Dear Anonymous Referee #2,

Thank you for your hard work and constructive comments on “Tomographic retrieval algorithm of OH concentration profiles using Double Spatial Heterodyne Spectrometers”. The helpful comments have substantially improved our paper. We agree with points raised and modify them in the revised version of the manuscript.

On behalf of the authors

Kind regards,

Yuan An

A Point-by-Point response to the reviewer’s comments

Reviewer: Abstract: The abstract only describes the method used in the paper. It should give more related results.

Reply: We agree with your points. Some related results are given in the revised version of the manuscript.

Reviewer: Page 2, Line 48: The introduction part of the OH radical method should be more summarized, including principle and results.

Reply: We agree with your points. The principle and results of OH radical methods have been summarized. We hope the entire modification is clearly described in the revised version of the new manuscript.

Reviewer: Page 7, line 210: The T2-1 and T2-2 position should be defined more clearly.

Reply: We apologize for the unclear expression about the T2-1 and T2-2 position. There are two T2 positions which are meeting the requirements of three-dimensional limb mode with the T1 position. We defined the satellite position on the same side to the time of T1 is defined as the position at the time of T2-1, and on the opposite side to the time of T1 is defined as the position at the time of T2-2.

Reviewer: Page 7, line 212: It should be the distance of satellite position fromT1 to T2-1 moment, and T1 to T2-2 moment.

Reply: We apologize for the unclear expression about these parts in the question. We change the sentence in question to “The distance of the satellite position at the time of T2-2 from the time of T1 is farther than the satellite position at the time of T2-1…” for more accurate expression.

Reviewer: Page 7, line 214: Consider replacing “cannot reflect the actual OH distributions” with “make bigger difference”.

Reply: Changed as suggested. Thank you very much! We change “…cannot reflect the actual OH distributions…” to “…make bigger difference…”.

Reviewer: Page 9, Line 237: Consider making the flow chart more clear and nice looking? Same with other flow charts in the essay.
Reply: We changed the flow chart as your suggestion. We hope the flow chart is clearer and nicer in the revised version of the manuscript.
Reviewer: Page 11, Line 297: The “Grating width” in the table 1 should have some unit.
Reply: Apologize for this mistake. The unit “line/mm” has added in the table.
Reviewer: Page 11, Line 304: Figure 8 should be replaced by a higher definition picture.
Reply: We apologize for this question. Figure 8 has replaced by a new figure which has the resolution of 600 dpi to be clearer.

Reviewer: Page 17, Line 467: Gave a lots description of the LSUV retrieval algorithm, which was not used in the essay, and did not explain the reason why it was incredible in lower atmosphere. The tomographic retrieval algorithm is an improved version of LSUV or two independent algorithms? Could you analyze the advantages of the tomographic retrieval algorithm by combining the inversion results of LSUV?
Reply: The LSUV retrieval algorithm have been used for many years. The tomographic retrieval algorithm and the LSUV are two independent algorithms. We use LSUV retrieval algorithm to highlight the advantages of the tomographic retrieval algorithm. We hope the entire modification is clearly described in the revised version of the manuscript.

Reviewer: Page 18, Line 487: when using the lookup table, the accuracy of the tomographic observed database will affect the OH results? Have you considered proper amount of the data? And the parameter setting of the influencing factors? The accuracy of this database is not described in this paper. And is there any other method to prove the accuracy of the database?
Reply: We analyzed the errors of forward model in Sect. 3 and have considered the parameters which affect the OH concentrations. So, we consider the tomographic observed database is accurate.

Reviewer: Page 20, Line 550: Consider giving more detailed description of the calculation of relative errors.
Reply: Changed as suggested. The descriptions of the calculation of relative errors have been added in the revised manuscript.

Reviewer: Page 23 line 627: the method of cubic spline interpolation is used to obtain the OH concentration that cannot use the lookup table. In essence, the interpolation result is not the actual measurement, and the error estimation of interpolation results in this paper may be inaccurate!
Reply: Thank you for your comment. Although an effective and reasonable lookup

table inversion algorithm have been constructed, it cannot be obtained the OH concentrations in all cases because 1) the size of lookup table is limited and 2) we cannot consider all situations when the sensor will work in the orbit. The interpolation methods and the threshold judgment methods are always used to solve such problems. A scientific and precise threshold in the threshold judgment method can be obtained according to a lot of OH concentrations data and experiments. However, the OH concentrations data are little now. The interpolation methods are chosen. The cubic spline interpolation method not only has higher stability but also can ensure the continuity and smoothness of the interpolation function under the premise of the ensuring the convergence compared with other interpolation methods like linear interpolation method and some others. The error estimation of the interpolation results is as accurate as possible. We will also improve the accuracy of the OH concentrations when the DSHS work officially in the future.

Reviewer: Page 25, Line 673,677,679,680: How were these errors calculated?
Reply: We apologize for being unclear about this part. The OH concentrations which are obtained by the LSUV algorithm have been given by Yibo Gao in his graduation thesis and some other papers by researchers. These errors in the question are calculated by the Eq (17) and the error transfer formula. The entire modification is given in the revised version. We hope it is clearly described.

Reviewer: Page 27, line 745: When will the DSHS used in the satellite and obtain the actual data for the OH measurement? The only way to verify the feasibility and correctness of the method is the deeply analysis of the obtained data.
Reply: We agree to this point deeply. The actual observed data is the key point. The DSHS and research results have been shown and communicated with the related departments. It has been included in the relevant satellite projects for 2020 to 2030.

Reviewer: The writing of the whole essay needs to be improved.
Reply: Thank you very much for your pertinent suggestion. Our English language do need to be improved. We have found a lot of phrases and grammatical errors through re-reading of the full manuscript carefully. All of the errors have been revised. I hope it is more accurate now on the English expression in the revised paper.

Dear Anonymous Referee #1,
Thank you for your careful work and constructive comments on “Tomographic retrieval algorithm of OH concentration profiles using Double Spatial Heterodyne Spectrometers”. The comments have substantially improved our paper a lot. We agree with points raised and modify them in the revised version of the manuscript.

On behalf of the authors
Kind regards,

Yuan An

A Point-by-Point response to the reviewer’s comments

General concerns

Reviewer: There are many points in this paper where the authors assert a value (e.g. for uncertainty) or conclusion (e.g. the tomographic retrieval is faster than the iterative retrievals) without providing a citation, clear chain of logic, or data to support those points. I list as many as I could find in the remainder of this document. These points need to be addressed so that readers can make their own assessment of these claims.
Reply: Thank you for your suggestions. We have considered your reminders in your document deeply and solved the problems. We hope the entire modification is clearly described in the revised version of the new manuscript.

Reviewer: What are the plans to test this algorithm in practice once the DSHS is operational. Balloon flights? Aircraft flights? Including this as a future direction would make clear what the next step is.
Reply: We agree to this point. The actual observed data is the key point for the algorithm. Some aircraft flights experiments have been done and the data is being analyzed. The DSHS and research results have been communicated with the related departments. It has been included in the relevant satellite projects for 2020 to 2030. We have added this part to what the next step is.

Specific comments

Sect. 2.2

Reviewer: Section 2.2 in this paper is intended to outline the observing strategy that this proposed instrument will follow in order to obtain spectroscopic information needed to infer the 3D OH fields. This is a key point that must be communicated clearly for the remote sensing community to evaluate whether this instrument is feasible. However, I found this section extremely difficult to read.

The main problem is it seems the authors are exploring different observing strategies and rejecting ones that will not accomplish their goals. It is not clear that this is what they are doing, and it is not clear how they determined whether a particular strategy meet their requirements, or even what those requirements are. I recommend this section be completely rewritten to improve its clarity, addressing the following points in order:

- First, clearly explain the criteria that a particular observing strategy must meet to be considered successful
- Second, explain how the strategies were evaluated to determine if they met these criteria
- Third, describe the final strategy that meets the necessary criteria
- Only then describe the alternate strategies considered and why they were rejected, making it absolutely clear that these are rejected strategies.

As an example of what makes this confusing, I point to lines 184–193:
It is not clear from this section that T2-1 and T2-2 are hypothetical positions, and the bolded text makes it sound like these are actual times when the satellite will make measurements.
I understand that, to some extent, it is necessary to explore these hypothetical options for different measurement times to explain the criteria **(e.g. line 194, "That means, the smaller distance between the P1 and R1 (or R2) is, the higher spatial resolution will be.")**. However, as written, it is unclear when hypothetical options are being considered and when the authors are describing the final option that is chosen.

Reply: We agree with your points deeply. We hope the entire modification is clearly described in the revised version of the manuscript especially the work mode of satellite.

Some more specific points of confusion:
Reviewer: p. 5, l. 141: "**The hierarchical detection of a series of observed radiances in the same target area comes true through the movement of the satellite platform**." Does "movement" refer to the satellite flying along its orbital path, or actively rotating to point the spectrometers at the target area?

Reply: We apologize for the unclear expression. The "movement" means the satellite fly along its orbital path.

Reviewer: p. 5, l. 139–140 vs. Fig. 3: The description of the two sensors as scanning along and across the satellite's orbit implies, to me at least, that the main axes of the telescopes are pointing parallel to the satellite's direction of flight, but Fig. 3 looks like the sensors are pointing to the right of the direction of flight.
– This is exacerbated by Table 3, which seems to indicate that there are different T2 points for different altitudes in the profile. Addressing the first point about describing how the satellite moves will help with this.
Reply: We agree with your points. The description about how the satellite moves will help to solve your question. The main axes of the telescopes are pointing parallel to the satellite's direction of flight. Fig. 3 shows that DSHS monitors the OH in the target area using the data of SHS1 at the time of T1 and SHS2 at the time of T2 together. However, the telescopes points to the forward of the direction of flight.

Reviewer: p. 6, l. 154–170: Since it's not clear how the satellite will move, it's very hard to understand what's going on here.
Reply: We have changed these parts in the revised version of paper. We hope the entire modification is clearly described in the revised version of the manuscript.

Reviewer: How long will the spectrometers record at T1 and T2? Is it very briefly, or is the spectrometer recording as it sweeps across the target zone?
Reply: The DSHS has the function of hierarchical imaging in the spatial dimensional. It can obtain the data at different heights at the same time without scanning the target area for obtaining the data. So, The DSHS record at T1 and T2 is very briefly.

Reviewer: Perhaps more generally, what is happening between T1 and T2? Is the satellite reorienting to get the appropriate pointing vector for the measurement at T2? Are the spectrometers on during this time?
Reply: The satellite needs to reorient for getting the appropriate vector for the measurement at the T2 moment. The spectrometers are on during this time.

Reviewer: Do I understand right that the point of Fig. 3b is that one cannot use data collected at only one time to retrieve the target area? If so, why not? That is not explained.
Reply: Your understanding is correct. If only we use the data at only one time. There are no intersection regions between the field of view slices of SHS1 and SHS2s' at the time of T1. The tomographic data cannot be obtained in this situation.

Reviewer: It's not obvious to me why the strategy shown in Fig. 3c of using SHS1 at T1 and SHS2 at T2 is better than using both SHS1 and SHS2 at both T1 and T2. Surely having SHS1-T1 + SHS2-T1 + SHS1-T2 + SHS2-T2 would provide a better constraint on the OH fields?

Reply: On the one hand, a lot of data can provide a better constraint but will cause redundancy. The inversion efficiency will reduce. On the other hand, SHS1-T1 + SHS2-T1 + SHS1-T2 + SHS2-T2 will have a lot of noise, it will make the signal-to-ratio low.

Reviewer: In Fig. 4, why are there red boxes (presumably representing the satellite) on both ends of the T2-x lines of sight but not the T1 line of sight?
Reply: We apologize for the drawing error. The red boxes at the one end of the T2-x lines of sight.

Retrieval algorithm (sect. 4.2)

Reviewer: Given that the tomographic retrieval described in this section is the major advance in this paper, more detail on how the retrieval works is necessary to fully evaluate it. Specifically, in Fig. 16: what precisely is meant by "**SHS1/SHS2 observed data at time of T1/T2**"—radiance at a single wavelength, a few selected wavelengths, or all wavelengths measured by the spectrometer?
Presumably it is not a single wavelength because that would make it extremely difficult to disentangle the effects of the background radiance from that of OH fluorescence. But if it is at all measured wavelengths, that seems like it would result in an extremely high dimensionality to deal with in the interpolation.
Then, in the first paragraph of sect. 5.1, the authors state that there are four databases, one per season. How are transitions between seasons handled? If it's an abrupt switch (say May 31st uses the spring database and June 1st uses the summer database), won't that introduce discontinuities to the retrieved OH time series?
For Fig. 17, how does the inversion result compare with the true profile that was used to simulate the radiances? Similarly, on line 570, the sentence "**The OH concentrations obtained by the LSUV retrieval algorithm are credible in the upper atmosphere,**" is extremely vague. What level of agreement do you consider "credible"? How large a sample size did you use to determine this?
Reply: Thank you for your comments. 1) The SHS1/SHS2 observed data at time of T1/T2 mean the radiance at all wavelengths measured by the DSHS. 2) We count a large number of samples in each grid in the day. It shows that the number of samples is not uniform in the grid, the number of samples in most grids is too small, even there is no profile data in some grids. The less profile data will order to a larger random error. Averaging method cannot reach the goal of smoothing profile and reducing errors. Therefore, the month is determined as the time resolution. We average the profile data in the grid to obtain the monthly average concentration profile data. So, it will not introduce discontinuities to the retrieved OH time series when an abrupt switch happens. 3) The OH concentrations obtained by the tomographic retrieval algorithm are the results of inversion process. The OH concentrations used to simulate the radiance are the parameter of forward process. These OH concentrations have different meaning and are not comparable. The conclusion of results obtained by the LUSV has been proven by a lot of manuscripts. We have added some citations in the revised version of the paper.

Other major concerns

Reviewer: Line 580: I don't quite understand "**the undisturbed parts are taken as the actual parameters, and the results based on these parts are the true OH fluorescence emission radiance. Then, some certain amounts as the disturbances are applied to the three parts above. The OH fluorescence emission radiance based on these parts is the incorrect results." What are the "undisturbed parts**"? Do you mean that you take the values of each of the atmospheric model, Doppler effect, and instrument calibration from the actual retrieval, and add some error to one at a time? That could be much clearer. How do you determine how much error to add? Do you do this for every OH concentration in the test set or a subset?
Reply: Apologize for these unclear parts in your questions above. Your understanding is exactly what I want to express. We take the values of the atmospheric model, Doppler effect and instrument calibration from the actual retrieval process. Some explanations are given in the following parts to solve the problems about how much errors we add. We do this for every OH concentration in the test subset because the conclusion is representative.
- line 597: "**...the amount of ozone profile for the disturbances is set as ±30%.**" Why 30%?
Reply: We referred to the relevant parameters of the instrument likes SHIMMER which is similar to the DSHS in this part due to the DSHS has not been worked officially. Apart from this, we did a lot of experiments according to the conclusions of the research of SHIMMER and found the 30% is the best threshold to disturb the ozone profile for analyzing the influence of the atmospheric model.

Reviewer: Line 652: "**We assumed the instrument calibration errors are ±5%...**" Why 5%? Please provide either a citation or the line of reasoning used to arrive at this value.
Reply: We apologize for this unclear description. The instrument calibration error is given by the manufacturing and design department of sensor. Some citations have been added in the revised version of the paper.

Reviewer: Line 657: "**It is about 0.32% for the six-dimensional cubic spline interpolation of OH concentrations, date parameters, longitude, latitude, solar zenith angle and azimuth angle after research and related experiment.**" What experiment did you do to determine the 0.32% value?
Reply: Apologize for the missing the experiments. We give a certain disturbance to different parameters to calculate the effect of the interpolation algorithm on the OH concentrations.

Reviewer: Line 741: "**The speed of traditional iterative retrieval algorithms is slow and will cost a lot of time.**" Can you quantify this? How much faster is the tomographic algorithm for, say, a single target area or a day's worth of data?

Reply: The traditional iterative retrieval algorithm will take several or even tens of hours to obtain OH concentrations due to the complex iterative process. The tomographic algorithm just needs minutes for obtaining the OH concentration due to the usage of the lookup table method. It is difficult to quantify how much fast is the tomographic algorithm absolutely. The results obtained by the traditional iterative retrieval algorithm in the lower atmosphere are unscientific but the OH concentrations obtained by tomographic algorithm in the same area are accurate. The tomographic algorithm can improve the accuracy of data while acquiring OH concentrations quickly.

Technical corrections

Points for clarification

Reviewer: Lines 252–255: What is meant by "**The SCIATRAN absorption spectrum database is completed based on the HITRAN 2014 database....**" Does this mean that the forward model kept the HITRAN 2012 database, but added the UV absorptions from HITRAN 2014? Or does the forward model use HITRAN 2014 instead of HITRAN 2012? Or something else?
Reply: We apologize for these imperfect descriptions. The SCIATRAN uses HITRAN 2012 by default. However, the HITRAN 2012 database does not include the OH absorption profiles in the ultraviolet band. We use the OH absorption profiles in the HITRAN 2014 to solve this problem. We upgraded the SCIATRAN in this part by using the HITRAN 2014.

Reviewer: In Eq. (10) where does N(j) come from, if Ng(j) is either from the OH database or the iterative update?
Reply: We apologize for these unclear descriptions. The N(j) is the result of iteration. It can get though the y_j and $N_g(j)$ when the converges meet the accurate requirement or the number of iterations exceeds the iteration number threshold.

Reviewer: Is Section 5.2.4 where we switch from discussion errors in the OH radiance to errors in the actual OH concentration? Please make it clearer that we are transitioning if so; the section title could be "**Analysis of total errors in the OH concentrations**" and add a first sentence like "**We now move from considering errors in the OH radiances to errors in the retrieved concentrations.**"
Reply: Changed as suggested. Thank you very much! We have changed the title of Section 5.2.4 and added a first sentence "The retrieved results must be considered after the discussion of the OH radiance".

Reviewer: Line 704: “**It is a great improvement to compare with the results of the LSUV retrieval algorithm which has a characteristic of iteration.**” What are the LSUV errors, or where in the paper are they listed? I found some of them later in the paragraph (line 715 and on), but why are they not in Table 7 for immediate comparison with the tomographic errors?
Reply: The OH concentrations obtained by the LSUV algorithm in the lower atmosphere are unsuitable for scientific research. These limit the applicability of LSUV algorithm. This manuscript focuses on the new algorithm, we use the table to emphasize the errors of new algorithm and consider it is more reasonable to use the text for comparing two algorithms.

Reviewer: Line 709: “**The corresponding accuracy ranges of MLS OH concentrations products are also given, and its useful height ranges are from 23 to 81 km.**” Where are these values given?
Reply: These values are given by the MLS data quality and description document which seems like the instructions of the data. It is given by the Jet Propulsion Laboratory to ensure the scientificity and accuracy.

Typographic comments

Reviewer: line 142: “**hierarchical detection**,” not sure what is meant by this.
Reply: We apologize for the unclear description about “**hierarchical detection**”. The DSHS will detect the altitudes from 15 to 85 km by the three-dimensional limb mode. This observation mode can obtain the OH data at the altitude of 15, 17, 19, 21,23 and the altitudes which follow this role. The atmosphere is divided into many layers. The hierarchical detection means the sensor can obtained the data in these layers like the multi-angle method.

Reviewer: line 228: “**The observed data received by the DSHS with an ultra-high spectrum resolution makes up by two parts in this research...**” does this mean that the data recorded by the spectrometers will be made up of two parts?
Reply: Correct. The data recorded by the DSHS is made up by two parts due to the ultra-high spectral resolution of spatial heterodyne spectroscopy. The atmospheric background radiance and the OH fluorescence emission radiance can be separated accurately for the following research.

Reviewer: line 247: Citation for the Lifbase software needed
Reply: Thank you for your suggestion about missing the citation for the Lifbase. The citation has been added in the revised version of the paper.

Reviewer: line 248: Citation for the Bremen atmospheric model needed
Reply: Thank you for your suggestion about missing the citation for the Bremen atmospheric model. The citation has been added in the revised version of the paper.

Reviewer: lines 236–275: there are several different ideas in one paragraph (the solar spectrum, the OH spectrum, the OH concentrations database). Please give each its own paragraph.
Reply: We agree with your points. These ideas have been separated clearly. We hope the entire modification is described in the revised version of the paper.

Reviewer: Eq. (5): units?
Reply: Apologize for missing the units. The Eq. (5) is the wavelength calibration equation. The δ_i is the symbol of intensity. So, the unit is phot^{-1}s^{-1}cm^{-2}nm^{-1}sr^{-1}.

Reviewer: line 429: "**However, it cannot be done to obtain OH concentrations in a converse way directly**"—should "converse" be "inverse"?
Reply: Apologize for this vocabulary mistake. We have changed the "converse" to "inverse" in this sentence.
Reviewer: Around line 575: it seems like there is a shift from comparing the iteration and lookup table methods to the formal error analysis of the lookup table method. This should be more clearly separated.
Reply: We agree with your points. These two parts in your question have been separated clearly in the revised version of paper. We hope the entire modification is clearly described.

Reviewer: line 646: "**The distributions of errors have the same tendency, but the relative errors of tomographic retrieval algorithm are smaller than LSUV retrieval algorithm.**" Does this mean that the distribution of errors due to the Doppler shift is similar between the LSUV and tomographic algorithm? As written, it sounds like the distribution of errors are similar to the relative errors.
Reply: Correct. The distribution of errors is similar between the LSUV and tomographic algorithm and is similar to the relative errors.

Reviewer: line 647: "**These indicate that tomographic retrieval algorithm can obtain more accurate OH concentrations although the Doppler effect cannot avoid**" The tomographic algorithm is more accurate than what? The LSUV algorithm? The Doppler effect cannot avoid what? Does this mean the Doppler effect cannot be avoided by either algorithm?
Reply: We apologize for being unclear about this. The OH concentrations which are obtained by the tomographic algorithm are more accurate than the results are obtained by the LSUV algorithm. The influence caused by the Doppler effect cannot avoid when the spectral resolution is extremely high when the sensors use the spatial heterodyne spectroscopy technology. So the Doppler effect can be avoided by some situations.

Reviewer: line 701: "**As the Table 7 indicates that the total errors of OH concentrations are increasing as the heights rise**." Since the errors only increase with altitude at first, perhaps instead: "**As Table 7 indicates, the total errors of OH concentrations initially increase with height...**"
Reply: Changed as suggested. Thank you very much.

Dear Hailiang Shi,

Thank you for your comments on the ‘Tomographic retrieval algorithm of OH concentration profiles using Double Spatial Heterodyne Spectrometers’. Your comments have substantially improved our paper.

The increasing Rayleigh scattering as the atmospheric background radiance is the major issue for the lower stratosphere. It is mainly subjected by the function of Rayleigh scattering, ozone absorption and OH self-absorption in this research. We add a table to show the intensity of simulated observation radiance and atmospheric background radiance at some tangent heights which are calculated by the modified SCIATRAN radiative transfer model in the Sect. 3. The errors of results are also given. The OH fluorescence emission radiance can be calculated by subtracting the atmospheric background radiance from the observation radiance. The errors of the inversion results which are given in the Sect. 5.2.4 show the data allow to inverse the accurate OH profile from 15 to 85 km.

On behalf of the authors
Kind regards,

Yuan An

A list of all relevant changes made in the manuscript

1. Page 1, Line 17: Change "…its…" to "…OH…"
2. Page 1, Line 22: Add "The error of the results obtained by the forward model is ±44.30% in the lower atmosphere such as 21 km height and decreases gradually until the limit of observation altitude ."
3. Page 1, Line 28: Add "The inversion results are given and the errors of them increase as the altitudes rise until about 41 km height then start to decrease. The errors of the inversion results reach the maximum about ±25.03% in the 41 km height and decrease to ±8.09% in the limited observation height. They are also small in the lower atmosphere which are ±12.96% in the 21 km height. In summary,"
4. Page 2, Line 46: Change "The Fluorescence Assay by Gas Expansion (FAGE), the Differential Optical Absorption Spectroscopy (DOAS) and the Chemical Ionization Mass Spectrometry (CIMS) are commonly used to measure the OH concentrations in the actual limited environments (Hard et al., 1984;Mauldin et al., 1998;Perner et al., 1976). In addition, the ^{14}CO oxidation method, the Scrubbing using the salicylic acid Technique and the Spin Trapping method are used to obtain the OH concentrations in the laboratory for some theoretical researches (Felton et al., 1990;Salmon et al., 2004;Watanabe et al., 1982). Apart from the six physical and chemical methods mentioned above, many researchers used the high-precision spectrum data from the ground-based instruments especially the Fourier Transform Ultraviolet Spectrometer (FTUVS) in the Table Mountain (Cageao et al., 2001;Cheung et al., 2008;Mills et al., 2002)." to "The Fluorescence Assay by Gas Expansion (FAGE) uses the 308 nm excitation mechanism to excite the OH radical continuously for generating the fluorescent signal to establish the relationship between the OH concentrations (Hard et al., 1984). The Differential Optical Absorption Spectroscopy (DOAS) obtain the OH concentrations because the absorption of OH follows the Lambert-Beer absorption law (Perner et al., 1976).The Chemical Ionization Mass Spectrometry (CIMS) collects ions of OH instead of the photons based on OH oxidation to obtain the OH concentrations (Mauldin et al., 1998) .These methods are commonly used to measure the OH concentrations in the actual limited environments and laboratories. In addition, the ^{14}CO oxidation method uses the $^{14}CO_2$ concentration, enrichment coefficient, reaction rate constant and reaction time to get the OH radical concentrations based on the OH oxidation (Felton et al., 1990). The Scrubbing using the salicylic acid Technique uses an acid and its production rate to obtain OH radical concentrations (Salmon et al., 2004). The Spin Trapping method uses electron spin trap and 4-OH-POBN to obtain the OH concentrations in the laboratory for some theoretical researches (Watanabe et al., 1982). Apart from the six physical and chemical methods mentioned above, many researchers used the high-precision spectrum data from the ground-based instruments especially the Fourier Transform Ultraviolet Spectrometer (FTUVS) in the Table Mountain: The OH $P_1(1)$ absorption spectrums which are measured by FTUVS are used to invert the OH concentrations (Cageao et al., 2001). The OH $Q_1(2)$ absorption spectrums which are measured by FTUVS are used as auxiliary spectrum to improve the accuracy of OH concentrations (Mills et al., 2002). An improve retrieval method is described that use an average method based on spectral fits to multiple lines weighted by line strength and fitting precision for obtain the OH concentrations. (Cheung et al., 2008)."

5. Page 4, Line 155: Change “The hierarchical detection of a series of observed radiance in the same target area comes true through the movement of satellite platform.” to “The hierarchical detection of a series of observed radiance in the same target area comes true when the satellite flies along its orbital path.”
6. Page 4, Line 157: Add “It means the sensor can obtain the data of each target height like multi-angle method.”
7. Page 8, Line 210: Add “satellite”
8. Page 8, Line 211: Add “satellite”
9. Page 8, Line 213: Change “…cannot reflect the actual OH distributions…” to “…make bigger difference…”
10. Page 8, Line 214: Change “However,…” to “Meanwhile,…”
11. Page 9, Line 221: Change the Figure 4
12. Page 11, Line 243: Change “…the atmospheric background radiance and the OH fluorescence emission radiance.” to “…the atmospheric background radiance is the result which the solar radiance is subjected to Rayleigh scattering and trace gas absorption and the OH fluorescence emission radiance is subjected to the OH fluorescence emission mechanism.”
13. Page 11, Line 247: Change the Figure 6
14. Page 11, Line 255: Change old paragraph to the new paragraph
15. Page 12, Line 261: Change old paragraph to the new paragraph
16. Page 12, Line 264: Add two citations “(Luque et al., 1999; Sinnhuber et al., 2009”
17. Page 12, Line 267: Change old paragraph to the new paragraph
18. Page 12, Line 267: Add “by default”
19. Page 12, Line 268: Change “The SCIATRAN absorption spectrum database is completed based on the HITRAN 2014 database which contains the OH absorption profiles to solve this problem.” to “The OH absorption profiles data in the ultraviolet band is added based on the HITRAN 2014 database into SCIATRAN to solve this problem.”
20. Page 12, Line 270: Change old paragraph to the new paragraph
21. Page 12, Line 291: Add “The simulated atmospheric background radiance is mainly subjected by the function of Rayleigh scattering, ozone absorption and OH self-absorption. It is calculated based on some parameters such as the spatial, temporal and observation geometries. An observed radiance profile is given as an example in the (27 °N,106 °E) area in the Fig. 7. The intensity of simulated observation radiance and atmospheric background radiance at some tangent heights are also given in the Table 1.”
22. Page 13, Line 300: Add a new Table 1
23. Page 14, Line 314: Add two units “(lines/mm)” in the Table 2
24. Page 14, Line 324: Change the Figure 8
25. Page 19, Line 402: Change the Figure 12
26. Page 20, Line 421: Add “The intensity of simulated observation radiance and atmospheric background radiance at some tangent heights are given in the Table 1. The OH fluorescence emission radiance can be calculated by subtracting the atmospheric background radiance from the observed radiance.”
27. Page 21, Line 452: Change “…a converse…” to “…an inverse…”
28. Page 22, Line 480: Add “The each parameters which constitute the database are accurate and the errors of them are analyzed in the Table 3. The method of constructing the database is

reasonable.”

29. Page 22, Line 485: Change the Figure 15
30. Page 23, Line 491: Add “…at all wavelengths…”
31. Page 23, Line 492: Add “…at all wavelengths…”
32. Page 24, Line 499: Change the Figure 16
33. Page 24, Line 504: Add “The profile data in the database is averaged to obtain the monthly average concentration profile data. So, it will not introduce discontinuities to the retrieved OH time series when an abrupt switch happens”
34. Page 26, Line 534: Change the Figure 18
35. Page 27, Line 554: Add “…,which can get though the y_j and $N_g(j)$ when the converges meet the accurate requirement or the number of iterations exceeds the iteration number threshold,…”
36. Page 28, Line 598: Change “The OH concentrations obtained by the LSUV retrieval algorithm are credible in the upper atmosphere. These results are the same as the results of MAHSRI and SHIMMER (Conway et al., 1999;Englert et al., 2010). However, the OH concentrations in the lower atmosphere such as below 30 km are unsuitable for scientific research. That is the reason why there are no OH concentrations in these regions from the MAHRSI and SHIMMER (Harlander et al., 2002).” to “The OH concentrations obtained by the LSUV retrieval algorithm are credible in the upper atmosphere which are the same as the results of MAHSRI and SHIMMER (Conway et al., 1999;Englert et al., 2010). However, the OH concentrations in the lower atmosphere such as below 30 km are unsuitable for scientific research because the interference factors like water vapor and ozone are too much. A single spatial heterodyne spectrometer with traditional limb mode cannot also obtain enough and high-quality data to invert the OH concentrations that will make the inversion results unscientific especially in the lower atmosphere.”
37. Page 28, Line 605: Add “The tomographic retrieval algorithm and the LSUV are two independent algorithms.”
38. Page 28, Line 611: Change “…the undisturbed parts are taken as the actual parameters, and the results based on these parts are the true OH fluorescence emission radiance. Then, some certain amounts as the disturbances are applied to the three parts above. The OH fluorescence emission radiance based on these parts is the incorrect results. The relative errors between the true and incorrect OH fluorescence emission radiance is calculated by the Eq. (17):…” to “1) some certain amounts as the disturbances are applied to the three parts above. The OH fluorescence emission radiance which obtained based on these parts is considered as the incorrect results. 2) the original parts are taken as the actual and undisturbed parameters, and the results based on these parts are the true OH fluorescence emission radiance. The relative errors between the true and incorrect OH fluorescence emission radiance is calculated by the Eq. (17):”
39. Page 31, Line 674: Change “The distributions of errors have the same tendency, but the relative errors of tomographic retrieval algorithm are smaller than LSUV retrieval algorithm. These indicate that tomographic retrieval algorithm can obtain more accurate OH concentrations although the Doppler effect cannot avoid.” to “The tendency of relative errors is same, but the relative errors of tomographic retrieval algorithm are smaller than LSUV retrieval algorithm. These indicate that tomographic retrieval algorithm can obtain more accurate OH concentrations than the LSUV retrieval algorithm although the errors caused by Doppler effect cannot avoid when the spectral resolution of DSHS is extremely high.”

40. Page 32, Line 681: Add "We assumed the instrument calibration errors are ±5% in this research after referring to the detector parameters due to the DSHS do not officially work (Song et al., 2009)"
41. Page 32, Line 683: Add "The cubic spline interpolation method not only has higher stability but also can ensure the continuity and smoothness of the interpolation function under the premise of the ensuring the convergence compared with other interpolation methods like linear interpolation method and some others. The errors estimation of the interpolation results is as accurate as possible."
42. Page 32, Line 689: Add "We give a certain disturbance to different parameters to calculate the effect of the interpolation algorithm on the OH concentrations"
43. Page 32, Line 694: Change "Analysis of total errors" to "Analysis of total errors in the OH concentrations"
44. Page 32, Line 695: Add "The errors of retrieved results must be considered after the discussions about the errors of OH radiance."
45. Page 34, Line 730: Change "As the Table 7 indicates that the total errors of OH concentrations are increasing as the heights rise." to "As Table 8 indicates, the total errors of OH concentrations initially increase with the heights rising."
46. Page 34, Line 733: Add "However, the OH concentrations obtained by the LSUV algorithm in the same area are unsuitable for scientific research mentioned in the Sect. 5.2."
47. Page 34, Line 735: Add "The relative errors of the OH concentrations obtained by LSUV algorithm are calculated by the Eq (17) and the error transfer formula mentioned above."
48. Page 34, Line 740: Change "The corresponding accuracy ranges of MLS OH concentrations products are also given, and its useful height ranges are from 23 to 81 km." to "The corresponding accuracy ranges and useful height ranges of MLS OH concentrations products are also given by the MLS data quality and description document."
49. Page 35, Line 773: Add "However, it is difficult to quantify how much fast is the tomographic algorithm absolutely. The traditional iterative retrieval algorithm will take several or even tens of hours to obtain OH concentrations due to the complex iterative process. The tomographic algorithm just needs minutes for obtaining the OH concentration due to the usage of the lookup table method. The results obtained by the traditional iterative retrieval algorithm in the lower atmosphere are unscientific but the OH concentrations obtained by tomographic algorithm in the same area are accurate."
50. Page 36, Line 813: Add "The actual observed data is the key point for this research. Some aircraft flights experiments have been done and the data is being analyzed. The DSHS and research results have been communicated with the related departments. It has been included in the relevant satellite projects for 2020 to 2030."
51. Page 38, Line 873: Add "Luque J, Crosley D R. LIFbase: Database and Spectral Simulation[J]. 1999"
52. Page 38, Line 885: Add "Sinnhuber, B.-M., Sheode, N., Sinnhuber, M., Chipperfield, M. P., and Feng, W.: The contribution of anthropogenic bromine emissions to past stratospheric ozone trends: a modelling study, Atmospheric Chemistry and Physics, 9, 2863–2871, https://doi.org/10.5194/acp-9-2863-2009, 2009."

Tomographic retrieval algorithm of OH concentration profiles using Double Spatial Heterodyne Spectrometers

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Abstract. The hydroxyl radical (OH) determines the atmospheric self-cleaning capability and is one of the significant oxidants in atmospheric photochemistry reactions. The global OH has been monitored by satellites with the traditional limb mode in the past decades. This observed mode can achieve high-resolution vertical OH data, but cannot obtain the enough horizontal OH data for inverting high-precision OH concentrations because OH has the high reactivity that makes OH concentrations extremely low and distributions complicated. The Double Spatial Heterodyne Spectrometers (DSHS) is designed in order to achieve more high-resolution and detailed OH data. This sensor can measure the OH by the three-dimensional limb mode to obtain the comprehensive OH data in the atmosphere. We propose a new tomographic retrieval algorithm here based on the simulated observation data due to the DSHS will work officially on the orbit in the future. We build up an accurate forward model that the main part of it is the SCIATRAN radiative transfer model which is modified according to the radiation transmission theory. The error of the results obtained by the forward model is ±44.30% in the lower atmosphere such as 21 km height and decreases gradually until the limit of observation altitude . We also construct the tomographic retrieval algorithm that the core is a look up table method. A tomographic observed database is built up through the atmospheric model, the spatial information (position of the target area and satellite position), the date parameters, the observation geometries, OH concentrations and simulated observation data. The OH concentrations can be found directly from it. If there are no corresponding query conditions in the tomographic observed database, the cubic spline interpolation is used to obtain the OH concentrations. The inversion results are given and the errors of them increase as the altitudes rise until about 41 km height then start to decrease. The errors of the inversion results reach the maximum about ±25.03% in the 41 km height and decrease to ±8.09% in the limited observation height. They are also small in the lower atmosphere which are ±12.96% in the 21 km height. In summary, the tomographic retrieval algorithm can obtain the more accurate OH concentrations even in the lower atmosphere where the OH data is not well and avoids the initial guess values for solving the iteration problems. Our research not only provides a scientific theory support for the construction of DSHS, but also gives a new retrieval algorithm idea for other radicals.

1 Introduction

The OH plays an important and initial role in atmospheric photochemistry reactions because of its strong oxidation It can remove many natural and anthropogenic compositions which are significant for the air quality, the ozone distributions even the climate change from the atmosphere (Stevens et al., 1994). OH mainly derives from the reactions of O(^{1}D), a photolytic product of ozone in the ultraviolet band, with water vapor in the middle and upper atmosphere. The removal of OH is affected by various compounds like nitrogen oxides, sulfur dioxide, carbon monoxide, methane and other volatile organic compounds (Lelieveld et al., 2016;Wolfe et al., 2019;Zhang et al., 2018). Therefore, on the one hand, the OH affects the photochemical and kinetic processes in the atmosphere and reflects the short-term and long-term climatic evolution processes in some respects. On the other hand, it is of great significance to enhance the understanding of the atmospheric physical and chemical processes.

It is a great challenge to monitor the OH in the atmosphere because of its low concentrations and strong activity. The Fluorescence Assay by Gas Expansion (FAGE) uses the 308 nm excitation mechanism to excite the OH radical continuously for generating the fluorescent signal to establish the relationship between the OH concentrations (Hard et al., 1984). The Differential Optical Absorption Spectroscopy (DOAS) obtain the OH concentrations because the absorption of OH follows the Lambert-Beer absorption law (Perner et al., 1976).The Chemical Ionization Mass Spectrometry (CIMS) collects ions of OH instead of the photons based on OH oxidation to obtain the OH concentrations (Mauldin et al., 1998) .These methods are commonly used to measure the OH concentrations in the actual limited environments and laboratories. In addition, the ^{14}CO oxidation method uses the $^{14}CO_2$ concentration, enrichment coefficient, reaction rate constant and reaction time to get the OH radical concentrations based on the OH oxidation (Felton et al., 1990). The Scrubbing using the salicylic acid Technique uses an acid and its production rate to obtain OH radical concentrations (Salmon et al., 2004). The Spin Trapping method uses electron spin trap and 4-OH-POBN to obtain the OH concentrations in the laboratory for some theoretical researches (Watanabe et al., 1982). Apart from the six physical and chemical methods mentioned above, many researchers used the high-precision spectrum data from the ground-based instruments especially the Fourier Transform Ultraviolet Spectrometer (FTUVS) in the Table Mountain: The OH P_1(1) absorption spectrums which are measured by FTUVS are used to invert the OH concentrations (Cageao et al., 2001). The OH Q_1(2) absorption spectrums which are measured by FTUVS are used as auxiliary spectrum to improve the accuracy of OH concentrations (Mills et al., 2002). An improve retrieval method is described that use an average method based on spectral fits to multiple lines weighted by line strength and fitting precision for obtain the OH concentrations. (Cheung et al., 2008). These methods are restricted by the features of the instruments which can provide accurate OH data in the limited lower atmosphere but are difficult to provide enough data for inverting OH concentrations from mesopause to tropopause. The development of satellite technologies carries out the global wide-scale detection of OH. Conway retrieved OH concentrations by the least squares fitting method until the success of the Middle Atmosphere High-Resolution Spectrometer Investigation (MAHRSI) which provided the first observed data of OH in the mesosphere with diffractions grating technology from space (Conway et al., 1999). A new interferometric technique called spatial heterodyne spectroscopy was applied to the Spatial Heterodyne Imager for

Mesospheric Radicals (SHIMMER) for reaching the higher spectral resolution on the space shuttle middeck with small size and no mobile optical components. This satellite sensor measured the OH solar resonance fluorescence by the limb mode from the low Earth orbit to obtain more accurate OH data. Englert used the same method as the MAHRSI to invert OH concentrations (Englert et al., 2008). Besides these sensors mentioned above, the 2.5-THz radiometer on the Microwave Limb Sounder (MLS) was designed to measure the thermal emission signal of OH from stratosphere to mesosphere because the spectral region around the pair of strong OH lines at 2.51 and 2.514 THz is clean relatively. The sensitive thermal emission data is used to invert the volume mixture ratio of OH concentrations, however the results have a poor signal to noise ratio in some atmospheric regions. Livesey used the standard optimal estimation method to obtain the OH concentrations from the calibrated MLS observed data (Level 1B) with the radiative transfer equation. He also corrected the noisy products by a separate task using the full radiance dataset as the relevant band after the main process (Livesey et al., 2006). Wang used the average orthogonal fitting method to handle with the MLS data for obtaining the OH concentrations, and compared the results with the ground-based FTUVS results in some areas. They matched very well (Wang et al., 2008). Damiani handled the MLS data of nighttime winter in the northern hemisphere from the latitude of 75° to 82° by regional mean method to research the change of OH concentrations in short-term (day) and long-term (weeks) dimension (Damiani et al., 2010).

The MAHRSI and SHIMMER measured the OH solar resonance fluorescence in the $\mathrm{A}^2\Sigma^+ - \mathrm{X}^2\Pi(0,0)$ band at the wavelength around 309 nm and proved that is the more possible and effective way to monitor atmospheric OH from space load at present. The traditional limb mode can obtain high-resolution vertical OH radiance profile which contains the information of OH concentrations at different tangent plane because of the benefit of spectral technology. However, the OH radiance which is measured at one field of view is the sum of the overall contributions in the corresponding fields of view, but the OH is inhomogeneous in fact. These will lead to some precision errors (Englert et al., 2010). Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, has done some researches on the DSHS in advance to improve the horizontal resolution of OH data and achieve the better structure of OH profiles. The DSHS, will work on the orbit at 500 km, is designed to measure the OH solar resonance fluorescence from 15 to 85 km by using the double spatial heterodyne spectrometers in orthogonal layout. The field of view angle is designed as 2 degree and the spectral resolution will reach 0.02 nm at least. This paper gives the OH interferogram which is emulated by a forward model and describes a new method called the tomographic retrieval algorithm which uses simulated DSHS data and theory of the look up table method to invert OH concentrations. The OH concentrations in the target area will be found out in the tomographic observed database which constructs by the atmospheric model, the spatial information, the date parameters, the observation geometries, OH concentrations productions and the simulated observation data. The retrieval algorithm can obtain more accurate OH concentrations in the middle and upper atmosphere especially in the lower atmosphere than the results which are obtained by traditional limb retrieval algorithms and can raise the inversion speed compared with the traditional limb retrieval algorithms.

2 Measurement strategy

2.1 The principle of spatial heterodyne spectroscopy

The DSHS is designed based on the principle of the spatial heterodyne spectrometer which the optical structure is shown in Fig. 1. The two planar mirrors in the traditional Michelson interferometer are replaced by the two diffraction gratings (grating 1, grating 2). The light, enters through aperture A, is collimated by the lens L1 and is split into two beams of coherent light of equal intensity by the beam splitter. One of the them is reflected by the beam splitter and incidents on the grating 1. The light is diffracted by the grating 1 and returns to the beam splitter. The other light incidents on the grating 2 through the beam splitter as well. It is diffracted by the grating 2 and reflects back to the beam splitter. The gratings are fixed which are placed at a Littrow angle θ with the orthogonal plane of the optical axis in the spatial heterodyne spectrometer system. The light incidents on the gratings by θ angle and diffracts back with θ angle at some wavenumbers. It is called the light of Littrow wavenumber σ_0. The two exiting wavefronts of light of σ_0 are diffracted by the gratings and perpendicular to the optical axis. The phase difference is zero and the interference fringe spatial frequency is zero that cannot form the interferogram. However, the light of non-Littrow wavenumber has an $\pm\gamma$ angle with the optical axis. The light forms interferogram which will image on the imaging detector by the optical imaging systems L2 and L3. So, the overlapping two wave surfaces have an angle of 2γ which are calculated by the Eq. (1) (Dohi and Suzuki, 1971):

$$\sigma[\sin\theta + \sin(\theta - \gamma)] = \frac{m}{d} \tag{1}$$

where the σ is the wavenumber of light, m is the order of diffraction, $1/d$ is the grating groove density. The angle between the light of arbitrary wavenumber and the exiting light of Littrow wavenumber is γ. The spatial frequency of two σ light is given by the Eq. (2):

$$f_x = 2\sigma\sin\gamma \approx 4(\sigma - \sigma_0)\tan\theta \tag{2}$$

For the input spectrum $\mathrm{B}(\sigma)$, the intensity on the imaging detector is given by Eq. (3):

$$I(x) = \int_0^\infty B(\sigma)\big(1 + \cos\big(2\pi(4(\sigma - \sigma_0)x\tan\theta)\big)\big)d\sigma \tag{3}$$

The spectral curve $\mathrm{B}(\sigma)$ can recover from interferogram $I(x)$ by Fourier transform algorithm.

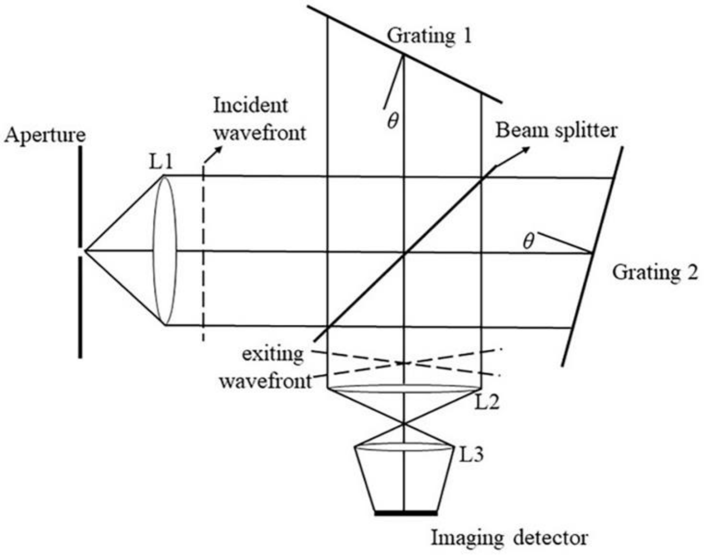

Figure 1. Schematic diagram of spatial heterodyne spectrometer.

Furthermore, the spatial heterodyne spectroscopy can acquire spatial distribution of one-dimensional spectral information through the two-dimensional detection technology. The scene in the field of view is divided into multiple field of view slices which are less or equal to the number of rows in the focal plane array by adding a cylindrical mirror to the front or rear optical system. The interferograms of each field of view slices are imaged on the corresponding detector rows respectively. The several rows on the detector correspond to a layered spectral information in a field of view. The corresponding spatial resolution unit on the detector can indicate the spatial information when the target signal appears in a certain range of line. These mean that the spatial heterodyne spectroscopy can obtain the radiance of atmosphere information at different altitudes simultaneously without scanning the atmosphere from a height to other height respectively. It is important for monitoring the OH to reduce the time of data acquisition because the OH is variable fast in the spatial and temporal dimension.

2.2 Instrument innovation

The spatial heterodyne spectroscopy technique, an ultra-high-resolution spectroscopy technique used in the DSHS, has the characteristics of high flux, no mobile part and small size. The DSHS which is shown in the Fig. 2 mainly consists of two spatial heterodyne spectrometers in the orthogonal layout to obtain the enough data for inverting the OH concentrations. It includes the telescope cylinder system, the collimating system, the information processing system, the imaging systems and some optical components else. One of the spatial heterodyne spectrometers which scans along the satellite working orbit is defined as SHS1, the other which scans across the satellite working orbit is defined as SHS2. The scanning direction of SHS2 is orthogonal to SHS1s' due to the special design of the orthogonal layout. The hierarchical detection of a series of observed radiance in the same target area comes true when the satellite flies along its orbital path. It means the sensor can obtain the data of each target height like multi-angle method.

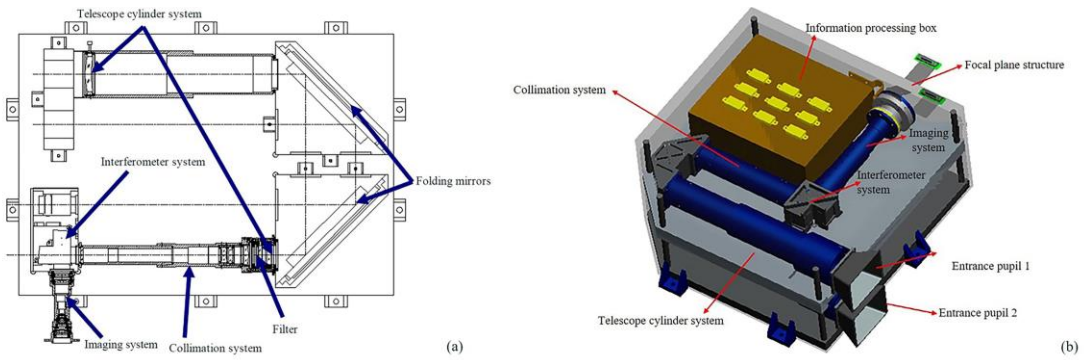

Figure 2. Illustration of the Double Spatial Heterodyne Spectrometers. the left part (a) shows the vertical view of the DSHS and the right part (b) shows the overall mold of the DSHS.

The DSHS will detect the altitudes from 15 to 85 km by the three-dimensional limb mode. It can obtain the observed data in the target area along the satellite working orbit and across the satellite working orbit respectively at the same time because the SHS1 and SHS2 are orthogonal to each other. The Figure 3 shows how the tomographic data is obtained in the process of satellite working. As the Fig. 3 (a) shows that there are no intersection regions between the field of view slices of SHS1 and SHS2s' at the time of T1. The tomographic data cannot be obtained in this situation. The three-dimensional structure of OH data is unable to be reconstructed. The data of SHS1 and SHS2 at different time will be used to get an intersection region for solving this issue. The different time which meet the requirements of three-dimensional limb mode of the DSHS (we define the first time as the time of T1 and the next time as the time of T2) is shown in the Fig. 3 (b). At the time of T1, the SHS1 and SHS2 finish a limb scanning in the vertical dimension (along the satellite working orbit) and the horizontal dimension (across the satellite working orbit), then the satellite platform will work into the next position at the time of T2 and finishes the next observation project. The line of sight at the time of T1 will intersect with the line of sight at the time of T2. The intersection region (target area) will form. However, the composition of tomographic data does not need all data from the SHS1 and SHS2 at the time of T1 and T2. Some data should be omitted. As the Fig. 3 (c) shows that the field of view slices of T2-SHS2 (a horizontal dimension spectrometer at the time of T2) are orthogonal to the field of view slices of T1-SHS1 (a vertical dimension spectrometer at the time of T1) in an intersection region. The data obtained by the SHS2 at the time of T1 and the SHS1 at the time of T2 will be omitted. Thus, the three-dimensional segmentation of the target area will complete. The three-dimensional division of the target atmosphere which consists of the intersection regions of SHS1 and SHS2 at different time is accomplished finally with the satellite platform moving forward continually. Different combinations of data for different intersection regions from two spatial heterodyne spectrometers at different time are used to invert OH concentrations.

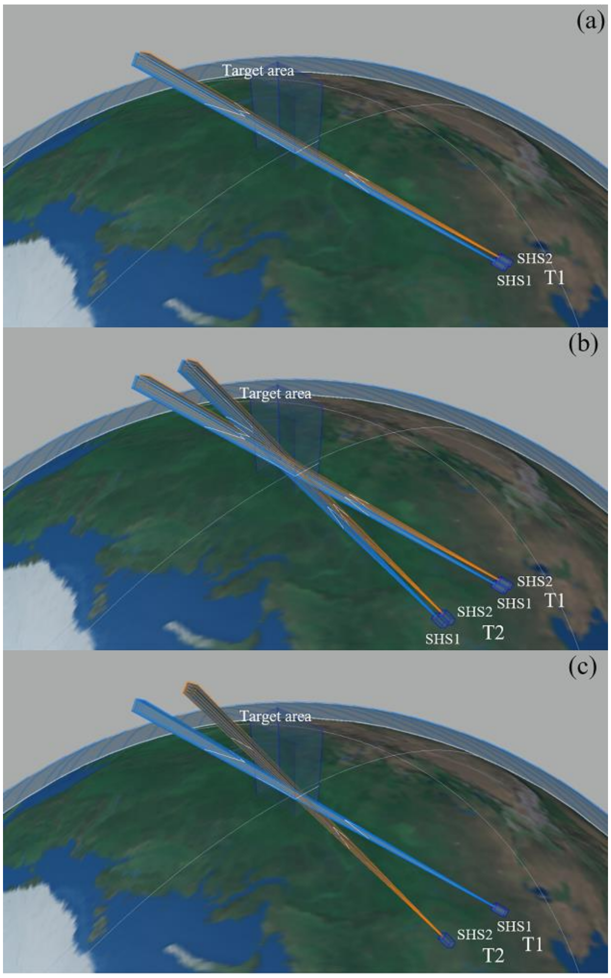

Figure 3. Three-dimensional limb mode of DSHS at the time of T1 and T2 (a). The DSHS consists of two spatial heterodyne spectrometers (SHS1 and SHS2). The data of target area cannot be obtained by DSHS at the time of T1 and T2 respectively (b). DSHS monitors the OH in the target area using the data of SHS1 at the time of T1 and SHS2 at the time of T2 together (c).

The scanning direction will not be completely vertical or parallel to the surface when the DSHS will be on the orbit, but will have an angle η. So, the satellite positions at the time of T2 where T2-SHS2 is orthogonal to T1-SHS1 in the intersection region are different with the η changing. In order to display and descript the satellite position of T2 conveniently, the η is set to 0° temporarily. However, a unified set of η is 30° in the relevant calculation during pre-research phase.
The satellite positions and observation geometries are given at the time of T1. The satellite positions at the time of T2 need to calculate for realizing the three-dimensional segmentation of the target atmosphere. It indicates that there are two satellite positions where the scanning directions of T2-SHS2 and T1-SHS1 are orthogonal to each other in the intersection region at the time of T2 based on the theory mentioned above. Figure 4 shows that there is a tangent point which is defined as P1 when the DSHS detects the atmosphere by the three-dimensional limb mode. The intersection region along the lines of sight of SHS1 (the blue line in the Fig. 4) and SHS2 are the observed data at the time of T1. There will be two lines of sight (the red lines in the Fig. 4) where the scanning direction of SHS2 is orthogonal to the SHS1s' under the premise that the working altitude of the satellite is

constant. The satellite position on the same side to the time of T1 is defined as the position at the time of T2-1, and on the opposite side to the time of T1 is defined as the position at the time of T2-2. The two lines of sight at the time of T2 (T2-1 and T2-2) and a single line of sight at the time of T1 form two intersection regions R1 and R2 which are symmetrical about P1 along the line of sight at the time of T1. Figure 5 shows relationship of the satellite positions at the time of T1 and T2 in the space. These two intersection regions are on the two sides of tangent point and are at the altitude of 500 km. The distance of the satellite position at the time of T2-2 from the time of T1 is farther than the satellite position at the time of T2-1 that means satellite needs more time to work from the position of the time of T1 to the position at the time of T2-2. The observed data will change a lot in this process and make bigger difference in the target area at the time of T1. The satellite position at the time of T2-2 should be omitted. Meanwhile, the satellite position at the time of T2-1 is near the position at the time of T1, the observed data at the position of the time of T2-1 can reflect the OH distribution in the target area better because the OH information does not change too much in the short working time of satellite. The distances between the intersection region and the tangent point determine the spatial resolution of the three-dimensional limb mode. That means, the smaller distance between the P1 and R1 (or R2) is, the higher spatial resolution will be.

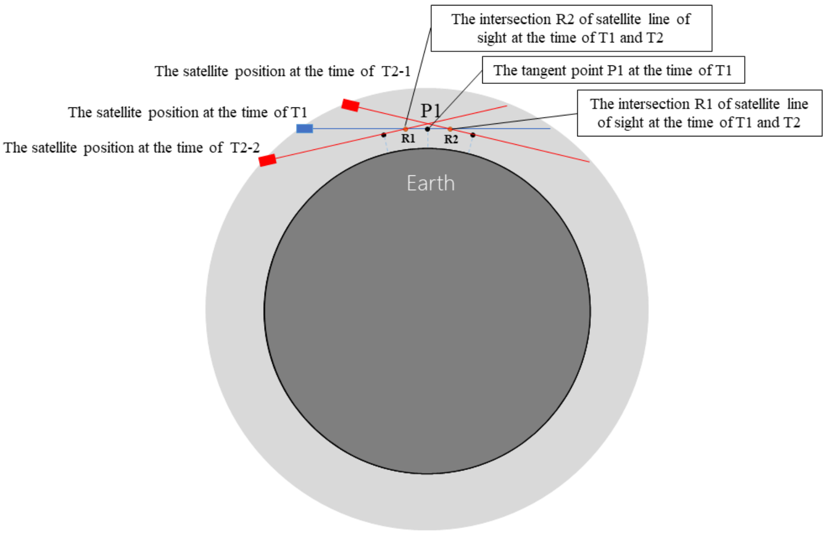

Figure 4. Limb geometry diagram for the three-dimensional limb mode at the time of T1 and T2. The blue line is the line of sight of SHS1. The red lines are the lines of sight of SHS2.

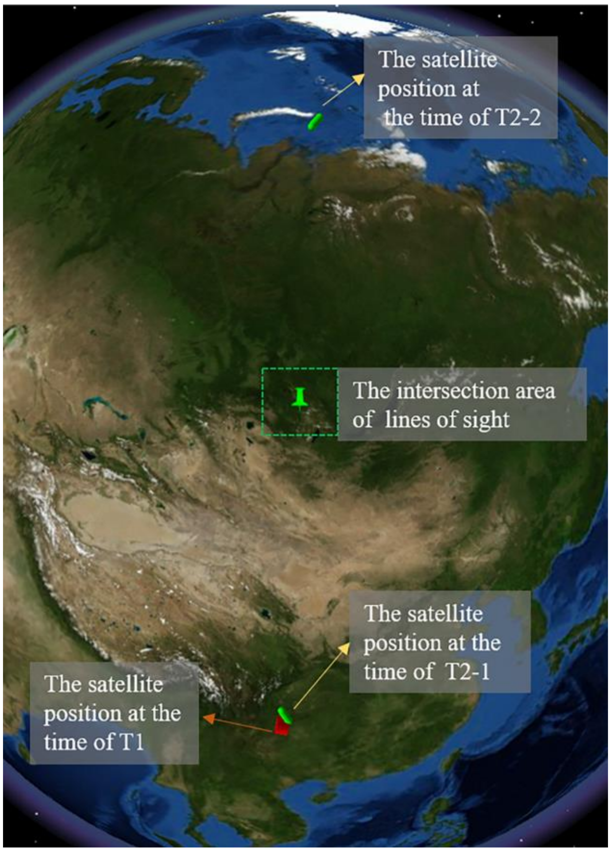

Figure 5. Relative location of DSHS by three-dimensional limb mode. There are two locations of satellite at the time of T2 which meet the requirement of three-dimensional limb mode. Map data: © Google Earth, Image IBCAO and Landsat/Copernicus.

So, the position of the time of T2-1 is picked up as the other satellite location where the field of view slices are orthogonal to each other at the intersection region. The OH concentrations at different heights are obtained by the combinations of observed data from the SHS1 at the time of T1 and SHS2 at the time of T2 in the intersection region. Although the observed data are different because the geometric position and the viewing angle are of difference between the time of T1 and T2 the OH concentrations in the target area will also be constant. The data forms and the structures of the observed data of the SHS1 and SHS2 are the same in three-dimensional limb mode. The distance between the intersection point R1 and tangent point P1 is presumed as 50 km which is used in the subsequent research.

3 Forward modeling

The forward model is used to express a physical process how the atmospheric state parameter is obtained by the sensors. A precise forward model is significant for the design of the instrument and the retrieval algorithm. The observed data received by the DSHS with an ultra-high spectrum resolution makes up by two parts in this research: the atmospheric background radiance and the OH fluorescence emission radiance: the atmospheric background radiance is the result which the solar radiance is subjected to Rayleigh scattering and trace gas absorption and the OH fluorescence emission radiance is subjected to the OH fluorescence emission mechanism. These data can be simulated by a forward model which is constructed by the flow chart in the Fig. 6.

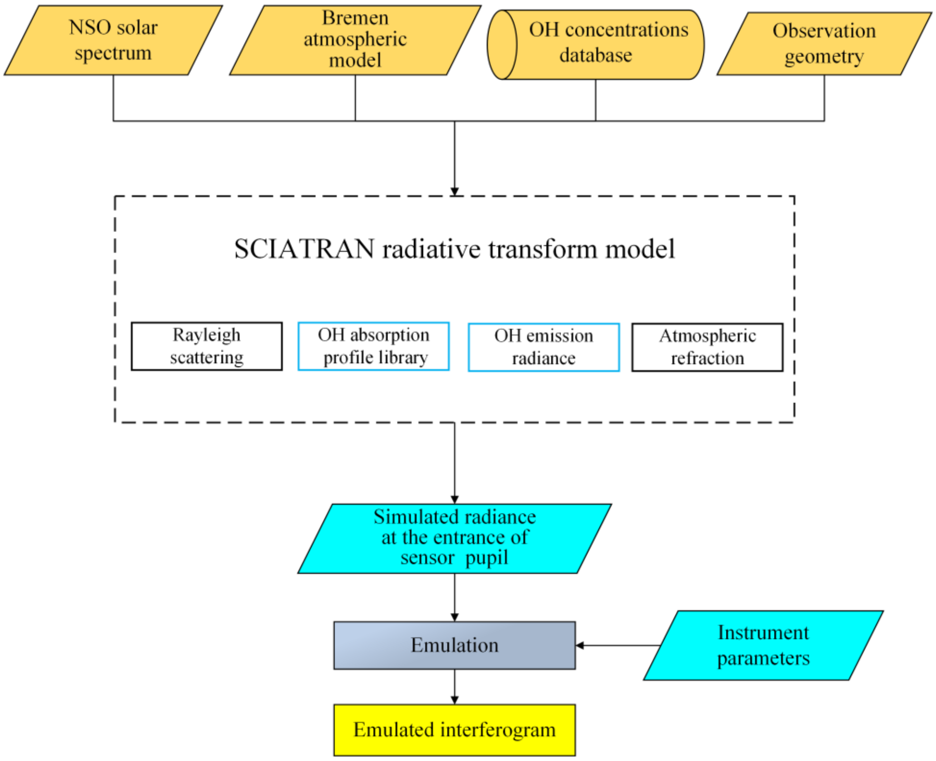

Figure 6. Flow chart of the Forward model.

The forward model is mainly constituted by a radiative transfer model called the SCIATARN which is developed for the Scanning Imaging Absorption spectrometer for Atmospheric CHartographY (SCIAMACHY). However, some parts of it need to modify for meeting the characters of the DSHS. First, the radiance source, the solar radiation, is considered in this forward model for the radiance transmission of OH in the middle and upper atmosphere because (1) OH fluorescence emission radiance will be generated from the excited state to the ground state when the OH is excited by the solar radiation. A high-precision calculation of the OH fluorescence emission radiance requires the high-resolution solar radiation, (2) the DSHS has a spectral resolution of 0.02 nm at least. The higher spectral resolution solar radiation will be used to get more realistic values. So, the National Solar Observatory (NSO) solar spectrum with resolution of 8.6×10^{-4} nm is used for this forward model. In addition to the NSO, the precision of the OH spectrum is also significant. SCIATRAN just takes the Rayleigh scattered radiance function, the ozone absorption function, the OH self-absorption function into account, but ignores the effect of OH fluorescence emission radiance. The Lifbase software is used to calculate the OH emission spectrum in a temperature ranges according to the Bremen atmospheric model (Luque et al., 1999; Sinnhuber et al., 2009). The OH emission spectrum database which put into the SCIATRAN as a source function is built up based on it. The corresponding OH spectral emission data can get from the database when the observed radiance is simulated at some conditions.

Next, the OH absorption spectrum database in the SCIATRAN bases on the HITRAN 2012 database by default which there are no OH absorption profiles in the ultraviolet band. The OH absorption profiles data in the ultraviolet band is added based on the HITRAN 2014 database into SCIATRAN to solve this problem.

Finally, the precise atmospheric composition parameters are the assurance to simulate the observed radiance received by the sensors. That means the high-quality OH concentrations database is needed when the radiance transfer is simulated accurately. For one thing, the MLS on the Aura has monitored the global OH for many years and the data can be acquired publicly at present. These data are used to build up the OH concentrations database. MLS was launched in15 July 2004 aboard on Aura, and kept offering consequent OH data until 2009. It was stopped monitoring from the November 2009 to the August 2011 in order to extend the service life of the sensor. It is restarted to monitor OH on the August and the September each year continually to obtain annual change trends of OH data. For another thing, the solar activity was relatively stable and maintained at a low level in the whole solar activity cycle from 2005 to 2009. The solar activity from 2010 to 2016 had just ended an active cycle, and the level of solar activity in the next few years would be similar to the situation from 2005 to 2009. Therefore, the OH concentrations data from 2005 to 2009 is a great reference for the research of atmospheric OH concentrations by the DSHS in the next few years. The MLS OH concentrations Level 2 geophysical production is used and averaged within a certain range for reducing the random errors to build up the OH concentrations database. The N32 Gaussian grid from European Centre for Medium-range Weather Forecasts is selected as the averaged methods due to the random errors of MLS OH concentrations in high latitude are much larger than them in low latitude. It is a lattice-level coordinate system for scientific model of spheres in earth science. The latitude and longitude zones in the grid are divided unconventionally. The latitude band intervals of the northern and southern hemispheres are symmetrical about the equator. The latitude band intervals and number of longitude bands on a latitude gradually decrease with the increase of the latitude to ensure that each grid area is approximately equal. In addition, the four seasons definition is used, the first quarter (March/April/May), the second quarter (June/July/August), the third quarter (September/October/November), the fourth quarter (December/January/February), as the temporal resolution for the OH concentrations database. The season is first quarter and the OH concentrations database is N32-seasonly database. The simulated atmospheric background radiance is mainly subjected by the function of Rayleigh scattering, ozone absorption and OH self-absorption. It is calculated based on some parameters such as the spatial, temporal and observation geometries. An observed radiance profile is given as an example in the (27 °N,106 °E) area in the Fig. 7. The intensity of simulated observation radiance and atmospheric background radiance at some tangent heights are also given in the Table 1.

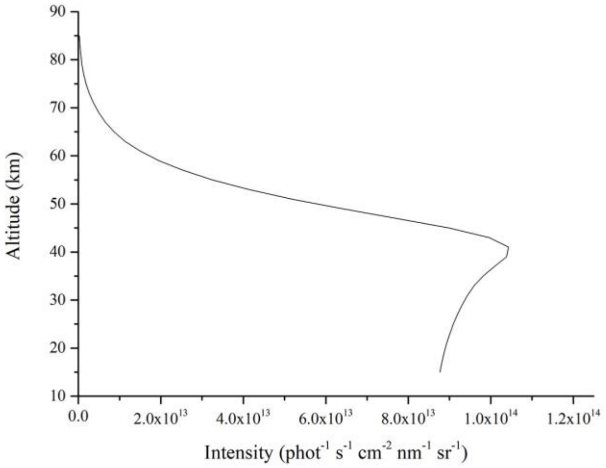

Figure 7. An observed radiance profile obtained by the modified SCIATRAN.

Table 1. The intensity of simulated observation radiance and atmospheric background radiance at some tangent heights

Altitude (km)	Intensity of simulated observation radiance (phot^{-1} s^{-1} cm^{-2} nm^{-1} sr^{-1})	Intensity of simulated atmospheric background radiance (phot^{-1} s^{-1} cm^{-2} nm^{-1} sr^{-1})
21	8.933×10^{13}	8.885×10^{13}
31	9.436×10^{13}	9.379×10^{13}
41	1.043×10^{14}	1.034×10^{14}
51	5.184×10^{13}	5.086×10^{13}
61	1.498×10^{13}	1.429×10^{13}
71	3.682×10^{12}	3.195×10^{12}
81	6.030×10^{11}	5.866×10^{11}

The results of modified SCIATRAN will be imaged in the image system which collects a complete data from all pixels on one line of CCD. The digital number (DN) values of each pixel in each row are generated by Eq. (4) at different tangent heights:

$$S_{CCD}(i,j) = \int_{y(j)}^{y(j+1)} \int_{x(i)}^{x(i+1)} \int_{0}^{\infty} B(\sigma,y)R(\sigma)\,(1+\cos\{2\pi[4(\sigma-\sigma_0)x\tan\theta]\})d\sigma dx dy$$

(4)

where i is the number of rows along the line, j is the tangent height from 15 to 85 km. The $x(i)$ and $y(j)$ is the areas where grating imaging image in the CCD, $B(\sigma,y)$ is the input radiance spectrum, $R(\sigma)$ is the instrument function, σ is the wavelength number and the θ is the Littrow angle. The interferogram will be got based on the instrument parameters in the Table 2.

Table 2. Instrument parameters for emulation

Parameters	value
Littrow length (nm)	306
Grating groove density (lines/mm)	1000
Numbers of interferogram samples	1024
Grating width (line/mm)	15
Littrow angle (°)	8.80

An emulated interferogram is shown in the Fig. 8. It is the two-dimensional observed radiance interferogram constructing by 1024 columns which indicate numbers of interferogram samples and 36 rows which indicate the observed tangent heights. The observed radiance gradually increases to reach the peak value which is the bright of interferogram with the altitude increases until the altitude around 40 km. The bright decreases gradually until the upper limit of the detection height.

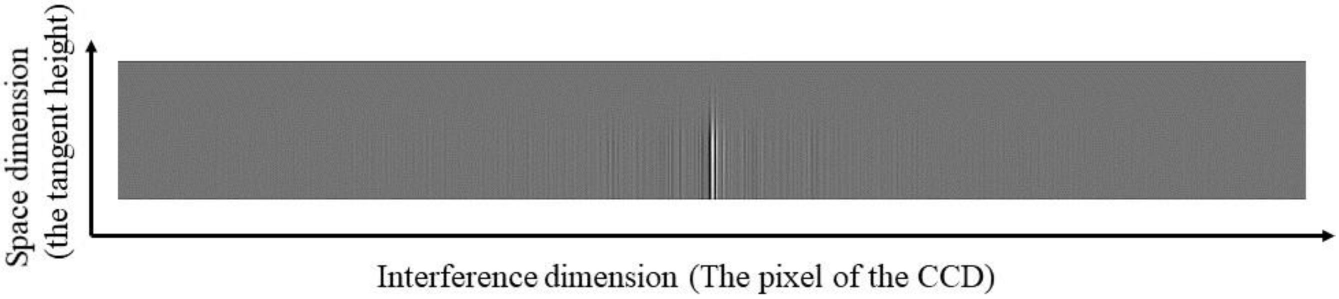

Figure 8. Emulated two-dimensional observed radiance interferogram.

The errors of this forward model mainly depend on three factors based on the modified parts above: the atmospheric model, the Doppler effect and the instrument calibration errors. First, the ozone in the atmospheric model is the influential factor which is considered in the atmospheric model because its absorption cannot ignore in the ultraviolet band. Second, the satellite has different heliocentric speeds when it flies over the area where the local solar time is noon or is around the morning faint line. The speed will lead to the shift of observed radiance at the ranges of wavelength. Third, there are many sources of errors in the process of converting electrical signals into radiance values. The instrument calibration errors are assumed as ±5% because the DSHS has not been officially working. The total errors of observed radiance are also given in the Table 3. Zhang had made the detailed analysis of errors and the reasons (Hong-hai et al., 2017).

Table 3. Total errors of observed radiance caused by different factors at some tangent heights

Tangent height (km)	Errors caused by the atmospheric model (%)	Errors caused by Doppler effect (%)	Errors caused by instrument calibration errors (%)	Total errors (%)
81	From -2.65 to 5.67	From -0.87 to 0.85	From -5 to 5	From -9.52 to 9.52
71	From -2.61 to 5.62	From -1.18 to 0.95	From -5 to 5	From -9.52 to 9.52

61	From -2.81 to 5.83	From -0.98 to 0.83	From -5 to 5	From -9.67 to 9.67
51	From -4.90 to 8.17	From -0.91 to 0.78	From -5 to 5	From -11.93 to 11.93
41	From -16.49 to 26.17	From -0.82 to 0.67	From -5 to 5	From -31.75 to 31.75
31	From -18.92 to 36.64	From -0.77 to 0.65	From -5 to 5	From -41.85 to 41.85
21	From -19.56 to 39.10	From -0.76 to 0.65	From -5 to 5	From -44.30 to 44.30

The DSHS is constituted by double spatial heterodyne spectrometers in the orthogonal layout. The distance between the intersection point and the tangent point is defined as 50 km and the angle between the spatial heterodyne spectrometer scanning direction and the vertical/horizontal direction is defined as 30°. The SHS2 positions at the time of T2 can be calculated according to the SHS1 observation geometry parameters at the time of T1 in the target area based on the detection theory of three-dimensional limb mode. The simulated observation radiance of SHS2 and SHS1 can be obtained by the modified SCIATRAN above. The partial associated geometric parameters of the time of T1 and the calculated geometric parameters of the time of T2 are given in the following Table 4.

Table 4. Locations and geometry parameters of SHS1 at the time of T1 at some tangent height and the corresponding parameters of SHS2 at the time of T2

Altitude (km)	The time of T1			The time of T2		
	Latitude (°)	Longitude (°)	Azimuth angle (°)	Latitude (°)	Longitude (°)	Azimuth angle (°)
21	54.501	121.483	321.98	55.071	121.534	321.041
31	54.501	121.483	321.98	55.117	121.303	321.045
41	54.501	121.483	321.98	55.163	121.073	321.050
51	54.501	121.483	321.98	55.210	120.844	321.056
61	54.501	121.483	321.98	55.255	120.616	321.062
71	54.501	121.483	321.98	55.31	120.389	321.070
81	54.501	121.483	321.98	55.346	120.163	321.078

The intersection region of the time of T1 and T2 mentioned above is (67.46 °N,86.618 °E). The observed radiance received by SHS1 at the time of T1 and received by SHS2 at the time of T2 is composed by a group of different satellite positions at tangent heights which shown in the Fig. 9.

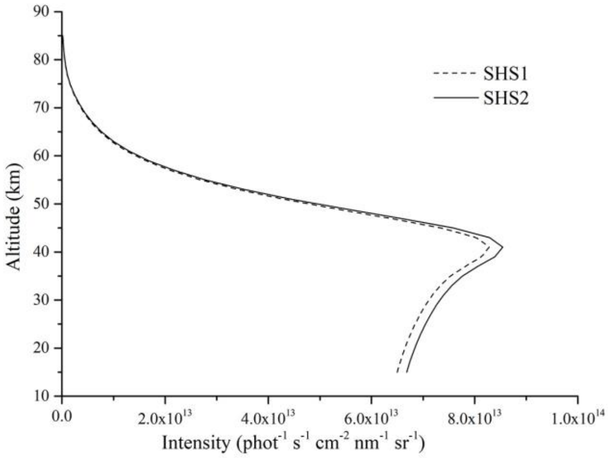

Figure 9. The simulated observation data in the intersection region at the time of T1 and T2. The dash line is the simulated observation data received by SHS1 at the time of T1 and the solid line is the simulated observation data received by the SHS2 at the time of T2.

Although the observation geometries and satellite positions are different at different time, both spatial heterodyne spectrometers scan the atmosphere by the same limb mode, the observed radiance profile still maintains the distribution in the range from 308 to 310 nm band. There are four global observed radiance databases for this research. The first quarter observed radiance is given as an example: the NSO solar spectrum is used as the radiance source ,the Bremen global atmospheric model, the first quarter OH concentrations data of N32-seasonly OH concentrations database and the geometric parameters of MLS OH production is also used to simulate the observed radiance in the range from 308 to 310 nm band at the time of T1 and T2. The results are shown in the Fig. 10 which the observed radiance received by SHS1 at the time of T1 and Fig. 11 which the observed radiance received by SHS2 at the time of T2.

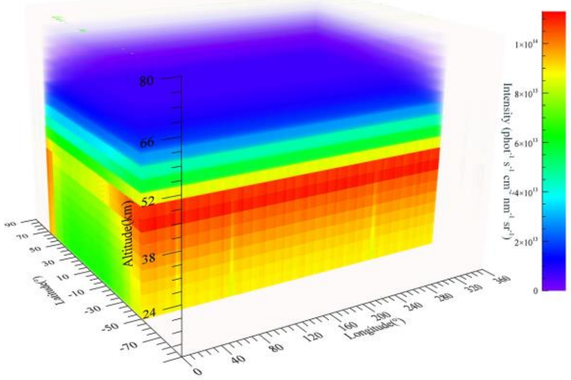

Figure 10. The simulated observation radiance received by SHS1 at the time of T1 under the different conditions

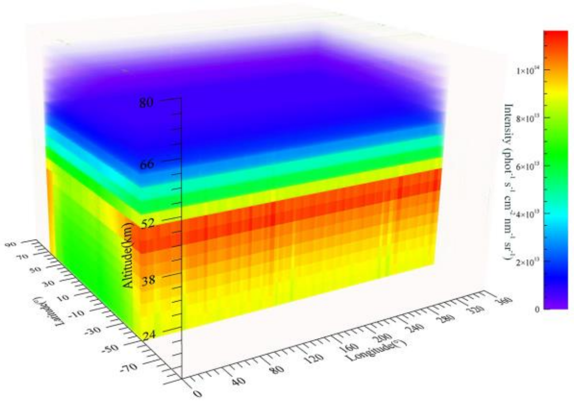

Figure 11. The simulated observation radiance received by SHS2 at the time of T2 under the different conditions

There are some differences between the Fig. 10 and the Fig. 11 because of the observed geometries and satellite positions. However, the whole observed radiance trends are same. The observed radiance increases from 8×10^{13} phot^{-1} s^{-1} cm^{-2} nm^{-1} sr^{-1} to 1.4×10^{14} phot^{-1} s^{-1} cm^{-2} nm^{-1} sr^{-1} until the altitude around 45 km and decreases to about 4×10^{13} phot^{-1} s^{-1} cm^{-2} nm^{-1} sr^{-1} with the altitudes increase. The lowest observed radiance is in the region around the 30 °N and gradually increases towards to the two poles in the altitude of 41 km. The reason for this trend is that the solar zenith angle is the minimum value around the 30 °N, and then increases towards to the two poles. At the altitude of 71 km, the observed geometries are not the only influencing factor and the distributions of OH concentrations are another one. The Rayleigh scattered radiance is weak and OH fluorescence emission radiance becomes strong. In addition to this one, the OH concentrations at the first quarter are low in the region around 60 °N that lead to the high radiance values appear in the southern hemisphere.

4 Inversion modeling

4.1 Interferogram pretreatments

The method of forming the interferogram by the spatial heterodyne spectroscopy is the spatial heterodyne modulation. The signal-to-noise ratio is susceptible to many factors, and the fine spectral lines of noisy signal can also be detected simultaneously. The interferogram needs to be pretreated for solving these problems and transformed to obtain the OH fluorescence emission radiance. The whole pretreatments are shown in the Fig. 12.

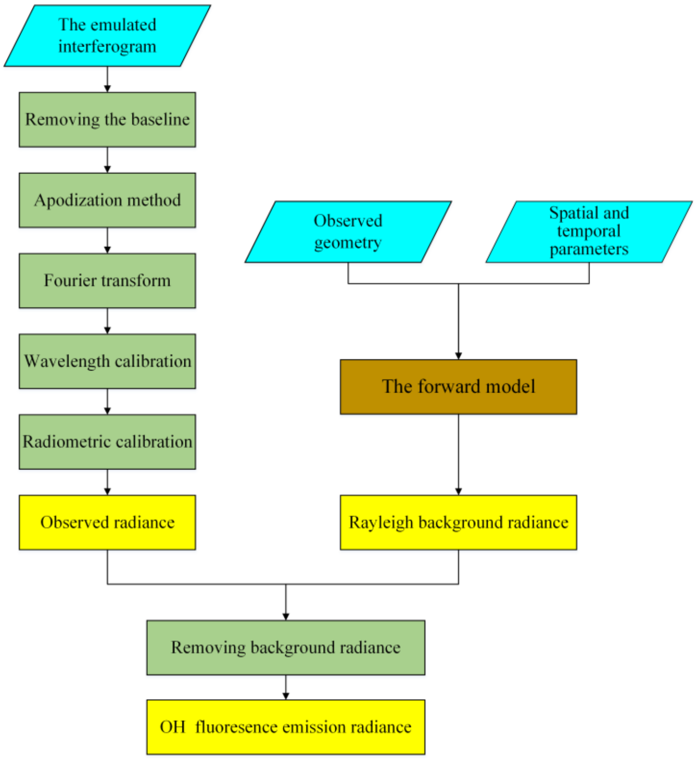

Figure 12. Flow chart of the interferogram pretreatments.

First, the interference data with low frequency baseline will make low frequency spurious signals in the process of the Fourier spectrum transform. The methods of removing the baseline mainly include the polynomial linear fitting to baseline, the first-order differential de-baseline at present. The function of first-order differential de-baseline method is the high-pass filtering. It is the most useful way to remove the baseline (Song et al., 2009). The interferogram obtained by the spatial heterodyne spectrometer is the interference data in the interval of finite optical path difference from -L to +L. That means an interferometric function forces a sudden drop to zero outside of this range. It will cause the interferogram to appear sharp discontinuities in the edge interval. The spectral profile has side lobes which the positive side lobes are the source of the false signal, and the negative side lobes will lead to the adjacent weak spectral signals to be submerged. The apodization method is used to mitigate the discontinuity of interferogram edge through multiplying the interferogram function by a progressive weight function. The interferogram after apodization is subjected to the Fourier spectrum transform for obtaining the corresponding spectrum. However, the high-spectral resolution observed radiance data which is simulated by the forward model mentioned above includes the atmospheric background radiance which must be identified and removed from the observed radiance data before the OH fluorescence emission radiance is obtained because: (1) it accounts for more than 95% of observed radiance that will make OH fluorescence emission radiance low, (2) some errors of atmospheric background radiance will transfer into the OH fluorescence emission radiance when the observed radiance of the interferogram form is recovered to the spectral form. The intensity of simulated observation radiance and atmospheric background radiance at some tangent heights are given in the Table 1. The OH fluorescence emission radiance can be calculated by subtracting the atmospheric background radiance from the observed radiance. The OH fluorescence emission radiance graph which is shown in the Fig. 13 can obtain after these steps and the Fourier spectrum transform can be done.

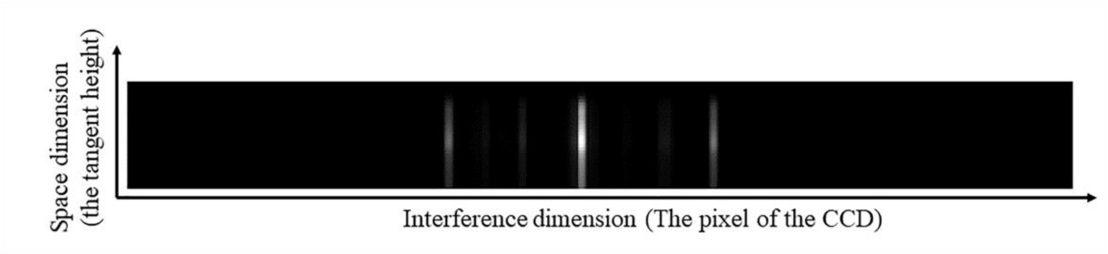

Figure 13. Graph of OH fluorescence emission radiance.

The fifth step is the wavelength calibration. The zero position of the spectral data points corresponds to the Littrow wavelength number in the Fourier transform spectrum, so the wavelength calibration equation is given by the Eq. (5):

$$\delta_i = -1.3166574i + 32679.738 \quad (5)$$

where i is from 0 to 1024. The δ_i is the symbol of intensity and the unit of it is phot^{-1}s^{-1}cm^{-2}nm^{-1}sr^{-1}.The sixth step is the radiometric calibration. The purpose of the radiometric calibration is to determine the quantitative relationship between the output signal of instrument and the spectral radiance because the spectrometer obtains the DN values directly. A liner fit method is used to simulate the different sets of known radiance values at each spectral data point for establishing a fitting relationship in the calibration process by Eq. (6):

$$S_\delta = B_\delta \cdot K_\delta + \varepsilon_\delta \quad (6)$$

where S_δ is the DN values at the wavelength number δ. B_δ is the assumed incident spectrum radiance. K_δ is the calibration factor and ε_δ is the deviation caused by other factors. A spectrum of OH fluorescence emission radiance is shown in the Fig. 14.

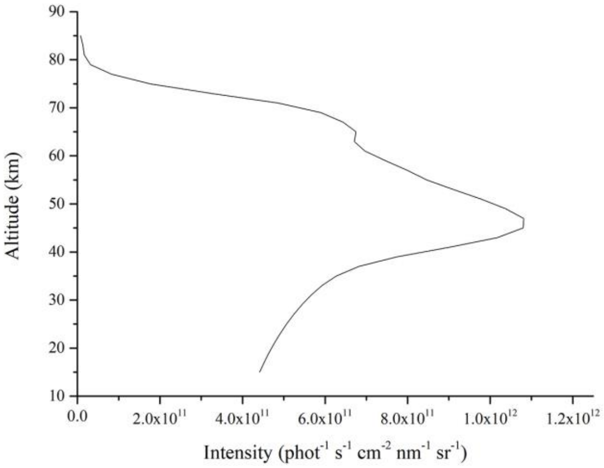

Figure 14. OH fluorescence emission radiance spectrum extracted from observed radiance.

4.2 Constructions of the retrieval algorithm

It is an inversion process to obtain the OH concentrations by the DSHS observed radiance. The observed radiance is simulated through the forward model in this research. However, it cannot be done to obtain OH concentrations in an inverse way directly. The atmospheric inversion problems are not an inversion of the forward model but use some related algorithms to estimate the atmospheric parameters for finding the best state parameters with the observed radiance data.

The DSHS consists by two spatial heterodyne spectrometers in the orthogonal layout for monitoring the OH from 15 to 85 km. It will provide large and complicated data. The traditional iterative retrieval algorithm will take more time to obtain the OH concentrations in this case because of the character of iteration. A new retrieval algorithm which is suitable for the DSHS needs to be constructed for inverting the accurate OH concentrations.

The precondition of the three-dimensional limb mode for monitoring the OH is the time interval must be relatively short, otherwise the OH concentrations will change, and the observed data cannot reflect the accurate OH concentrations. The distance between the satellite positions at the time of T1 and T2 is approximately 66 km when the intersection point is assumed as 50 km away from the tangent point mentioned in the Sect. 2. So, the intervals between those points are very small. Additionally, the OH concentrations do not change much in a very short time according to MLS products. Short interval time between the time of T1 and T2 and small change of OH concentrations in a very short time support the theoretical premise of the new retrieval algorithm and make DSHS possible to detect atmosphere through the three-dimensional limb mode. An algorithm called tomographic retrieval algorithm is constructed to invert OH concentrations from 15 to 85 km based on the theory above. The core of this retrieval algorithm is look up table method. The most important step for it is the construction of the tomographic observed database. The main influencing factors for making up the tomographic observed database contain the atmospheric model, the observation geometries (the solar zenith angle and the relative azimuth angle), the spatial information (position of the target area and satellite position) and date parameters, the OH concentrations and the simulated observation data. The solar zenith angle and the relative azimuth angle have a greater influence on the observed radiance. The spatial information and date parameters will influence the OH concentrations and the atmospheric model. The solar zenith angle is changed between 0° to 100° and the relative azimuth angle is changed between 0° to 180°. The corresponding OH concentrations are found out according to the date parameters and spatial information such as the season, latitude and longitude of the target area from the OH concentrations database which has been built up in the Sect.3. The OH concentrations are changed in the ranges between 50% to 150% times amount of the original corresponding one at different tangent height for more possible situations. Many combinations of various state parameters can be generated according to these changes. The observed data received by DSHS in these combinations is simulated through the forward model mentioned above. The tomographic observed database can be built up for the tomographic retrieval algorithm. The each parameters which constitute the database are accurate and the errors of them are analyzed in the Table 3. The method of constructing the database is reasonable. The flow chart is shown in the Fig. 15 and a subset of rows of the tomographic observed database is given in the Table 5 which the SZA is the solar zenith angle, the AA is the relative azimuth angle, the MultiND_height is the tangent height.

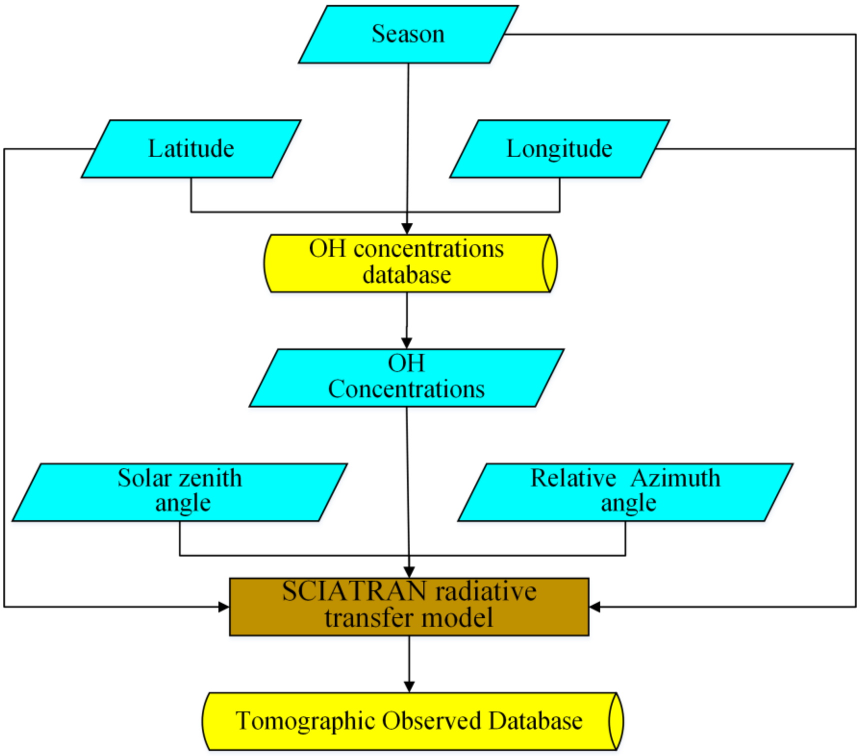

Figure 15. Construction of the tomographic observed database.

Table 5. A subset of rows of the tomographic observed database

N	Latitude (°)	Longitude (°)	SZA (°)	AA (°)	MultiND_height (km)	Season
1	54.240	125	0	0	15	1
2	54.240	125	0	0	15	2
3	54.240	125	0	0	15	3

4	54.240	125	0	0	15	4
5	54.240	125	0	0	17	1
6	54.240	125	0	0	17	2
7	54.240	125	0	0	17	3

Based on the satellite positions, the observation geometries, the date parameters, the observed radiance at all wavelengths of SHS1 at the time of T1 and the corresponding observed radiance at all wavelengths of SHS2 at the time of T2, the OH concentrations in the target area can be found in the tomographic observed database directly. If there is no corresponding query condition in the database, the cubic spline interpolation is used to calculate the OH concentrations. The cubic spline interpolation not only has the higher stability than other interpolation algorithms, but also can maintain the continuity and smoothness of the interpolation function under the premise of convergence. Figure 16 shows the flow chart that the OH concentrations obtain by the tomographic retrieval algorithm.

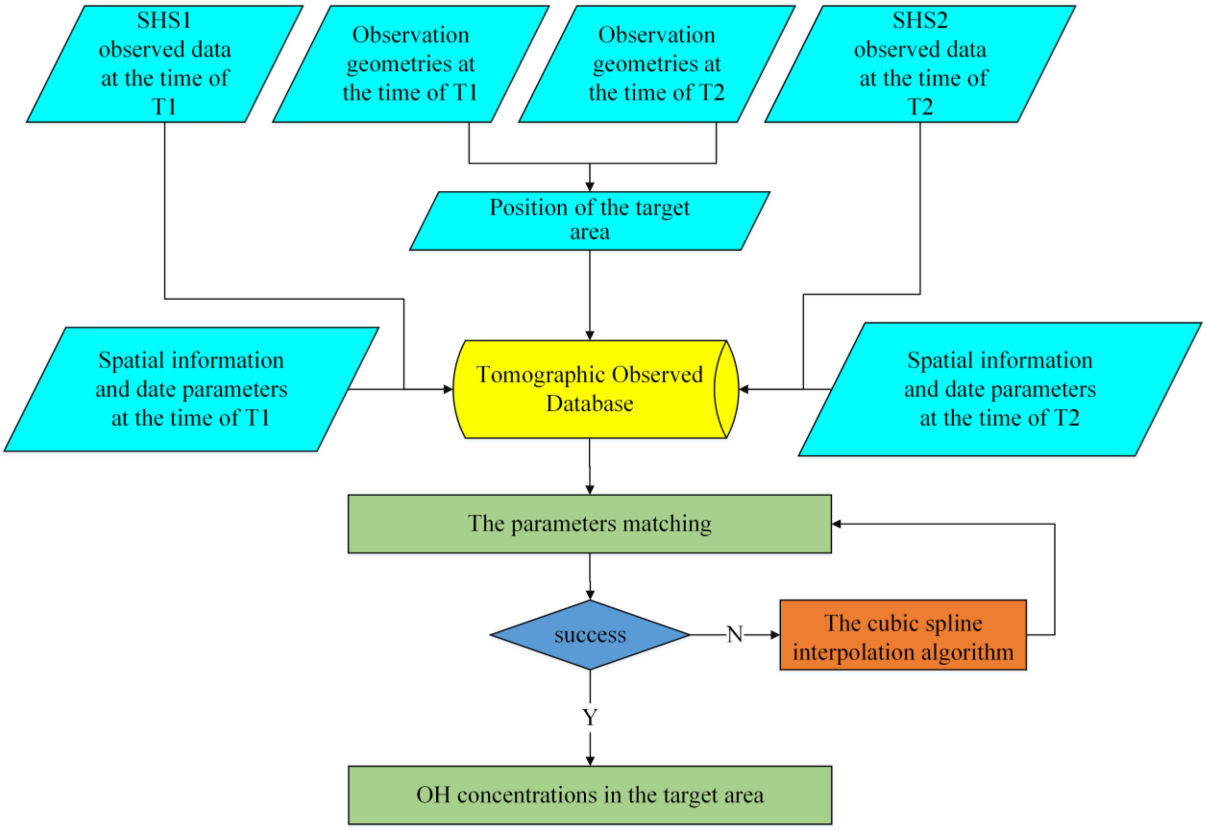

Figure 16. Flow chart of the tomographic retrieval algorithm. The OH concentrations can be obtained from the tomographic observed database or by the cubic spline interpolation algorithm.

5 Results and discussion

5.1 Inversion results

There are four tomographic observed databases based on the four seasons. The profile data in the database is averaged to obtain the monthly average concentration profile data. So, it will not introduce discontinuities to the retrieved OH time series when an abrupt switch happens. Here, the spring season tomographic observed database is used as an example to prove the feasibility and superiority of the tomographic retrieval algorithm. The OH concentrations are obtained in the (67.46 °N, 86.168 °E) through satellite positions, observation geometries, date parameters which are given by the Table 5. If there is a corresponding query condition, the OH concentrations can be given directly from the tomographic observed database, otherwise the OH concentrations will be calculated by the cubic spline interpolation. The inversion results are shown in the Fig. 17.

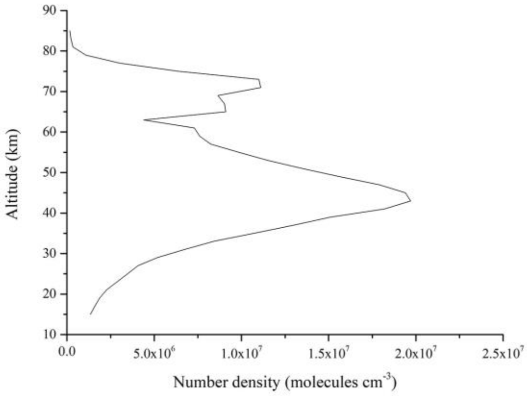

Figure 17. Inversion results in the target area using the tomographic retrieval algorithm.

The results indicate that a maximum peak value of OH concentrations are about 2.0×10^{7} molecules cm^{-3} appear around the 45 km height as the heights increase which closely relates to the ozone concentrations in the stratopause. A valley value of OH concentrations about 5.0×10^{6} molecules cm^{-3} appear around 65 km. As the height continues to rise, the second maximum peak value of OH concentrations about 1.0×10^{7} molecules cm^{-3} appears around the 70 km height which is mainly affected by the water vapor concentrations in the mesosphere. The OH concentrations above the 75 km height and below the 25 km is very low (about 2.5×10^{6} molecules cm^{-3}). In general, the OH concentrations increase with the altitudes rise until around 40 km height and reach the valley value around 65 km. The second maximum value peak which is affected by the water vapor concentrations appears around 75 km, then the OH concentrations continue to decrease until the limited altitudes of detection.

5.2 Discussion

We also constructed an iterative retrieval algorithm to compare with the tomographic retrieval algorithm here for reflecting the outstanding advantages of tomographic retrieval algorithm. The schematic diagram of iterative inversion is shown in the Fig. 18. The simulated radiance can be obtained under the condition which an initial OH concentrations guess can get from the OH concentrations database. The residuals of the simulated and observed data will be calculated to judge the convergence. The initial OH concentrations guess values are the inversion results if the residual meets the accurate requirements of research. Otherwise, the retrieval algorithm will be used to obtain the inversion results.

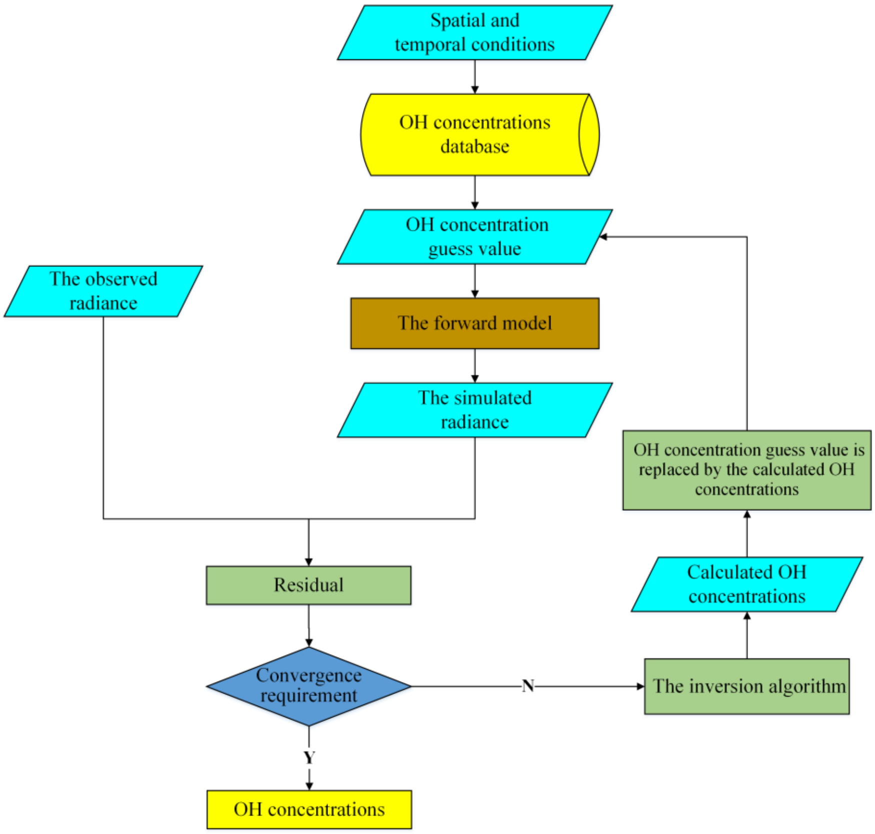

Figure 18. Schematic diagram of iterative inversion.

The LSUV (Limb scan of Scattered UV radiation) retrieval algorithm (Aruga and Heath, 1988;Aruga and Igarashi, 1976) is modified to invert OH concentrations at each tangent height. The tangent height is defined as *m* and the target area is divided into *n* atmospheric layers from the bottom to the top. One-order approximate Taylor expansion is used to establish a linear relationship between the OH concentrations and observed radiance at each atmospheric layer by Eq. (7):

$$F_i = [\sum_{j=1}^{n} D_{ij} \cdot y_j]\varepsilon_i \qquad \begin{cases} i = 1, \dots, n \\ j = 1, \dots m \end{cases} \tag{7}$$

where ε_i ,the inversion coefficient, is approximately 1 when the OH concentrations are closed to the guess OH concentrations. The F_i is the difference between the observed data and simulated data by Eq. (8) at the *i* layer:

$$F_i = I(h_i) - I_s(h_i), i = 1,2, \ldots, m$$

(8)

The D_{ij} which is given by Eq. (9) is the partial derivative of radiance data at the *i* tangent height:

$$D_{ij} = \left[\frac{\partial F_i(y_j)}{\partial y_j}\right]_{y_j=0} = \frac{I_s{}^*\left(h_i,\frac{\Delta y_j}{2}\right)-I_s{}^*(h_i,-\frac{\Delta y_j}{2})}{\Delta y_j}$$

(9)

The y_j in the Eq. (10) is relative error between the OH concentrations $N(j)$,which can get though the y_j and $N_g(j)$ when the converges meet the accurate requirement or the number of iterations exceeds the iteration number threshold, and the guess concentrations $N_g(j)$:

$$y_j = \frac{[N(j)-N_g(j)]}{N_g(j)} \quad j = 1,2, \ldots, n$$

(10)

The approximate OH concentrations at each tangent height $(Q)_i$ can be got based on the Eq. (10) through the Eq. (11):

$$(Q)_i = \frac{F_i}{\sum_{j=1}^{n} D_{ij}}$$

(11)

The approximate OH concentrations $(Q)_i$ at *i* tangent height is determined by the OH concentrations on several atmospheres near the *i*. The weight function P_{ij} is established from the D_{ij} by Eq. (12):

$$P_{ij} = \frac{D_{ij}}{\sum_{j=1}^{n} D_{ij}}$$

(12)

where P_{ij} is the proportion of the contribution of OH concentrations on the radiance of each tangent height in each layer. The accurate relative errors of the OH concentrations and guess OH concentrations in each layer y_j can be obtained from P_{ij} by Eq. (13):

$$y_j = \frac{\sum_{i=1}^{m} Q_j \cdot P_{ij}}{\sum_{i=1}^{m} \cdot P_{ij}}$$

(13)

However, the difference between the OH concentrations and guess OH concentrations is large in the actual satellite detection, the ε_i is not 1, the ${\varepsilon_i}^*$ is used to replace it by Eq. (14):

$${\varepsilon_i}^* = \frac{F_i({y_1}^*, {y_2}^*, \ldots, {y_n}^*)}{\sum_{j=1}^{n} D_{ij} {y_j}^*}$$

(14)

Finally, the precise relative error y_j between the OH concentrations and the guess OH concentrations can be obtained. The OH concentrations $N(j)$ can get though the y_j and $N_g(j)$. The convergence error R_i^l between the observed data $I(h_i)$ and simulated data $I_s(h_i)$ at *i* layer which based on the guess OH concentrations $N_g(j)$ is calculated by Eq. (15):

$$R_i^l = \frac{I(h_i) - {I_s}^{(l)}(h_i)}{{I_s}^{(l)}(h_i)} = \frac{F_i^{(l)}}{{I_s}^{(l)}(h_i)}$$

(15)

where l is the number of iterations. The average residual R^l based on the R_i^l at different tangent heights can be calculated by Eq. (16):

$$R^l = \sqrt{\frac{\sum_{i=1}^{m} \{R_i^l\}^2}{m}}$$

(16)

The threshold of average residual and the numbers of iteration are set based on the inversion experience. The iteration result which meets the precision requirement is the final result, otherwise the prior $N_g(j)$ will be replaced by the $N(j)$ which is calculated by the inversion algorithm and the iteration process will continue until the residual converges meet the accurate requirement or the number of iterations exceeds the iteration number threshold. The iteration stops when the average residual decreases below 0.005 or the number of iterations reaches 15 times in this research.

The observed radiance which is simulated by the forward model are used by the LSUV retrieval algorithm to invert OH concentrations. When the iteration results meet the accurate requirements of research, the OH concentrations can be obtained. The OH concentrations obtained by the LSUV retrieval algorithm are credible in the upper atmosphere which are the same as the results of MAHSRI and SHIMMER (Conway et al., 1999;Englert et al., 2010). However, the OH concentrations in the lower atmosphere such as below 30 km are unsuitable for scientific research because the interference factors like water vapor and ozone are too much. A single spatial heterodyne spectrometer with traditional limb mode cannot also obtain enough and high-quality data to invert the OH concentrations that will make the inversion results unscientific especially in the lower atmosphere. That is the reason why there are no OH concentrations in these regions from the MAHRSI and SHIMMER (Harlander et al., 2002).

The tomographic retrieval algorithm and the LSUV are two independent algorithms. The tomographic retrieval algorithm effectively avoids the constraints of the initial guess values and does not take a lot of time to iterate for obtaining the results in the inversion progress. The errors of this retrieval algorithm just include the errors caused by the interpolation method and the observed radiance errors which are used in this algorithm. The observed radiance errors are affected by the atmospheric model, the Doppler effect and the instrument calibration errors. The OH fluorescence emission radiance which is separated from the observed radiance is the most important part for the OH concentrations, so the contributions of each part to the OH fluorescence emission radiance is analyzed by the following method: 1) some certain amounts as the disturbances are applied to the three parts above. The OH fluorescence emission radiance which obtained based on these parts is considered as the incorrect results. 2) the original parts are taken as the actual and undisturbed parameters, and the results based on these parts are the true OH fluorescence emission radiance. The relative errors between the true and incorrect OH fluorescence emission radiance is calculated by the Eq. (17):

$$Err = \frac{[I]_{err} - [I]_{act}}{[I]_{act}} \times 100\%$$

(17)

where the $[I]_{err}$ is the incorrect OH fluorescence emission radiance and the $[I]_{act}$ is the true OH fluorescence emission radiance.

5.2.1 Influence of the atmospheric model

The atmosphere environment is complicated. Although, the atmospheric model used in this research is scientific and reasonable, it cannot reflect the accurate atmospheric condition which leads to some errors between true OH fluorescence emission radiance and incorrect OH fluorescence emission radiance. These will cause an error when the OH concentrations are looked up or calculated. Many factors can affect the OH fluorescence emission radiance in the atmospheric model such as some physical parameters like temperature, pressure and some atmospheric composition like nitrogen dioxide and formaldehyde. Here, the main composition we consider is ozone which is the source of OH and mainly affects the radiance transmission in the ultraviolet band. The original ozone profiles are assumed as the actual atmospheric state parameter and the amount of ozone profile for the disturbances is set as ±30%. The relative errors of the OH fluorescence emission radiance caused by the atmospheric model is calculated by Eq. (17) and are shown in the Fig. 19. The relative errors of the OH fluorescence emission radiance at some tangent height are also given in the Table 6.

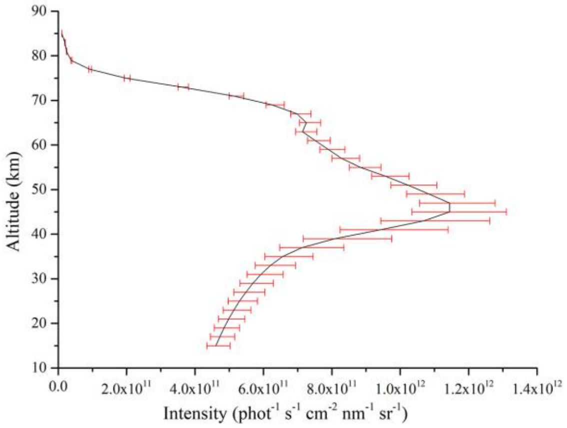

Figure 19. Relative errors of OH fluorescence emission radiance caused by the atmospheric model in the tomographic retrieval algorithm. The red error bars indicate the relative errors caused by the atmospheric model.

Table 6. Relative errors of OH fluorescence emission radiance caused by the atmospheric model at some tangent heights

Tangent height (km)	Relative errors caused by the atmospheric model (%)
81	From -2.71 to 5.72
71	From -2.72 to 5.75
61	From -2.96 to 5.92
51	From -4.92 to 8.19
41	From -12.57 to 20.92
31	From -6.78 to 10.98
21	From -6.00 to 9.51

The relative errors caused by the atmospheric model indicate that the atmospheric model have a strong impact on the OH fluorescence emission radiance especially in the lower atmosphere where the ozone concentrations are high. The errors caused by ±30% uncertainty of the atmospheric model have a peak value from -12.57 % to 20.92 % at the altitude of 41 km. It also appears in the LSUV retrieval algorithm by the same calculation method which is from -12.19 % to 20.28 %. The concentrations of ozone gradually decrease with the relative errors decreasing to about -3% to 5% when the heights increase. The effect the of atmospheric model is smaller in the upper atmosphere. The distribution of errors has the same tendency in the LSUV retrieval algorithm that the relative errors increase and then gradually decrease after reaching the peak value altitude. These mean the relative errors caused by the atmospheric model are inevitable. If the more actual atmospheric model is used, the more accurate OH fluorescence emission radiance may be obtained which is the key for the retrieval algorithm.

5.2.2 Influence of the Doppler effect

The satellite has the different heliocentric speeds on its working orbit around the earth at different times. It will cause a pseudo-random offset of observed data within ±5×10^{-4} nm at the range of wavelength. The observed radiance is calculated within ±5×10^{-4} nm disturbance at the wavelength. The OH fluorescence emission radiance which is separated from the observed radiance is too weak. A little Doppler effect will cause a great influence on the weak OH fluorescence emission radiance. The magnitude of wavelength shift caused by the Doppler effect is much smaller than the spectral resolution. “Finding peak” is used to find a peak value which needs to be larger than the values on its sides. It is impossible to correct the offset by “Finding Peak” method. However, “Taking peak” is defined to find the highest peak value in the OH fluorescence emission radiance. The “Taking Peak” is used for reducing the impact of the Doppler effect. Therefore, the OH fluorescence emission radiance at the peak position is used instead of the wavelength from 308 nm to 310 nm when calculating the relative errors caused by the Doppler effect at a single tangent height. The relative errors of the OH fluorescence emission radiance caused by the Doppler effect is calculated by Eq. (17) and are shown in the Fig. 20. The relative errors of OH fluorescence emission radiance caused by the Doppler effect at some tangent heights are also given in the Table 7.

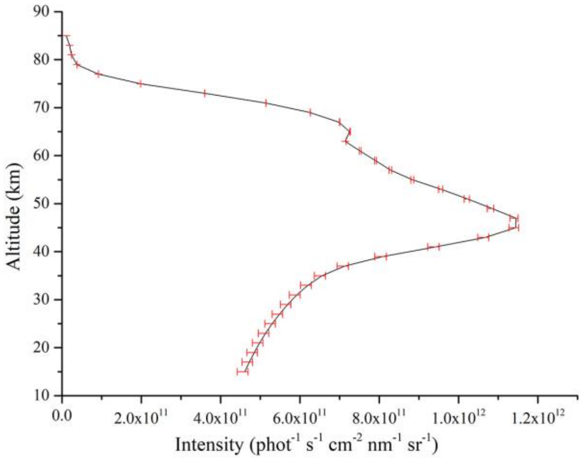

Figure 20. Relative errors of OH fluorescence emission radiance caused by the Doppler effect in tomographic retrieval algorithm. The red error bars indicate the relative errors caused by the Doppler effect.

Table 7. Relative errors of OH fluorescence emission radiance caused by the Doppler effect at some tangent heights

Tangent height (km)	Relative errors caused by the Doppler effect (%)
81	From -0.39 to 0.35

71	From -0.20 to 0.31
61	From -0.11 to 0.34
51	From -0.88 to 0.41
41	From -2.16 to 0.93
31	From -3.14 to 1.44
21	From -3.71 to 1.62

The relative errors of OH fluorescence emission radiance caused by the Doppler effect decrease when the altitudes rise. The tendency of relative errors is same, but the relative errors of tomographic retrieval algorithm are smaller than LSUV retrieval algorithm. These indicate that tomographic retrieval algorithm can obtain more accurate OH concentrations than the LSUV retrieval algorithm although the errors caused by Doppler effect cannot avoid when the spectral resolution of DSHS is extremely high.

5.2.3 Influence of other factors

There are some other errors exist apart from the errors above. For one thing, there are various sources make some errors between the radiance received by the sensors and the actual radiance in the instrument calibration process. The instrument calibration errors will cause an error of the inversion result. We assumed the instrument calibration errors are ±5% in this research after referring to the detector parameters due to the DSHS do not officially work (Song et al., 2009). For another, the error caused by the interpolation method is the main factor we need to calculate. The cubic spline interpolation method not only has higher stability but also can ensure the continuity and smoothness of the interpolation function under the premise of the ensuring the convergence compared with other interpolation methods like linear interpolation method and some others. The errors estimation of the interpolation results is as accurate as possible. The OH concentrations can be obtained by the tomographic retrieval algorithm according to the atmospheric model, spatial information, date parameters, observation geometries and observed radiance. The interpolation errors are also the other error factors which effect the accuracy of OH concentrations. We give a certain disturbance to different parameters to calculate the effect of the interpolation algorithm on the OH concentrations. It is about 0.32% for the six-dimensional cubic spline interpolation of OH concentrations, date parameters, longitude, latitude, solar zenith angle and azimuth angle after research and related experiment. Therefore, 0.32% is taken as the error of the interpolation algorithm in the tomographic retrieval algorithm.

5.2.4 Analysis of total errors in the OH concentrations

The errors of retrieved results must be considered after the discussions about the errors of OH radiance. The theoretical basis for calculating the total errors of inversion results is the error transfer formula which is always used to calculate the indirect measurement error (Hong et al., 2009).The total errors based on a single error factor often have an asymmetric distribution. The corresponding standard error according to the single each error factor should be calculated if error transfer formula is used. It is calculated according to the B-class standard uncertainty evaluation method. The relationship between input parameters and results can be expressed by the Eq. (18):

$$[I] = f(x_1, x_2, x_3, \dots, x_n)$$
(18)

where $x_1, x_2, x_3, \dots, x_n$ are the input parameters and $[I]$ indicates the inversion results. Equation (19) can be got when the error Δx_i is considered and the does a Taylor expansion:

$$[I] + \Delta[I] = f(x_1, x_2, x_3, \dots, x_n) + \sum_{i=1}^{n} \frac{\partial f}{\partial x_i} \Delta x_i$$

(19)

The maximum value of relative error can be calculated by the Eq. (20):

$$\frac{\Delta[I]}{[I]} = \sum_{i=1}^{n} \left| \frac{\partial f}{\partial x_i} \right| \frac{\Delta x_i}{[I]}$$
(20)

where the right part of equal sign means the errors caused by $x_1, x_2, x_3, \dots, x_n$. The error transfer formula can be derived from the Eq. (20) when the errors are squared. The total errors RSS can be calculated by the "square root" method with the Eq. (21):

$$RSS = \sqrt{\sum_{i=1}^{n} \left\{ \frac{\partial[I]}{x_i} \frac{\Delta x_i}{[I]} \right\}^2}$$
(21)

The total errors of the results caused by each factor can be obtained by the Eq. (21). The total errors of the inversion results caused by each error factor are calculated which are shown in the Fig. 21 and the total errors of inversion results at some tangent heights are given in the Table 8.

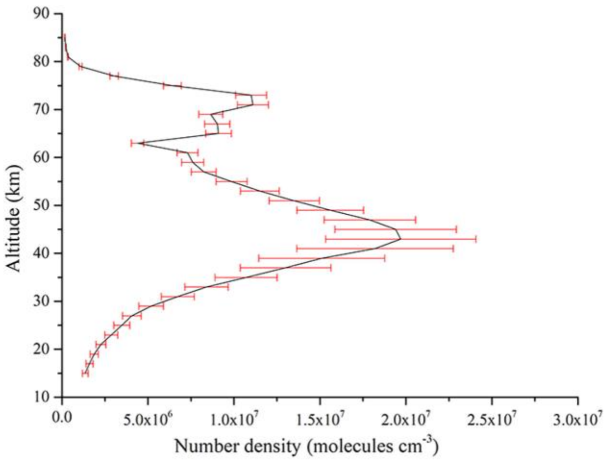

Figure 21. Total errors of inversion results caused by some factors mentioned above. The red error

bars indicate the relative errors caused by some factors mentioned above.

Table 8. Total errors of inversion results at some tangent heights

Tangent height (km)	Total errors of inversion results (%)
81	From -8.09 to 8.09
71	From -8.11 to 8.11
61	From -8.31 to 8.31
51	From -10.83 to 10.83
41	From -25.03 to 25.03
31	From -14.27 to 14.27
21	From -12.96 to 12.96

As Table 8 indicates, the total errors of OH concentrations initially increase with the heights rising. It reaches the maximum value range from -25.03 % to 25.03 % at the 41 km height, and then decreases from -8.09 % to 8.09 % as the heights continue to rise until the limited height. The total errors of the inversion results in the lower atmosphere are small especially. However, the OH concentrations obtained by the LSUV algorithm in the same area are unsuitable for scientific research mentioned in the Sect. 5.2. It is a great improvement to compare with the results of the LSUV retrieval algorithm which has a characteristic of iteration. The relative errors of the OH concentrations obtained by LSUV algorithm are calculated by the Eq (17) and the error transfer formula mentioned above. The initial guess values have a great influence on the iterative speed and the inversion results for the iterative retrieval algorithm. A realistic initial guess value can achieve higher iteration efficiency and more accurate inversion results. The initial guess values used in the LSUV retrieval algorithm are obtained from the OH concentrations database through the date parameters and spatial information such as the season, latitude and longitude of the target area. The corresponding accuracy ranges and useful height ranges of MLS OH concentrations products are also given by the MLS data quality and description document. Therefore, the data accuracy is assigned at the upper limited height to altitude greater than the upper limit of MLS data altitude and at the lower limited height to the altitude smaller than the lower limit of MLS data altitude. The initial guess values can be obtained by using the data accuracy to perform the positive and negative turbulence for the initial OH concentrations values. A minimum positive value is given to the data less than zero for the physical meaning when a negative value may occur in the negative direction. The two initial guess values are used in the LSUV retrieval algorithm separately. Although, the initial guess values above are chosen scientifically and reasonably, the inversion results are unsuitable for scientific research in the lower atmosphere that the relative errors caused by the initial guess values reach from -95.39 % to 545.44 %. Besides the initial guess values, the inversion results are also affected through the errors caused by the atmospheric model, the Doppler effect and the instrument calibration error. The relative errors of inversion results obtained by the LSUV retrieval algorithm are small from 30 km to 85 km. The total errors of inversion results are from -24.83 % to 24.83 % in the altitude of 31 km and then decrease gradually until the altitude of 51 km. A little increase occurs from the altitude of 60 km to 70 km and reaches from -37.01 % to 37.01 % at the 81 km. However, the inversion results are bad in the lower atmosphere. The total errors of inversion results are from-553.16 % to 553.16 % in the altitude of 21 km. The total errors of inversion results are 2 times larger than tomographic retrieval algorithm at some heights like 71 and 61 km, however, the total errors of inversion results are 4.625 times larger than the errors of the tomographic retrieval algorithm at 21 km height especially. The application altitudes of the LSUV retrieval algorithm are limited. The main reasons lead to these unscientific inversion results are two parts. The OH fluorescence emission radiance, the main factor for the retrieval algorithm, is not strong enough due to the ozone optical depth becomes large in the lower atmosphere. That will lead to the observed radiance does not come from the tangent area but the near field. The lower atmospheric environment is complicated. The human activity will produce a lot of substances which will join in the atmospheric environment by the atmospheric convection motion. The aerosol also distributes here. These will lead to a bad detection accuracy. So, the useful altitudes of the LSUV retrieval algorithm based on the data of single spatial heterodyne spectrometer are limited. The DSHS can counteract the errors by the two spatial heterodyne spectrometers because of its special optical design which the inversion results of retrieval algorithm have proved. These analyses above also indicate that the tomographic retrieval algorithm is a feasible retrieval algorithm for the atmosphere from 15 to 85 km. The look up table method avoids the intervention of initial guess values effectively by establishing a tomographic observed database of multi-dimensional variables and is understood easily. It also avoids the complicated iterative optimization process, and the OH concentrations can be obtained directly from the tomographic observed database. These make the speed of inversion process fast compared with the LSUV retrieval algorithm. The tomographic retrieval algorithm is a suitable retrieval algorithm for the three-dimensional limb mode of DSHS as well. The three-dimensional limb mode provides big and numerous observed data. The speed of traditional iterative retrieval algorithms is slow and will cost a lot of time. However, it is difficult to quantify how much fast is the tomographic algorithm absolutely. The traditional iterative retrieval algorithm will take several or even tens of hours to obtain OH concentrations due to the complex iterative process. The tomographic algorithm just needs minutes for obtaining the OH concentration due to the usage of the lookup table method. The results obtained by the traditional iterative retrieval algorithm in the lower atmosphere are unscientific but the OH concentrations obtained by tomographic algorithm in the same area are accurate. The tomographic retrieval algorithm solves these problems and improves the efficiency of retrieval algorithm which is important for the OH concentrations. The LSUV retrieval algorithm can invert OH concentrations well under the conditions that the OH fluorescence emission radiance is strong and the interference factors like water vapor and ozone are little. However, a single spatial heterodyne spectrometer with traditional limb mode cannot obtain enough and high-quality data to invert the OH concentrations that will make the inversion results unscientific especially in the lower atmosphere. The initial guess values play a significant role in the LSUV retrieval algorithm due to the feature of the iteration. The determination of the initial guess values involves many factors and there is no best way to determine the initial guess values at present. This feature is particularly pronounced at the lower atmosphere where OH fluorescence emission radiance is less sensitive to OH concentrations.

In summary, the LSUV retrieval algorithm based on the iterative method can invert the OH concentrations well in the higher atmosphere like mesosphere but cannot obtain the accurate OH concentrations in the lower atmosphere like the bottom of stratosphere. Many factors lead to this result like the atmospheric model, the limit of initial guess values and something else. The tomographic retrieval algorithm using the look up table method can obtain the accurate OH concentrations from stratosphere to the mesopause with the feature of the three-dimensional limb mode. This retrieval algorithm not only saves the time of inversion process, but also avoids the problem that the OH concentrations are unsuitable for scientific research in the lower atmosphere.

6 Conclusions

The OH is the key oxidant in the atmosphere and has a great influence on the atmospheric photochemistry process. The DSHS based on the spatial heterodyne spectroscopy will monitor the OH with the three-dimensional limb mode in the future. A forward model is constructed to simulate the observed data of DSHS accurately. A new retrieval algorithm for obtaining the OH concentrations is also proposed based on the simulations of the forward model. The distinctive features of this algorithm are the usage of a look up table method. The MLS OH concentrations products and N32 Gaussian grid are used to construct the OH concentrations database of four seasons. The observed radiance is obtained by the forward model. The other factors like spatial information and observation geometries are also simulated according to the characteristic of DSHS for the tomographic retrieval algorithm. The tomographic observed database is established based on the parameters above. The OH concentrations in the target area are obtained through finding in the tomographic observed database directly. The cubic spline interpolation method is also used to obtain the OH concentrations without the corresponding query conditions. The errors are also analyzed caused by the atmospheric mode, the instrument calibration error, the Doppler effect and interpolation algorithm. The results show that the tomographic retrieval algorithm is not only faster compared with the LSUV retrieval algorithm, but also improves the inversion precision of the OH concentrations especially in the lower atmosphere where the OH concentrations are sensitive.

There are still some problems to be solved when the DSHS will officially work in the future. The performances of two spatial heterodyne spectrometers are inevitably different because they are affected by the temperature, humidity and electromagnetic environment during manufacturing although the bi-orthogonal structure is theoretically identical in the design parameters. The instrument calibration errors of the instrument are only considered to be 5% at present. This part will be optimized according to the actual working condition after the instrument is officially working. The actual observed data is the key point for this research. Some aircraft flights experiments have been done and the data is being analyzed. The DSHS and research results have been communicated with the related departments. It has been included in the relevant satellite projects for 2020 to 2030.

Code/Data availability. All code and data can be obtained from the corresponding author upon request.

Author contribution. YA and JM designed the study. YA and GB performed the simulations and carried out the data analysis. JM, WX, and XW provided useful comments on the paper. YA prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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