Point-By-Point Reply to Referee Comment 2 from Anonymous Referee #1

1: Specific comments

Reviewer Comment (1-1)
-p.1 l.12-14: Please specify also the relative values corresponding to the given absolute differences.

Author's response (1-1)
We appreciate your suggestion. We added the relative values corresponding to the absolute differences.

Author's changes in the manuscript (1-1)
-Page 1, Line 16:
Added "(within 10% between 30 and 40 km. There's a larger discrepancy below 30 km.)"

-Page 1, Line 18:
Added "(12 – 15 %)"

Reviewer Comment (1-2)
- p.2 l.43-46: Could you please explicitly comment the differences between the results of the comparisons mentioned here (between SMILES v2 and MLS / ACE-FTS, by Sugita et al., 2013) and the results of your validation study? It would be interesting to add such a comment in the conclusion section, where you discuss your results.

Author's response (1-2)
Thank you for pointing it out. This earlier paper by Sugita et al., 2013 used the v215 SMILES data and limited to an altitude of around 30 km in the polar region. In contrast, this study used the v300 data and performed global comparison in the stratosphere and mesosphere. We added a note regarding the features of this paper.

Author's changes in the manuscript (1-2)
-Page 2, 3, Line 51, 53:
Added "In contrast to the previous study, this study used v300 SMILES data and provided a global comparison, including the stratosphere and mesosphere."

Reviewer Comment (1-3)
- p.4. l.76: Do not forget to specify in the text that you are talking about the daily number of observations.

Author's response (1-3)
Thank you for your comment. We mentioned that we are talking about the number of observations per day.

Author's changes in the manuscript (1-3)
-Page 3, Line 83:
"the number of observations" → "the number of observations per day"

Reviewer Comment (1-4)

- p.4 l.92: Could you give some information about the a priori data used in the retrieval process: what is the source of this data set? Does it depend on latitude and time? Etc.

Author's response (1-4)

Thank you for pointing it out. The U.S. standard atmosphere is used as the a priori profile in SMILES version 3.0.0 retrieval. And the a priori profile is used separately in the polar, equatorial, mid-Summer, and mid-winter regions. We described the treatment of the a priori profile in the manuscript.

Author's changes in the manuscript (1-4)

-Page 6, Line 106 – 108:

Added "We used the U.S. standard atmosphere profiles as the a priori state (xa). They are used separately for polar, equatorial, summer mid-latitude, or winter mid-latitude regions (US-Standard 1976)."

(US-Standard, P.: U.S Standard Atmosphere 1976, Tech. rep., U.S Government Printing Office Washington DC, 1976)

Reviewer Comment (1-5)

- p.6 l.102: "We assumed that the natural isotopic abundance of H35Cl/HCl was 0.7576. . .": Please explain where this value comes from. A citation should be added here.

Author's response (1-5)

We appreciate you pointing it out. We added Berglund and Wieser (2011) to the citation.

Author's changes in the manuscript (1-5)

-Page 7, Line 116:

"was 0.7576" → "was 0.7576 by Berglund and Wieser., 2011"

(Berglund, M. and Wieser, M. E.: Isotopic compositions of the elements 2009 (IUPAC Technical Report), Pure and Applied Chemistry, 83, 397360– 410, https://doi.org/https://doi.org/10.1351/PAC-REP-10-06-02, 2011)

Reviewer Comment (1-6)

- p.6-7 Sect.3: Please discuss the differences and/or similarities between the three subperiods under consideration or change Fig. 4 to show only the results averaged over the whole SMILES operational period. I do not understand the point in dividing the comparisons between SMILES and SD-WACCM into three different time periods if this is not discussed.

Author's response (1-6)

We are grateful for your kind comments. We changed Fig. 4 to show only the results averaged throughout the SMILES operational period.

Author's changes in the manuscript (1-6)

-Page 9, Figure 4: The figure was improved.

-Page 9, Figure 4, caption:

"rows" → "panel"

Removed "in three time range"

- Page 7, Line 119, 120:

"Left panel: Time range between October 16, 2009 and December 15, 2009. Middle panel: Between December 16, 2009 and February 15, 2010. Right panel: Between February 16, 2010 and April 17, 2010."

→ "The time period is between October 16, 2009 and April 17, 2010."

Reviewer Comment (1-7)
- p.6 l.110-112: An explanation about the HCl vertical and latitudinal distribution is missing. Please describe the chemical and physical mechanisms controlling it, or at least comment on the current state of knowledge about that. (This could be added either here or in the introductory section.)

Author's response (1-7)
We appreciate you pointing it out. We explained the chemical and physical mechanism that produced the distribution of HCl.

Author's changes in the manuscript (1-7)
-Page 7,8 , Line 127 – 131:
Added "In the lower and middle stratosphere, HCl is generated by the reaction of Cl with CH_4 and HO_2 and transported by circulation (e.g. Brewer-Dobson circulation). The HCl abundance is balanced by production ($Cl + HO2 \rightarrow HCl + O_2$) and loss ($HCl + OH \rightarrow Cl + H_2O, HCl + h\nu \rightarrow H + Cl$) in the upper stratosphere and mesosphere. Near the mesopause, the photodissociation becomes the dominant reaction, and the HCl abundance decreases with height (Brasseur and Solomon, 2005)."

Reviewer Comment (1-8)
- p.9 Eq.3: Even if it is obvious for most readers, N should be explicitly defined.

Author's response (1-8)
Thank you for pointing it out. We defined "N" as the number of coincidence.

Author's changes in the manuscript (1-8)
-Page 9, Line 157:
Added "N(z) is the number of coincidences at an altitude z"

Reviewer Comment (1-9)
- p.9 Tab.3: That could be helpful to include the vertical resolution and the altitude range covered by each of these instruments.

Author's response (1-9)
Thank you for your comment. We included the vertical resolution and the altitude range of each instruments in Table 3.

Author's changes in the manuscript (1-9)
-Page 10, Table 3:
Added the row of "vertical resolution" and "altitude range".

Reviewer Comment (1-10)
- p.9 l.140: Regarding the MLS vertical resolution, you should explicitly say that it is of the same order as that of SMILES, in order to highlight the fact that the observed differences in the profiles are not due to differences in vertical resolution. (Same comment about the comparison with ACE in Sect. 4.2.)

Author's response (1-10)
Thank you for pointing it out. We mentioned the vertical resolution to highlight the fact that the observed differences in the profiles are not due to differences in vertical resolution. Also, we added a reference to ACE-FTS version 4.0 retrieval.

Author's changes in the manuscript (1-10)
-Page 10, Line 169, 170:
Added "The vertical resolution of MLS is of the same order as that of SMILES."

-Page 11, Line 195:
Added "(Boone et al., 2020)"
(Boone, C., Bernath, P., Cok, D., Jones, S., and Steffen, J.: Version 4 retrievals for the atmospheric chemistry experiment Fouriertransform spectrometer (ACE-FTS) and imagers, Journal of Quantitative Spectroscopy and Radiative Transfer, 247, 106 939,https://doi.org/https://doi.org/10.1016/j.jqsrt.2020.106939, http://www.sciencedirect.com/science/article/pii/S0022407319305916, 2020.)

-Page 12, Line 197:
Added "The vertical resolution of ACE-FTS is of the same order as that of SMILES."

Reviewer Comment (1-11)
- p.10 l.148-149: There are however changes with latitude observed below 35 km. Please describe them.

Author's response (1-11)
Thank you for your comment. We added a description on this.

Author's changes in the manuscript (1-11)
-Page 10, Line 178, 179:
Added "While below 35 km of altitude, several areas of large differences can be identified, especially in the equatorial region."

Reviewer Comment (1-12)
- p.10 l.155-156: The water vapor effect should be explained (maybe not here, but earlier in the paper, when explaining the retrieval process).

Author's response (1-12)
We appreciate your valuable point of view. We agreed with your suggestion and removed the statement as follows. As for the discussion of the water vapor effect, we removed it because it was not verified enough. The reason for describing water vapor is the treatment of the continuum in the SMILES retrieval algorithm. In the SMILES retrieval algorithm, we do not retrieve the continuum simultaneously when deriving the species abundance, but rather we give it as a parameter. The continuum can also be seen in observed spectra at about 30 km in Fig.1. The effect of this continuum is also mentioned in the SMILES ozone validation paper (Kasai et al., 2013). On these grounds, we described it as a water vapor effect, but due to a lack of quantitative discussion, we removed it in this paper. We're glad you pointed that out.

Author's changes in the manuscript (1-12)
-Page 10, Line 184 – 186:
Removed "There is a possibility that this difference was caused by water vapor. The SMILES HCl profile

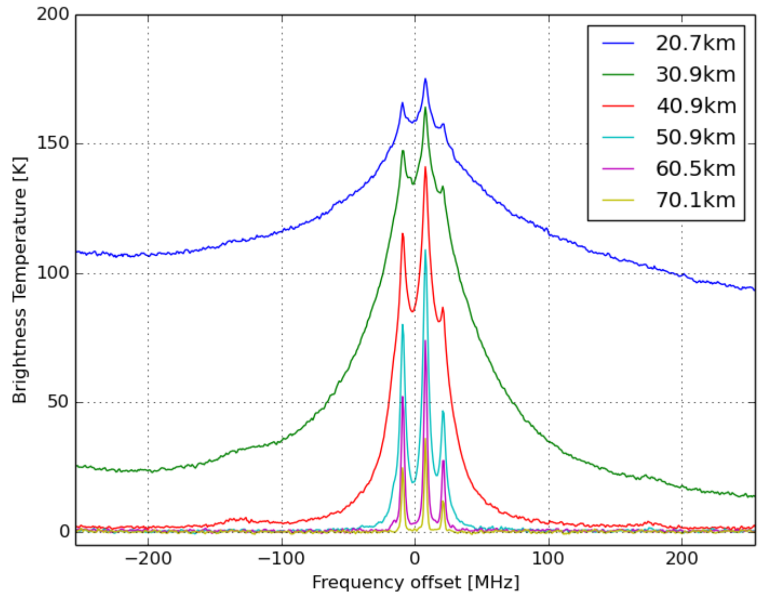

Figure 1: $H^{35}Cl$ spectra observed by SMILES.

was retrieved without considering water vapor effect and the influence of the water vapor was thus one of the possible results of the latitudinal difference."

Reviewer Comment (1-13)
- p.10 l.166: "MAD" has not been defined.

Author's response (1-13)
We are grateful for your kind advice. MAD is the acronym for Median Absolute Deviation.

Author's changes in the manuscript (1-13)
- Page 12, Line 198:
"3-MAD" → "3 times the median absolute deviation (3-MAD)

Reviewer Comment (1-14)
- p.12 l.192: Please specify the geographic coordinates of the Kiruna station.

Author's response (1-14)
Thank you for your comment. We added the information about the geographic coordinates of the Kiruna station.

Author's changes in the manuscript (1-14)
-Page 16, Line 225, 226:
"in terms of geolocation and time on January 24, 2010 over Kiruna." → "in terms of geolocation over Kiruna (67.8 N° 20.4 E°) and time on January 24, 2010."

Reviewer Comment (1-15)
- p.13 Fig.7: It is good that information about the variability of the differences is given in panels (B), by the representation of $\pm$ 1 σ. However, I wonder why this information is not given for difference profiles shown in panels (A), as well as in Fig. 9 (A).

Author's response (1-15)
We are grateful for your kind advice. We added the representation of $\pm$ 1 σ in panels (A) according to your comment.

Author's changes in the manuscript (1-15)
-Page 13, Figure 7, panels (A) & -Page 15, Figure 9, panels (A) :
Figure corrections and changes.

Reviewer Comment (1-16)
- p.18 Eq.10: I guess that Δb_0 is the uncertainty on model parameters. Please define it explicitly.

Author's response (1-16)
Thank you for pointing it out. As you mentioned, Δb_0 is the uncertainty on parameters. We defined it in the manuscript.

Author's changes in the manuscript (1-16)
-Page 18, Line 258:
"where I is the inversion function and b_0 is the vector of model parameters." $\rightarrow$ "where I is the inversion function, b_0 is the vector of model parameters and Δb_0 is the uncertainty on model parameters"

Reviewer Comment (1-17)
- p.18 l.234: "about 0.9 ppbv at 50 km" This value is inconsistent with what is shown in Fig. 11. Please correct.

Author's response (1-17)
We corrected the value to 0.09 ppbv following your comment.

Author's changes in the manuscript (1-17)
-Page 18, Line 268:
"about 0.9 ppbv" $\rightarrow$ "about 0.09 ppbv"

Reviewer Comment (1-18)
- p.18-20, Sect. 5.2: Please give more information about the temperature data used in the retrieval process, for the three instruments under consideration. Has the temperature been retrieved from measurements performed by the instrument itself or has, in some cases, external data been used? Discuss the quality of these T data sets, comment on their accuracy. Are there some validation studies that could give an indication as to which ones of the SMILES, MLS and ACE-FTS temperature profiles are closer to the true atmospheric temperature? Such additional information would be helpful for future users to know which of these three HCl data sets is likely the most realistic in the upper stratosphere / lower mesosphere. Knowing more about the temperature data used in the SMILES retrieval procedure would also be useful to better estimate the quality of the SMILES HCl data set in higher altitude regions, where

measurements from other instruments are not available.

Author's response (1-18)
We are grateful for your kind advice. The details of the a priori temperature profile for SMILES, MLS and ACE-FTS retrieval were added.

Author's changes in the manuscript (1-18)
-Page 4, Line 93 – 99 :
Added "The temperature profile used in the SMILES version 3.0.0 retrieval was synthesized, assuming the hydrostatic equilibrium, using the Goddard Earth Observing System, Version 5 (GEOS-5) reanalysis meteorolog-ical datasets and the climatology based on the Aura/MLS measurements (Kuribayashi et al., 2017). The GEOS-5 datasets were used in the upper troposphere and stratosphere, and the Aura/MLS datasets were used in the mesosphere and lower thermosphere, respectively. The temperature profile was also retrieved using the ozone transition in the SMILES version 3.0.0 retrieval process, but it was not applied to the retrieval of atmospheric species including HCl, except for ozone, to avoid systematic error propagation issues (SMILES-NICT, 2014)."

-Page 20, Line 289 – 300:
Added "The a priori temperature profile used in the SMILES retrieval procedure is based on the GEOS-5 profile in the stratosphere and MLS retrieved profile above the mesosphere.The altitude limit of the MLS temperature profile is 0.001 hPa, with a vertical resolution of 6 - 14 km and a precision of 1.2 – 3.6 K per profile. MLS uses GEOS-5 up to 1 hPa as with SMILES. For pressures smaller than 1 hPa, the COSPAR International Reference Atmosphere (CIRA-86) is used as a priori temperature information (with a loose constraint) in the MLS retrieval procedure (Schwartz et al., 2008). The altitude range and vertical resolution of the CIRA-86 profile are ground to 120 km and 2 km, respectively (Fleming et al., 1990). The temperature value retrieved by MLS is 10 K lower than the a priori profile on average in some areas for pressure values smaller than 1 hPa, based on earlier version validation studies (Schwartz et al., 2008). The ACE-FTS retrieval procedure uses the retrieved temperature profile as the a priori between 18 – 125 km. The vertical resolution of ACE-FTS retrieved temperature is 3 – 4 km. The temperature values retrieved by ACE-FTS are less than 10 K larger than the MLS derived temperatures. (Schwartz et al., 2008). These types of difference are also seen in the comparison results performed here.".

Reviewer Comment (1-19)
- p.21, Fig.12: The quality of this figure needs to be improved. The legends are barely readable. It is confusing that panel (A) does not have the same vertical scale as the other ones. Also, it would be clearer to use the same colour code or line styles in both panels (C) and (D).

Author's response (1-19)
Thank you for your comment. We revised the altitude scale and improved the figure resolution that according to your comment.

Author's changes in the manuscript (1-19)
-Page 21, Figure 12 :
Figure improvements and corrections

2: Technical corrections.

Reviewer Comment (2-1)

- p.1 l.2: Change "has been" to "is".

Author's response (2-1)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-1)
-Page 1, Line 2:
"has been" → "is"

Reviewer Comment (2-2)
- p.1 l.12: Change "well agreed" to "agreed well".

Author's response (2-2)
Thank you for your comment. We revised that according to your comment.

Author's changes in the manuscript (2-2)
-Page 1, Line 16:
"well agreed" → "agreed well".

Reviewer Comment (2-3)
- p.1 l.18: "concentration" add an "s".

Author's response (2-3)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-3)
-Page 2, Line 23:
"concentration" → "concentrations".

Reviewer Comment (2-4)
- p.2 l.30: "HALOE HCl" remove "HCl".

Author's response (2-4)
We appreciate you pointing it out. We revised that according to your comment.

Author's changes in the manuscript (2-4)
-Page 2, Line 35:
"HALOE HCl" → "HALOE"

Reviewer Comment (2-5)
- p.2 l.48-49: Incomplete sentence (no verb).

Author's response (2-5)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-5)
-Page 3, Line 54:
"SMILES observation of" → "we examined"

Reviewer Comment (2-6)
- p.3 l.58: "observation" add an "s".

Author's response (2-6)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-6)
-Page 3, Line 64:
"observation" → "observations"

Reviewer Comment (2-7)
- p.3 l.60: Reword (suggestion "SMILES operational period started on October 12, 2009 and ended on April 21, 2010.")

Author's response (2-7)
We are grateful for your kind advice. We revised that according to your advice.

Author's changes in the manuscript (2-7)
-Page 3, Line 66, 67:
"The period of the SMILES observation was from October 12, 2009 to April 21, 2010." → "SMILES operational period started on October 12, 2009 and ended on April 21, 2010."

Reviewer Comment (2-8)
- p.3 l.4: "observation" add an "s". "Kasai et al. (2013)" add "by Kaisai et al. . . "

Author's response (2-8)
We appreciate you pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-8)
-Page 3, Line 70:
"observation" → "observations"

-Page 3, Line 71:
"Kasai et al. (2013)" → "by Kasai et al. (2013)"

Reviewer Comment (2-9)
- p.4 l. 68: Change "and" to "or".

Author's response (2-9)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-9)
-Page 3, Line 75:
"and" → "or"

Reviewer Comment (2-10)

- p.4 l.92: "y is THE observed spectrum".

Author's response (2-10)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-10)
-Page 6, Line 105:
"y is observed spectra" → "y is the observed spectra"

Reviewer Comment (2-11)
- p.6 Fig.2, caption: Change "spectrum" to "spectra" (three times). (Same comment about the caption of Fig. 3.)

Author's response (2-11)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-11)
-Page 6, Figure 2, caption:
"spectrum" → "spectra"

Reviewer Comment (2-12)
- p.6 l.105: "the altitudeS 50 km-90 km" or "the altitude range 50 km-90 km".

Author's response (2-12)
We appreciate you pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-12)
-Page 7, Line 119:
"altitude of $50\,km - 90\,km$" → "altitudes $50\,km - 90\,km$"

Reviewer Comment (2-13)
- p.6 l.110-112: Reword (suggestion "The HCl vertical distribution shows an increase with altitude with a maximum below the stratopause, approximately constant values between [. . .], and a decrease with altitude from the mesopause to. . .")

Author's response (2-13)
We are grateful for your kind comment. We revised that according to your advice.

Author's changes in the manuscript (2-13)
-Page 7, 8, Line 124, 125:
"The HCl vertical profile showed an increasing with the altitude increased below the stratopause ($\sim$ 45 km), approximately constant between" → "The HCl vertical distribution shows an increase with altitude with a maximum below the stratopause ($\sim$ 45 km), approximately constant values between"

-Page 7, Line 126, 127:
"decreased with the altitude increasing" → "a decrease with altitude"

Reviewer Comment (2-14)

- p.6 l.114: "panelS (B)"

Author's response (2-14)
Thank you for pointing it out. We changed the figure 4 (B) to one.

Reviewer Comment (2-15)
- p.7 l.122: "observationS"

Author's response (2-15)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-15)
-Page 8, Line 144:
"observation" → "observations"

Reviewer Comment (2-16)
- p.8 Fig.4, caption: ". . . within latitude bins of 10°."

Author's response (2-16)
Thank you for your advice. We revised that according to your advice.

Author's changes in the manuscript (2-16)
-Page 9, Figure 4, caption:
"a latitude bin of" → "latitude bins of"

Reviewer Comment (2-17)
- p.9 l.132: "previous work ON MLS observations"

Author's response (2-17)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-17)
-Page 10, Line 161:
"previous work of the MLS observations" → "previous work on MLS observations"

Reviewer Comment (2-18)
- p.10 l.147: "increases" remove the "s".

Author's response (2-18)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-18)
-Page 10, Line 176:
"increases" → "increase"

Reviewer Comment (2-19)
- p.10 l.154: "at below 30 km" remove "below".

Author's response (2-19)
We appreciate you pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-19)
-Page 11, Line 184:
"at below 30 km" → "at 30 km"

Reviewer Comment (2-20)
- p.10 l.156: Change "was" to "is".

Author's response (2-20)
Thank you for your advice. We removed the sentence containing this. See Author's response (1-12) for more information.

Reviewer Comment (2-21)
- p.10 l.156: "one of the possible results" Do you mean "one of the possible causes"?

Author's response (2-21)
We appreciate you pointing it out. We removed the sentence containing this. See Author's response (1-12) for more information.

Reviewer Comment (2-22)
- p.10 l.157&170: Change "less" or "lower".

Author's response (2-22)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-22)
-Page 11, Line 188 & Page 12, Line 202
"less" → "lower"

Reviewer Comment (2-23)
- p.10 l.169: "945" There is a mistake. This value is different from the one given in Table 3.

Author's response (2-23)
We appreciate you pointing it out. We have revised the values to be correct.

Author's changes in the manuscript (2-23)
-Page 12, Line 201:
"945" → "935"

Reviewer Comment (2-24)
- p.10 l.172: Change "tropic region" to "tropical region".

Author's response (2-24)
Thank you for your advice. We revised that according to your advice.

Author's changes in the manuscript (2-24)
-Page 12, Line 205:
"tropic region" → "tropical region"

Reviewer Comment (2-25)
- p.10 l.173: "altitude" written twice in a row.

Author's response (2-25)
Thank you for pointing it out. Removed one of the two "altitude".

Author's changes in the manuscript (2-25)
-Page 12, Line 205:
"altitude altitude" → "altitude"

Reviewer Comment (2-26)
- p.11 l.177: "conformed" Do you mean "confirmed"?

Author's response (2-26)
Thank you for pointing it out. The correct word is "Confirmed". We fixed it.

Author's changes in the manuscript (2-26)
-Page 12, Line 210:
"conformed" → "confirmed"

Reviewer Comment (2-27)
- p.12 Fig.6: Adding "SMILES" and "MLS" as a title for the left and middle panels would make the figure clearer.

Author's response (2-27)
We are grateful for your kind comment. We revised that according to your advice.

Author's changes in the manuscript (2-27)
-Page 12, Figure 6: Added a title for panels

Reviewer Comment (2-28)
- p.13 Fig.7, caption: Change "Eq (4)" to "Eq (3)".

Author's response (2-28)
Thank you for pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-28)
-Page 13, Figure 7, caption:
"Eq (4)" → "Eq (3)"

Reviewer Comment (2-29)

- p.18 l.226: “valueS”

Author’s response (2-29)
Thank you for your comment. We revised that according to your advice.

Author’s changes in the manuscript (2-29)
-Page 18, Line 261:
“value” → “values”

Reviewer Comment (2-30)
- p.18 l.234: Change “between the altitude region of 30 and 60 km” to “between 30 and 60 km”.

Author’s response (2-30)
Thank you for your comment. We revised that according to your advice.

Author’s changes in the manuscript (2-30)
-Page 18, Line 268:
“between the altitude region of 30 and 60 km” → “between 30 and 60 km”

Reviewer Comment (2-31)
- p.19 l.249: “synthesizes” reword. “lower smaller” remove smaller.

Author’s response (2-31)
Thank you for pointing it out. We revised the sentence.

Author’s changes in the manuscript (2-31)
-Page 19, Line 282 – 285:
“Negative value of the jacobian means that higher temperature synthesizes lower smaller brightness temperature spectrum, thus, increases the HCl abundance in the retrievals calculation to compensate for the underestimation of the synthesized spectrum due to the temperature higher than the true value.”
→ “ A negative jacobian value means that highertemperatures induce a lower brightness temperature spectrum, thus increasing the HCl abundance in the retrieval to compensate for this underestimation. ”

Reviewer Comment (2-32)
- p.20 l.269: Change “a had” to “had a”.

Author’s response (2-32)
Thank you for your comment. We revised that according to your advice.

Author’s changes in the manuscript (2-32)
-Page 20, Line 320, 321:
“showed that the SMILES HCl a had” → “shows that the SMILES HCl has a”

Reviewer Comment (2-33)
- p.20 l.270: Change “were” to “was”.

Author’s response (2-33)
Thank you for pointing it out. We revised the sentence included this point.

Author's changes in the manuscript (2-33)
-Page 20, 21, Line 320 – 323:
"our theoretical error analysis showed that the SMILES HCl a had negative bias of 0.2–0.25 ppbv at 55 km which were consistent with the difference from the MLS and ACE-FTS within the 1 σ standard deviation."
→ "our theoretical error analysis shows that the SMILES HCl has a negative bias of at most 0.25 ppbv between 40 and 60 km; remaining difference between SMILES and MLS or ACE-FTS can be explained by the standard deviation in the comparison result."

Reviewer Comment (2-34)
- p.20 l.273-275: Reword (see previous comment about l.110-112).

Author's response (2-34)
We are grateful for your kind comment. We revised that according to your advice.

Author's changes in the manuscript (2-34)
-Page 21, 22, Line 329 – 333:
Added "In the lower and middle stratosphere, HCl is generated by the reaction of Cl with CH_4 and HO_2 and transported by circulation (e.g. Brewer-Dobson circulation). The HCl abundance is balanced by production ($Cl + HO2 \rightarrow HCl + O_2$) and loss($HCl + OH \rightarrow HCl + H_2O$, $HCl + h\nu \rightarrow H + Cl$) in the upper stratosphere and the mesosphere. Above the mesopause, the photodissociation becomes the dominant reaction and the HCl abundance decreases."

Reviewer Comment (2-35)
- p.20 l.281: "coincidenceS"

Author's response (2-35)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-35)
-Page 22, Line 338:
"coincidence" → "coincidences"

Reviewer Comment (2-36)
- p.21 Fig.12, caption: Change "dash" to "dashed"

Author's response (2-36)
Thank you for your comment. We revised that according to your advice.

Author's changes in the manuscript (2-36)
-Page 21, Figure 12, caption:
"dash" → "dashed"

Reviewer Comment (2-37)
- p.21 l.284: Change "The negative bias" to "A negative bias".

Author's response (2-37)
Thank you for your comment We revised that according to your advice.

Author's changes in the manuscript (2-37)
-Page 22, Line 341:
"The negative bias" → "A negative bias"

Reviewer Comment (2-38)
- p.21 l.295: "improvement of THE retrieval algorithm".

Author's response (2-38)
We appreciate you pointing it out. We revised that according to your advice.

Author's changes in the manuscript (2-38)
-Page 22, Line 355, 356:
"further improvement of retrieval algorithm" → "potential improvement in the SMILES retrieval algorithms"

3: Other minor changes in the manuscript.

Author's changes in the manuscript (3-1)
-Page 8, Line 141, 142:
Moved "Differences of HCl abundance between SMILES and SD-WACCM are shown in the panel (C)." from page 8, Line 133, 134 to page 8, Line 141, 142

Author's changes in the manuscript (3-2)
-Page 8, Line 144, 145:
Moved "The SMILES HCl abundance agrees well with SD-WACCM simulation (within 0.1 ppbv) in the stratosphere and middle mesosphere." from page 8, Line 134, 135 to page 8, Line 144, 145

Author's changes in the manuscript (3-3)
-Page 10, Line 180 & 182:
"Panels" → "Panel"

Author's changes in the manuscript (3-4)
-Page 18, Line 273:
"above 40 km" → "between 40 − 60 km"

Author's changes in the manuscript (3-5)
-Page 19, Line 281:
"with perturbation" → "with a perturbation"

Author's changes in the manuscript (3-6)
-Page 19, Line 281:
Removed "the"

Author's changes in the manuscript (3-7)
-Page 19, Line 286, 287:
"The vertical profile of temperature used for the retrieval procedure of" → "The temperature profiles used in the retrievals by"

Author's changes in the manuscript (3-8)

-Page 20, Line 288:
Removed “the”

Author’s changes in the manuscript (3-9)
-Page 20, Line 303:
“MLS and ACE-FTS” → “MLS and ACE-FTS comparisons”

Author’s changes in the manuscript (3-10)
-Page 20, Line 307, 308 :
“temperature profile used in retrieval calculation” → “the temperature profiles used in the retrievals”

Author’s changes in the manuscript (3-11)
-Page 20, Line 314, 315:
“SMILES and ACE-FTS was explained by the uncertainty in γ_{air} and temperature profile used in the retrievals.” → “SMILES and ACE-FTS can be explained by the uncertainty in γ_{air} and the temperature profiles used in the SMILES retrievals.”

Author’s changes in the manuscript (3-12)
-Page 20, Line 317, 318:
“The 1 % difference of γ_{air} might cause the HCl abundance increase about 0.03 ppbv at 55 km” → “A 1 % difference in γ_{air} might cause the HCl abundance to increase by about 0.03 ppbv”

Author’s changes in the manuscript (3-13)
-Page 22, Line 334:
“at altitude 30 to 70 km” → “for altitudes between 30 and 70 km.”

Author’s changes in the manuscript (3-14)
-Page 22, Line 335, 336:
“the comparison study with other instrument measurements and the theoretical error analysis”
→ “comparisons versus other measurements, and supported by a theoretical error analysis”

Author’s changes in the manuscript (3-15)
-Page 22, Line 337:
“with the” → “, versus”

Author’s changes in the manuscript (3-16)
-Page 22, Line 340, 341:
Added “the”
“with in the difference of” → “with differences within”

Author’s changes in the manuscript (3-17)
-Page 22, Line 342, 343:
“compared to the MLS and ACE-FTS” → “in comparisons versus MLS and ACE-FTS HCl profiles.”

Author’s changes in the manuscript (3-18)
-Page 22, Line 344, 345:
“We estimated the total error based on the perturbation method and error due to the uncertainty in the atmospheric temperature profile used in the retrieval calculation.”
→ “We estimated the total error for SMILES HCl based on the perturbation method and considering the uncertainties in atmospheric temperature profiles used in the retrievals.”

Author’s changes in the manuscript (3-19)

-Page 22, Line 350, 351:
"the temperature profile were capable of totally contributing up to 40 – 50 % of the negative bias in 50 – 60 km altitudes." →
"the temperature profile are capable of contributing a total of 40 – 50 % of the SMILES HCl negative biases at 50 – 60 km."

Author's changes in the manuscript (3-20)
-Page 22, Line 357, 358:
Removed "quantitative estimations of the"

Author's changes in the manuscript (3-21)
-Page 22, Line 358, 359:
"Further observations and model studies regarding HCl abundance including upper atmosphere are needed to understand the source and sinks"
→ "Further observations and model studies are needed to better understand the sources and sinks, "

Author's changes in the manuscript (3-22)
-Page 14, Fig. 8 & Page 15, Fig. 9 & Page 21, Fig. 12:
We changed the color of the ACE-FTS plot from red to green.

Author's changes in the manuscript (3-23)
-Page 14, Line 212 & Page 14, Fig. 8, caption & Page 15, Fig. 9, caption & Page21, Fig. 12, caption:
"red" → "green"

Validation of the vertical profiles of HCl over the wide range of the stratosphere to the lower thermosphere measured by SMILES

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Abstract. Hydrogen chloride (HCl) is the most abundant (more than 95 %) among inorganic chlorine compounds Cl_y in the stratosphere. The HCl molecule ~~has been~~ is observed to obtain long-term quantitative estimations of total budget of the stratospheric anthropogenic chlorine compounds. In this study, we provided HCl vertical profiles at altitudes of 16–100 km using the superconducting submillimeter-wave limb-emission sounder (SMILES) from space. The HCl vertical profile from the upper troposphere to the lower thermosphere is reported for the first time from SMILES observations; the data quality is quantified by comparison with other measurements and via theoretical error analysis. We used the SMILES Level-2 research product version 3.0.0. The period of the SMILES HCl observation was from October 12, 2009 to April 21, 2010, and the latitude coverage was 40°S–65°N. The average HCl vertical profile showed an increase with altitude up to the stratopause (∼45 km), approximately constant values between the stratopause and the upper mesosphere (∼80 km), and a decrease from the mesopause to the lower thermosphere (∼100 km). This behavior was observed in the all latitude regions, and reproduced by the Whole Atmosphere Community Climate Model in specified dynamics configuration (SD-WACCM~~model.~~).We compared the SMILES HCl vertical profiles in the stratosphere and lower mesosphere with HCl profiles from ~~MLS~~ Microwave Limb Sounder (MLS) on the Aura satellite, as well as from Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS) on SCISAT and from ~~TELIS~~ TEraheltz and submilimeter LImb Sounder (TELIS) (balloon-borne).The TELIS observations were performed using the superconductive limb emission technique, as used by SMILES. The globally averaged vertical HCl profiles of SMILES ~~well agreed~~ agreed well with those of MLS and ACE-FTS within 0.25 and 0.2 ppbv between 20 and 40 km ~~, respectively.~~ (within 10% between 30 and 40 km, There's a larger discrepancy below 30km.), respectively. The SMILES HCl concentration was smaller than those of MLS and ACE/FTS as the altitude increased from 40 km, and the difference was approximately 0.4–0.5 ppbv (12 – 15%) at 50–60 km. The difference between SMILES and TELIS HCl

observations was about 0.3 ppbv in the polar winter region between 20 and 34 km, except near 26 km. SMILES HCl error sources that may cause discrepancies with the other observations are investigated by a theoretical error analysis. We calculated errors caused by the uncertainties of spectroscopic parameters, instrument functions, and atmospheric temperature profiles. The jacobian for the temperature explains the negative bias of the SMILES HCl concentration concentrations at 50–60 km. The HCl vertical profile from the upper troposphere to the lower thermosphere is reported for the first time from SMILES observations; the data quality is quantified by comparisons with other measurements and via theoretical error analysis.

1 Introduction

Hydrogen chloride (HCl) is the most abundant species of all total inorganic chlorine compounds Cl_y in the stratosphere. More than 95 % of Cl_y exists as HCl at about 1 hPa (∼50 km) (see Froidevaux et al. (2008)). Thus, HCl has been used for long-term quantitative estimates of the total budget of stratospheric chlorine (WMO, 2010). Stratospheric HCl vertical profiles have been observed globally by several satellites. The halogen occultation experiment (HALOE) on the Upper Atmosphere Research Satellite measured the stratospheric HCl profiles continuously from late 1991 through November 2005 (Russell III et al., 1996). Comparisons of HCl profiles obtained by the Aura Microwave Limb Sounder (MLS) and the Atmospheric-Chemistry-Experiment Fourier-Transform Spectrometer (ACE-FTS) have been reported by Froidevaux et al. (2008) and Mahieu et al. (2008). These profiles agreed within 5% (0.15 ppbv) at 0.5 hPa (∼53 km). The Aura/MLS and ACE-FTS HCl concentrations were larger (by 10% – 20%) than those from HALOE HCl (WMO, 2010). (WMO, 2010). Balloon-borne remote sensing in polar winter region was performed using the TErahertz and submillimeter LImb Sounder (TELIS) from 2009 to 2011 (Birk et al., 2010). Xu et al. (2018) reported the error analysis and comparison of some trace gas profiles provided by TELIS.

The superconducting submillimeter-wave limb-emission sounder (SMILES) was launched in 2009 to observe the atmospheric compositions of ozone and species related to the stratospheric ozone destruction cycle including HCl (Kikuchi et al., 2010). The SMILES mission is a joint project of the Japan Aerospace Exploration Agency and the National Institute of Information and Communications Technology (NICT). The SMILES is an instrument used to conduct limb observations from the International Space Station (ISS) using super-sensitive 4 K superconducting receivers in the submillimeter-wave regions (625 and 649 GHz bands). The non-sun-synchronous circular orbit of the ISS gave us an opportunity for observations of HCl profiles at different local times. The unprecedented low noise of the SMILES instrument provided a sensitivity 10 times superior to those of previous microwave/sub-millimeter limb emission instruments used to observe HCl spectra from space (Kikuchi et al., 2010). The SMILES observations were used to reveal small abundances of atmospheric species in the stratosphere and mesosphere (e.g. Sato et al. (2017); Yamada et al. (2020))

A comparison of the HCl profiles inside the Antarctic vortex has been performed for November 19—24, 2009 using the SMILES, Aura/MLS, and ACE-FTS data (Sugita et al., 2013). The SMILES HCl values (version 2.1.5 of NICT products) agreed within 10 % (0.3 ppbv) with those of the MLS and ACE-FTS HCl data between the potential-temperature levels of 450 and 575 K and 425 and 575 K, respectively. In contrast to the previous study, this study used version 3.0.0 SMILES data and provided a global comparison, including the stratosphere and mesosphere. The HCl climatology using SMILES NICT version 2.1.5 was reported by Kreyling et al. (2013).

In this study, SMILES observations of we examined HCl vertical profiles from 16–100 km, including the upper mesosphere and lower thermosphere (MLT) region. We used the SMILES NICT Level-2 product version 3.0.0 (v300), which was released in late 2012 (http://smiles.nict.go.jp/pub/data/index.html). HCl vertical profiles from the upper troposphere to the lower thermosphere are reported for the first time. We perform a validation of the SMILES HCl vertical profiles by comparisons with the corresponding global model results of SD-WACCM, satellite observations from MLS and ACE-FTS, balloon-borne observations from TELIS and we provide a SMILES HCl error analysis.

The SMILES HCl observations and retrieval procedure are described in Sect. 2. The HCl vertical profile derived by the SMILES product and comparisons of HCl distribution between 40°S–60°N with SD-WACCM are shown in Sect. 3. Results of HCl comparisons between SMILES and other instruments are described in Sect. 4. An estimation of the systematic and random errors is presented in Sect. 5. Section 6 describes our conclusions.

2 SMILES HCl observationobservations

The SMILES instrument had been attached to the Japanese Experimental Module (JEM) on the ISS, since September 2009. The period of the SMILES observation was from SMILES operational period started on October 12, 2009 to and ended on April 21, 2010. The ISS has a non-sun-synchronous circular orbit with an inclination angle of 51.6° with respect to the equator. The height of the ISS changed slowly over the observational period ranging 340–360 km. We tilted the line-of-sight at a 45° angle in the direction of the forward movement of the ISS to observe the northern polar region. The SMILES parameters are summarized in Table 1. The details of the SMILES observation observations are described in Kikuchi et al. (2010) and in an ozone-validation paper by Kasai et al. (2013), respectively.

Herein, we briefly describe the SMILES atmospheric observations in terms of the HCl spectra. The SMILES had three frequency bands in the submillimeter-wave regions, Band A (624.32–625.52 GHz), Band B (625.12–626.32 GHz), and Band C (649.12–650.32 GHz). Each single-scan provided a combination of two out of three of the frequency bands: Bands A+B, C+B, and or C+A. The combinations changed on a daily basis, as described in Kikuchi et al. (2010). The rotational transitions of two HCl isotopologues were located in Bands B and A for $H^{35}Cl$ at 625.9 GHz and $H^{37}Cl$ at 624.9 GHz, respectively. Two acousto-optical spectrometers (AOSs) were equipped in the SMILES instrument. The spectral resolution obtained by AOS1 and AOS2 was 0.8 MHz. The band configuration for each HCl isopologues had three patterns as follows:,

a) $H^{37}Cl$ observation by Band-A with AOS1,

b) $H^{37}Cl$ observation by Band-A with AOS2,

c) $H^{35}Cl$ observation by Band-B with AOS2.

Band-B was always associated with AOS2.

Figure 1 shows the number of observations per day obtained for $H^{35}Cl$ (Band-B) and $H^{37}Cl$ (Band-A) for 5° latitude bins during the SMILES observational period. It should be noted that the sampling is not homogeneously distributed for the

Table 1. SMILES specification

Parameter	Characteristics
Orbit	Non-sun-synchronous orbit
	Inclination angle 51.6°
	Altitude 340–360 km
	$\sim$ 90-min orbital period
Latitude coverage	38°S-65°N(nominal)
Measurement geometry	Limb scan
Number of scans	1630 scans per day
Integration time	0.47 s
Frequency range	624.32–625.52 GHz (Band-A)
	625.12–626.32 GHz (Band-B)
	649.12–650.32 GHz (Band-C)
Receiver system	SIS*1 mixer and HEMT*2 amplifiers
Spectrometer	Acousto-Optical Spectrometers
	(AOS1 and AOS2)
Frequency resolution	0.8 MHz
System noise temperature	$\sim$ 350 K

*1: Superconductor-Insulator-Superconductor, *2: High-Electron-Mobility Transistor

SMILES observations. The SMILES instrument was sometimes not in operation, for example, when the ISS was boosted up from a low to a high altitude, and the solar panels disturbed the observational line-of-sight. The SMILES observational latitude range was 65°N–38°S in nominal operations, and changed to 38°N–65°S when the ISS performed a yaw maneuver of 180°, which happened three times during the 7-month-long observational period.

We used the SMILES NICT Level 2 product version 3.0.0 (v300) for this study. The general method of v300 for the HCl retrieval is similar to that of the Level 2 product version 2.1.5 (v215) (Kasai et al., 2013). The major updates from the v215 product are as follows: 1) An improvement in the spectrum calibration, particularly for the gain non-linearity of the receiver system (Ochiai et al., 2013), 2) An improvement in the accuracy of the tangent height estimation (Ochiai et al., 2013), and 3) An update of the temperature retrieval. ~~The~~ The temperature profile used in the SMILES version 3.0.0 retrieval was synthesized, assuming the hydrostatic equilibrium, using the Goddard Earth Observing System, Version 5 (GEOS-5) reanalysis meteorological datasets and the climatology based on the Aura/MLS measurements (Kuribayashi et al., 2017). The GEOS-5 datasets were used in the upper troposphere and stratosphere, and the Aura/MLS datasets were used in the mesosphere and lower thermosphere, respectively. The temperature profile was also retrieved using the ozone transition in the SMILES version 3.0.0 retrieval process, but it was not applied to the retrieval of atmospheric species including HCl, except for ozone, to avoid systematic error propagation issues (SMILES-NICT, 2014). The level 1b spectrum data version 008 was used for the SMILES

Table 2. Spectroscopic line parameters measured in the laboratory and used in the processing.

Species	Line frequency [MHz]	Air-broadening [MHz/Torr] /temperature dependence	Frequency shift [MHz]
	625 901.6627[a]		
$H^{35}Cl$	625 918.7020[a]	3.39/0.72[b]	0.145[b]
	625 931.9977[a]		
	624 964.3718[a]		
$H^{37}Cl$	624 977.8059[a]	3.39/0.72[b]	0.145[b]
	624 988.2727[a]		

a: Laboratory measurements by Cazzoli and Puzzarini (2004). b:Baron et al. (2011)

NICT Level 2 product v300 (Ochiai et al., 2013). The spectroscopic parameters for the HCl retrieval were based on Cazzoli and Puzzarini (2004) and Laboratory measurement, as summarized in Table.2. We used only the $H^{35}Cl$ data because the intensities of the $H^{37}Cl$ spectra were weaker than those of $H^{35}Cl$, as shown in Fig. 2.

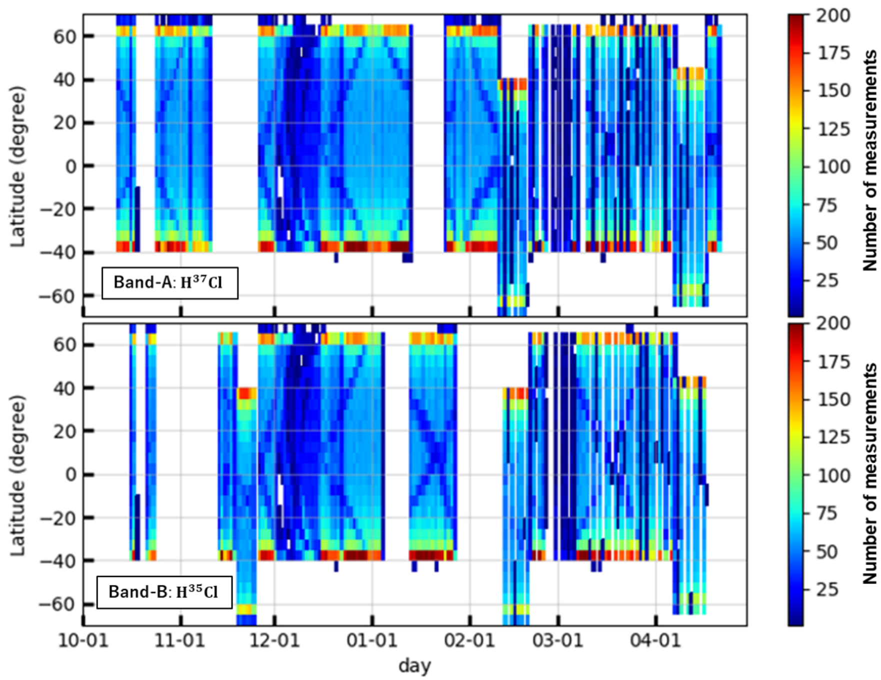

Figure 1. Number of HCl observations for a 5° latitude bin in 1 day for the range of SMILES observational latitudes and period (October 12, 2009 – April 21, 2010). The top and bottom panels show the number of observations from Band-A ($H^{37}Cl$) and Band-B ($H^{35}Cl$), respectively.

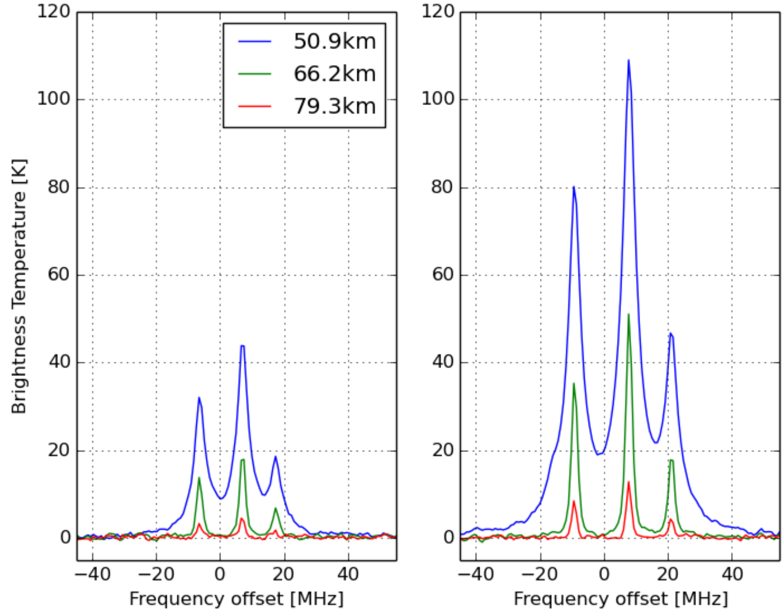

Figure 2. Left: An example of the $H^{37}Cl$ ~~spectrum~~ spectra observed by SMILES from a single scan measurement. Tangent heights are approximately 50, 65, and 80 km. Center frequency is 624.98 GHz. Right: An example of the $H^{35}Cl$ ~~spectrum~~ spectra for the same condition as for the $H^{37}Cl$ ~~spectrum~~spectra. Center frequency is 625.92 GHz.

The criteria for the selection of the $H^{35}Cl$ profiles used in this study are as follows: (1) χ^2 less than 0.8. χ^2 is defined by

$$\chi^2 = [\mathbf{y} - \mathbf{F}(\mathbf{x})]^{\mathbf{T}} \mathbf{S}_{\epsilon}^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x})] + [\mathbf{x} - \mathbf{x}_{\mathrm{a}}]^{\mathbf{T}} \mathbf{S}_{\mathrm{a}}^{-1} [\mathbf{x} - \mathbf{x}_{\mathrm{a}}] \tag{1}$$

$\mathbf{y}$ is the observed spectrum, $\mathbf{S}_{\epsilon}$ is the covariance matrix of the measurement noise, $\mathbf{x}_{\mathrm{a}}$ is the a priori state, $\mathbf{S}_{\mathbf{a}}$ is the covariance matrix of $\mathbf{x}_{\mathrm{a}}$, and $\mathbf{F}(\mathbf{x})$ is the forward model depending on the state vector $\mathbf{x}$. We used the U.S.standard atmosphere profiles as the a priori state ($\mathbf{x}_{\mathrm{a}}$) (US-Standard, 1976). They are used separately for polar, equatorial, summer mid-latitude, or winter mid-latitude regions.The forward model is essentially the atmospheric radiative transfer and the instrument model. (2) measurement response, MR, between 0.8 and 1.2 for each vertical grid. MR is defined by

$$\mathrm{MR}[\mathrm{i}] = \sum_{\mathrm{j}} \mathbf{A}[\mathrm{i},\mathrm{j}] \tag{2}$$

$\mathbf{A}$ is the averaging kernel matrix. A value of MR near unity indicates that most of the information in the retrieval results is provided by observations. A low value of MR indicates that the retrieval results are largely influenced by the a priori state and are forced to be identical to a priori values.

An example $H^{35}Cl$ profile and its averaging kernel are presented in Fig. 3 for the single scan spectrum observation at latitude of 38.3° N, longitude of 12.7°E, and solar zenith angle (SZA) of 160.9° on 15 November 2009. We assumed that the natural isotopic abundance of $H^{35}Cl$/HCl was 0.7576 by Berglund and Wieser (2011) for these retrievals . The peak values of the averaging kernels for the SMILES $H^{35}Cl$ profiles exceeded 0.8 in the height range of 16 km to 90 km (∼ 100 hPa to 0.001 hPa) as shown in Fig. 3. The full width at half maximum (FWHM) of the averaging kernels is approximately 3 –4 km at altitudes of 18 km–50 km and becomes greater than 5 km for the altitude of altitudes 50 km–90 km.

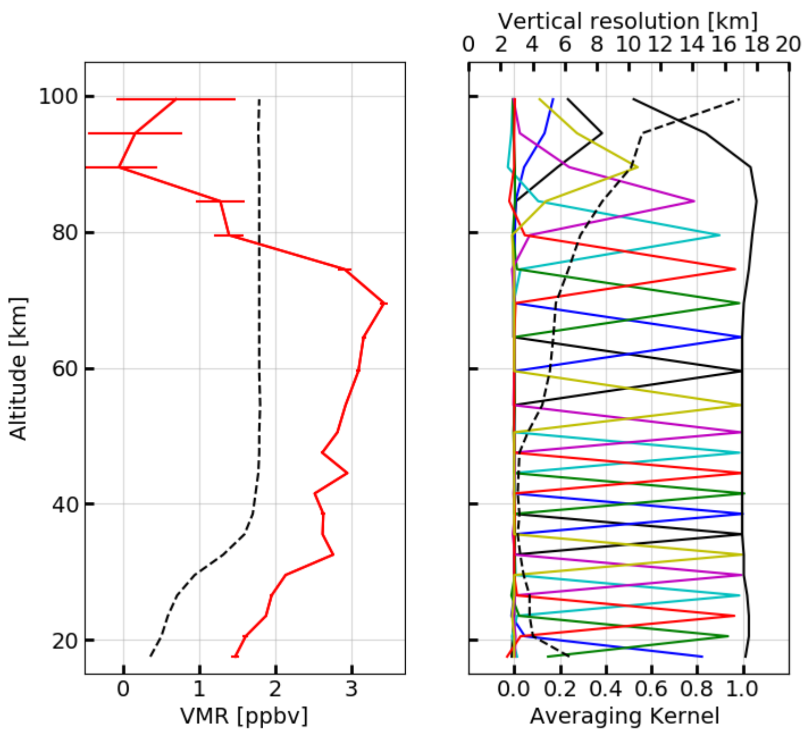

Figure 3. Left: An example of a HCl profile retrieved from the $H^{35}Cl$ spectrum on November 15, 2009 at a latitude of 38.3°N, longitude 12.7°E, and SZA of 160.9°. The solid red line indicates the retrieved HCl volume mixing ratio (VMR), and the error bar is the root sum square of the smoothing and measurement errors. The dashed black line is an a priori HCl profile used for the retrieval procedure. The right panel shows the averaging kernel,the measurement response (indicated by the thick black line) and the FWHM of the averaging kernel profile (indicated by the thick dashed black line).

3 Vertical and latitudinal distribution of SMILES HCl

In this section, we provide the HCl vertical profiles derived from the SMILES NICT Level 2 product v300. Figure 4 (A) shows the HCl zonal mean distribution in the latitudinal range from 40°S to 60°N. The observation periods are time period is between October 16, 2009–December 15, 2009 (Left), December 16, 2009–February 15, 2010 (Middle), and February 16, 2010–April and April 17, 2010 (Right). 2010. The HCl vertical profile showed an increasing with the altitude increased distribution shows an increase with altitude with a maximum below the stratopause (∼45 km), approximately constant values between the stratopause and the upper mesosphere (∼80 km), and ~~decreased with the altitude increasing~~ a decrease with altitude from the mesopause to the lower thermosphere (∼100 km). In the lower and middle stratosphere, HCl is generated by the reaction of Cl with CH_4 and HO_2 and transported by circulation (e.g. Brewer-Dobson circulation). The HCl abundance is balanced by production ($\mathrm{Cl + HO2 \rightarrow HCl + O_2}$) and loss ($\mathrm{HCl + OH \rightarrow Cl + H_2O, HCl + h\nu \rightarrow H + Cl}$) in the upper stratosphere and mesosphere. Near the mesopause, the photodissociation becomes the dominant reaction, and the HCl abundance decreases with height (Brasseur and Solomon, 2005).

This behavior was reproduced by the Whole Atmosphere Community Climate Model version 4 (WACCM4) in specified dynamics configuration (SD-WACCM) (Lamarque et al., 2012; Limpasuvan et al., 2016), see the panel (B). ~~Differences of HCl abundance between SMILES and SD-WACCM are shown in the panel (C). The SMILES HCl abundance agrees well with SD-WACCM simulation (within 0.1 ppbv) in the stratosphere and middle mesosphere. At 70 – 100km, the SMILES HCl abundances are about 0.4 – 1.3 ppbv larger than in the SD-WACCM simulation. For these comparisons, the SD-WACCM profiles are interpolated linearly for each SMILES observation point and time from the nearest model positions and times.~~ In the specified dynamics configuration, the simulated meteorological fields in the troposphere and stratosphere are constrained to the Global Modeling and Assimilation Office Modern-Era Retrospective Analysis for Research and Applications (Rienecker et al., 2011). The horizontal resolution of SD-WACCM simulation is 1.9° and 2.5° for latitude and longitude, respectively. The vertical grid is from the ground to 150 km with 88 levels, and the time resolution is 3 hours. Differences of HCl abundance between SMILES and SD-WACCM are shown in the panel (C). For these comparisons, the SD-WACCM profiles are interpolated linearly for each SMILES observation point and time from the nearest model positions and times. The panel (D) shows the number of selected SMILES ~~observation.~~ observations. The SMILES HCl abundance agrees well with SD-WACCM simulation (within 0.1 ppbv) in the stratosphere and middle mesosphere. At 70 – 100km, the difference of the HCl VMR between SMILES and WACCM in the upper mesosphere and lower thermosphere is 0.4 – 1.0 ppbv, which is larger than the total systematic error in the SMILES HCl VMR estimated by quantitative error analysis (0.1 – 0.4 ppbv). Thus, the difference is significant. It might be caused by a combination of the SMILES observation and the WACCM model uncertainties. The underestimation of the model could be because of uncertainties in the HCl photodissociation and the reaction with OH radical, which are dominant destruction mechanisms of HCl for the mesosphere/thermosphere (Brasseur and Solomon, 2005).

4 Comparison of SMILES HCl profile with those obtained using other instruments

We performed a comparison between SMILES HCl products and other datasets obtained using the MLS on the Aura satellite, the ACE-FTS on the SCISAT satellite, and the balloon-borne TELIS instrument. The characteristic of instruments and datasets are summarized in Table 3. The mean absolute difference between SMILES and the other instrument is defined as

$$\Delta_{\mathrm{abs}}(z) = \frac{1}{N(z)} \sum_{i=1}^{N(z)} x_c(z) - x_s(z), \quad (3)$$

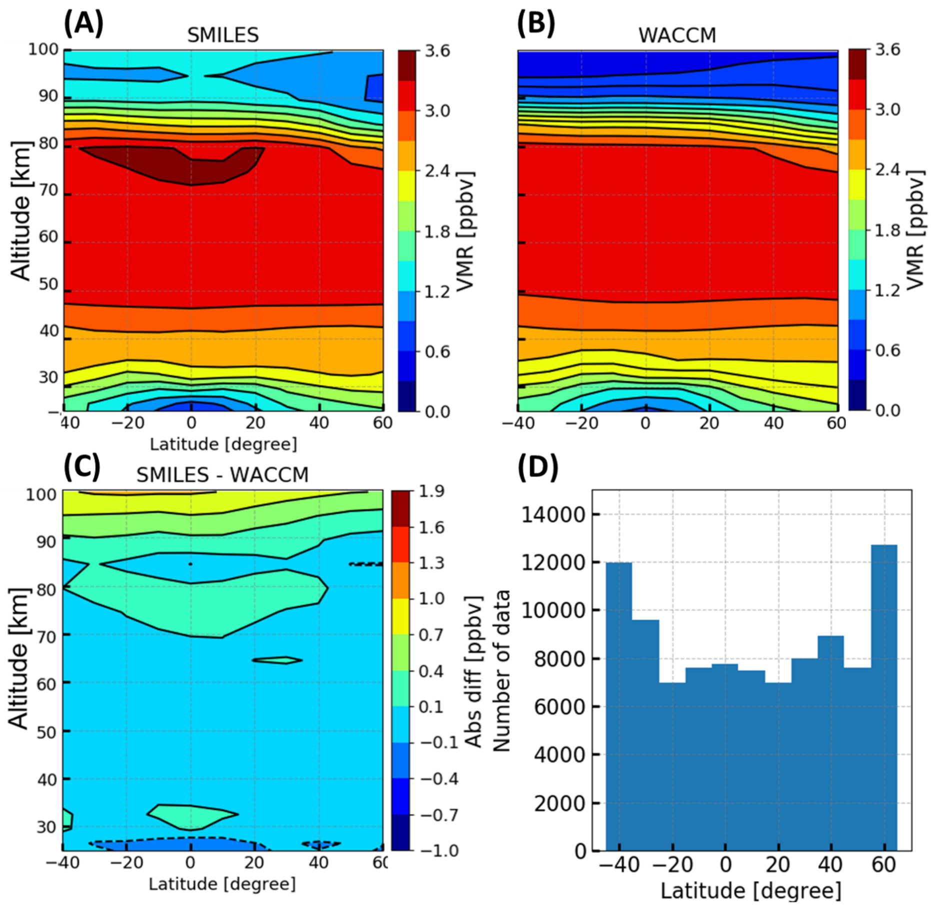

Figure 4. Zonal mean distribution of HCl from SMILES, SD-WACCM and their differences (SMILES - SD-WACCM) are shown in ~~rows~~ panel (A), (B), and (C), respectively. Each panel displays distributions as a function of latitude (x-axis) and altitude (y-axis)~~in three time range~~. The latitude range covers between 40°S and 60°N. The number of HCl profiles observed by SMILES is described in panel (D) ~~row~~ as a histogram versus latitude. ~~Left panel: Time range~~ The time period is between October 16, 2009 and ~~December 15, 2009. Middle panel: Between December 16, 2009 and February 15, 2010. Right panel: Between February 16, 2010 and~~ April 17, 2010. The data ~~was~~ is averaged within ~~a~~ latitude ~~bin~~ bins of 10°.

where $x_s(z)$ and $x_c(z)$ are the HCl volume mixing ratio (VMR) at an altitude z for SMILES and the other instrument, respectively. $N(z)$ is the number of coincidence at an altitude z.

The mean relative difference (in percentage) is given by

$$\Delta_{\mathrm{rel}}(z) = \frac{1}{N(z)} \sum_{i=1}^{N(z)} \frac{x_c(z) - x_s(z)}{(x_c(z) + x_s(z))/2} \times 100. \tag{4}$$

The coincidence criteria between the SMILES observations and those of the other instruments was set to within 2° for latitude, 8° for longitude, and 5 h for time, following the previous work ~~of the~~ on MLS observations (Froidevaux et al., 2008).

Table 3. Characteristics of instruments and data sets used in the comparison.

Instrument &Platform	Measurement type -	Time period overlapped with SMILES (yyyy/mm/dd)	Data ~~version~~ version	Number of ~~coincident observations~~ coincidence	Altitude range
MLS Aura	Limb emission sub-mm/microwave	2010/01/24 - 2010/01/27 (Band-13)	4.2	4356	100 – 0,32 hPa
ACE-FTS SCISAT	Solar occultation mid IR	2009/10/16 - 2010/04/17	4.0	935	7 – 63 (Equator) 6 – 59 km (Polar)
TELIS Balloon borne	Limb emission submm/THz	2010/01/24	3v02	4	16 – 34 km

4.1 Comparison with Aura/MLS

The Aura satellite has a sun-synchronous orbit with an ascending node at 1:45 PM local time (Waters et al., 2006). The measurements cover latitudes from 82°S to 82°N and provide approximately 3500 vertical profiles each day. We used version 4.2 of the Aura/MLS HCl profiles for these comparisons. The Aura/MLS team provided two HCl products one from spectral band 13 (HCl-640-B13) and one from band 14 (HCl-640-B14). The HCl-640-B13 product is available for the pressure region between 100 hPa and 0.32 hPa. The single profile precision of HCl-640-B13 is less than 0.8 ppbv (25%) in the stratosphere, and the estimated accuracy is approximately 0.3 ppbv (10%) (Livesey et al., 2018).

The vertical and horizontal resolutions are 3–5 km and 200–400 km, respectively. The vertical resolution of MLS is of the same order as that of SMILES. We used the recommended parameters "Status," "Quality," and "Convergence" for screening the Aura/MLS data based on Livesey et al. (2018). The good profiles were selected using (1) Quality < 1.2, (2) Convergence > 1.05, and (3) Status = Even. The SMILES profiles were linearly interpolated onto the MLS pressure grid for the comparisons.

Figure 5 shows a comparison of the HCl vertical profiles derived from SMILES and Aura/MLS in the SMILES latitude range. The total number of coincident profile pairs was 4356. The HCl profiles from the SMILES measurements agreed with those of MLS within 0.25 ppbv between 20 and 40 km (50–3 hPa), while large biases about 0.4 ppbv were confirmed between 40 and 55 km (3–0.32 hPa). The differences between HCl profiles ~~increases~~ increase from the stratosphere to the mesosphere. Figure 6 shows the zonal mean absolute differences averaged for 5° latitude bins. The differences did not change with latitude in the altitude region between 35–55 km (7–0.32 hPa). While below 35 km of altitude, several areas of large differences can be identified, especially in the equatorial region.

Figure 7 displays more detailed features of the differences between the SMILES and MLS profiles. ~~Panels~~Panel (A) of Fig. 7 show the averaged HCl vertical profiles of SMILES (blue) and MLS (red), and the absolute difference in each latitude region (40°S–20°S, 20°S–20°N, 20°N–50°N, and 50°N–65°N). ~~Panels~~Panel (B) show the latitudinal distribution averaged in 5° bins for SMILES (blue) and MLS (red) at 30, 40, and 50 km. The error bar shows the 1 σ variability for each latitude grid. In the equatorial regions, the SMILES HCl profiles were 0.5 ppbv larger than those of MLS at ~~below~~ 30 km (10 hPa). ~~There is a possibility that this difference was caused by water vapor. The SMILES HCl profile was retrieved without considering water vapor effect and the influence of the water vapor was thus one of the possible results of the latitudinal difference.~~ On the other hand, the HCl profiles from the SMILES measurements were 0.4 ppbv ~~less~~ lower than those of MLS in all latitude regions at 50 km.

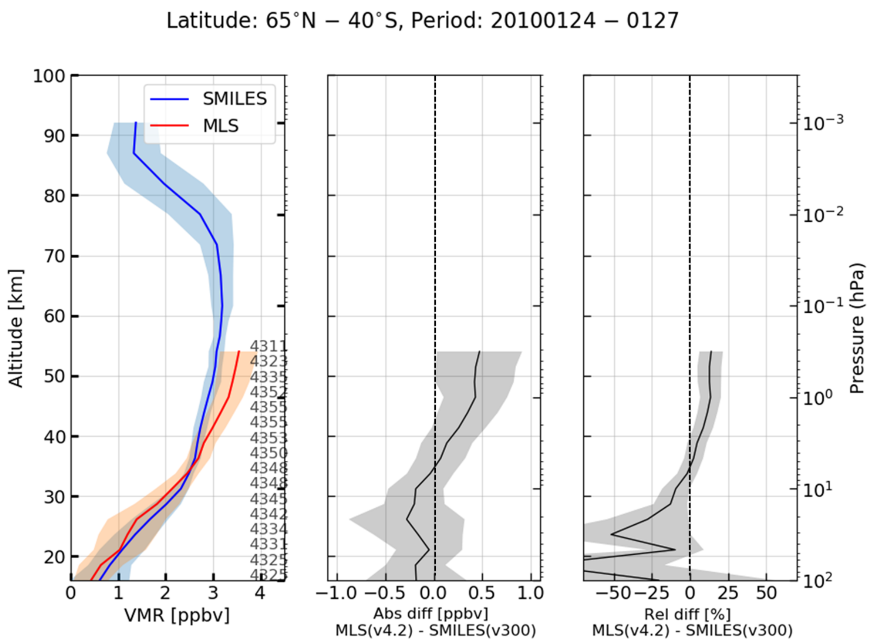

Figure 5. Comparison between SMILES and MLS profiles from January 24, 2010 to January 27, 2010. Left: mean HCl VMR values (solid lines) and 1 σ (shaded areas) for SMILES and MLS. The blue and red lines indicate the SMILES HCl profiles and MLS profiles, respectively. Middle: The absolute difference between the SMILES and MLS profiles calculated using Eq (3). Right: The relative difference between the SMILES and MLS profiles calculated using Eq (4).

4.2 Comparison with SCISAT/ACE-FTS

The ACE-FTS is an instrument mounted on the Canadian SCISAT satellite. SCISAT moves along an orbit at a 650 km altitude and is inclined at 74° to the equator (Bernath et al., 2005). The ACE comprises two instruments: the Fourier Transform Spectrometer (ACE-FTS) and the Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation (ACE-MAESTRO). The HCl observation has been performed using ACE-FTS. The ACE-FTS observes solar occultation spectra in the infrared spectral region (750−4400 cm^{-1}) with a high spectral resolution (0.02 cm^{-1}). We used ACE-FTS HCl profiles from the version 4.0 software, which is the latest data version ~~.~~ (Boone et al., 2020). The vertical resolution of the

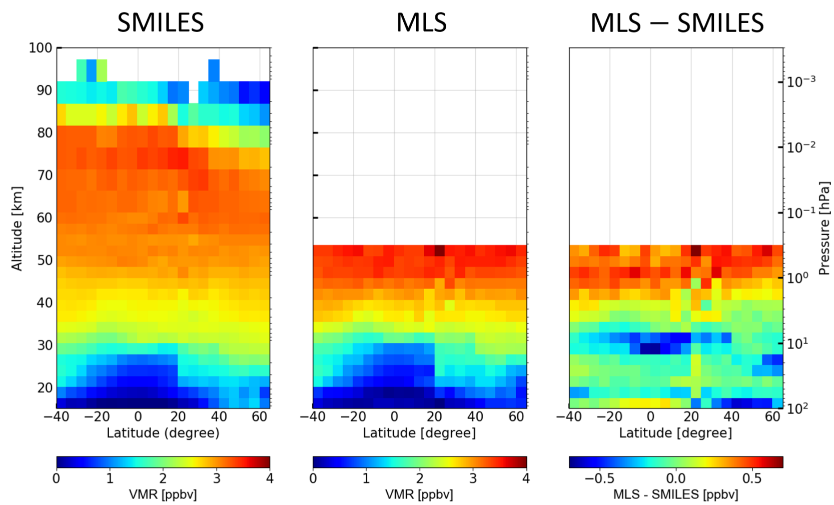

Figure 6. Left: SMILES zonal mean HCl profiles versus latitude. Profiles that satisfied coincidence criteria between SMILES and MLS for January 24 – 27, 2010 were used here. Middle: Zonal mean HCl profiles from MLS. Right: The absolute difference of the matched pairs versus latitude.

ACE-FTS HCl retrieval is 3–4 km, and the values from the retrieval grid are interpolated onto the 1-km grid, using a piecewise quadratic method (Bernath et al., 2005). The vertical resolution of ACE-FTS is of the same order as that of SMILES. In this study, we used the data within ±3 times the median absolute deviation (3-MAD) from the median to remove significantly large positive and negative biases based on Boone et al. (2019).

Figure 8 presents the comparison of the HCl vertical profiles between SMILES and ACE-FTS in the latitude range of 40°S–65°N. The total coincident data number was ~~945.~~ 935. The SMILES HCl profiles agreed with those of the ACE-FTS by 0.2 ppbv between 20 and 40 km (50–2 hPa), while 0.5 ppbv (15%) ~~less~~ lower than those of ACE-FTS above 40 km. This negative bias in the SMILES HCl concentration was confirmed as in the case of the MLS–SMILES comparison. The results of the comparison with the ACE-FTS for each latitudinal region are presented in Fig. 9 (A) in a manner similar to Fig. 7 (A). ~~In the tropic~~ In the tropical region, a large discrepancy was observed at an altitude ~~altitude~~ of 25 km because of relativity poor sampling number. The HCl profiles observed using the SMILES were about 0.5 ppbv (15%) less than those of ACE-FTS for each latitudinal region above 50 km.

The period of the SMILES $H^{35}Cl$ observation (from Oct 16, 2009 to Apr 17, 2010) was covered by the ACE-FTS observation period, while the MLS Band-13 observation period had an ~~overlapped~~ overlap of only 4 days. We analyzed the difference between SMILES and ACE-FTS for each month and ~~conformed~~ confirmed the seasonal variation of the bias. Figure 9 (B) shows a seasonal variation of the difference between the SMILES and ACE-FTS profiles in the northern hemisphere (from

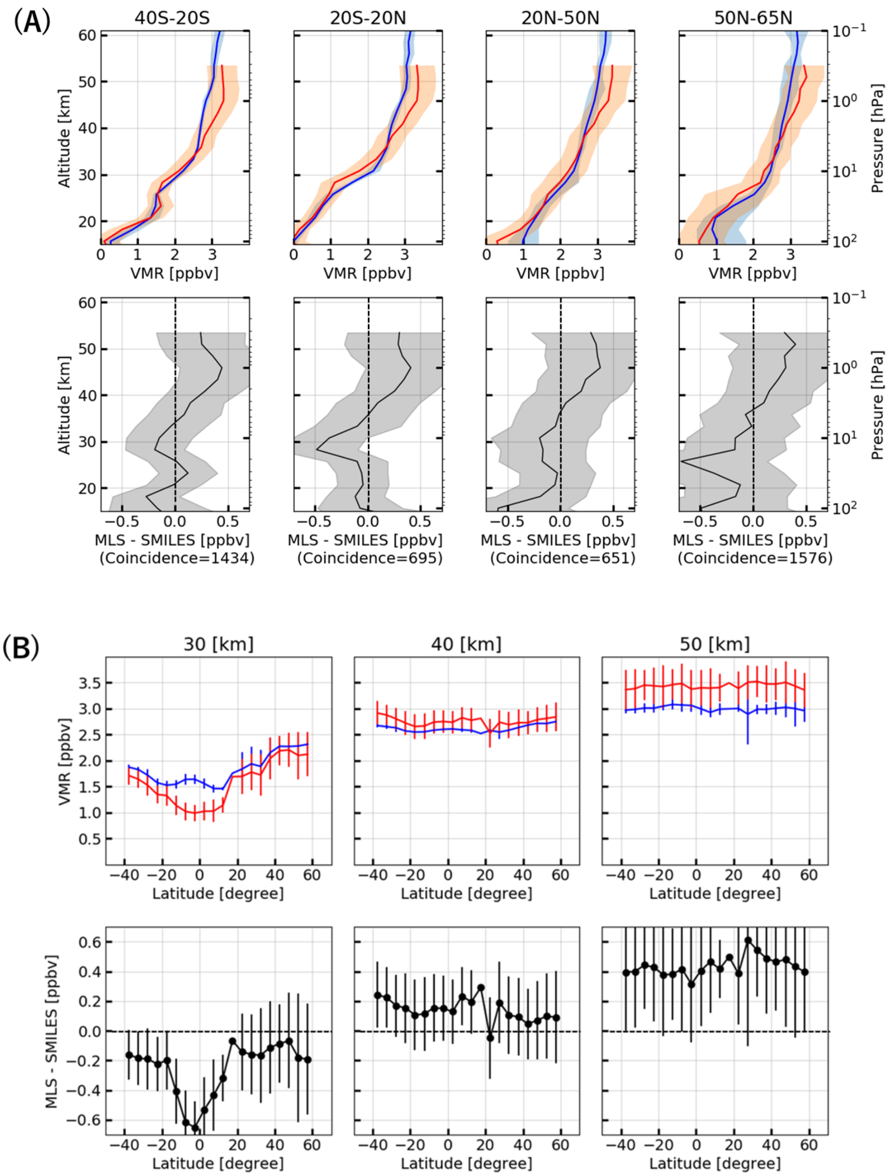

Figure 7. (A):Mean HCl profiles, 1 σ uncertainty based on variability (shaded region) and the absolute differences for each latitude region. Upper row: The blue and red lines indicate the mean profiles from SMILES and MLS for 40°S-20°S, 20°S-20°N, 20°N-50°N, and 50°N-65°N (left to right panels). Lower row: The absolute difference between the SMILES and MLS profiles calculated using Eq (43) for each latitude region. (B): Latitudinal variation of SMILES and Aura/MLS HCl profiles at three altitude levels. The profiles were averaged for each 5° bin. Upper row: Mean of SMILES (blue) and MLS (red) profiles for each altitude level. The error bars indicate 1 σ uncertainties. Lower row: The means and 1 σ of the absolute differences are displayed.

30°N–65°N latitude range). The upper panels in Fig. 9 (B) show a mean of the SMILES (blue) and ACE-FTS (~~red~~green) profiles for each month at three altitude levels. The difference for every month, shown in the lower panels, was represented by the mean for each month at each altitude, and the error-bar showed the 1 σ standard deviation. No significant seasonal dependence of the difference was observed. The difference of HCl value from these measurements was consequently about 0.5 ppbv at 50 km.

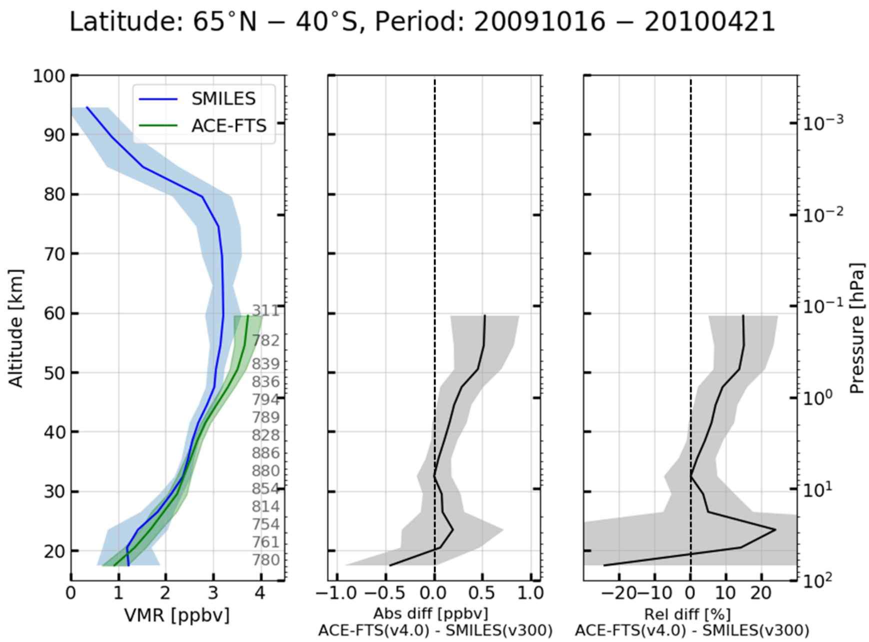

Figure 8. Comparison between SMILES and ACE-FTS profiles from October 16, 2009 to April 17, 2010. Left: The mean HCl VMR values (solid lines) and 1 σ (shaded areas) for SMILES and ACE-FTS. The blue and ~~red~~ green lines are the SMILES and ACE-FTS HCl profiles, respectively. Middle: The absolute difference between the SMILES and ACE-FTS profiles calculated using Eq (3). Right: The relative difference between the SMILES and ACE-FTS profiles calculated using Eq (4).

4.3 Comparison with balloon-borne instrument TELIS

The TELIS (Birk et al., 2010) is a balloon-borne THz/sub-millimeter-wave spectrometer with superconductive 4K technology similar to that of SMILES. TELIS was one of the instruments used in the balloon observation campaigns for three winters (2009–2011) in Kiruna, Sweden. Limb observations were performed from a flight altitude of 30–35 km, toward the stratosphere and the upper troposphere with 1.5–2 km vertical sampling. TELIS was used to observe $H^{37}Cl$ at the transition frequency of 1873.4 GHz in the 1.8 THz channel (Level-1b version 3v02), which corrected non-linearity problems in the radiometric calibration using a quadratic term for each frequency segment. The details on the TELIS 1.8 THz channel and its L1 data processing are shown in Suttiwong (2010). In this study, we compared two $H^{37}Cl$ profiles obtained by TELIS (observation

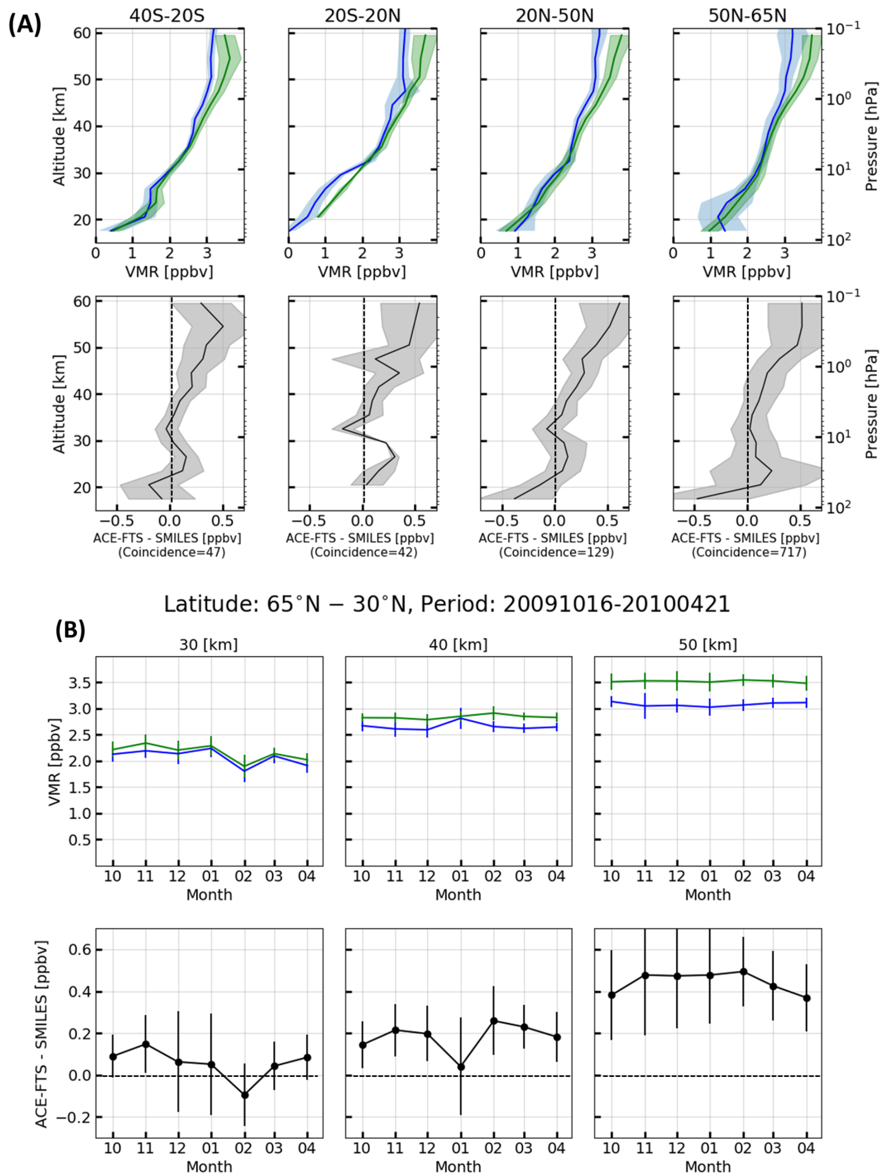

Figure 9. Figure-(A): The mean of HCl values, 1 σ and the absolute difference for each latitudinal region are shown. Upper row: The blue and ~~red~~ green lines indicate the SMILES HCl profiles and ACE-FTS profiles for each latitudinal region, 40°S-20°S, 20°S-20°N, 20°N-50°N, and 50°N-65°N from the left side. Lower row: The absolute difference between the SMILES and ACE-FTS profiles calculated using Eq (3) for each latitudinal region. Figure-(B): Seasonal variation in the SMILES and ACE-FTS HCl profiles at three altitude levels. Latitudinal range is 30°N – 65°N. Upper row: Monthly mean of SMILES (blue) and ACE-FTS (~~red~~green) HCl profiles for each altitude level. The error bars correspond the 1 σ. Lower row: The mean and 1 σ uncertainty of the absolute difference.

number 20044 and 21537) with the SMILES profiles (Band-B and identifiers 761 and 762) in terms of geolocation over Kiruna (67.8 N° 20.4 E°) and time on January 24, 2010 over Kiruna. 2010. The total error of the $H^{37}Cl$ profile derived from the TELIS measurement was estimated to be 0.25–0.5 ppbv, with a vertical resolution of 3 km (Xu et al., 2018).

Figure 10 shows a comparison of the HCl vertical profiles of SMILES and TELIS. The left, middle and right panels show the SMILES and TELIS HCl vertical profiles, their absolute difference, and their relative difference, respectively. We converted the $H^{37}Cl$ amount to HCl using the natural isotopic abundance ($H^{37}Cl/HCl = 0.2424$). The SMILES and TELIS HCl profiles agreed well, within 0.5 ppbv from 17 km to 34 km. A depletion in the HCl profile was observed below 25 km in both the SMILES and TELIS observations. This result is considered to have been caused by chlorine activation in the polar vortex (Webster et al., 1993). (Webster et al., 1993; Wegner et al., 2016). Both the SMILES and TELIS profiles agree well in general, but the TELIS profiles are larger than the SMILES profiles above 32 km (8 hPa). The $H^{37}Cl$ line is still rather strong at higher altitudes, and the dominant error source of the TELIS data stems from the non-linearity in the calibration process, which shows that even a small uncertainty may result in significant errors in the retrieval . (Xu et al., 2018).

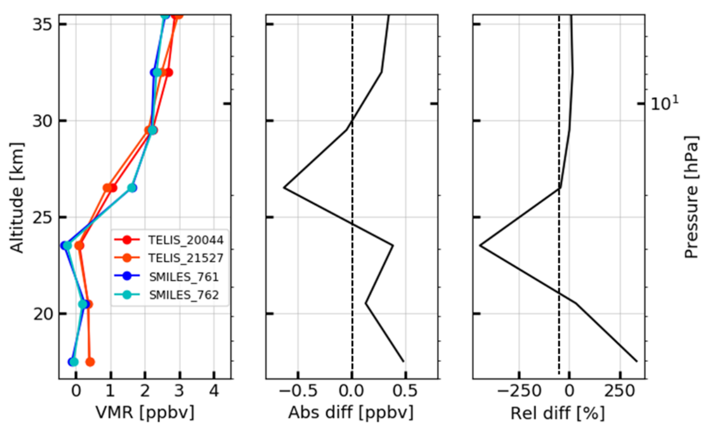

Figure 10. Differences between SMILES and TELIS HCl observations are shown for January 24, 2010. Two of the SMILES observations were taken from closer points, where the latitude, longitude, and SZA were 64.3°N, 30.6°E, and 84.6°SZA and 64.8°N, 38.2°E, 86.2° SZA, respectively. Left: Each profile obtained by SMILES and TELIS instruments. Middle: The absolute difference between the SMILES and TELIS profiles calculated using Eq (3). Right: The relative difference between the SMILES and TELIS profiles calculated using Eq (4).

Table 4. Summary of the HCl comparison study between SMILES and the other instruments

Altitude [km]	Δx_{MLS} [ppbv]	$\Delta x_{\mathrm{ACE-FTS}}$ [ppbv]	$\Delta x_{\mathrm{TELIS}}$ [ppbv]
60	—	0.5	—
50	0.4	0.4	—
40	0.2	0.15	—
30	-0.2	0.05	0.4 [a]
20	-0.1	-0.02	0.5 [a]

5 Theoretical error analysis

We have evaluated the total error in the HCl vertical profiles observed by SMILES, and we discussed the cause of the bias observed in the comparison study, see Sect. 4.

5.1 Estimation of total error

We employed a perturbation method to estimate the total error of the SMILES HCl profile. The details of the perturbation method of the SMILES error analysis have been described in Kasai et al. (2006) and Sato et al. (2014), Kasai et al. (2013), and Sato et al. (2012) for ozone isotopes, ozone, and ClO, respectively. We assumed an averaged HCl profile within the coincidence with MLS in the south mid-latitude region (20°S – 40°S) as a reference. The total error ($\mathbf{E}_{\mathrm{total}}$) for each altitude grid was calculated by

$$\mathbf{E}_{\mathrm{total}}[\mathrm{i}] = \sqrt{\mathbf{E}_{\mathrm{noise}}[\mathrm{i}]^2 + \mathbf{E}_{\mathrm{smooth}}[\mathrm{i}]^2 + \mathbf{E}_{\mathrm{param}}[\mathrm{i}]^2}, \tag{5}$$

where $\mathbf{E}_{\mathrm{noise}}$ is the error due to spectrum noise, $\mathbf{E}_{\mathrm{smooth}}$ is the smoothing error, and $\mathbf{E}_{\mathrm{param}}$ is the model parameter error. $\mathbf{E}_{\mathrm{noise}}$ and $\mathbf{E}_{\mathrm{smooth}}$ were calculated by

$$\mathbf{E}_{\mathrm{noise}}[\mathrm{i}] = \sqrt{\mathbf{S}_{\mathrm{noise}}[\mathrm{i},\mathrm{i}]} \tag{6}$$

$$\mathbf{S}_{\mathrm{noise}} = \mathbf{D}\mathbf{S}_{\epsilon}\mathbf{D}^{\mathbf{T}} \tag{7}$$

and

$$\mathbf{E}_{\mathrm{smooth}}[\mathrm{i}] = \sqrt{\mathbf{S}_{\mathrm{smooth}}[\mathrm{i},\mathrm{i}]} \tag{8}$$

$$\mathbf{S}_{\mathrm{smooth}} = (\mathbf{U}-\mathbf{A})\mathbf{S}_{\mathrm{a}}(\mathbf{U}-\mathbf{A})^{\mathbf{T}} \tag{9}$$

where $\mathbf{D}$ is the contribution function, and $\mathbf{U}$ is the unit matrix.

We calculated the errors due to the uncertainties of the spectroscopic parameters and the instrument functions to calculate $\mathbf{E}_{\mathrm{param}}$. The error $\mathbf{E}_{\mathrm{param}}$ was calculated by

$$\mathbf{E}_{\mathrm{param}} = I(\mathbf{y}_{\mathrm{ref}}, \mathbf{b}_0 + \Delta\mathbf{b}_0) - I(\mathbf{y}_{\mathrm{ref}}, \mathbf{b}_0) \tag{10}$$

where I is the inversion function~~and~~ , $\mathbf{b}_0$ is the vector of model parameters ~~.~~ and $\Delta\mathbf{b}_0$ is the uncertainty on model parameters. The $\mathbf{y}_{\mathrm{ref}}$ is the reference spectrum calculated using the reference profile. The details of the estimation of the total error are described in Sato et al. (2012). The error sources and perturbations for the model parameter used in this study are summarized in Table 5. These parameters and perturbation ~~value~~ values are based on Sato et al. (2014). The uncertainties of the spectroscopic parameters of the O_3 transition 625.37 GHz were included to estimate the error due to the interference from the line shape of O_3 spectrum. The calibration error was not considered in this study because the latest L1b data version 008 was used and Sato et al. (2014) reported that the error due to the spectrum calibration in this L1b data was insignificant.

Table 5. Error sources and their perturbation values for this study.

Error source	Uncertainty
Spectroscopic parameters of the $H^{35}Cl$ transition at 625.92 GHz	
Line intensity	1 %
Air-pressure broadening coefficient (γ_{air})	3 %
Temperature dependence of γ_{air} ($\mathrm{n}_{\mathrm{air}}$)	10 %
Spectroscopic parameters of the O_3 transition at 625.37 GHz	
γ_{air} of the O_3 transition (γ_{air} (O_3))	3 %
$\mathrm{n}_{\mathrm{air}}$ of the O_3 transition ($\mathrm{n}_{\mathrm{air}}$ (O_3))	10 %
Instruments	
Antenna beam pattern (Antenna)	2 %
AOS response function (AOS)	5 %

The perturbation value was based on Sato et al. (2014).

The estimated errors in the SMILES HCl v300 product are presented in Fig. 11. The total model parameter error, labeled as “Param”, was calculated by a root-sum-square (RSS) of all model parameter errors. The three spectroscopic parameters, line intensity, air-pressure broadening coefficient (γ_{air}), and its temperature dependence ($\mathrm{n}_{\mathrm{air}}$) were dominant error sources below 30 km. The γ_{air} was a major error source between ~~the altitude region of~~ 30 and 60 ~~-~~km, which was about ~~0.9~~ 0.09 ppbv ($\sim$ 3%) at 50 km. At altitudes of 60–90 km, the largest error source was the AOS response function, and its peak value reached 0.38 ppbv ($\sim$12%) at 70 km.

5.2 Discussion: Cause of the negative bias of the SMILES HCl vertical profile

In Sect. 5.2, we discuss the cause of the negative bias of the SMILES HCl vertical profile, i.e., approximately 10 % less than those of Aura/MLS and ACE-FTS especially at the altitudes ~~above~~ between 40 – 60 km. Such a large bias could not be explained by the total error estimated by the perturbation method described in Sect. 5.1. We further investigated the cause of this bias by difference of temperature profile used in the retrieval calculation. The temperature profile used for the retrieval procedure of SMILES was ~~smaller~~ lower than those of MLS and ACE-FTS particularly in the upper stratosphere ~~, mesosphere, and lower thermosphere~~and mesosphere. We estimated the difference of the retrieved HCl vertical profile, Δx, due to the difference of

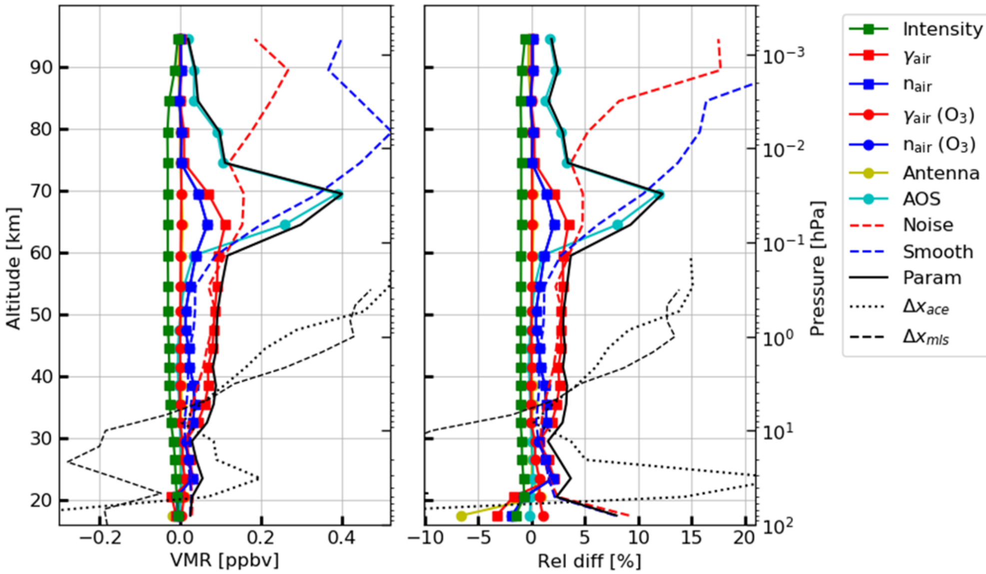

Figure 11. Summary of the errors for a single scan observation. The left and right panels, respectively, show the absolute difference and relative difference, which were estimated using the perturbation method. The green marker indicates an error due to the uncertainty in line intensity ("Intensity"). The red and blue square show errors for the air broadening coefficient ("γ_{air}"), and its temperature dependence ($\mathrm{n_{air}}$) of the $H^{35}Cl$ transition. The red and blue circles show errors for "γ_{air}" and $\mathrm{n_{air}}$ of the O_3 transition ("γ_{air} (O_3)", "$\mathrm{n_{air}}$ (O_3)"). The yellow and cyan symbols show errors for the antenna beam pattern ("Antenna"), and the AOS response function ("AOS"). The red and blue dashed lines indicate $\mathbf{E}_{\mathrm{noise}}$ ("Noise") and $\mathbf{E}_{\mathrm{smooth}}$ ("Smooth"), respectively. The black solid lines ("Param") indicate the total model parameter errors, obtained from the RSS of all the estimated parameter errors. The black dotted and dashed lines indicate the difference between SMILES and MLS, and between SMILES and ACE-FTS.

the temperature profile, ΔT, as follows.

$$\Delta x = \mathrm{D}\mathrm{K}_T \Delta T, \quad \mathrm{D} = \frac{\partial x}{\partial y}, \quad \mathrm{K}_T = \frac{\partial y}{\partial T} \tag{11}$$

The jacobian K_T indicates the sensitivity of the spectral brightness temperature (y) with reference to changes of the temperature (T). Here we synthesized the jacobian with a perturbation of 0.5 K. Figure 12 (A) shows the jacobian as a function of ~~the~~ tangent height. Here the minimum value of the column of the K_T matrix is plotted. ~~Negative value of the jacobian~~ A negative jacobian value means that higher ~~temperature synthesizes lower smaller~~ temperatures induce a lower brightness temperature spectrum, thus ~~, increases~~ increasing the HCl abundance in the retrieval ~~calculation~~ to compensate for ~~the underestimationof the synthesized spectrum due to the temperature higher than the true value.~~

~~The vertical profile of temperature used for the retrieval procedure of~~ this underestimation. The temperature profiles used in the retrievals by SMILES, MLS, and ACE-FTS and their differences, MLS−SMILES, ACE-FTS−SMILES, are shown in

~~the~~ panels (B) and (C) in Fig. 12, respectively. The vertical profile of temperature of SMILES is approximately 5-10 K ~~smaller~~ lower than those of both MLS and ACE-FTS ~~at the altitude 50–70km. The panel (D)~~ for altitudes between 50 and 60 km. The a priori temperature profile used in the SMILES retrieval procedure is based on the GEOS-5 profile in the stratosphere and MLS retrieved profile above the mesosphere.The altitude limit of the MLS temperature profile is 0.001 hPa, with a vertical resolution of 6 - 14 km and a precision of 1.2 – 3.6 K per profile. MLS used GEOS-5 up to 1 hPa as with SMILES. For pressures smaller than 1 hPa, the COSPAR International Reference Atmosphere (CIRA-86) is used as a priori temperature information (with a loose constraint) in the MLS retrieval procedure (Schwartz et al., 2008). The altitude range and vertical resolution of the CIRA-86 profile are ground to 120 km and 2 km, respectively (Fleming et al., 1990). The temperature value retrieved by MLS is 10 K lower than the a priori profile on average in some areas for pressure values smaller than 1 hPa (Schwartz et al., 2008), based on earlier version validation studies (Schwartz et al., 2008). The ACE-FTS retrieval procedure uses the retrieved temperature profile as the a priori between 18 – 125 km. The vertical resolution of ACE-FTS retrieved temperature is 3 – 4 km. The temperature values retrieved by ACE-FTS are less than 10 K larger than the MLS derived temperatures. (Schwartz et al., 2008). These types of difference are also seen in the comparison results performed here.

Panel (D) in Figure 12 shows the Δx due to the difference of the temperature profile. ~~The difference of the temperature profile caused increase HCl abundance by~~ This temperature difference caused an increase in SMILES HCl of 0.12 and 0.20 ~~ppbv at about 55 km for~~ ppbv at 50-60 km for the MLS and ACE-FTS comparisons, respectively.

In the comparison study, see Sect. 4, we confirmed the negative bias in the SMILES HCl vertical profile of 0.4±0.38 and 0.5±0.28 ppbv ~~at 55~~between 40 – 60 km for MLS and ACE-FTS, respectively. We estimated the total error by the perturbation method and investigated the temperature profile used in the retrieval calculation, in order to investigate the cause of this negative bias. The largest error sources were the uncertainties in γ_{air} of the $H^{35}Cl$ transition and ~~temperature profile used in retrieval calculation~~the temperature profiles used in the retrievals. We assumed ~~the~~ a 3 % uncertainty in γ_{air} ~~and it might cause approximately~~ which could lead to a -0.1 ppbv ~~negative bias at 55~~bias in the 40 – 60 km ~~. The difference in~~ region. In addition to the error due to the γ_{air} coefficients, the effect of temperature differences should be taken into account at altitudes above 50 km. The gradual increase in bias is caused by the difference in altitude at which these two errors become more pronounced. The difference in temperature profiles used in the retrieval between SMILES and ACE-FTS caused ~~approximately~~ a negative bias of about 0.2 ~~ppbv negative bias . Totally~~ppbv at 50-60 km.. In summary, 0.3 in 0.5 ppbv (60 %) negative bias between SMILES and ACE-FTS ~~was~~ can be explained by the uncertainty in γ_{air} and ~~temperature profile~~ the temperature profiles used in the ~~retrieval~~SMILES retrievals. The effect of γ_{air} error on the negative bias between SMILES and MLS is less than that of SMILES and ACE-FTS, since the SMILES and MLS observed the same $H^{35}Cl$ transition lines and the values of the γ_{air} are consistent within approximately 1 % (3.39 MHz/Torr for SMILES and 3.42 MHz/Torr for MLS (Drouin, 2004)). ~~The~~ A 1 % difference ~~of~~ in γ_{air} might cause the HCl abundance ~~increase~~ to increase by about 0.03 ppbv ~~at 55 km~~ according to our error analysis with a perturbation method. ~~The~~ About 40 % (0.15 ~~in~~ out of 0.4 ppbv~~(approximately 40 %)~~) of the negative bias between SMILES and MLS ~~was explained. Conclusively~~can be explained. Therefore, our theoretical error analysis ~~showed~~ shows that the SMILES HCl ~~a had~~ has a negative bias of ~~0.2–0.25~~at most 0.25 ppbv ~~at 55~~between 40 and 60 km~~which were~~

consistent with the difference from the MLS and ; remaining difference between SMILES and MLS or ACE-FTS within the 1 σ standard deviation . can be explained by the standard deviation in the comparison result.

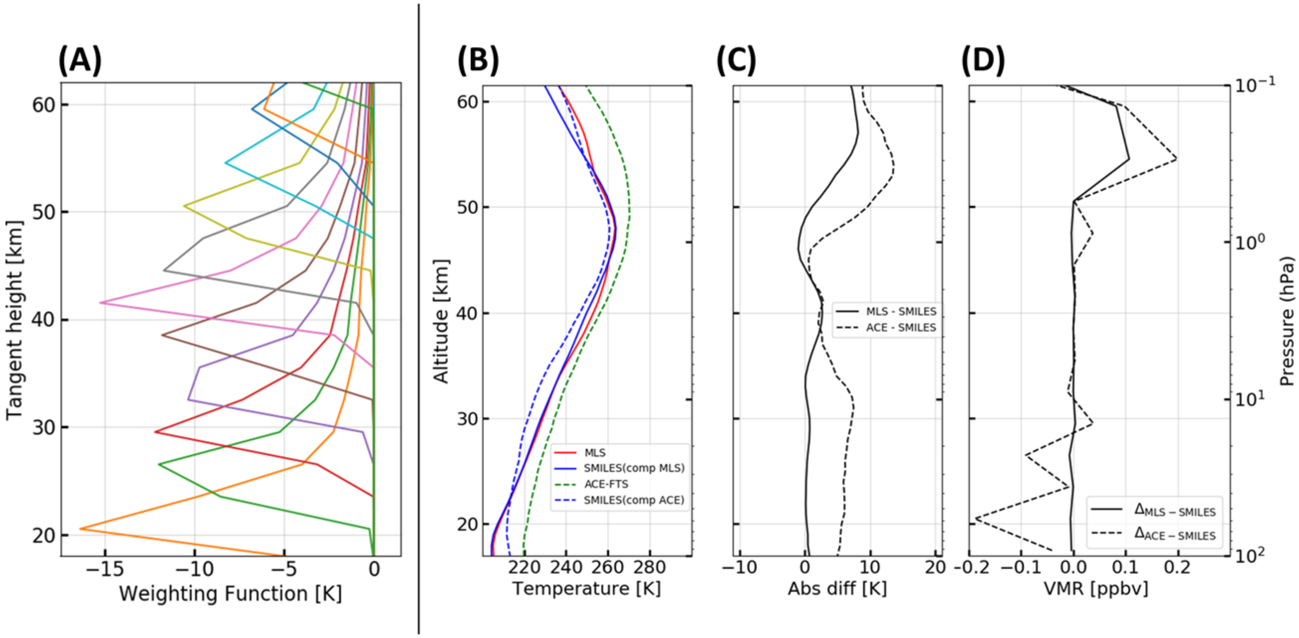

Figure 12. (A) The dashed lines represent the jacobian for the temperature calculated using the mean profile from the SMILES winter mid-latitude region (20°S–40°S). The units of the jacobian (x-axis) are Kelvin. (B) Temperature profile of each instruments. The blue and red solid lines indicate the mean SMILES and MLS temperature profiles within the coincidence criteria used for the HCl comparisons. The blue and red dash green dashed lines indicate the SMILES and ACE-FTS temperature profiles for the same coincidence criteria. (C) The absolute difference of temperature profiles between SMILES and other instruments calculated using Eq (3 (D) The difference in HCl profiles calculated using Eq (11).

6 Conclusion

In this study, the HCl vertical profile in a wide range from the upper troposphere to the lower thermosphere was reported for the first time using the SMILES NICT Level 2 data product v300. The HCl vertical profile showed distribution shows an increase with the altitude increased altitude with a maximum below the stratopause (∼45 km), approximately constant values between the stratopause and the upper mesosphere (∼80 km), and decreased with the altitude increased a decrease with altitude from the mesopause to the lower thermosphere (∼100 km). In the lower and middle stratosphere, HCl is generated by the reaction of Cl with CH_4 and HO_2 and transported by circulation (e.g. Brewer-Dobson circulation). The HCl abundance is balanced by production ($Cl + HO2 \rightarrow HCl + O_2$) and loss($HCl + OH \rightarrow HCl + H_2O$, $HCl + h\nu \rightarrow H + Cl$) in the upper stratosphere and the mesosphere. Above the mesopause, the photodissociation becomes the dominant reaction and the HCl abundance decreases. This behavior was reproduced by the SD-WACCM model, and the SMILES HCl vertical profile agreed well with the SD-WACCM model within ±0.1 ppbv ~~at altitude~~ for altitudes between 30 ~~to~~ and 70 km.

The data quality of the SMILES HCl vertical profile was quantified by ~~the comparison study with other instrument measurementsand the~~ comparisons versus other measurements, and supported by a theoretical error analysis. We compared the SMILES HCl vertical profiles with well validated data of two satellite instruments, Aura/MLS and ACE-FTS, as well as a balloon-borne instrument, TELIS, at their temporal-spatial ~~coincidence.~~ coincidences. The SMILES HCl profiles at 20–40 km showed good agreements, within less than 0.25±0.3 (1σ) and 0.20±0.2 (1σ) ppbv~~with the~~ , versus MLS and ACE-FTS, respectively. The comparison with the TELIS in the polar winter region at 20–34 km showed similar behavior ~~within the difference of~~ with differences within 0.3 ppbv, which is the same order of magnitude as the systematic error of the TELIS data. ~~The~~ A negative bias (<0.5 ppbv) of the SMILES HCl profiles from 40 to 60 km altitudes was observed ~~compared to the~~ in comparisons versus MLS and ACE-FTS ~~.~~ HCl profiles.

We estimated the total error for SMILES HCl based on the perturbation method and ~~error due to the uncertainty in the atmospheric temperature profile~~ considering the uncertainties in atmospheric temperature profiles used in the ~~retrieval calculation~~retrievals. The dominant contributions to the systematic errors were from the air broadening parameter (0.09 ppbv) and the AOS response function (0.38 ppbv) at ~~30–60~~30 - -60 km and 60–100 km altitudes, respectively. The uncertainty in the temperature profile used in the retrieval calculation caused ~~the~~ a negative bias of 0.12 ~~and~~ to 0.20 ppbv ~~at about 55~~ between 50 – 60 km, which was 30% and 40% of the HCl abundance difference between SMILES and MLS, and SMILES and ACE-FTS, respectively. The uncertainties of the air broadening parameter and the temperature profile ~~were capable of totally contributing up to~~ are capable of contributing a total of 40 – 50 % of the ~~negative bias in 50–60~~SMILES HCl negative biases at 50 – 60 km~~altitudes. Totally.~~ In summary, our theoretical error analysis showed that the ~~SMILES HCl had~~ HCl profiles had a negative bias of 0.20 ~~–0.25–~~ 0.25 ppbv at ~~55~~50 – 60 km~~which were~~ , which is consistent with the ~~difference from the~~ observed differences versus MLS and ACE-FTS ~~within the~~ profiles within 1 ~~σ~~ standard deviation. The spectroscopic parameters of the HCl transitions and the temperature profile above ~~stratosphere~~ the stratopause are key parameters for ~~further improvement of retrieval algorithm.~~ potential improvements in the SMILES retrieval algorithms.

The observation of HCl abundances in the upper atmosphere is important to investigate the long-term ~~quantitative estimations of the~~ total budget of anthropogenic chlorine in the Earth's atmosphere. Further observations and model studies ~~regarding HCl abundance including upper atmosphere~~ are needed to ~~understand the source and sink~~better understand the sources and sinks, transport processes, and chemical reactions related to HCl.

Data availability. The SMILES data is available at http://smiles.nict.go.jp/pub/data/index.html.

The MLS data is available at https://disc.gsfc.nasa.gov/datasets?page=1&keywords=AURA%20MLS or see https://mls.jpl.nasa.gov/data/

The ACE-FTS data is available at http://www.ace.uwaterloo.ca/instruments_acefts.php

The details on the TELIS 1.8 THz channel and its L1 data processing are shown in https://elib.dlr.de/66749/ "Development and characterization of the balloon borne instrument TELIS (TErahertz and submillimeter LImb Sounder): 1.8 THz receiver Suttiwong, Nopporn (2010)" and/or https://elib.dlr.de/97249/ “Inversion for Limb Infrared Atmospheric Sounding Xu, Jian (2015)"

The WACCM data is available at https://www2.acom.ucar.edu/gcm/waccm

Author contributions. SN designed the study and performed the analysis. YK designed the study and provided the SMILES data. TOS provided the code for the error analysis and contributed to data analysis and interpretation. TY, TF, and KK contributed to the data analysis and reviewed the manuscript. LF, NJL, KAW, JX, and FS provided MLS, ACE-FTS, and TELIS information and knowledge of HCl in the atmosphere and helped review the manuscript. YJO and VL performed and provided the WACCM simulations, and helped review the manuscript.. NK and TM supervised writing of and reviewed the manuscript.

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