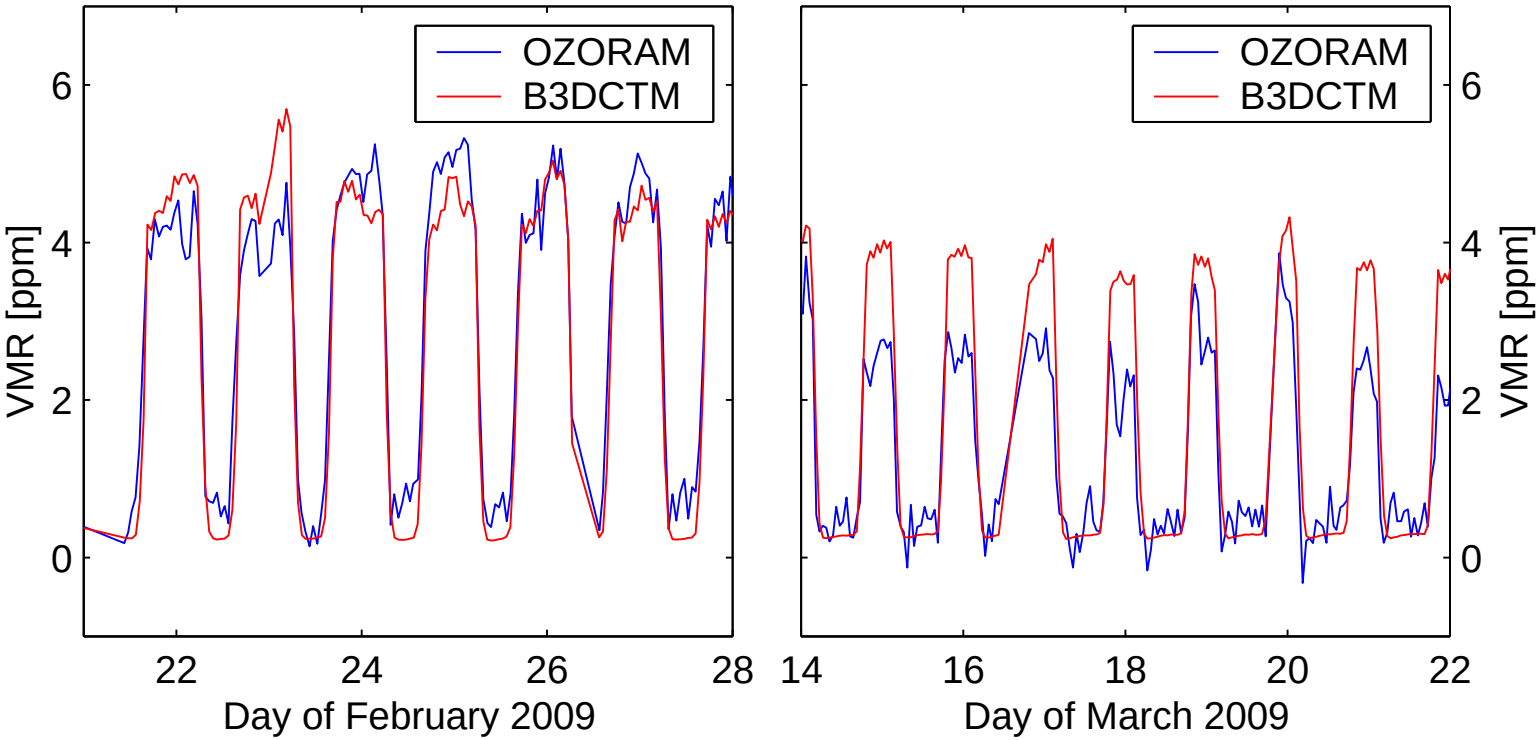

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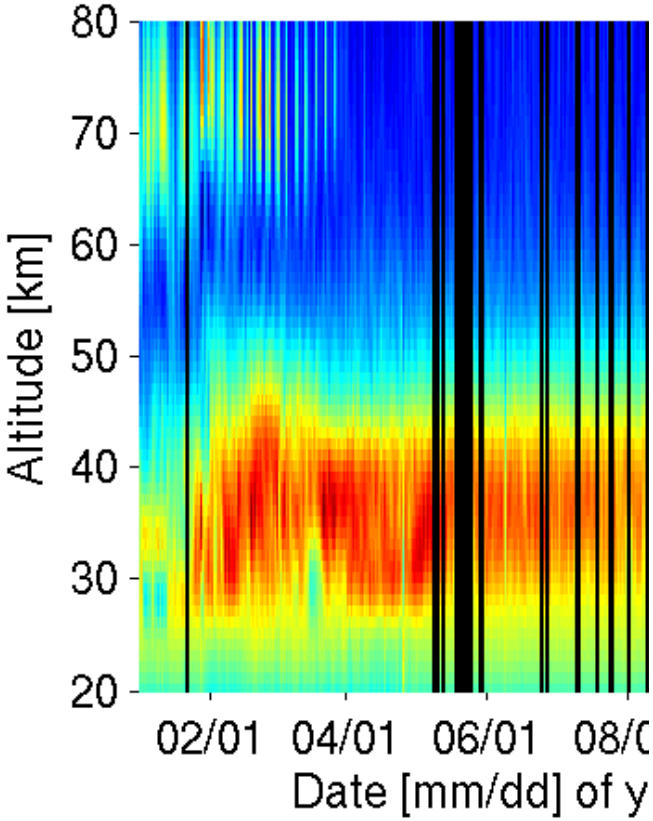
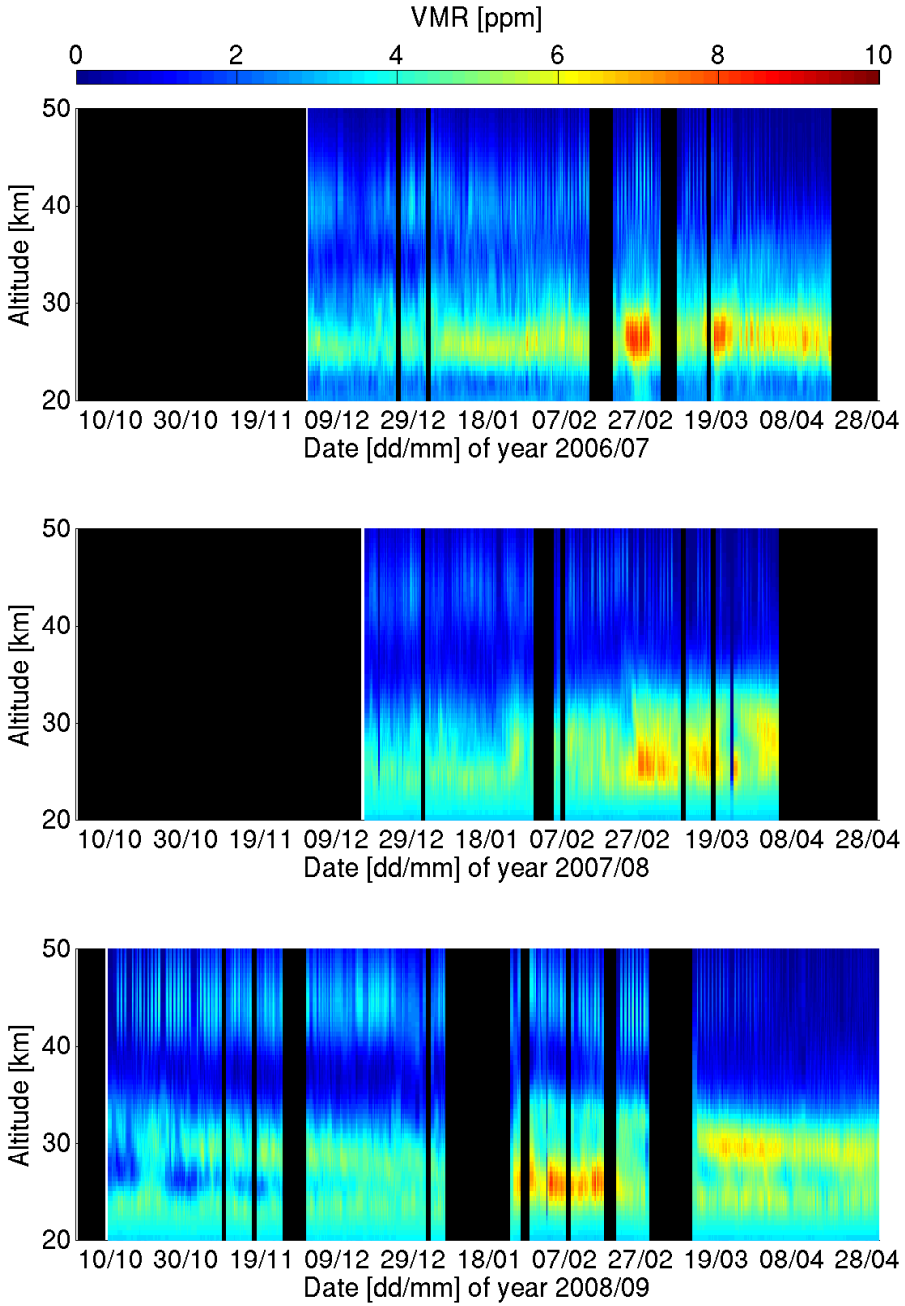
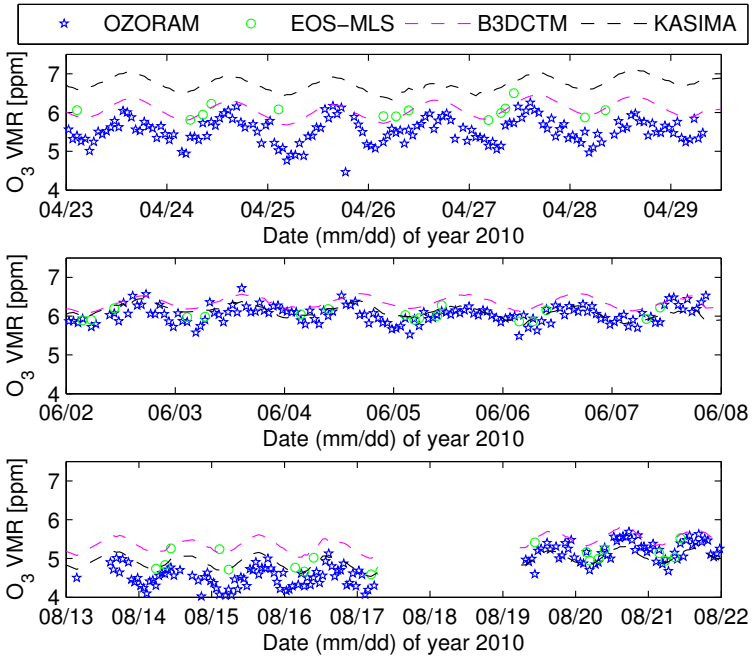




Overview



- measurements at 37 km altitude
- clearly discernible diurnal cycle
- MLS data do not cover a full diurnal cycle

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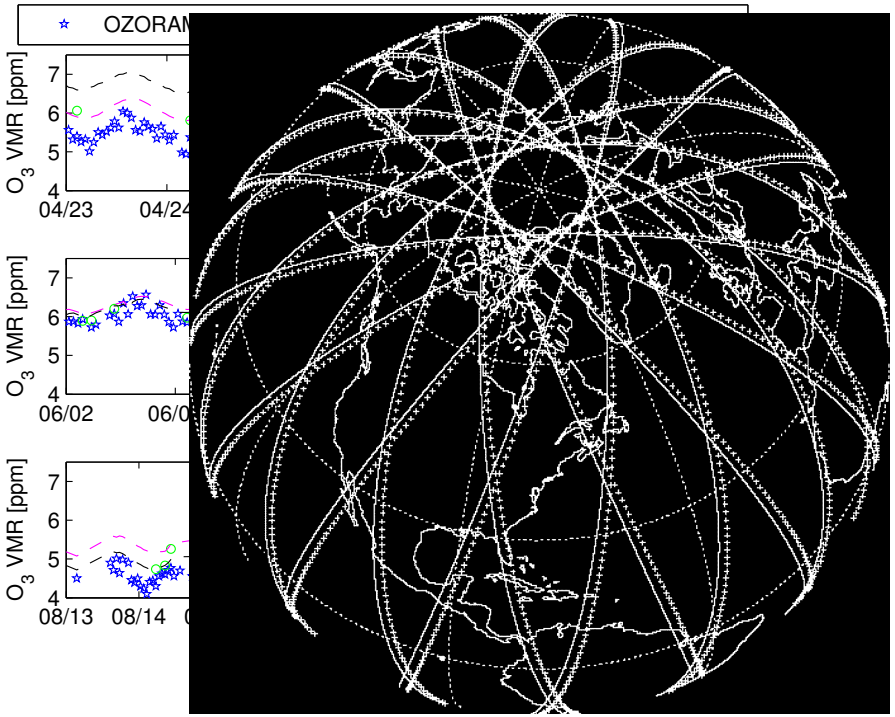

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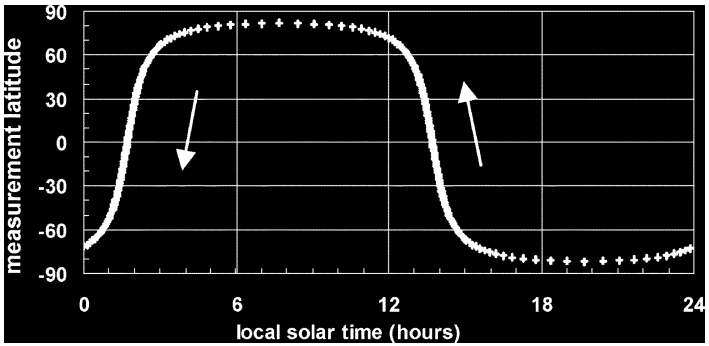

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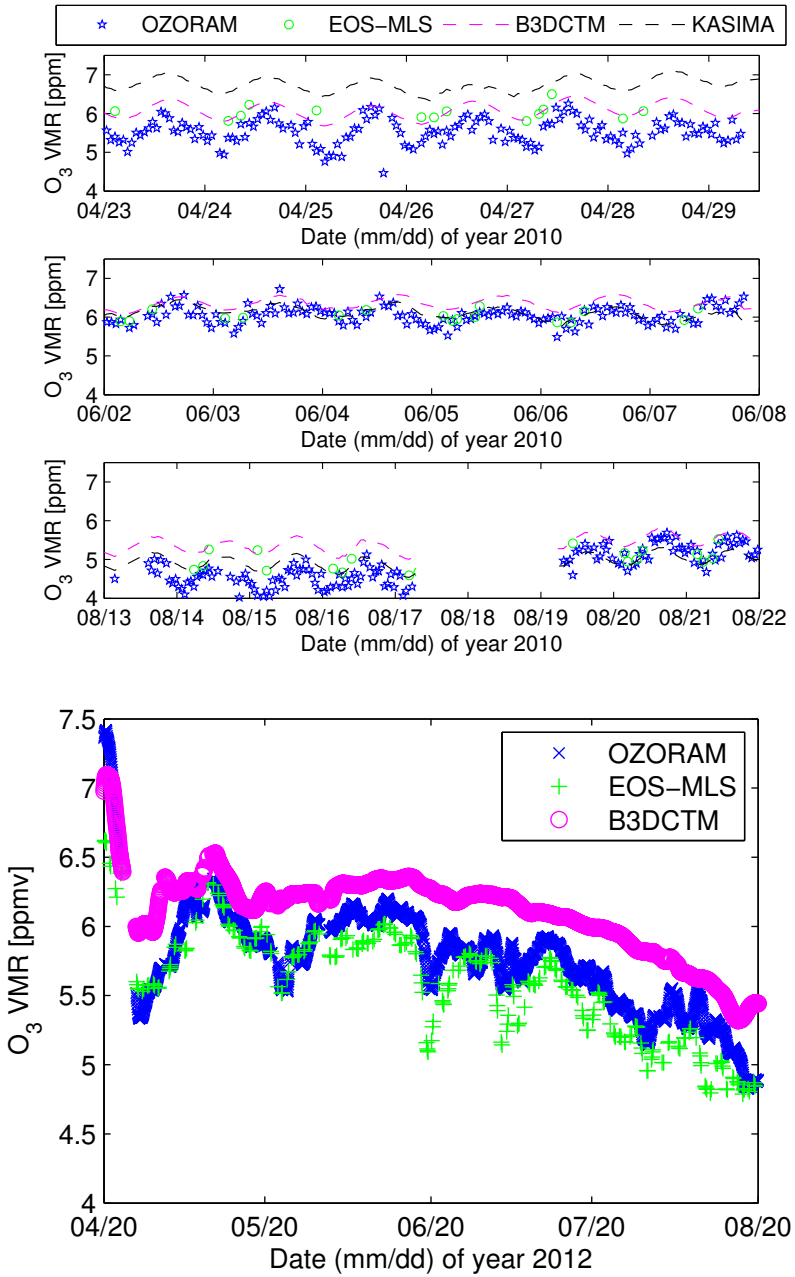


- measurements at 37 km altitude
- clearly discernible diurnal cycle
- MLS data do not cover a full diurnal cycle
- O3 depletion in KASIMA model stronger than in measurements
- O3 depletion in KASIMA model weaker than in measurements
- modelled diurnal cycle smaller in spring

Chemistry of O_3 in the upper stratosphere I

$$\text{J2: } \mathrm{O_2} + \mathrm{h}\nu(\lambda \leq 242.4\mathrm{nm}) \longrightarrow \mathrm{O}(^3\mathrm{P}) + \mathrm{O}(^3\mathrm{P}) \quad (1)$$

$$\mathrm{O}(^3\mathrm{P}) + \mathrm{O_2} + \mathrm{M} \longrightarrow \mathrm{O_3} + \mathrm{M} \quad (2)$$

$$\mathrm{O}(^3\mathrm{P}) + \mathrm{O_3} \longrightarrow 2\,\mathrm{O_2} \quad (3)$$

Ozone is photo-dissociated by

$$\text{J3: } \mathrm{O_3} + \mathrm{h}\nu(\lambda \geq 320\mathrm{nm}) \rightarrow \mathrm{O_2}(^3\Sigma^-_\mathrm{g}) + \mathrm{O}(^3\mathrm{P}) \quad (4)$$

$$\text{J3A: } \mathrm{O_3} + \mathrm{h}\nu(\lambda \leq 320\mathrm{nm}) \rightarrow \mathrm{O_2}(^1\Delta_\mathrm{g}) + \mathrm{O}(^1\mathrm{D}) \quad (5)$$

- O_3, $O(^3P)$ and $O(^1D)$ form Ox-family
- lifetime of Ox-family longer than its constituents
- During polar day is Ox chemically controlled

Chemistry of O_3 in the upper stratosphere II

Catalytic removal of Ox in the upper stratosphere:

$$\mathrm{NO} + \mathrm{O_3} \longrightarrow \mathrm{NO_2} + \mathrm{O_2} \quad (6)$$

$$\mathrm{NO_2} + \mathrm{O}(^3\mathrm{P}) \longrightarrow \mathrm{NO} + \mathrm{O_2} \quad (7)$$

$$\mathrm{NET}: \mathrm{O_3} + \mathrm{O}(^3\mathrm{P}) \longrightarrow 2\,\mathrm{O_2} \quad (8)$$

The main source of NO in the stratosphere is the reaction

$$\mathrm{N_2O} + \mathrm{O}(^1\mathrm{D}) \longrightarrow 2\,\mathrm{NO} \quad (9)$$

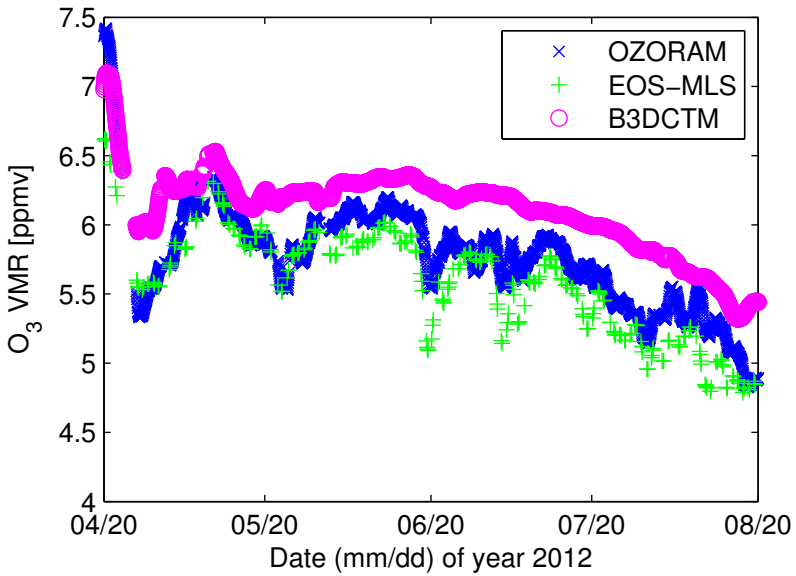
which also removes N_2O from the stratosphere. The reactions

$$\mathrm{N_2O} + \mathrm{O}(^1\mathrm{D}) \longrightarrow \mathrm{N_2} + \mathrm{O_2} \quad (10)$$

$$\mathrm{N_2O} + \mathrm{h}\nu(\lambda \leq 200\mathrm{nm}) \longrightarrow \mathrm{O}(^1\mathrm{D}) + \mathrm{N_2} \quad (11)$$

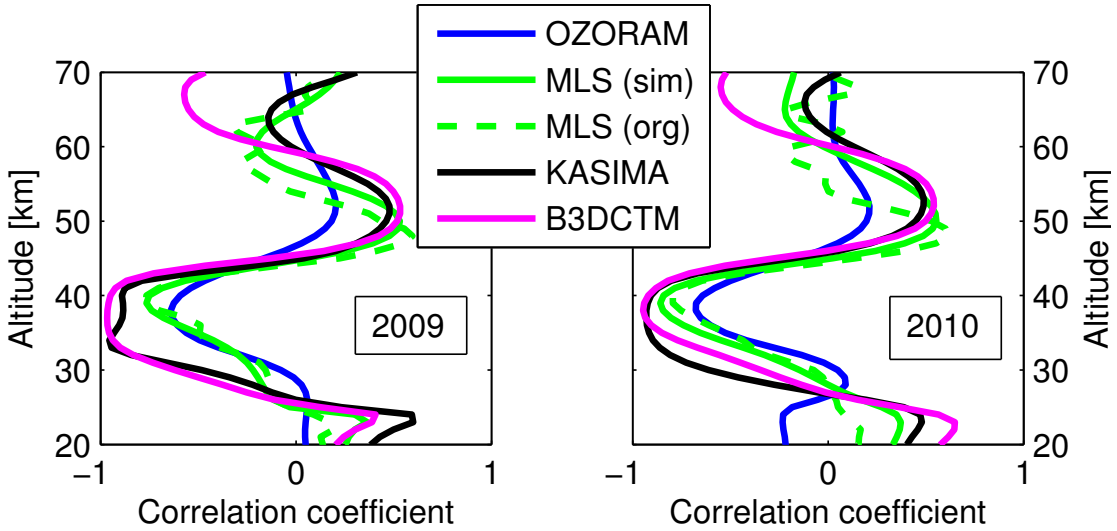
removes N_2O from the stratosphere and produces Ox

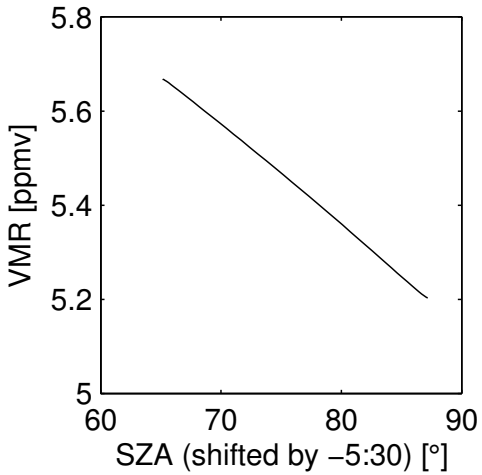
O_3 variation during polar day



- Variance in O_3 only partly covered
- O3 depletion in B3DCTM smaller

Diurnal variation of O_3-chemistry

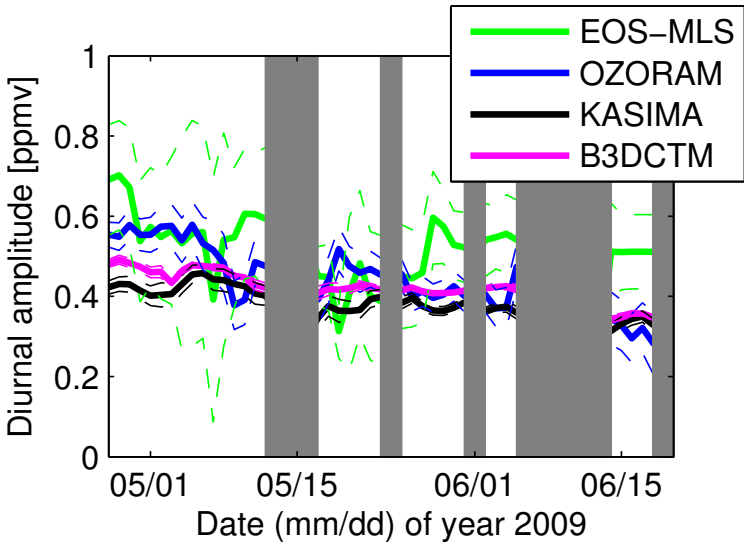


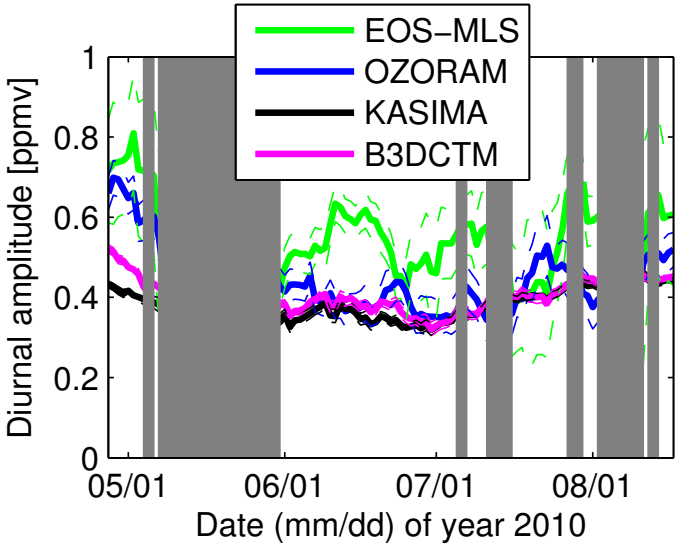


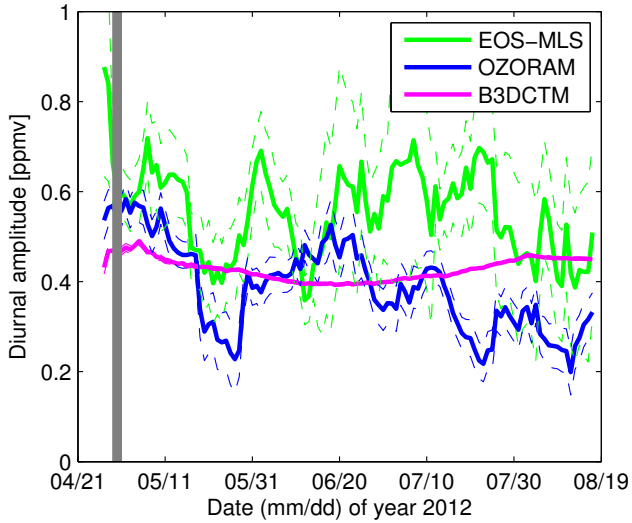
- photolysis the natural candidate for the diurnal cycle
- diurnal variation of O_3 linear to the SZA (lagging 5h) behind:

$$x_{O_3} = a * SZA_{t-5h} + b$$

The diurnal amplitude of O_3



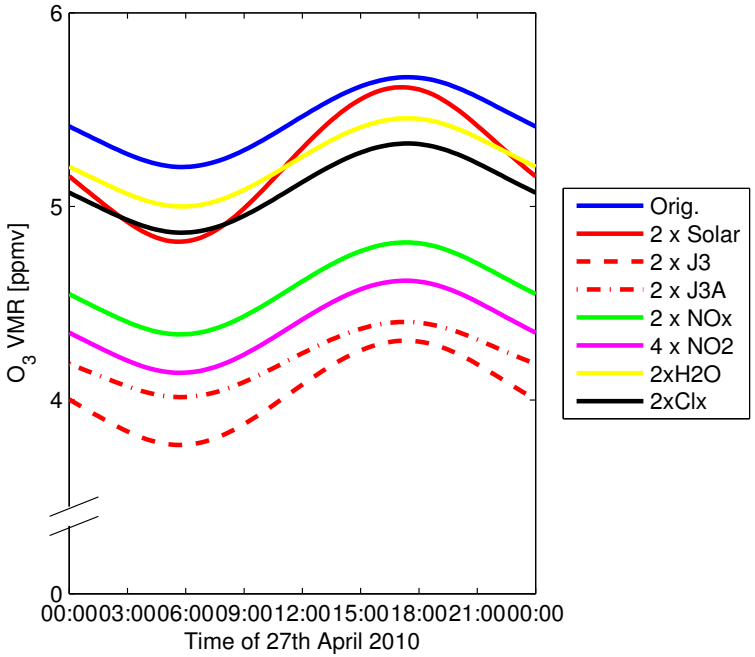




- measured diurnal amplitude higher and more variable than modelled one.

The diurnal amplitude of O_3

parameter increased	diurnal amplitude [ppmv]	diurnal amplitude normalized	influence on O3 VMR
none	0.46	0.08	–
Solar irradiation	0.79	0.15	decrease
NO_2	0.48	0.1	decrease
NOx	0.48	0.1	decrease
Clx	0.46	0.09	
H_2O	0.45	0.08	
*J*2	0.9	0.1	
*J*3	0.5	0.13	
*J*3*	0.38	0.09	
J_{N2O}	0.46	0.1	
Diurnal amplitude measured and modele			
OZORAM	0.65	0.13	
B3DCTM	0.52	0.09	
KASIMA	0.43	0.07	



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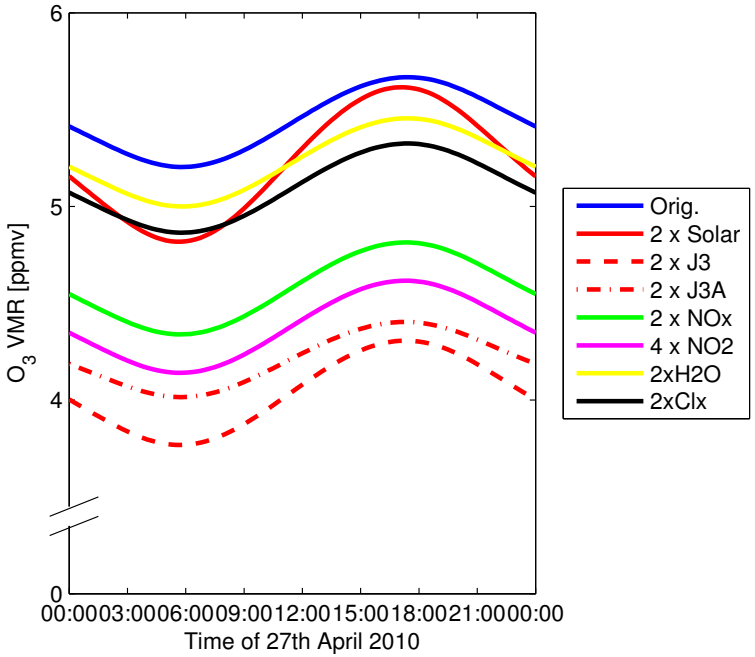

Summary

- ground based millimeterwave radiometry of O_3 suitable for short term observations
- O_3 strongly correlated to SZA wioth a time lag of 5 h.
- measured O_3 diurnal cycle higher in spring than modelled one and more variable throughout the year.
- O_3 depletion not well matched in both models
- possible reason wrong photolysis constants