Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2019-261-AC1, 2019

Atmospheric Measurement Techniques Discussions

Interactive comment on "Validation analysis of deriving acetonitrile (CH_3CN) profiles by observations of SMILES from the International Space Station, in the stratosphere and lower mesosphere" *by* T. Fujinawa et al.

T. Fujinawa et al.

fujinawa.tamaki@nies.go.jp

Received and published: 12 December 2019

Dear Anonymous Reviewer #1,

We deeply appreciate for valuable comments and suggestions. Please find the manuscript with several revisions. Answers to your comments/questions are given by point by point. We hope that current manuscript is significant for the publication in Atmospheric Measurement Techniques.

Sincerely yours, Tamaki Fujinawa.

Printer-friendly version

Discussion paper

General comments 1-1

The level of English in the manuscript could use some work, starting with the title. It is readable, but the phrasing is occasionally not quite correct. There are too many instances for me to provide line-by-line corrections.

Answer to general comments 1-1

We appreciate your valuable comment. As you mentioned, we have checked the grammar and phrasing again.

Major comment 1-2

I have some issues with the background discussion on CH3CN. While it is all technically correct that the molecule is associated with biomass burning and pollution and can be used as a tracer for biomass burning events, most biomass burning activity occurs in the troposphere but the measurements reported here are in the stratosphere. Some of the discussion is not entirely relevant to what they are measuring. They report the observation of a seasonal maximum of CH3CN in the upper stratosphere in February supposedly resulting from a peak in biomass burning during the time period from December to March. Age of air in the upper stratosphere is the order of a few years, but they appear to be suggesting they are measuring enhanced CH3CN in the upper stratosphere almost immediately. It should take a few years for young tropospheric air containing enhanced CH3CN to make its way to that atmospheric region. Intense fires can inject biomass burning products directly into the stratosphere (pyro-convection), but I don't think that is what they propose is happening here. CH3CN levels would be anomalously high in that case, and I expect the effect would not extend into the upper stratosphere. It would be confined to the lower stratosphere. The authors may want to rethink their interpretation taking transport times to the upper stratosphere into consideration.

Answer to major comment 1-2

We are grateful for pointing this out and for your valuable comment. In our understanding, we could assume that CH3CN is almost emitted from biomass burning into the atmosphere and there is no major source of CH3CN in the stratosphere. In addition, it is likely that there is seasonality of CH3CN in lower stratosphere from 28 km to 36 km as can be seen in Figure 7, even if the error bars (one sigma standard deviation) were taken into account. Therefore, we thought that the enhancement which can be seen in Figure 7 would be caused by biomass burning event. However, as you mentioned, our description about the enhancement might cause a misunderstanding that biomass burning plume emitted during the time period from December to March was immediately transported to the stratosphere. Therefore we revised some parts of the sentences as follows to make it clear.

Revisions to major comment 1-2

Lines 5-6: "We estimated the systematic and random errors to be ∼5.8 ppt (7.8 %) and 25 ppt (60 %) for a single observation at 15.7hPa, respectively, in the Tropics, where the CH3CN measurements are enhanced." was replaced by

→

Lines 5-6: "We estimated the systematic and random errors to be ∼5.8 ppt (7.8 %) and 25 ppt (60 %) for a single observation at 15.7hPa, respectively, in the Tropics.".

Lines 198-199: ", which is consistent with our understanding that most biomass burning occurs from December–March." was removed.

Lines 198-199: "These seasonal maximum CH3CN levels measured in February are consistent with the yearly high period of BB events from December–March." was removed.

Major comment 1-3

In Figure 7, for altitudes below 40 km, the systematic high bias of results from AOS1 relative to results from AOS2 are quite evident. The authors make the statement that

the differences between the AOSs is due to “sensitivity differences.” I do not know what that means. The phrasing would suggest something like signal-to-noise ratio, but I would expect that to yield increased variability and not a systematic offset, unless perhaps the retrieval is being “pulled” more toward the a priori in the optimal estimation analysis because of the reduced signal-to-noise ratio. Is it possible to provide a few extra words to explain what is meant by sensitivity differences? It might be instructive to see the signal from the two different AOSs under similar measurement conditions, to see why one is yielding a larger VMR than the other.

Answer to major comment 1-3

We appreciate your valuable comment. The “sensitivity differences” indicate inherent sensitivity differences between the two AOSs derived from instrumental characterization determined when manufacturing. As you pointed out, only using of the phrase “sensitivity differences” was unclear. Therefore, we added an additional explanation as below.

Revisions to major comment 1-3

Line 135 : “Note that the sensitivity differences indicate inherent sensitivity differences between the two AOSs derived from instrumental characterization determined when manufacturing.” was added.

Major comment 1-4

In Figure 9, the top VMR two points from MLS are negative, which contributes to the strong magnification of the discrepancies between the two instruments in this altitude region. Negative VMRs have no physical meaning. You should probably point this out when discussing the discrepancies. This is an item in your favor.

Answer to major comment 1-4

We appreciate you pointing this out. As you mentioned, it shows the exaggeration of the discrepancies between the two observation. We added a sentence as below to

point that out.

Revisions to major comment 1-4

Line 180 : “It should be noted that the discrepancies between the two instruments were exaggerated at upper pressure levels from 2.8\,hPa although negative values of CH3CN VMR derived from MLS have no physical meaning.” was added.

Major comment 1-5

How was the a priori state constructed? If MLS results went into generating the a priori used in the optimal estimation analysis, extra care might be required in using comparisons to MLS as part of a validation effort.

Answer to major comment 1-5

We appreciate your valuable comment. We used the results from version 5.2 of the Goddard Earth Observing System Model (GEOS-5.2) as a priori information (e.g., O$_3$ VMR profile, temperature and pressure profile). Therefore, we think it is not required to provide extra care about it. However, we added an explanation about preparing a priori state as you pointed out as below.

Revisions to major comment 1-5

Line 57 : “This version of L2r product was derived from the Level-1b (L1b) version 008 calibrated spectra, which used version 5.2 of the Goddard Earth Observing System Model (GEOS-5.2) as a priori information (e.g., O3 VMR profile, temperature and pressure profile) (Rienecker et al., 2008).” was added.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-261, 2019.