Response to Reviewer Comments

We wish to thank the reviewer for his careful comments. We have addressed these comments in the revised manuscript, including the reviewer's high-level comments:

- We have removed US-centric discussions throughout the manuscript.
- We now discuss the shortcomings of various datasets to address the concern that not all satellite data products are of equal quality.
- We have clarified the overall focus of the manuscript.

The reviewer's comments are in black and our responses are in red.

Reviewer: Maarten Krol

This opinion paper communicates views on an important subject: the use of current and future satellite data to constrain global troposphere OH. Given the central role of OH in removing pollution (including CH_4), this is an important opinion paper that aims to advise the upcoming 2027-2037 Earth Science Decadal Survey (ESDS) for the National Aeronautics and Space Administration (NASA).

The paper contains valuable views. However, the paper is rather wordy (saying sensible things), and it would help to provide some more structural elements like tables listing available and future missions. The figures presented in the paper seem to merely advertise personal work, rather than helping the reader to grasp the research agenda concerning OH-proxies.

Scattered around the paper, different observing strategies are mentioned (Polar orbiting, Geostationary, UV/vis, NIR, IR, lighting observations, etc.). Here it would really help to provide a table of existing strategies to monitor OH proxies, right at the beginning of the paper.

In that sense, the order is rather odd. I would expect first an overview of the role of OH in tropospheric chemistry, and historic developments e.g (Lelieveld et al., 2004, 2006). Currently, some of this information is provided later in the paper (in the "thought experiment"). This order might be OK for an opinion paper, but I was distracted by the lack of proper introduction of the subject.

The focus of this opinion piece is on a new area of research that involves using satellite data to constrain spatio-temporal variations on OH, which has not been achieved before. The focus is not to serve as an overview of tropospheric OH or of satellite datasets. In that vein, the introduction is devoted to discussing the strengths and limitations of non-satellite methods to constrain OH and to highlight the potential of satellite-based methods. To help make this distinction clearer, we added the following sentence to the end of the first paragraph of the introduction:

> "The reader is referred to, for instance, Lelieveld et al. (2016) and Fiore et al. (2024) for comprehensive overviews of the importance of OH in tropospheric chemistry as well as discussions of key uncertainties in its global chemical budgets and estimates of past global trends and variations of its atmospheric abundance."

Table 1 summarizes important instruments for data continuity of various observables. We focus on satellite instruments that provide near complete spatial coverage of the troposphere, neglecting instruments that provide data of, for instance, the upper troposphere (e.g., MLS) and geostationary orbits. Therefore, we do not discuss every potential instrument, just the ones that provide the best quality data and near global coverage. We do not list potential future instruments given that space agency priorities and budgets change and because of the history of satellites failing to reach orbit during launch; we have clarified this in Table 1.

The figures that we show are illustrative and support our recommendations – they are not meant to advertise our own work.

- Figure 1 shows OH trends from one space-based approach simply to give the reader an idea of what the approach may provide.
- Figure 2 shows that there is potential of data product refinement through retrieval algorithm development.
- Figure 3 shows that additional research is necessary to assess whether or not a satellite proxy of HO_2 is required to constrain two of the sources of tropospheric OH (Table 1).
- Figure 4 shows that clear-sky/low cloud observations introduce a bias in the tropospheric OH estimate from satellite-based cases, indicating that methods need to be developed to account for OH in cloudy environments.

What is also a missing element in the paper is a thorough discussion about the quality of the products. Observing isoprene, H2O2, and formaldehyde is exiting, but using these quantitively is a different game. The same holds for tropospheric ozone. They author mention the 2006 Ziemke approach, and indeed since then not much happened to reliably determine tropospheric ozone. The reason is that it is simply a difficult problem. Although mentioned at places, the authors should be more specific what approaches are feasible to improve on the quality of the OH proxies. For tropospheric ozone, for instance, multi-wavelength satellite observations (IR, vis/Uv) could bring the scientific community further (there are existing studies in this field). As written now, the paper seems to argue that "all" mentioned proxies are of the same quality. Maybe adding a column in table one about the current accuracy would help to guide future needs for scientific research.

Yes, we agree that adding discussion on the quality of satellite data products is necessary. To address this, we modified the fourth column in Table 1, as suggested, to include additional information. Please see the revised table at the end of this response.

We also added text to Section 5.2.1:

> "An important caveat is that the satellite data products in Table 1 are not of equal quality and require additional research and/or technological development to improve their utility for indirectly constraining OH. A quantitative assessment of each data product's quality should be performed using independent suborbital observations when available (*Section 3*). Such an assessment would benefit from intercomparable uncertainty characterization between the different data products; however, this is not currently done. Therefore, *we recommend the use of a common set of reporting standards to be applied for uncertainty*

characterization of satellite data products (e.g., von Clarmann et al., 2020), which will allow for a more intercomparable assessment of the utility of each satellite data product for indirectly constraining tropospheric OH."

We also added the italicized text above as a recommendation in Section 6.

Concerning the comment about multi-wavelength satellite observations, the original text discusses the potential of these data in Section 4.4 and calls out the need for further research in the recommendations in Section 6.

I understand the US-central approach, since this paper aims to inform the 2027-2037 Earth Science Decadal Survey (yet, to be published in a European journal?). However, some developments in Europe could help the US developments, and have played a vital role.

Agreed. We have revised wording to be less US centric – to highlight that international cooperation has been integral to the development of the current global observing strategy and certainly will be true going forward. Here is specifically how we revised the text in the Introduction:

From:

"Our recommendations are intended to inform efforts to prioritize observational needs, including the upcoming 2027-2037 Earth Science Decadal Survey (ESDS) for the National Aeronautics and Space Administration (NASA), National Oceanic and Atmospheric Administration (NOAA) and United States Geological Survey (USGS) and the broader effort as proposed by Waliser & KISS Continuity Study Team (2024) "for a greater and more impactful US contribution to the global satellite observing system."

To:

"Our recommendations are intended to inform international efforts to prioritize observational needs."

Saying "international" includes all nations (i.e., beyond the US and Europe) with Earth-observing satellite programs.

We also removed the US-centric discussion of Terra, Aqua and Aura at the end of Section 5.2.2. The new text reads:

> "Continuity is assured for most satellite proxies (Table 1) and we currently have the ability to constrain adequately or partially most of the sources and sinks of tropospheric OH (Table 1) with the satellite-based approaches (Section 2) from late 1990s/early 2000s to the present. As discussed in Section 5.2.1, the primary exception is related to VOCs (e.g., isoprene, which became available only in 2012)."

As an example of the US-central approach of the paper, I would like to point to: As another example, discussion surrounding continuity of the NO2 VCD (Section 5.1.1) started with OMI (launched in 2004), though such observations actually began in 1996 with ESA's Global Ozone Monitoring Experiment.

Section 5.1.1, which is a case study for satellite data continuity, is focused on the afternoon-orbit satellites, which are largely supported by the U.S., except for OMI, which is a Dutch-Finnish contribution to the NASA Aura satellite mission. Section 5.2.2, which is an example of satellite data products for constraining past OH, is focused on the morning-orbit satellites, which are supported by European efforts. The relevant original text in Section 5.2.2 is:

> "As another example, discussion surrounding continuity of the NO_2 VCD (Section 5.1.1) started with OMI (launched in 2004), though such observations actually began in 1996 with ESA's Global Ozone Monitoring Experiment (GOME; 1995-2011; Burrows et al., 1999), which was followed by SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY; 2002-2012), GOME-2 instruments on the Meteorological Operational (METOP) satellites (METOP-A, 2006-2021; -B, since 2012; and -C, since 2018). All these ESA instruments were/are in morning orbits."

We added here a reference to Boersma et al. (2018) as the reviewer suggested.

Other European elements that are missing are the ESA efforts in the AQ4ECV program (e.g. for NO2, CH2O). Focus should be on synergy, e.g. with the developments of satellites in geostationary orbits (TEMPO, GEMS, S4).

In the original text, we discuss the potential of international geostationary satellites in Section 5.1.3 "Spatio-Temporal Coverage" for constraining tropospheric OH.

In the final recommendations I miss the further development of techniques (ML, data-assimilation, …) that help the interpretation of the data. Models are needed because they act as an integrating operator that moves around the longer-lived gases towards the "next" satellite observation. Moreover, models provide first guess profiles for retrievals, and may fill in unobserved parts of the atmosphere. Observing system development should be paired with model developments and the developments of techniques to integrate satellite data in models. In that sense, again, no mention is made of the European Copernicus program, which in my opinion is a missed opportunity to guide the US developments.

Agreed. To address this concern, we've added the following text to the end of Section 2:

> "The robustness of constraining OH with these satellite-based approaches is not limited by, for instance, the ML or data assimilation methods at the present time, but instead by the quality of the data used as input to these methods and limitations in our current understanding of the chemistry and emissions that influence tropospheric OH (e.g., Table 2 of Fiore et al., 2024). Therefore, the recommendations in the sections that follow focus on further development of the satellite-based approaches as well as improvement to the quality of satellite data and independent validation products."

Concerning improving CTMs, Section 5.3 discusses the need for process-based diagnostics to better constrain CTMs. Specifically, a recommendation in Section 6 is: "We also recommend additional investment in research … to develop process-based diagnostics using satellite and suborbital observations to improve the representation of key atmospheric processes in CTMs that influence OH."

Minor comments (see also annotated PDF).

Figure1: Although interesting, this paper should not be used to present new results. At most, it could be used as an uncertain exploration of newly developed techniques. Now there is no uncertainty quantification, the legend fails to mention "tropical ocean".

The figure is simply meant to give the reader an indication of the spatio-temporal variability of inferred OH. In this context, the figure is appropriate for an opinion piece.

We added to the end of the caption "This figure is an adaptation of Figure 2 from Anderson et al. (2024).", which indicates that it is not a new result. Anderson et al. (2024) showed results for boreal fall. Figure 1 of this paper shows all four seasons. In addition, "tropical" was added to the caption as suggested.

Line 30: suborbital observing strategy: unclear (in an abstract).

"Suborbital" is now defined at first use in both the Abstract and Introduction.

Line 36: "will be an integral part of a comprehensive observing strategy"

Maybe "should be an integral part of a comprehensive observing strategy." Is better?

The sentence is now:

"Suborbital observations (i.e., data collected from non-satellite platforms, such as aircraft, balloons, and buildings) are required for information difficult to obtain from space and for validation of satellite-based OH estimates; therefore, they should be an integral part of a comprehensive observing strategy."

Line 70: contributes significantly to the overall uncertainty in the budget, interannual variability, and trends of CH_4 (Saunois et al., 2020).

I agree here with uncertainties in the budget, but the interannual variability and trend in methane are well constrained by observations. Of course, the impact of OH on trend and variability in methane is large (what you intend to write), but this phrasing suggests that trend in methane itself is uncertain.

Removed ", interannual variability, and trends".

Line 164: "In contrast, decreases in CO can lead to decreased OH and longer CH_4 lifetimes."

I guess this should be "increased OH" and "shorter CH_4 lifetimes".

Corrected.

Lelieveld, J., Brenninkmeijer, C., Joeckel, P., Isaksen, I., Krol, M., Mak, J., Dlugokencky, E., Montzka, S., Novelli, P., & Peters, W. (2006). New Directions: Watching over tropospheric hydroxyl (OH). Atmospheric Environment, 40(29), 5741–5743. https://doi.org/10.1016/j.atmosenv.2006.04.008

Lelieveld, J., Dentener, F. J., Peters, W., & Krol, M. C. (2004). On the role of hydroxyl radicals in the self-cleansing capacity of the troposphere. Atmospheric Chemistry and Physics, 4, 2337–2344.

As suggested, the above papers were added to the introduction.

Comments from the annotated manuscript provided by the reviewer:

Line 216 of the original manuscript: "CH_4 observations could be useful given that there are space-based observations." The reviewer commented: "This is a strange sentence. CH4 is a major sink of OH, but well-mixed (thus a good estimate can be obtained)." To address this comment, we removed the sentence. We also added the following sentence in Section 5.2.1 when discussing OH sinks (Line 488 of original text):

> "CH_4 is relatively well mixed throughout the troposphere, therefore we could assume a tropospheric distribution informed by in situ observations."

Line 456 of the original manuscript: "…lightning is an important source of NO_x in the middle and upper troposphere (Allen et al., 2021), which modulates OH there (e.g., Fiore et al., 2006)." The reviewer noted: "True, but this would require a model, or ML techniques to relate these observations to OH?" We modified the text to:

> "Lightning is an important source of NO_x in the middle and upper troposphere (Allen et al., 2021), which modulates OH there (e.g., Fiore et al., 2006). Satellite observations of lightning flash counts (e.g., over the Americas from the Geostationary Lightning Mapper, GLM, aboard the NOAA Geostationary Operational Environmental Satellite-16, GOES-16; over Europe, Africa and Middle East from EUMETSAT Meteosat Third Generation – Imager 1 (MTG-I1) Lightning Imager) may be useful to constrain the vertical distribution of NO_2 using ML or a CTM to relate flash counts to NO_2 concentrations."

Line 470 of the original text: The reviewer commented: "I guess this source is not relevant: H2O2 likely produces OH mostly in the lower atmosphere...." We addressed this point as indicated by the underlined text and also in Table 1:

> "There are satellite observations from Atmospheric Chemistry Experiment (ACE) for H_2O_2, the fourth source (i.e., H_2O_2+hv), in the mid to upper troposphere<u>, though this source primarily occurs in the lower troposphere (e.g., Spivakovsky et al., 2000)</u>; these observations are sparse so multi-year averages are required to obtain seasonal, zonally-averaged distributions (Allen et al., 2013)."
>
> Table 1: "Instrument and/or retrieval algorithm development required<u>, including to obtain observations in the lower troposphere</u>."

Line 490 of the original text: The reviewer commented: "I am not convinced that tropospheric O3 products are of sufficient quality (given teh overhead stratospheric ozone layer)....here TIR-UV/vis approaches may help (there are references!)". To address this comment, we modified Table 1. See the revised table at the end of this response.

Line 548 of the original text: The reviewer said: “Again, one should differentiate between "ability to observe" and "quantify". One could argue that this will imporve after time, but quantitative use of satellite data is currently only possible for some proxies, such as H2O, NO2, UV-radiation, ...but not for tropospheric ozone, VOCs, etc.”

Agreed. See our response above to a similar comment.

Line 567 of the original text: The reviewer said: “I would say that drought induced fires (e.g. 2018 Indonesia, 2010 Russia, etc. are equally important.” The text has been revised from:

> “For instance, climate indices, such as the Multivariate El Niño–Southern Oscillation (ENSO) Index (MEI) and indicators of drought, correlate well with long-term (e.g., monthly, seasonal) variations in tropospheric constituents (e.g., CO, NO_2, O_3, isoprene, water vapor) that influence OH (e.g., Oman et al., 2011; Oman et al., 2013; Wells et al., 2020; Anderson et al., 2021; Anderson et al., 2023; Shutter et al., 2024) as well as OH itself, primarily through NO_x emissions associated with lightning (Turner et al., 2018).”

To:

> “For instance, climate indices, such as the Multivariate El Niño–Southern Oscillation (ENSO) Index (MEI) and indicators of drought, correlate well with long-term (e.g., monthly, seasonal) variations in tropospheric constituents (e.g., CO, NO_2, O_3, isoprene, water vapor) that influence OH (e.g., Oman et al., 2011; Oman et al., 2013; Turner et al., 2018; Wells et al., 2020; Anderson et al., 2021; Anderson et al., 2023; Shutter et al., 2024) as well as OH itself.”

The last phrase of the original text was left over from a previous version of the sentence and should have been removed.

Line 594 of the original text: The reviewer said: “TROPOMI CO should be mentioned here.” The revised text is:

> “There may be viable ways to infer some of the OH drivers over clouds (e.g., NO_2, Marais et al., 2018; Marais et al., 2021; CO, Landgraf et al., 2016), but further study is needed.”

Line 603 of the original text: The reviewer wrote: “maybe wise to mention that also heterogeneous chemical processes are less well understood/quantified.”

We added this uncertainty in the introduction where we discuss uncertainties in OH.

Revised Table 1:

	Sources [a]	% of Total	Satellite Proxies (wavelengths)	Satellite Proxy Limitations & Potential Improvements	Short-Term Data Continuity? [h]

		Sources [a]			
1	$O(^1D)+H_2O_{(v)}$	33%	Stratospheric O_3 VCD (UV/Vis) for $O(^1D)$. Water vapor (IR).	No major limitations.	Stratospheric O_3 VCD (yes) - e.g., TROPOMI. Water vapor (yes) - e.g., CrIS.
2	$NO+HO_2$	30%	NO_2 VCD (UV/Vis). Research is needed to identify a proxy for HO_2.	Needs improved SNR where NO_2 VCDs are low (Buscela et al., 2013). Lightning flash counts may provide information on the vertical distribution over some continents (e.g., NOAA GeoXO Lightning Mapper (LMX); MTG-I1 Lightning Imager)	NO_2 VCD (yes) - e.g., TROPOMI.
3	O_3+HO_2	14%	Tropospheric O_3 VCD (UV/Vis). Multispectral products (*Section 4.4*). Research is needed to identify a proxy for HO_2.	Needs accurate stratosphere-troposphere separation of total column O_3 VCD (Ziemke et al., 2006). Research is needed to determine the potential of multispectral products (e.g., TROPOMI/CrIS), which may provide information on vertical distribution.	Tropospheric O_3 VCD (yes) - e.g., TROPOMI, OMPS.
4	$H_2O_2+h\nu$	10%	H_2O_2 (IR).	Instrument and/or retrieval algorithm development required, including to obtain observations in the lower troposphere. ACE data of H_2O_2 are sparse; zonal averages of multiple years	H_2O_2 (no).

				are required to obtain near-global coverage in the mid to upper troposphere (Allen et al., 2013).	
5	OVOCs[b], ROOH[c]+h*v*	13%	HCHO, glyoxal (UV/Vis). Numerous potential VOCs (TIR).	Needs improved SNR where HCHO VCDs are low. VOC instrument (TIR) and retrieval algorithm development required. Research required to determine the suitability of these VOCs for constraining this source.	HCHO, glyoxal VCD (yes) - e.g., TROPOMI. Numerous VOCs (yes for some, but not others) - e.g., CrIS.
	HONO[d] + h*v*	–	HONO (UV/Vis; TIR).	Observations primarily for intense wildfire plumes that reach altitude. Data are very noisy. Instrument and/or retrieval algorithm development required.	HONO (yes) - e.g., TROPOMI.
	Sinks [a]	% of Total Sinks [a]			
1	$OH+HO_y$[e]	18%	Tropospheric O_3 VCD (UV/Vis). H_2O_2 (IR). Assume constant distribution of H_2.	See source #4 above.	See source #3 above.
2	$OH+CH_4$	12%	CH_4 (IR).	Needs (1) better SNR to detect variations in high background concentration and (2) improved	CH_4 (partially, yes) - e.g., TROPOMI (over

				sensitivity to near-surface.	land, glint over ocean).
3	OH+CO	39%	CO (IR). Multispectral products (*Section 4.4*).	Need improved near-surface sensitivity. Research is needed to determine the potential of multispectral products (e.g., TROPOMI/CrIS), which may provide information on vertical distribution.	CO VCD (yes) - TROPOMI, CrIS.
4	OH+other C_1 VOC [f]	15%	Methanol.	VOC instrument (TIR) and retrieval algorithm development required. Research required to determine the suitability of these VOCs for constraining this sink.	Numerous VOCs (yes for some, but not others) - e.g., CrIS.
5	OH+C_{2+} VOC [g]	14%	Isoprene. PAN, etc.	VOC instrument (TIR) and retrieval algorithm development required. Research required to determine the suitability of these VOCs for constraining this sink.	Numerous VOCs (yes for some, but not others) - e.g., CrIS.

[a]Reproduced from Table 1 of Lelieveld et al. (2016), except neglecting two minor sinks. [b]OVOCs = oxygenated VOCs, such as acetone and acetaldehyde. [c]ROOH = organic peroxides, such as CH_3OOH. [d]HONO is not explicitly listed as a source of OH in Lelieveld et al. (2016), though it is an important source in some environments (Theys et al., 2020; Fredrickson et al., 2023). [e]HO_y = H_2, O_3, H_2O_2, radical–radical reaction. [f]VOC with one C atom (excluding methane), including methanol, C1-reaction products. [g]VOC with ≥2 C atoms, C2+ reaction products. [h]Defined in *Section 5.1.1*. We focus on current satellite instruments that provide near complete spatial coverage of the troposphere, neglecting instruments that provide data of, for instance, the upper troposphere (e.g., MLS) and geostationary orbits. We do not list potential future instruments given that space agency priorities and budgets change and because of the history of satellites failing to reach orbit during launch.