Response to the Referee #2:

Referee' comments (black) and Author Responses (blue):

(The changes in the paper are underlined in this response.)

The authors present a new method to infer the stratospheric photolysis rate of NO_2 using satellite (MIPAS) measurements. The photolysis rate coefficient determines the diurnal variation of NOx photochemistry. The results agree well with model predictions. This work provides the first observations-based validation of the role of albedo in driving polar photochemistry.

The scientific questions addressed by the paper are certainly within the scope of ACP. The title clearly reflects the contents of the paper, the abstract provides a concise and complete summary of the work, the authors provide proper credit to related work by other groups and clearly indicate their new contribution. The presentation is generally well structured and clear, and the language and mathematical notation is adequate.

My main concern is that the description of the approach is not sufficiently complete to allow their reproduction by others. For example, how do the authors collocate the model horizontal and vertical grid with those of the measurements? It is stated that "Model values for December 2009 at the same times and location as the satellite data are selected to compare with the satellite data", but what are the actual spatial and temporal collocation criteria? Further, is any kind of interpolation (temporal, horizontal and/or vertical) done subsequently to match the grids up? This is critical information that is missing in the paper. Are aerosols or clouds considered in the comparisons? This is relevant because the authors use a four-stream radiative transfer model, which may not be accurate enough (especially in the UV/Visible spectral regions) when aerosols or clouds are present. The retrieval accuracies may also degrade in these scenarios.

Thank you for the comments. We have improved the description of the methods.

The model's satellite profile algorithm outputs constituents or rates (e.g., NO concentration, or Jno_2) at the nearest available latitude, longitude, and local time to those of each observation. The model resolution for this study is ~2 degrees in the horizontal; therefore, the spatial resolution accuracy would be within +/- 100km in each horizontal direction (+/- 140km along the diagonal direction). The vertical resolution is dependent on the model vertical resolution. The model chemistry time step is 30 minutes; therefore, the temporal resolution is +/- 15 minutes.

The photolysis routine is based on a lookup table approach (Kinnison et al., 2007). This approach does not include clouds or aerosols in the radiative transfer. However, there is a cloud correction factor applied to the total photolysis rate based on Chang et al., 1987 (equations 12-14). This photolysis cloud correction approach is discussed in Brasseur et al., 1998. There is no correction factor included for aerosols. In this paper, we didn't exclude the data when aerosols and clouds are present.

Chang, J. S., R. A. Brost, I. S. A. Isaksen, S. Madronich, P. Middleton, W. R. Stockwell, and C. J. Walcek, Three-Dimensional Eulerian Acid Deposition Model: Physical Concepts and Formulation, J. Geophys. Res., vol. 92, NO. D12, 14,681-14,700, 1987.

Brasseur, G. P., D. A., Hauglustaine, S. Walters, P. J. Rasch, J.-F. Muller, C. Granier, and X. X. Tie, MOZART, a global chemical transport model for ozone and related chemical tracer: Model description, J. Geophys. Res., vol. 103, NO. D21, 28,265-28,290, 1998.

Kinnison, D. E., G. P. Brasseur, S. Walters, R. R. Garcia, F. Sassi, B. A. Boville, D. Marsh, L. Harvey, C. Randall, W. Randel, J. F. Lamarque, L. K. Emmons, P. Hess, J. Orlando, J. Tyndall, and L. Pan, Sensitivity of chemical tracers to meteorological parameters in the MOZART-3 chemical transport model, J. Geophys. Res., 112, D20302, doi:10.1029/2006JD007879, 2007.

Therefore, we added the description of the satellite profile algorithm as follows:

Model values for December 2009 at the same times and locations as the satellite data are selected by the satellite profile algorithm to compare with the satellite data, and denoted "Model". The satellite profile algorithm outputs constituents (e.g., J_{NO_2} and NOx concentrations) at the nearest latitude, longitude, and local time to the observation.

Furthermore, to ensure better clarity and accuracy in this paper, we have expanded the descriptions of data processing and data sources.

Data processing: The data from the satellite was averaged daily and zonally (Because the specific latitudes of the satellite data vary somewhat from one orbit to another, we bin the data using a two-degree interval). Then we calculate the four-day running mean, which is shown in Fig. 2 and Fig. 3.

Reaction rate constants sources: The k_{NO+O_3}, k_{NO+ClO}, k_{O+NO_2} and their uncertainties are from JPL (Burkholder et al., 2015), and k_{O+O_2+M} and its uncertainty are from International Union of Pure and Applied Chemistry (IUPAC; Atkinson et al., 2004).

Figure descriptions: To ensure clear visual distinction for each point, black outlines are applied around them.

The uncertainties were also discussed in more detail, please see uncertainty part below.

This is relevant because the authors use a four-stream radiative transfer model, which may not be accurate enough (especially in the UV/Visible spectral regions) when aerosols or clouds are present. The retrieval accuracies may also degrade in these scenarios.

Regarding your inquiry about the adequacy of the 4-stream discrete ordinates, we conducted a brief sensitivity study to validate the accuracy of the four-stream radiative transfer model. In this test, we compare the performance of 2, 4, 8, 16 stream radiative transfer, under both high and low albedo scenarios. The Delta-Eddington 2 stream model is the fastest model, and the Discrete Ordinates 4 stream model is used in WACCM. We also use Discrete Ordinates 8 stream model and Discrete Ordinates 16 stream model, which is the most accurate and serves as the standard for this test.

Subsequently, we calculated the Jno_2 at 30 km under clear sky conditions without the presence of aerosols or clouds using these different radiative transfer models. The Jno_2 and the difference with 16-stream model, under surface albedo conditions of 0.1 and 0.9, are presented in Table 1 and Table 2, respectively.

Table 1. The Jno_2 and the errors of 2, 4, 8, 16 stream models under the surface albedo of 0.1

sza	2-stream	4-stream	8-stream	16-stream	Err2 (%)	Err4 (%)	Err8 (%)
0	1.26E-02	1.30E-02	1.30E-02	1.30E-02	-3.07	-0.31	-0.23
10	1.26E-02	1.30E-02	1.30E-02	1.30E-02	-3.15	-0.23	-0.15
20	1.25E-02	1.29E-02	1.29E-02	1.30E-02	-3.63	-0.31	-0.23
30	1.23E-02	1.28E-02	1.28E-02	1.29E-02	-4.20	-0.31	-0.23
40	1.21E-02	1.27E-02	1.27E-02	1.27E-02	-5.03	-0.39	-0.24
50	1.18E-02	1.24E-02	1.25E-02	1.25E-02	-5.92	-0.40	-0.24
60	1.13E-02	1.20E-02	1.21E-02	1.21E-02	-6.77	-0.58	-0.25
70	1.06E-02	1.14E-02	1.14E-02	1.15E-02	-7.41	-0.87	-0.35
80	9.49E-03	1.01E-02	1.02E-02	1.03E-02	-7.48	-1.46	-0.29
90	6.49E-03	6.64E-03	6.73E-03	6.78E-03	-4.34	-2.11	-0.72

Note that sza is solar zenith angle and the error is the difference with 16-stream model, which serves as the standard for this test.

Table 2. The Jno_2 and the errors of 2, 4, 8, 16 stream models under the surface albedo of 0.9

sza	2-stream	4-stream	8-stream	16-stream	Err2 (%)	Err4(%)	Err8(%)
0	2.28E-02	2.21E-02	2.21E-02	2.22E-02	3.12	-0.32	-0.23
10	2.26E-02	2.19E-02	2.19E-02	2.20E-02	2.96	-0.32	-0.23
20	2.20E-02	2.14E-02	2.14E-02	2.14E-02	2.57	-0.28	-0.19
30	2.09E-02	2.05E-02	2.05E-02	2.05E-02	1.80	-0.34	-0.24
40	1.95E-02	1.92E-02	1.93E-02	1.93E-02	0.78	-0.41	-0.26
50	1.77E-02	1.77E-02	1.77E-02	1.78E-02	-0.56	-0.45	-0.28
60	1.56E-02	1.58E-02	1.59E-02	1.59E-02	-2.14	-0.57	-0.31
70	1.32E-02	1.37E-02	1.37E-02	1.38E-02	-3.92	-0.80	-0.29
80	1.06E-02	1.10E-02	1.12E-02	1.12E-02	-5.54	-1.34	-0.27
90	6.61E-03	6.73E-03	6.83E-03	6.88E-03	-3.91	-2.11	-0.73

WACCM uses the 4 stream discrete ordinates model. Errors in Jno_2 at 30 km computed with a 4 streams model, relative to 16 streams model, are less than 1% for sza<75, reaching about 2% at sza=90. The error is always negative, causing a slight underestimation of the true value. Two-stream errors are substantially larger, positive or negative, and can reach 7 or 8 %. In this test, we found that the accuracy of the 4-stream model is nearly equivalent to that of the 16-stream model, indicating that the 4-stream model provides sufficient accuracy for our purposes.

The work does present an "observations-based check on the role of albedo in driving polar photochemistry", but this result alone would only provide an incremental improvement to existing scientific understanding. It would be a lot more revealing if the authors could figure out under what conditions the models do not work so well (scenarios with aerosols and/or clouds?).

In this paper we introduce a novel approach to derive precise J_{NO_2} values based on satellite data. This paper focuses on the methodology, which is totally different from model. In the model, taking TUV model as example, it is based on radiative transfer principles. By accounting for the vertical distribution of atmospheric constituents, the TUV model can calculate the intensity and spectral distribution of UV and visible radiation at different altitudes. Then based on photochemical data (such as from JPL), calculating the Jno_2 values. Our satellite method is grounded in the steady state assumption. We calculate the Jno_2 values using satellite data through Equations 2 and 3. The key message of our paper is that using two totally different methods, Jno_2 calculated by satellite data and Jno_2 from the model are consistent with each other, significantly increasing the confidence in the existing Jno_2 values. Moreover, future studies could verify other photolysis rates using our method. In addition, an implication of the paper is conceptual: Obtaining photolysis rates through satellite data. The benefits of obtaining photolysis rates through satellite data are significant: We can obtain global uninterrupted data, similar to species concentration data from satellites, which can be used for more refined research. We can also test and confirm the impact of surface albedo, an important factor in atmospheric photolysis and chemistry, tested here with the contrast from the Antarctic to lower latitudes. Indeed, due to the uncertainty of satellites, the temporal and spatial resolutions for obtaining effective Jno_2 are currently very low. In this paper, a point represents the average of the data in the same latitude in four days. But with the advancement of satellite sensors and retrieval methods, the temporal and spatial resolution will become smaller and smaller, which supports the further research for better understanding the Jno_2.

In summary, our paper not only significantly increases the confidence in the existing Jno_2 knowledge, and introduces a method that can be extended to other photolysis rates, but also presents a new promising concept that obtaining photolysis rates through satellite.

This also leads to the issue of uncertainty quantification. There is no mention of error characteristics in the paper. This is critical for satellite-based retrievals. Without knowledge of the retrieval errors, it is very hard to make any evaluations about the quality and/or robustness of the results. For example, the statement that "However, in the stratosphere below about 33 km [O] has a small effect on JNO2 (less than 8.1 percent)" is meaningless unless it is contrasted with errors in JNO2 itself. The authors do report precisions for the various species. These could probably be used to obtain precisions for the photolysis rate estimates.

Thank you for the comments. We agree with your insights regarding the significance of uncertainty in this study. We therefore added uncertainties for J_{NO2} in Figure 3 and Figure S1, along with descriptions and discussions about uncertainty in the paper.

The uncertainty is based on the Equation (2) and (3):

$$J_{NO_2} = \frac{[NO]}{[NO_2]} \times \left(k_{NO+O_3} \times [O_3] + k_{NO+ClO} \times [ClO]\right) - k_{O+NO_2} \times [O]$$

$$[O] = \frac{J_{O_3} \times [O_3]}{k_{O+O_2+M} \times [O_2] \times [M]}$$

We considered the uncertainty of $[NO]$, $[NO_2]$, $[O_3]$, $[ClO]$, k_{NO+O_3}, k_{NO+ClO}, k_{O+NO_2} and k_{O+O_2+M} based on the available uncertainty of each term as given in the literature. In the calculation of the uncertainty, we considered the accuracies of different species because each point in Figure 3 is the average of several hundred data points and random error is much smaller than systematic error (accuracy).

The accuracies of different species and their sources are now described in this paper as follows:

In this paper, we used the NO, NO_2, O_3, ClO, temperature and pressure data from V8 MIPAS retrievals performed with the IMK/IAA level 2 processor. The retrieval of temperature was reported by Kiefer et al. (2021). For NO retrieval, the method considered the populations of excited NO states (Funke et al., 2005). This implies that photolysis of NO_2 is included in the retrieval priors. However, retrieved NO is only weakly dependent on prior knowledge of J_{NO_2} values (10-15%). In our calculations, according to Eq. (2) and (3), NO, NO_2 and O_3 play comparable roles in calculation of J_{NO_2}, reducing the impact of prior knowledge on the final results. Therefore, prior knowledge of J_{NO_2} will have a small effect on our findings as long as prior knowledge of J_{NO_2} is not completely incorrect. The NO retrieval was documented by Funke et al. (2023). These authors reported an accuracy of 8-15% for altitudes of 20 to 40 km. Regarding O_3, Kiefer et al (2023) reported an accuracy of 3-8% in the altitude region of interest. The retrievals of NO_2 and ClO were described by Funke et al. (2005) and von Clarmann et al. (2009), respectively, with accuracies of 0.2-0.8 ppbv for NO_2 and total error of more than 35% for ClO. However, please note that these papers refer to older data versions. Accuracy estimates for V8 ClO and NO_2 are not yet available but the values quoted here were used as a rough guideline.

The reaction rate constants of different species and their total errors were described in this paper as follows:

The k_{NO+O_3}, k_{NO+ClO}, k_{O+NO_2} and their uncertainties are from JPL (Burkholder et al., 2015), and k_{O+O_2+M} and its uncertainty are from International Union of Pure and Applied Chemistry (IUPAC; Atkinson et al., 2004).
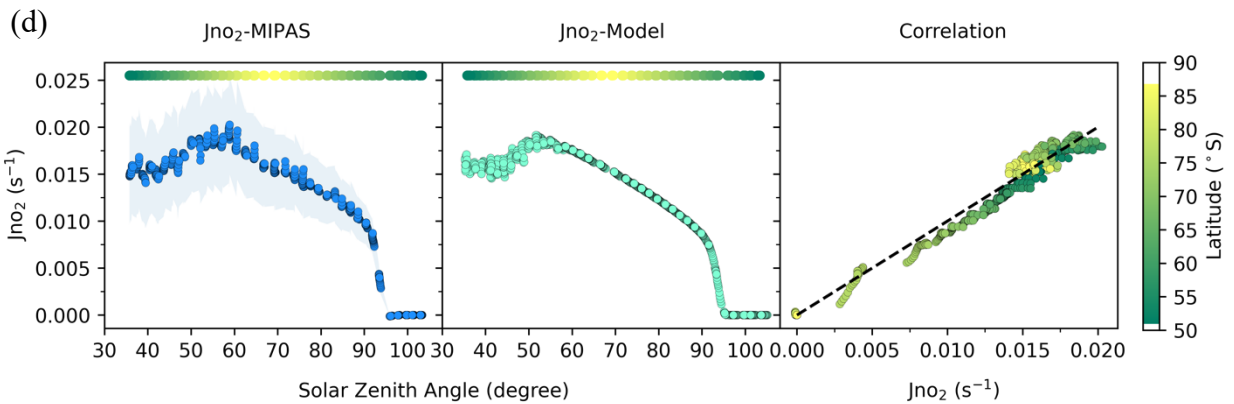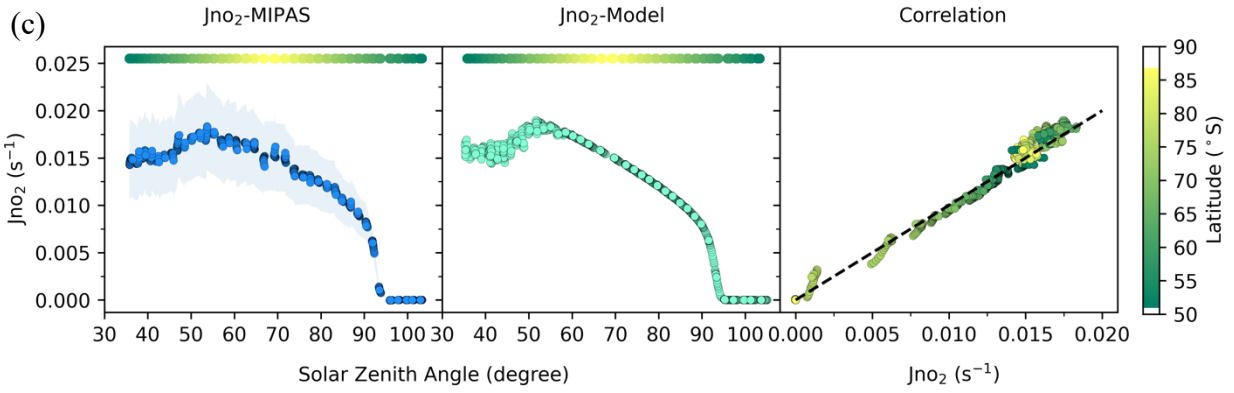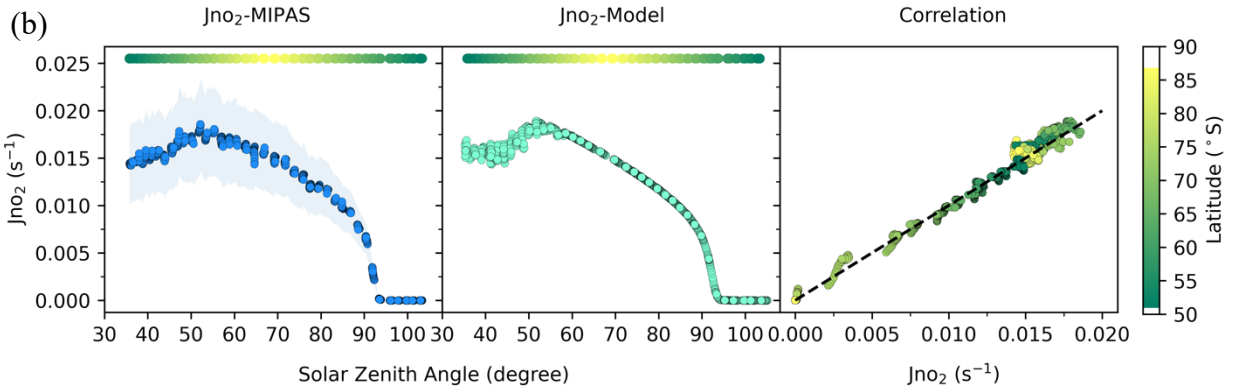
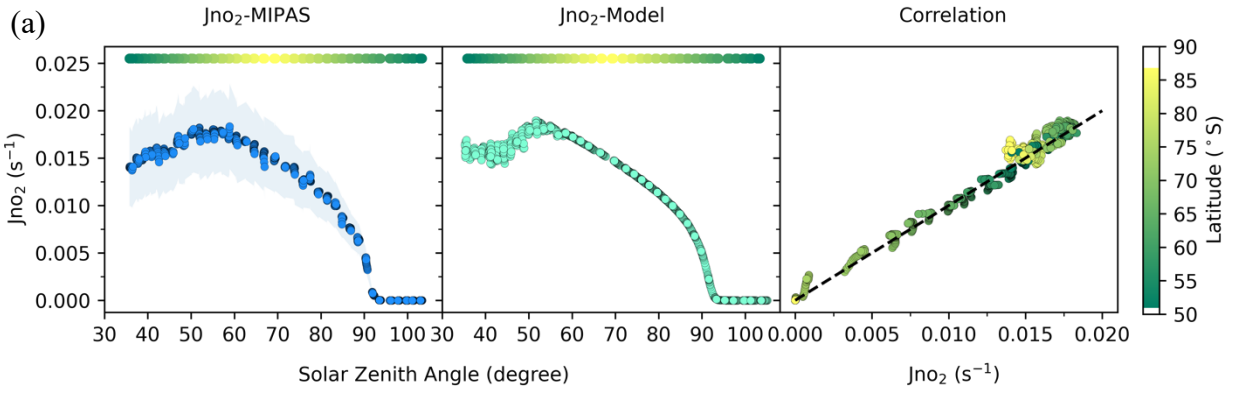Figure 3 after adding error bars is as follows:

Figure 3. The J_{NO_2} in 50° S-90° S from MIPAS and the model at different altitudes. (a) 23 km (b) 28 km (c) 33 km (d) 38km. The color bar represents the latitude of the data points at the same solar zenith angle. In the correlation plots, the abscissa is J_{NO_2}-MIPAS and the ordinate is the J_{NO_2}-Model and the slope of dashed line is 1. To ensure clear visual distinction for each point, black outlines are applied around them.

The uncertainties are about 20% for all altitudes from 20-40 km. There is no dominant term for uncertainty. [NO], [NO2], [O3], [ClO], k_{NO+O_3}, k_{NO+ClO}, k_{O+NO_2} and k_{O+O_2+M} all impart errors that cannot be ignored. Compared with Jno2 at other altitudes, the uncertainties at 38 km are bigger. This is because at 38 km, we need to consider ClO in calculation, which is associated with large error.

A few statements need references:

Thanks. Added the references as following.

"However, in the stratosphere below about 33 km [O] has a small effect on JNO2 (less than 8.1 percent)."

However, in the stratosphere below about 33 km [O] has a small effect on J_{NO_2} calculation (less than 8.1 percent) due to its low concentration (Johnston and Podolske, 1978).

"ClO can similarly be ignored when altitudes are less than 35 km, where ClO concentrations are small"

ClO can similarly be ignored when altitudes are less than 35 km, where ClO concentrations are small (Sagawa et al., 2013)

"HO2 and BrO both can react with NO but they are not measured by MIPAS and their contributions to the partitioning between NO and NO2 are negligibly small at the altitudes considered here."

HO2 and BrO both can react with NO but they are not measured by MIPAS and their contributions to the partitioning between NO and NO2 are negligibly small at the altitudes considered here (Del Negro et al., 1999).

Overall, the paper has potential for publication after the changes listed above are made.