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Validation of methane tropospheric column observed by FTIR at Tsukuba with airplane sampling

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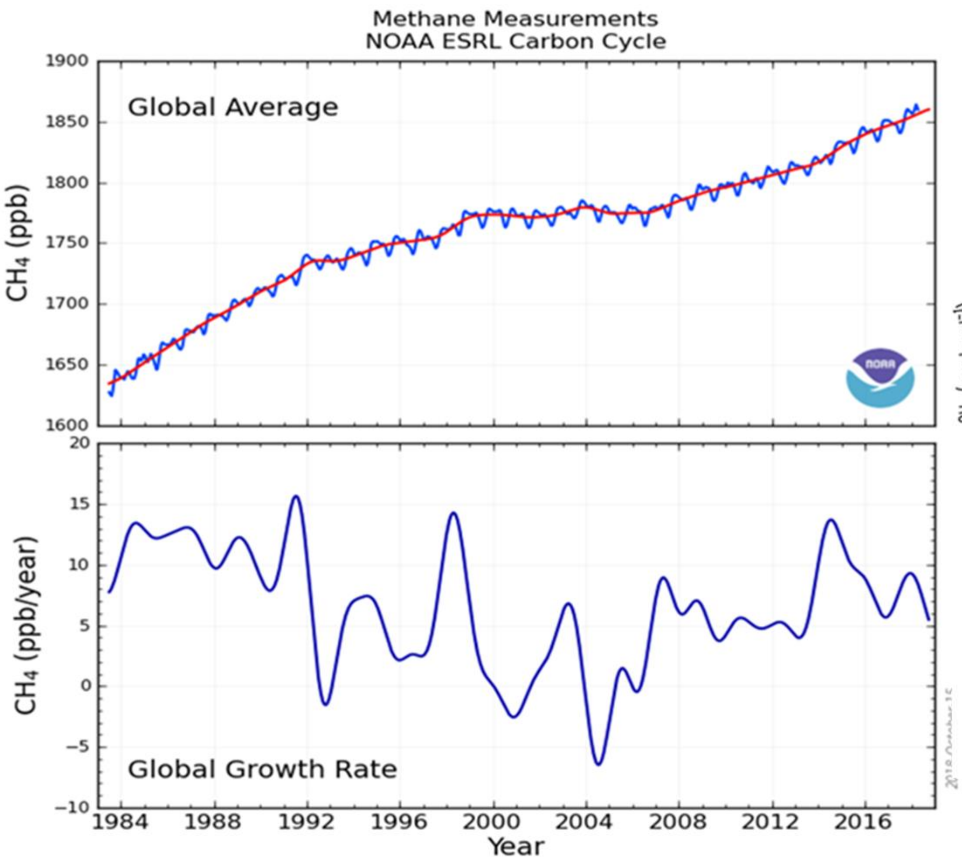
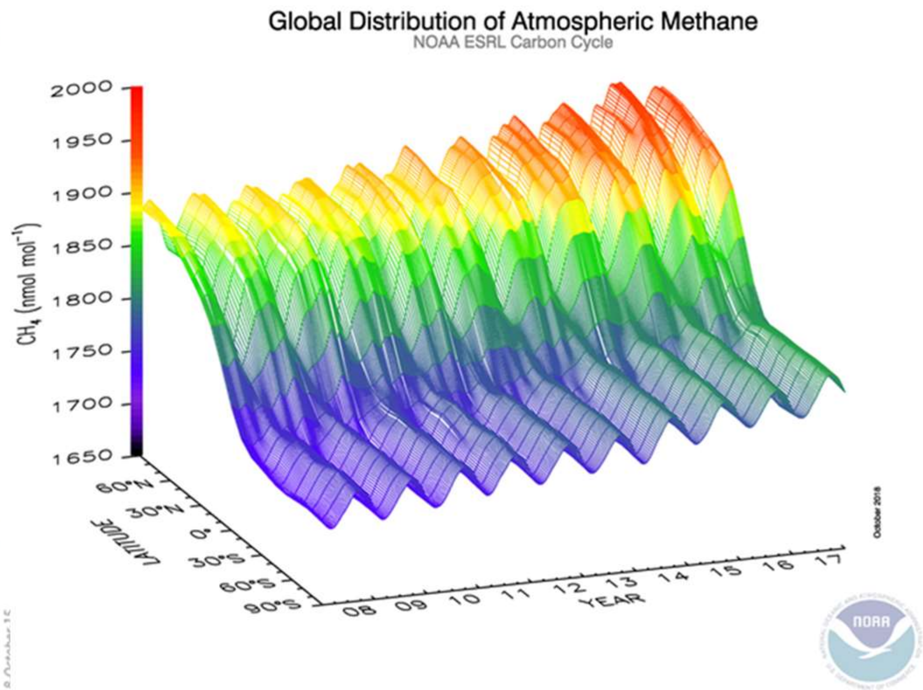
1. Introduction



Global Average of CH_4 concentration and growth rate



[National Oceanic and Atmospheric Administration / Earth System Research Laboratory (NOAA ESRL) Carbon Cycle website]

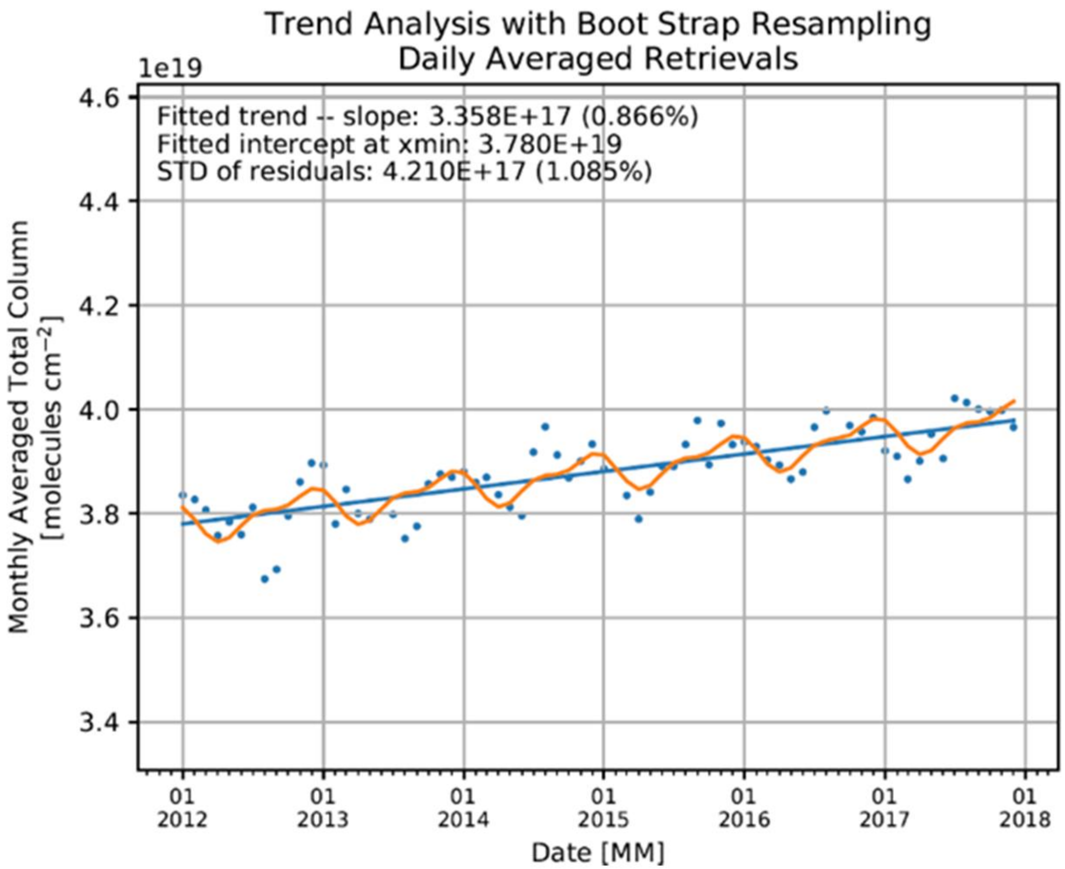
The growth rate varies year by year.
There are many sources (wetland, livestock, fossil fuels, biomass burning, etc.)
The cause of the variability isn't understand well.

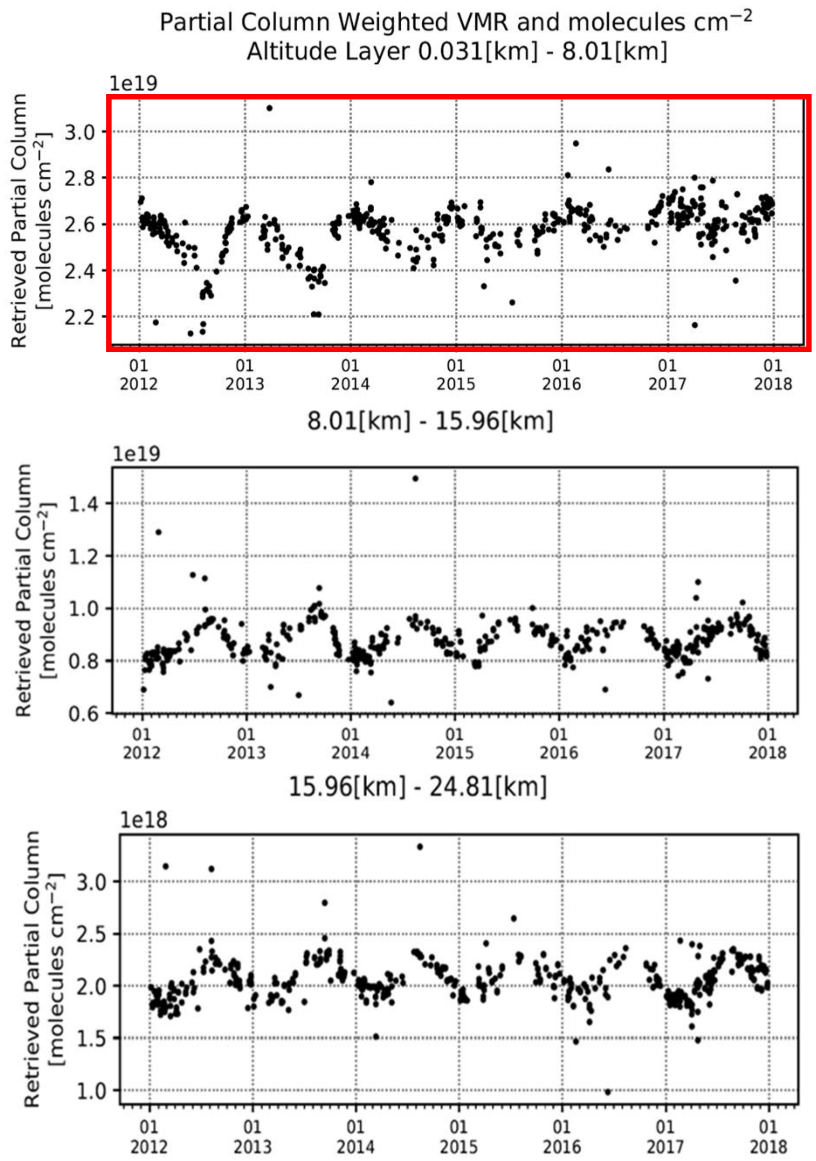
Spatial distribution is also varies with latitude.

90% of the sink is the reaction with OH.

It is important to monitor CH_4 concentration continuously in global scale.

1. Introduction





We can get 3 independent information from FTIR analysis because the DOFs for the CH_4 retrieval is around 3.
Tsukuba result (above) indicate that the increase occurs in the troposphere.

But the validation for the partial columns isn't enough.

Purpose of this study

1. Compare some of fitting parameters for methane retrieval from Tsukuba spectra and optimize them.
2. Validate the retrieved tropospheric columns of methane by the comparison with airplane sampling results.

* Validation for the spectra taken at high humidity station (Tsukuba) will be useful for the analysis in the other stations.

2. Instrument (FTIR)

Bruker 125HR at Tsukuba
（36.1^{o}N, 140.1^{o}E, 31 m A.S.L.）

Period for this analysis
Jan. 2012 - Dec. 2017

Measured spectra: 518

Spectra taken with Filter #3 were used
(Wavenumber range: 2400-3200cm^{-1})

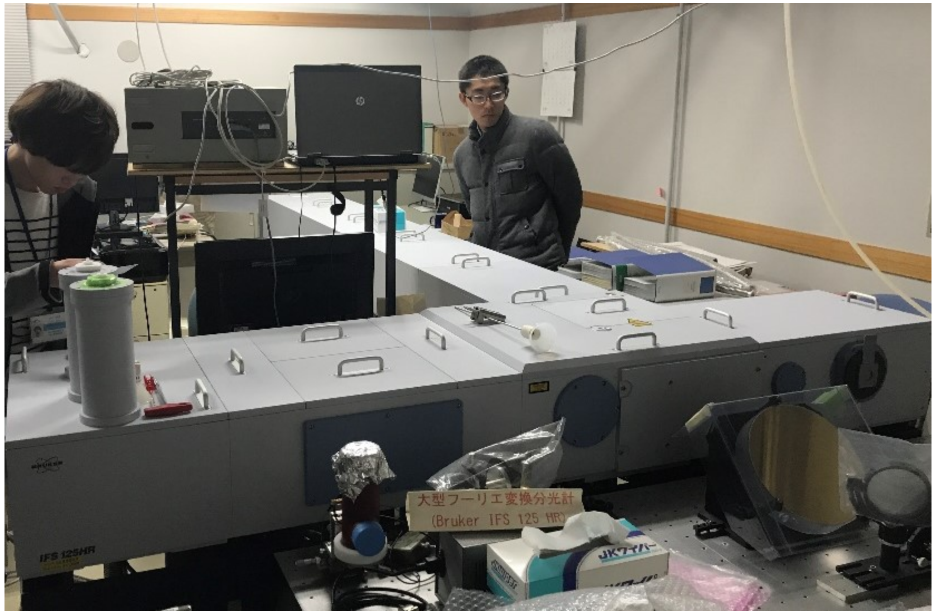
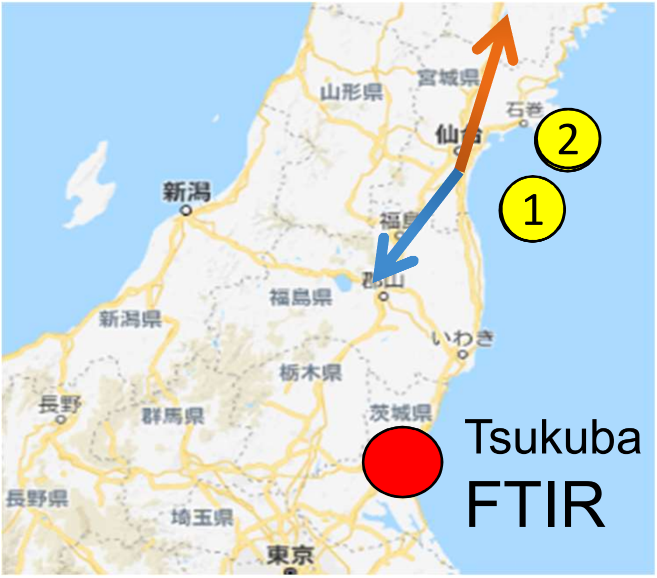


Figure1. FTIR（Bruker 125HR）

2. Airplane sampling



Areas of airplane samplings

<u>Airplane sampling (once/month)</u>

2012-2017: Total 66 observations

<u>Cessna Altitude: 0 – 4 km   14 points</u>

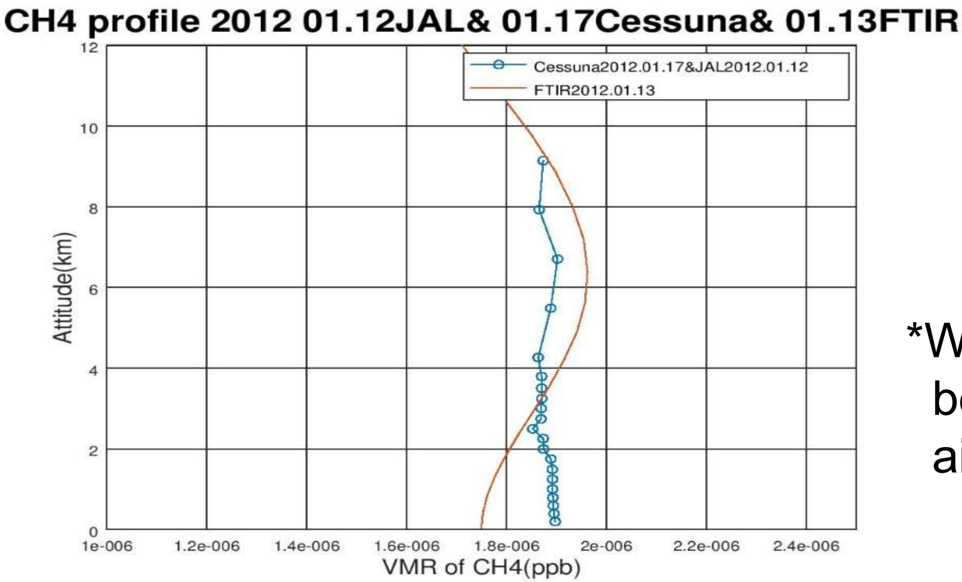
① Jan. 2012 – Mar. 2015 (Fukushima)

② Apr. 2015 – Dec. 2017 (Ishinomaki)

<u>JAL   Altitude: 4 – 9 km   7 points</u>

↗ Jan. 2012 – Jan. 2017 (Sendai – Chitose)

↙ Feb. 2017 – Dec. 2017 (Sendai – Osaka)



Sample for methane profiles observed by FTIR and airplane sampling

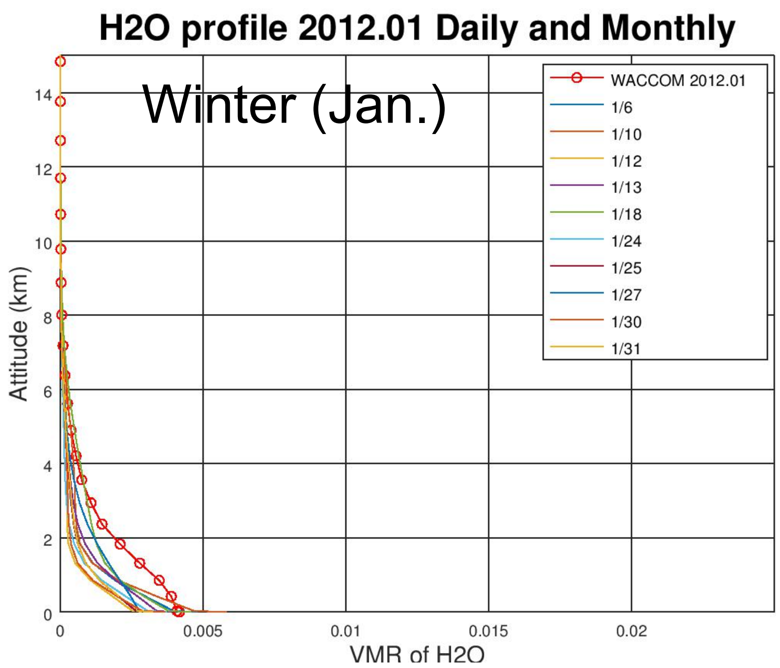
Observed by Center for Atmospheric and Oceanic Studies (CAOS), Tohoku University

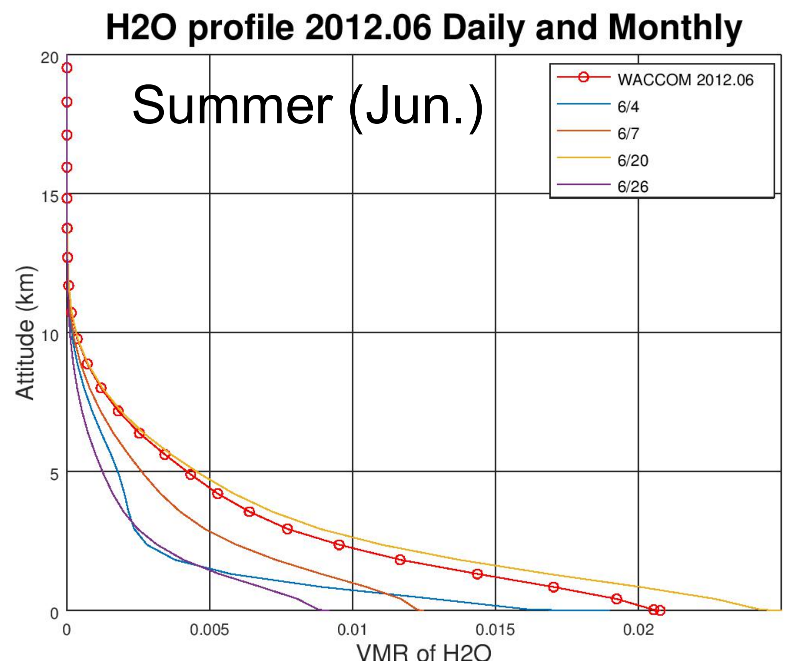
Accuracy: <u>0.003ppm</u>

*We compared the results by monthly mean because the dates and places of FTIR and airplanes were different.

3. Comparison of fitting parameters

Initial profile of Water vapor





Seasonal and daily variation of water vapor at Tsukuba is very large

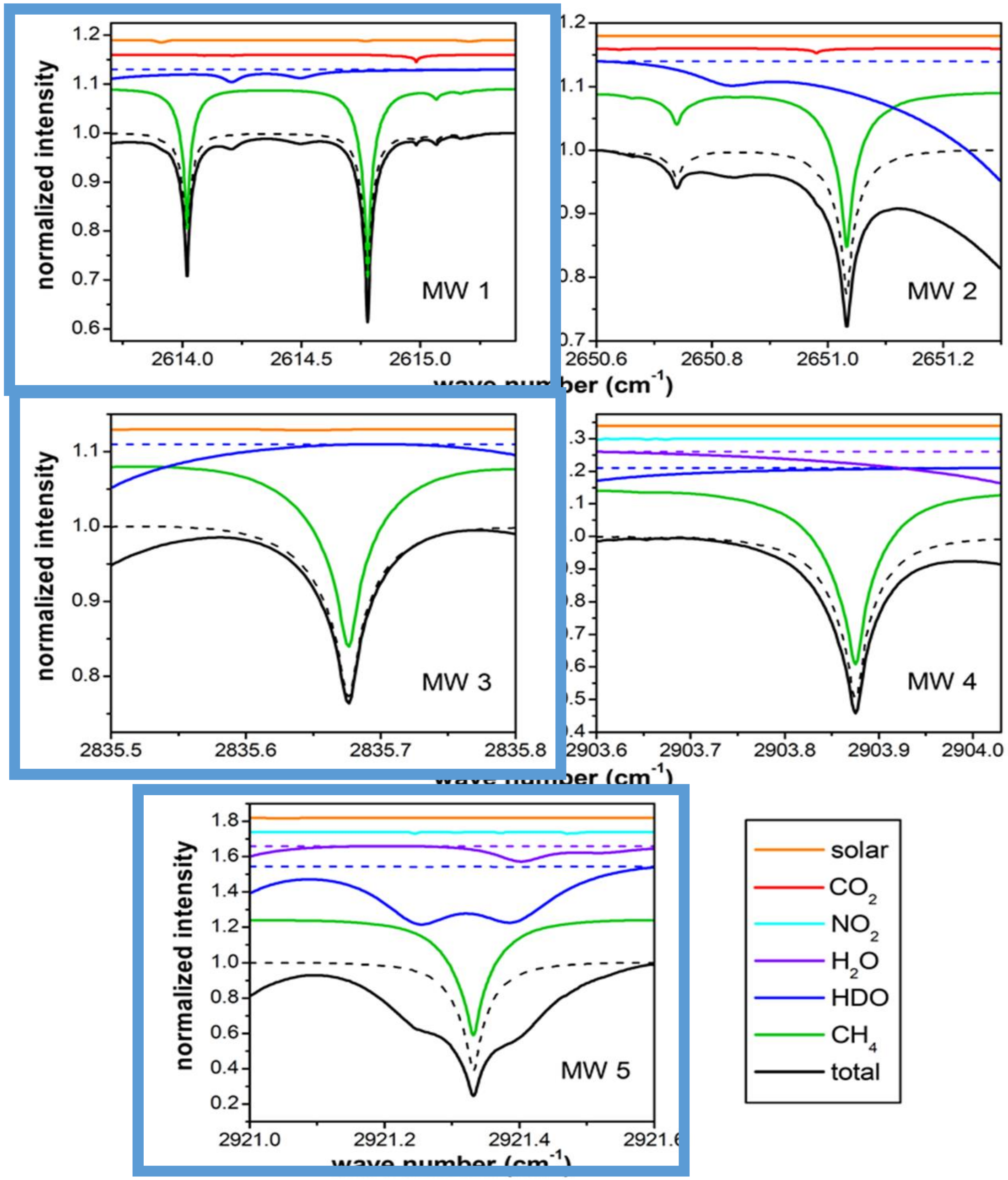
⟹ Initial profile is important

We tested two kind of initial profiles for Water vapor

v1：WACCM monthly average

v5：Daily average of pre-retrieved profiles using the spectra take the same day

Selection of Micro Windows



Sussmann et al., [2011] recommends to use 3 micro windows (MW) of 5 MWs for methane retrieval at the high humidity station.

We tested two kinds of MWs

3MW:

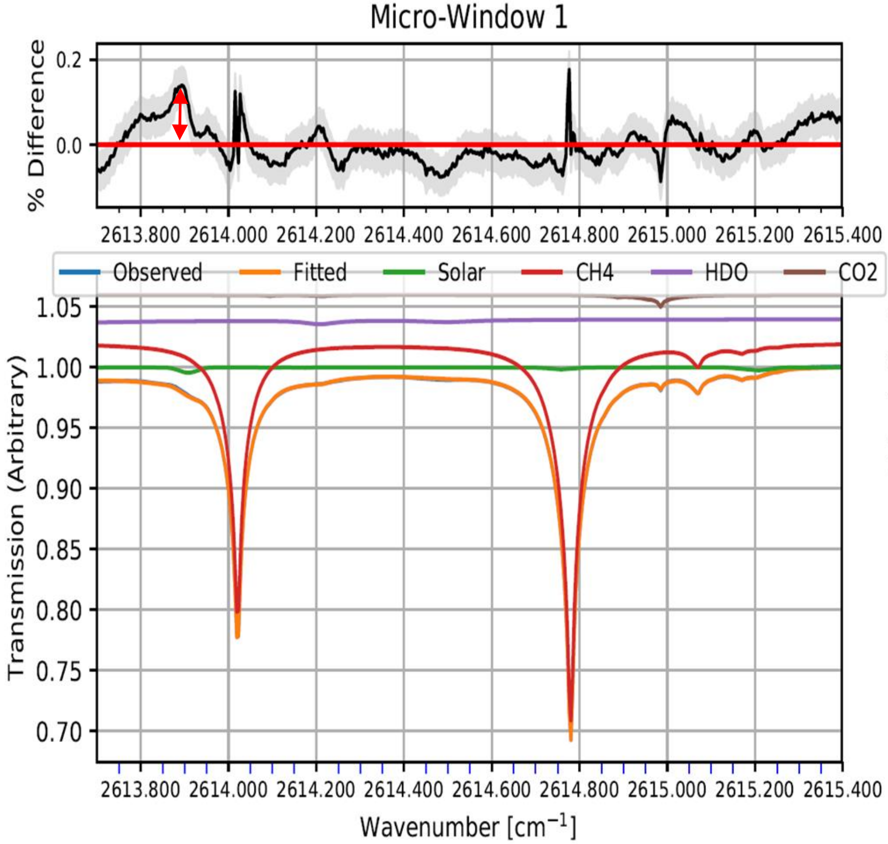
MW1, MW3, and MW5

5MW:

All 5 MWs

RMS and DOFs as a indicator for fitting quality

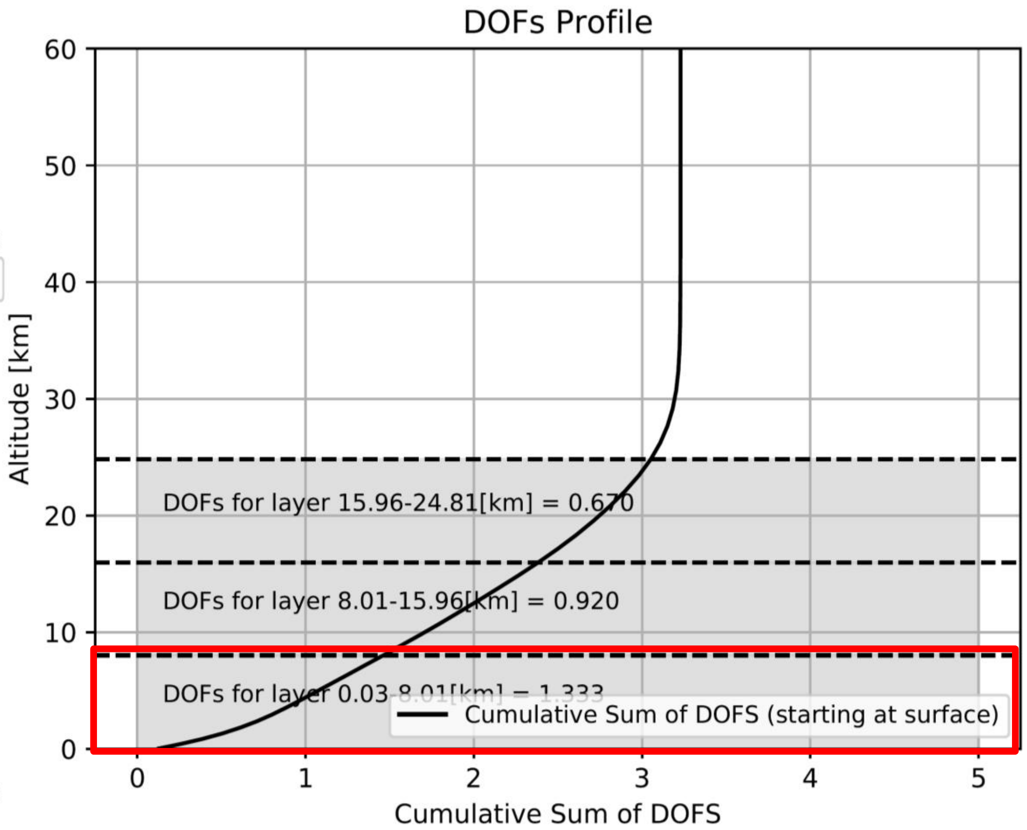
RMS: Root Mean Square Residual



Sample of the spectral fitting for CH_4

Smaller RMS is better

DOFs: Degree of Freedoms



Sample of the DOFs for CH4 retrieval

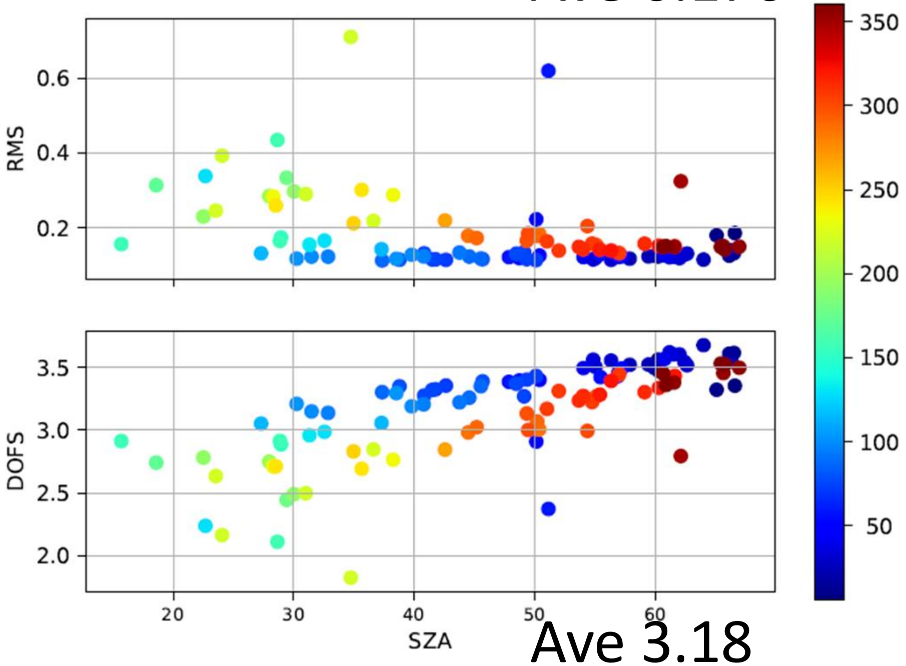
Larger DOFs is better

*The comparisons were performed for all spectra taken in 2012 (one year).

Results: 3MW v1 vs. 5MW v1

3MW v1

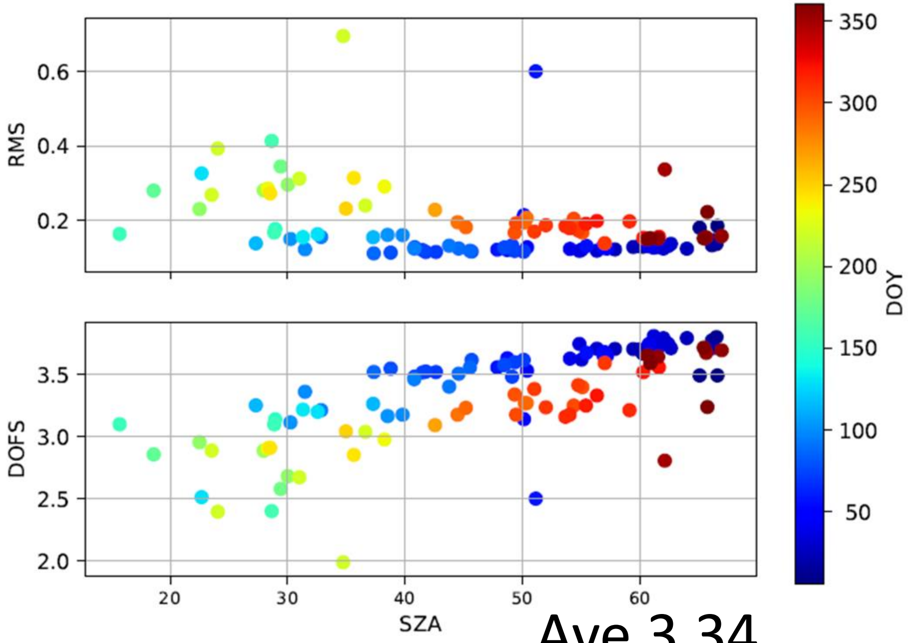
Ave 0.176



Ave 3.18

5MW v1

Ave 0.185



Ave 3.34

RMS

3MW is better than 5MW → 3MW has higher-quality information
3MW is less affected by water vapor

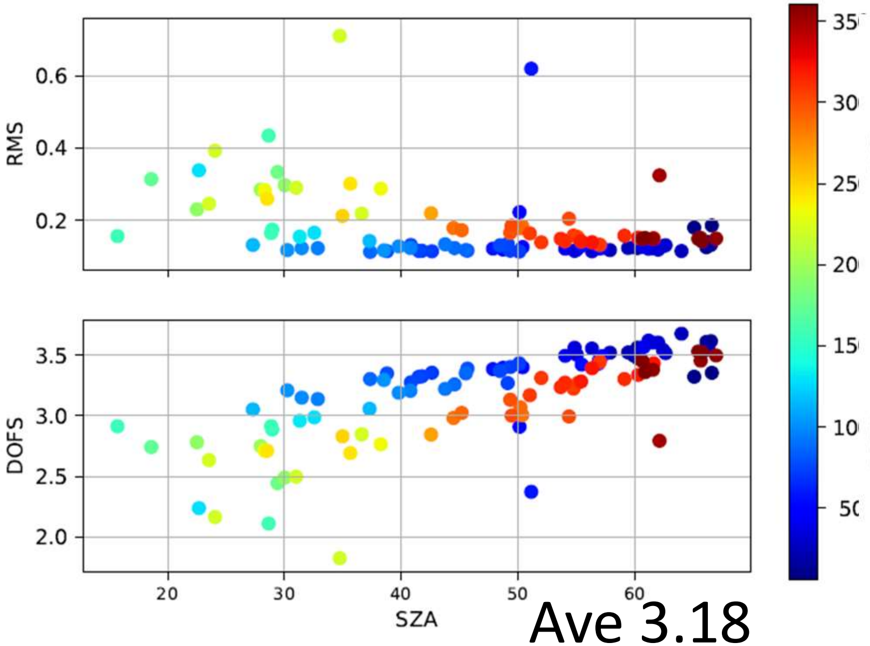
DOFs

5MW is better than 3MW → 5MW has more information

Results: 3MW v1 vs. 3MW v5

3MW v1

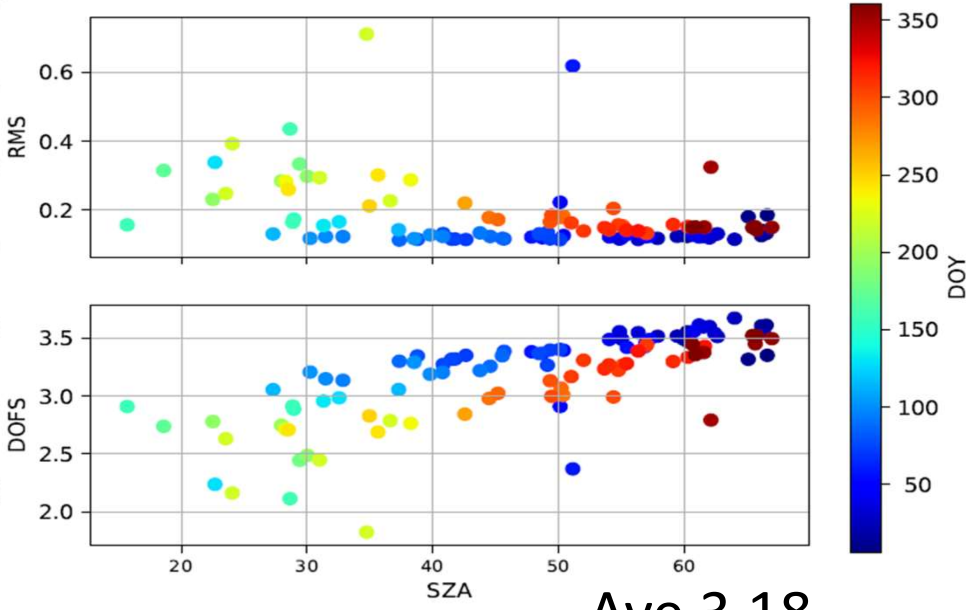
Ave 0.176



Ave 3.18

3MW v5

Ave 0.176



Ave 3.18

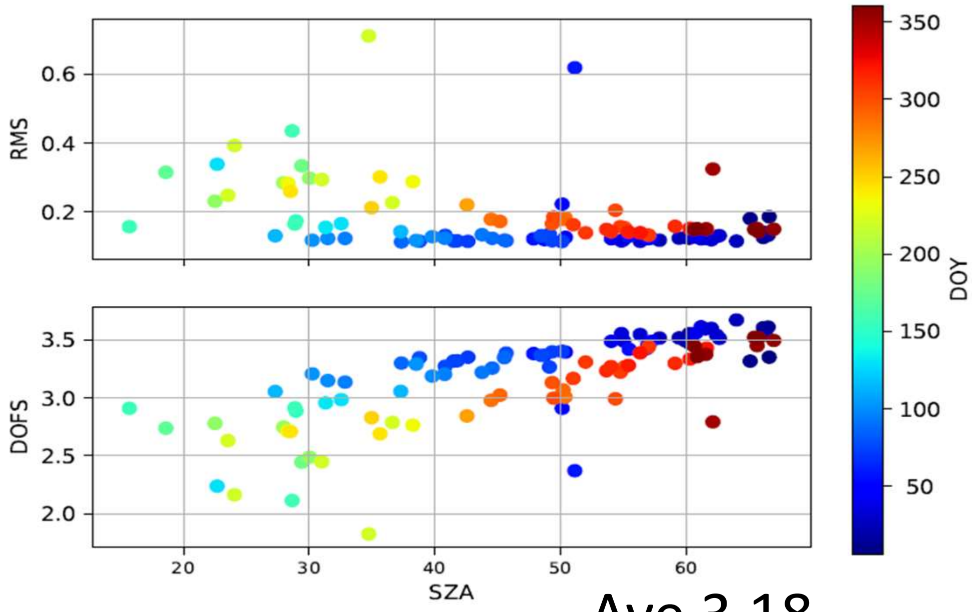
The changes in RMS & DOFs are very small.
(The annual averages are same.)

⇒3MW is hard to be affected by water vapor

Results: 3MW v5 vs. 5MW v5

3MW v5

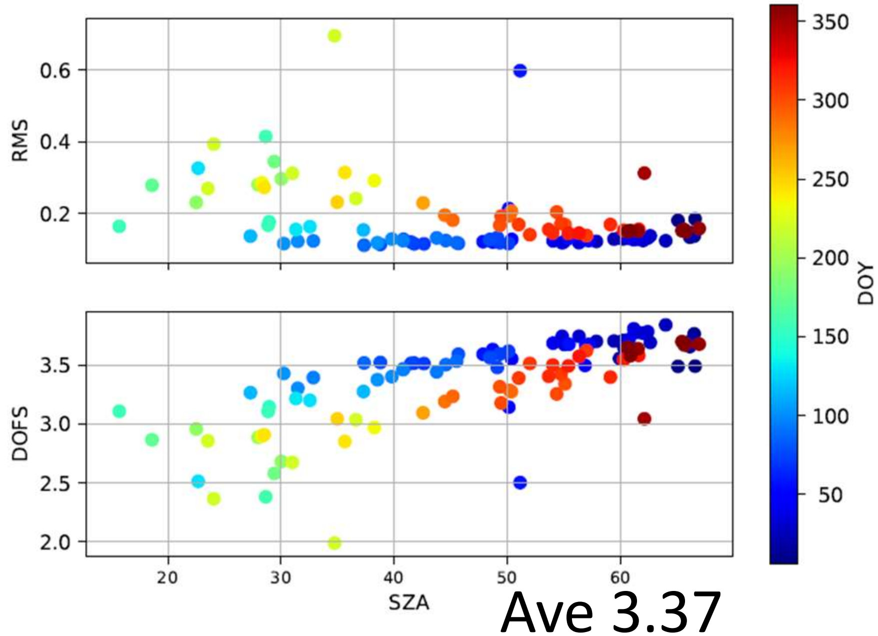
Ave 0.176



Ave 3.18

5MW v5

Ave 0.180



Ave 3.37

RMS

3MW is better than 5MW ⇒ 3MW has higher-quality information

DOFs

5MW is better than 3MW ⇒ 5MW has more information

Summary for comparison

Parameters set	Averaged RMS (2012)	Averaged DOFs (2012)
3MW v1	0.176	3.18
3MW v5	0.176	3.18
5MW v1	0.185	3.34
5MW v5	0.180	3.37

MWs

- Fitting is better for 3MW than 5MWs (RMS is smaller for 3MW)
- 5MW has more information than 3MW (DOFs is larger for 5MW)

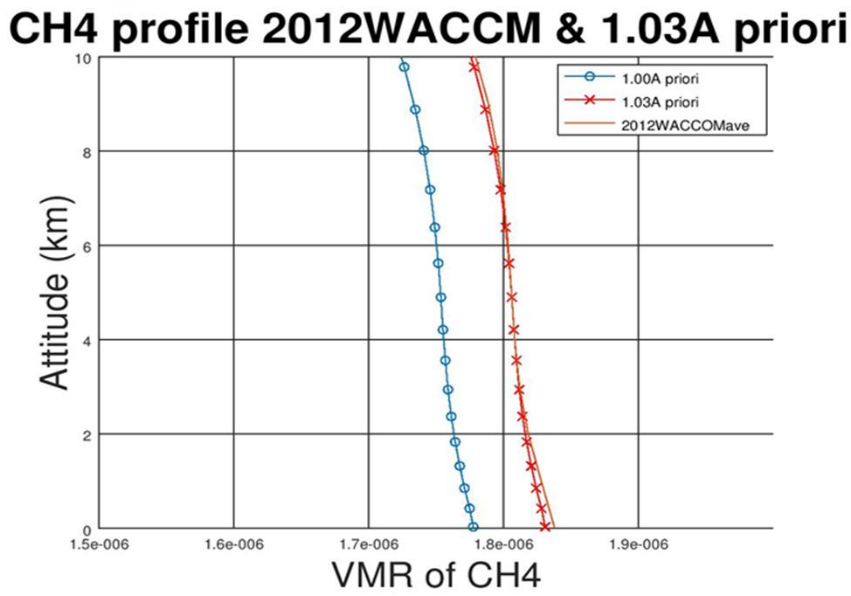
Initial profile of water vapor

- No difference for 3MW
- v5 is better for 5MW as indicated by both RMS and DOFs

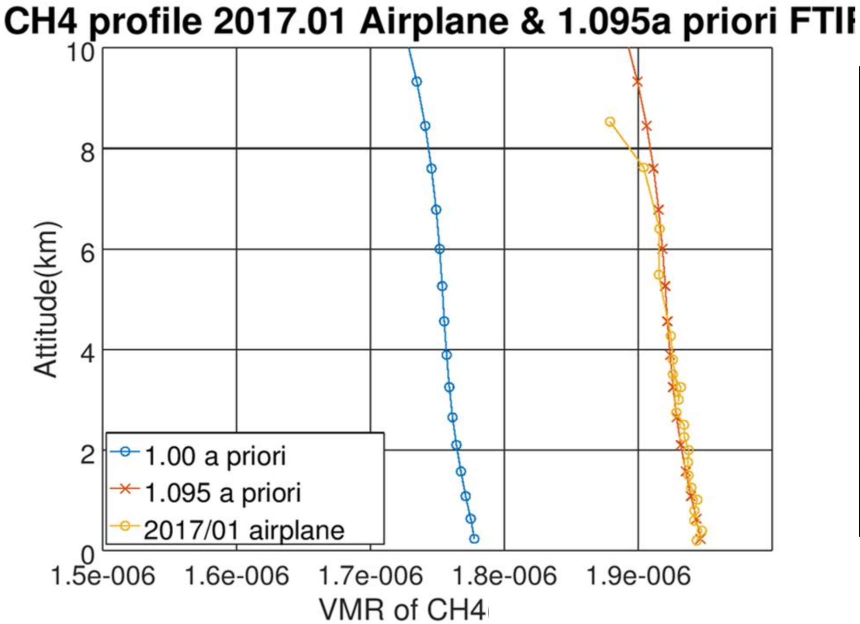
Hereafter, we use 3MW v1, 3MW v5, and 5MW v5.

(5MW v1 isn't used for the validation by airplane sampling results.)

Dependence for initial profiles of CH_4



	CH_4 Partial column [0-8km] ratio for a priori 1.03 / 1.00 2012 average (103 observations)	Standard deviation
3MW v1	1.00010 (+0.01%)	0.00078
3MW v5	1.00006 (+0.01%)	0.00062
5MW v5	0.99996 (-0.00%)	0.00045



	CH_4 Partial column [0-8km] ratio for a priori 1.095 / 1.00 2017 average (103 observations)	Standard deviation
3MW v1	0.9998 (-0.02%)	0.0023
3MW v5	0.9999 (-0.01%)	0.0025
5MW v5	0.9998 (-0.02%)	0.0018

* 1.00 a priori = WACCM 40 years mean

<u>Dependence for initial profiles of CH_4 is negligible</u>

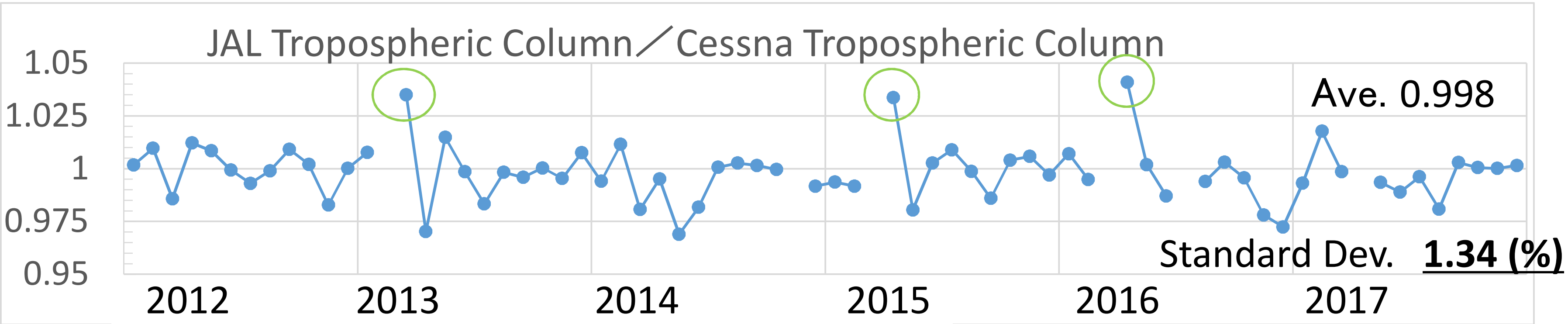
4. Validation of the tropospheric columns by airplane sampling results

Uncertainty of tropospheric columns derived from airplane sampling

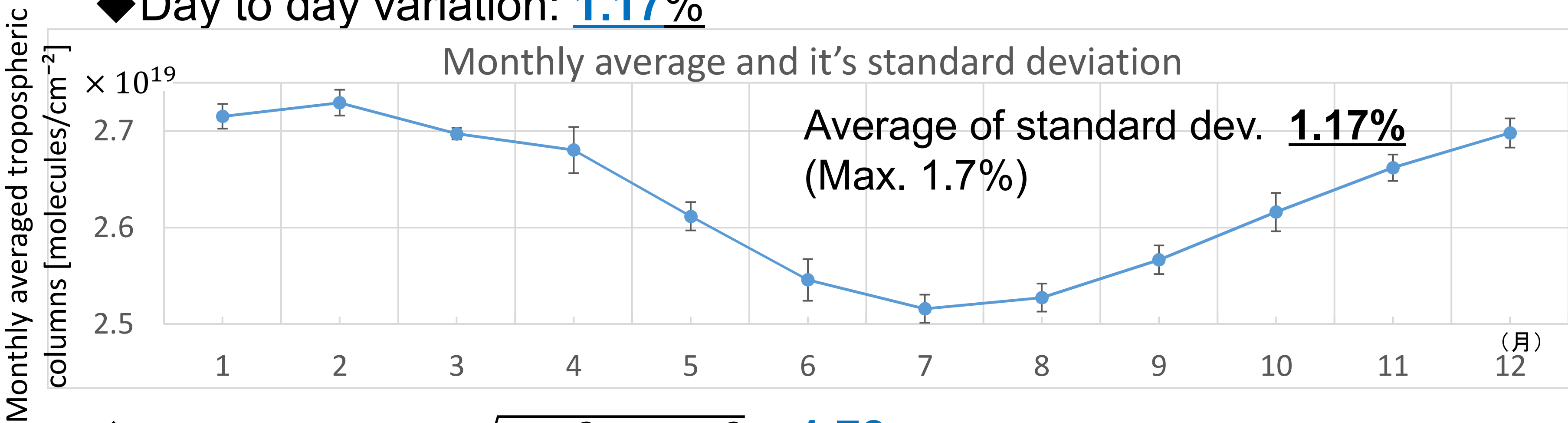
- ◆uncertainty of mixing ratio: **0.16%**
- ◆uncertainty from using temp. and pres. profiles at different place **1.34 %**

Temp. and pres. profiles were taken from NCEP Reanalysis for Cessna or JAL sampling points.

* Hereafter, temp. and pres. profiles for JAL sampling points are used. (Note that there are 4% differences in spring)



◆Day to day variation: **1.17%**



◆Total uncertainty: $\sqrt{1.34^2 + 1.17^2} = $ **1.78%**

Uncertainty of tropospheric columns derived from FTIR

◆Day to day variation from the average of the standard deviation of monthly mean tropospheric columns derived from FTIR

1.3-1.4%

(This value includes the uncertainty of the FTIR measurement (0.8%)

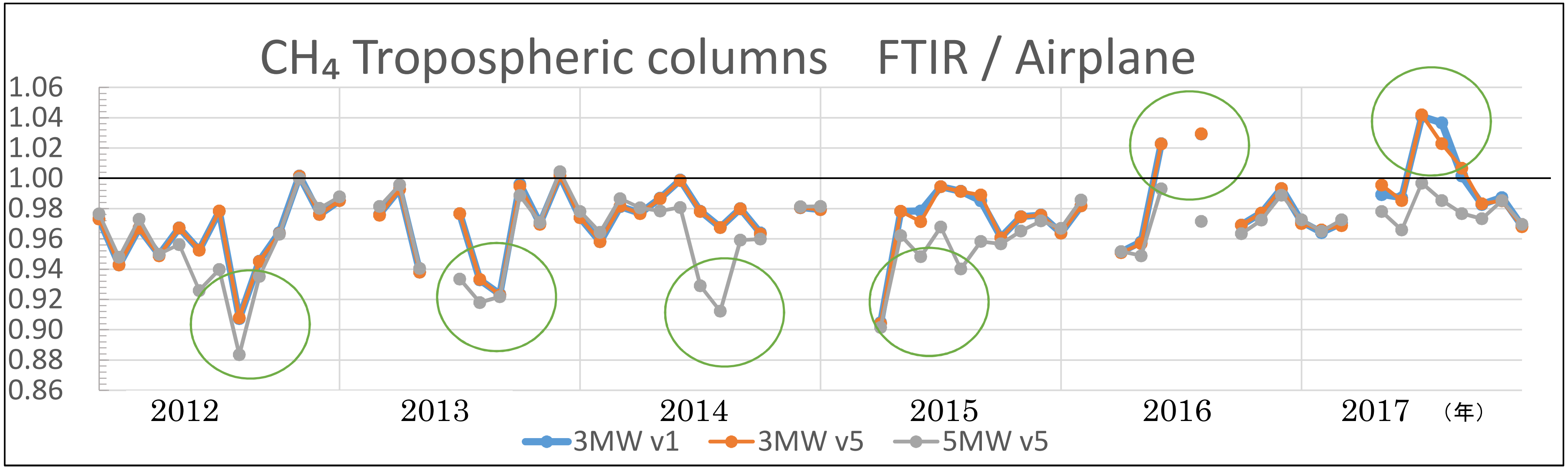
Relative uncertainty between FTIR and airplane sampling

（Airplane1.8%　FTIR 1.4%）　$\sqrt{1.8^2 + 1.4^2} = 2.3\%$

↓

If the difference between the tropospheric columns derived from FTIR and those derived from airplane sampling is smaller than this value (2.3%), the accuracy of the tropospheric columns derived from FTIR is enough.

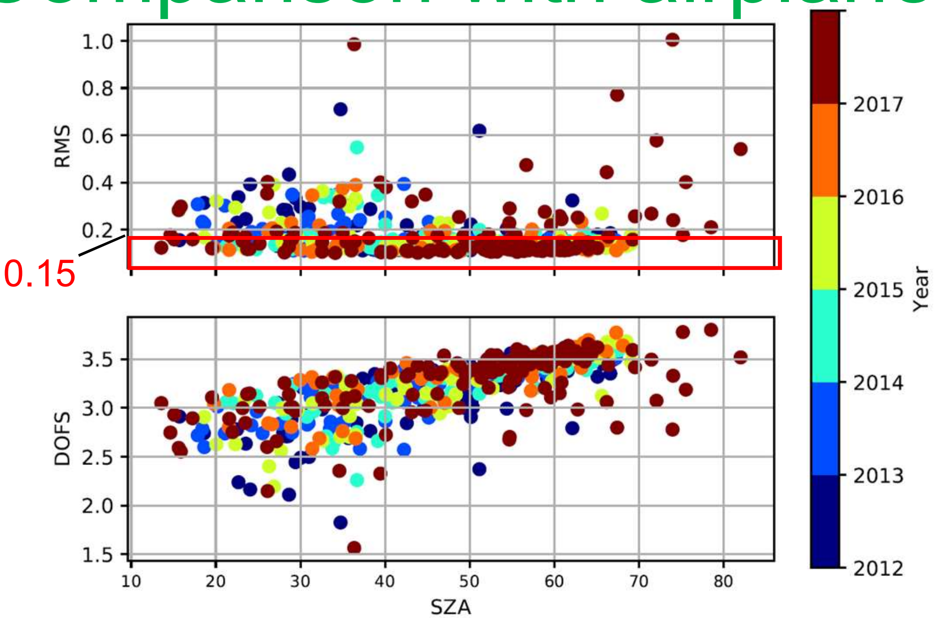
Comparison with airplane sampling (All data)



	3MW v1	3MW v5	5MW v5
Average	0.975	0.975	0.964
Standard Dev.	0.0252	0.0250	0.0245

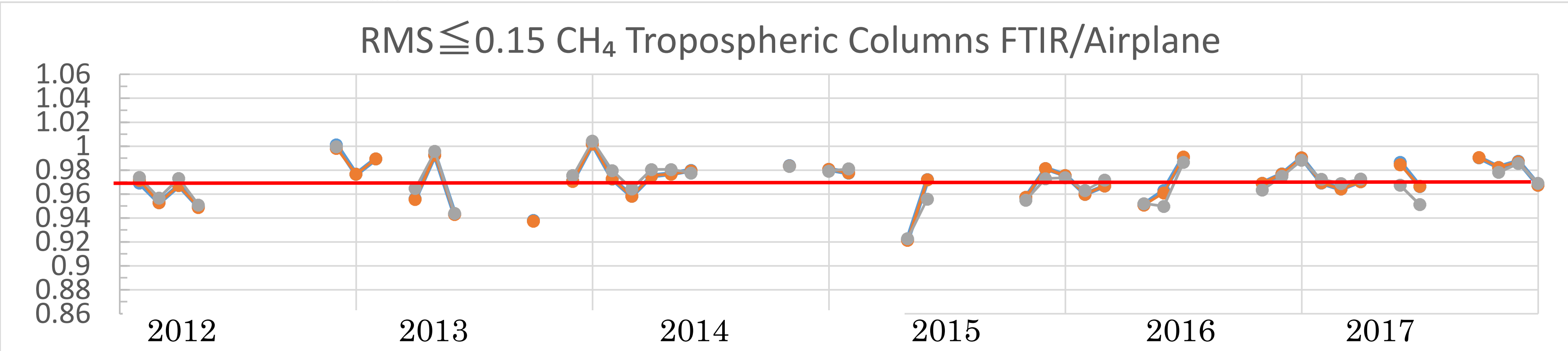
○The difference becomes large mainly in summer (5-10%)
○FTIR underestimate the column.
○The standard deviations of the difference is larger than relative uncertainty (2.3%). ⇨ FTIR doesn't agree with airplane

Comparison with airplane sampling (small RMS data)



3MW v1: 518 spectra → 331 spectra
3MW v5: 518 → 330
5MW v5: 518 → 297

	3MW v1	3MW v5	5MW v5
Average	0.971	0.971	0.971
Standard Dev.	0.0165	0.0165	0.0159



○FTIR underestimate the column (2.9%).

○The standard deviations of the difference are 1.6-1.7% for all parameter sets and smaller than relative uncertainty (2.3%). ⇨ FTIR agrees with airplane

○Summer data were deleted by large RMS….

Bias by the difference of the observation points

Comparison with airplane sampling at Tsukuba

airplane sampling at Tsukuba (observed by NIES)

4 observation (2 in Jan. 2013 and 2 in Jan. 2014)

Altitude: 0-9km 8 points

FTIR / Tsukuba Airplane	3MWv1	3MWv5	5MWv5
2013年1月 0.031-8.01km	0.996	0.996	0.999
2014年1月 0.031-8.01km	0.985	0.985	0.989

FTIR / Sendai Airplane	3MWv1	3MWv5	5MWv5
2013年1月 0.031-8.01km	0.985	0.985	0.988
2014年1月 0.031-8.01km	0.974	0.974	0.978

The differences between FTIR and Tsukuba airplane for above months are all **1.1**% smaller than those between FTIR and Sendai airplane. This **1.1**% difference maybe due to latitudinal gradient of CH_4 concentration.

The average bias of **2.9**% will be partly explained by this latitudinal gradient. Other possible reasons are the uncertainty of the line intensity etc.

5. Conclusion

1. We compared some of fitting parameters for methane retrieval from Tsukuba spectra.

・Dependence for initial profiles of CH_4 is negligible.

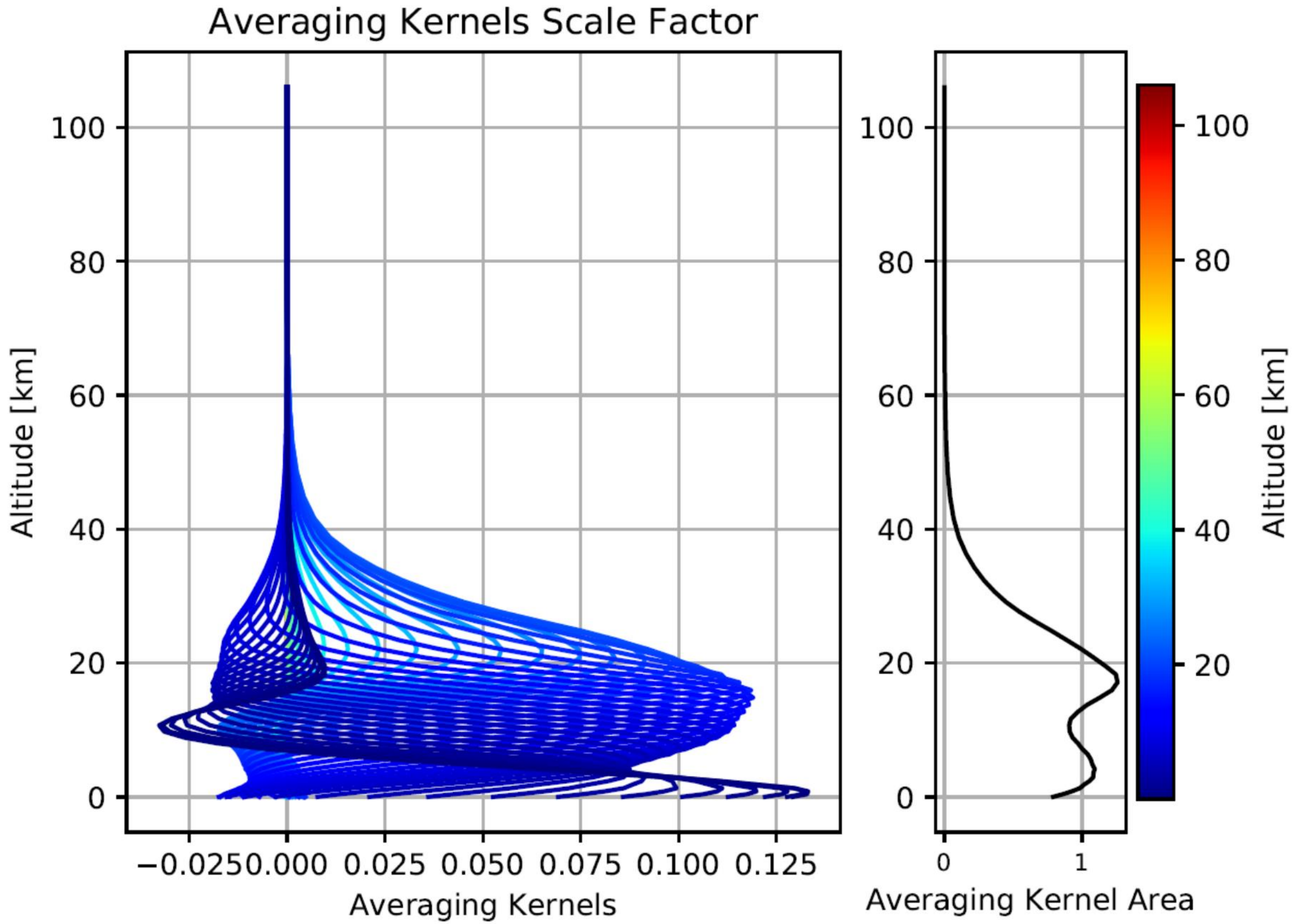
・Among 4 parameter sets (3MW v1, 3MW v5, 5MW v1, and 5MW v5), only 5MW v1 is worse than others.

2. We validated the retrieved tropospheric columns of methane by the comparison with airplane sampling results.

・FTIR agrees with airplane within the relative uncertainty (**2.3**%) when the retrieved results was limited by the RMS value of 0.15.

・There is a negative bias of **2.9**%.

・There are no significant differences among the 3 parameter sets.


Averaging Kernels Scale Factor
Altitude [km]
100
80
60
40
20
0
−0.025
0.000
0.025
0.050
0.075
0.100
0.125
Averaging Kernels
0
1
Averaging Kernel Area
Altitude [km]