I would like to thank the reviewers for their comments which have helped us to improve the manuscript and make it a more useful work for other researchers. I have addressed their comments as follows (indicated in blue as in the revised manuscript).

Reviewer 1

The author introduces a new method to detect optically thin high clouds from twilight observations at a single wavelength. The method is based on previous work by the same author where they used the color index calculated from the ratio at two wavelengths. It can often be confirmed by visual experience after sunset, when optically thin cirrus clouds light up in bright red and become visible, and the reason for this coloring is nicely explained in the manuscript. The new method which uses only one wavelength rather than the ratio of two, is clearly explained. Sensitivities to all relevant parameters are tested. The method is based on a fast single scattering model which is verified by an accurate Monte Carlo model. I recommend publication of the manuscript after considering the following minor points:

-The description is quite long, and the main points of the manuscript are sometimes hidden by the wealth of equations and information. One way to improve that would be to shorten the first section which deals with the two-wavelengths method and focus on the new part, the single wavelength method.
*Author response: Section 2 (The color index approach) has been substantially shortened by removing the former Figure 1 together with its associated discussion and by condensing the description of the conventional two-wavelength CI approach. These modifications place greater emphasis on the new single-wavelength formalism developed in this work.

-line 85: it is argued that cloud layers extending over hundreds of km are unrealistic. This is relevant because of the long path of the radiation through the atmosphere. While it is certainly correct that the same cloud extending over hundreds of km is unrealistic, but the long pathlength in fact implies that the detection at one place can be affected by high clouds several hundred kilometres away. This would have an effect on the detection efficiency of the method because the absence of an observation of a cloud above the observer could either be caused by (1) actually no cloud being there, or (2) a distant cloud blocking the path of the radiation on it's long way to the observer.
*Author response: We agree with the reviewer that, because of the long twilight optical path, clouds located several hundred kilometres away may also influence the radiation reaching the observer. However, the objective of the present work is to investigate the physical origin of the CI maximum under the simplified assumption of a single localized cloud. Accounting for multiple cloud layers distributed along the solar path would require prior knowledge of their locations and optical properties, which are generally unknown in practical observations. To make this assumption explicit, the corresponding sentence has been revised as follows:

'*Assuming that the cloud under study is the only cloud present in the atmospheric scene, treating it as a spherical shell rather than as a localized layer would imply that, for SZA >90, the direct solar radiation interacts twice with the cloud (see Fig. 2)*'

-Figure 1: The normalisation of the CI curves helps to better plot them on the same axis? In principle I would prefer the non-normalised curves because they would illustrate the "red-ness" of the sky. Also the sharp drop is puzzling. It is explained later in the text but it would help to have a brief explanation of the reason for the sharp drop towards larger SZA.

*Author response: We thank the reviewer for this comment. Following the reviewer's earlier recommendation to simplify Section 2 and focus on the new methodology introduced in this work, Figure 1 has been removed from the revised manuscript. Consequently, this comment is no longer applicable.

-line 215, eq 16: true, but a hint that this directly follows from the evaluation of the derivative of the ratio in eq (4) would be helpful.
*Author response: The text has been revised to explicitly indicate that Eq. (16) is obtained by differentiating the ratio defined in Eq. (4) with respect to the SZA and setting the derivative equal to zero. This makes the origin of Eq. (16) more explicit.

-Figure 7: I was a bit surprised that water vapor was not included. Water vapor has strong absorption bands in particular in the right red block which is nearly free of absorption by NO2 and O3. Most of the water vapor is located in the atmospheric boundary layer between 0 and 2km but the zenith radiance has to pass through it in any case.
*Author response: We thank the reviewer for this comment. We agree that, as originally written, the text accompanying Fig. 6 of the revised mauscript could give the impression that NO2 and O3 constitute the only atmospheric absorbers relevant to CI retrievals. This was not the intended message. The purpose of Fig. 6 of the revised mauscript is simply to illustrate how gaseous absorption can complicate the selection of the two wavelengths used to compute the CI, rather than to provide an exhaustive list of all relevant absorbers. The text has therefore been revised to clarify this point, explicitly stating that, depending on the atmospheric conditions and the vertical distribution of the absorbing species, other gases, such as H2O, may also need to be considered. In particular, the sentence:

'*Figure 6 illustrates this limitation by showing the absorption cross sections of NO2 (green) and O3 (blue), derived from Burrows et al. (1998) and Molina and Molina (1986).*'

Has been replaced by:

'*Figure 6 illustrates this limitation by showing, as representative examples, the absorption cross sections of NO2 (green) and O3 (blue), derived from Burrows et al. (1998) and Molina and Molina (1986).'*

And at the end of the paragraph, immediately after "*which limits the available wavelength combinations for CI-based retrievals.*", the following sentence has been added:

'*It is important to note that the absorbers shown in Fig. 6 are representative of the atmospheric conditions considered in Gomez-Martin et al. (2021). Under different atmospheric conditions, other absorbers, such as H2O, may also need to be taken into account if their vertical distribution extends sufficiently high to influence the CI-based retrievals.*'

-Figure 12: It wasn't very clear to me here that you look for the crossing between the Rayleigh and the total curve to identify cloud height. Maybe it is mentioned in the text and I missed it but you may mention it directly when you describe Figure 12 in the text.
*Author response: We agree with the reviewer that this point was not sufficiently clear in the original manuscript. The paragraph describing Fig.12 has been revised to clarify that "The retrieved value of SZA_{max} is determined by the intersection between the cloud and Rayleigh relative-variation curves." Please see the revised manuscript.

-In the Conclusions I was wondering if you could give a lower threshold for the optical thickness of the clouds to define a detection limit? You study a number of optical thicknesses up to 0.3 but I'm not sure about the lower end.
*Author response: We agree that the minimum detectable cloud optical depth is an important parameter from an observational point of view. However, such a detection threshold depends on the characteristics of the observing instrument and therefore cannot be defined universally. To avoid possible confusion, the Conclusions have been revised to explicitly state that the single-scattering approximation becomes increasingly accurate as the cloud optical depth decreases, while the comparison with the Monte Carlo model demonstrates that it remains valid for cloud optical depths up to $\tau C \approx 0.3$. In particular, the text:

'*The single-scattering methodology (SSM) was compared with a reference Monte Carlo (MC) radiative transfer model previously used for twilight studies in the atmospheres of the Earth, Mars, and Titan. The comparison shows that the single-scattering approximation provides similar results for cloud optical depths up to $\tau c \approx 0.3$.*'

has been replaced by:

'The single-scattering methodology (SSM) was compared with a reference Monte Carlo (MC) radiative transfer model previously used for twilight studies in the atmospheres of the Earth, Mars, and Titan. As expected, the single-scattering approximation becomes increasingly accurate as the cloud optical depth decreases, while maintaining a good agreement with the Monte Carlo model for cloud optical depths up to $\tau C \approx 0.3$.'

Reviewer 2

The manuscript presents a simplified formulation to reproduce and understand the behavior of the color index (CI) at high solar zenith angles (SZA) in the presence of optically thin high-altitude clouds, such as polar stratospheric clouds, without the need to solve the full radiative transfer problem. Based on the improved mechanistic understanding, a new so-called Rayleigh-referenced CI is introduced, defined as the ratio between the measured zenith intensity at a single wavelength and the corresponding intensity expected from a purely Rayleigh-scattering atmosphere. The method is verified and tested across a range of relevant parameters using comprehensive spherical radiative transfer simulations.

The work aligns well with the scope of AMT, and I recommend publication after addressing the following minor points.

General comments

-The manuscript is submitted by a single author, yet the text consistently uses the first-person plural ("we"). To enhance clarity and conform to standard academic conventions, I recommend adopting a more neutral tone throughout. For example, in line 7:
"In this work, we introduce …." -> "In this work, …. is introduced."
*Author response: The manuscript has been revised throughout to replace the first-person plural with neutral or passive constructions.
-The definition of z_dark (ll. 117–119 and 180–187) is not entirely clear. As I understand it, z_dark refers to the lowest altitude illuminated by the Sun. However, the text describes it as an

upper threshold. Please clarify the exact meaning of z_dark and its usage in the formalism. If possible, include z_dark in Fig. 1 to improve illustration and interpretation.
*Author response: We thank the reviewer for this comment. The original definition of z_{dark} was indeed ambiguous and has been replaced by the following clearer description:

'*Here, z_{top} denotes the altitude of the top of the atmosphere, while z_{dark}, which depends on the SZA, denotes the lowest altitude that still receives direct solar illumination during twilight. Below z_{dark}, direct solar radiation is completely blocked by the Earth's curvature.*'

-Given that high-altitude clouds have primarily been studied in remote regions using the color index method, how strongly does the absorption by NO2 (and also O3, H2O) affect the cloud height retrieval if the chosen wavelength pair is suboptimal? Please comment on the expected magnitude of such effects (Sect. 3.4 / Fig. 7).
*Author response: The impact of gaseous absorption on the retrieved SZAmax depends on whether the absorption modifies the CI signal close to, or far from, the SZA at which the maximum occurs. This, in turn, depends on several factors, including the cloud altitude, the selected wavelengths, the gaseous absorption optical depth, and the vertical distribution of the absorbing species.
To illustrate this behaviour, several tests were performed using different combinations of λ1 and λ2, assuming the presence of a gas that absorbs at λ1 but not at λ2. The total gaseous absorption optical depth was varied between 0.01 and 0.1, and a Gaussian vertical distribution centred at 20 km with a geometrical thickness of 10 km was assumed. Although the resulting modifications of the CI signal can be substantial, depending on the gaseous absorption optical depth, the position of the SZA maximum may remain almost unchanged. This is because SZAmax is determined by the intersection between the relative SZA variations of the signals at the two wavelengths. Therefore, if the gaseous absorption does not significantly modify these relative variations in the vicinity of this intersection, only a small displacement of SZAmax is expected.
Figure 1 shows representative simulations of the CI with and without gaseous absorption for a cloud layer located at 18 km and a total gaseous absorption optical depth of 0.05. In all cases, λ1 is fixed at 400 nm, while λ2 = 800, 700, 600 and 500 nm (different panels of the figure). These examples show that introducing the absorbing layer can substantially modify the shape and amplitude of the CI signal. However, the corresponding variation of SZAmax depends strongly on the selected wavelengths. In particular, the smaller the spectral separation between λ1 and λ2, the larger the displacement of SZAmax. For the 400/800 nm combination, the displacement is only about 0.01°, whereas it increases to approximately 0.05°, 0.21°, and 0.43° for the 400/700 nm, 400/600 nm, and 400/500 nm combinations, respectively.
It is also important to note that increasing gaseous absorption reduces the absolute radiance reaching the instrument at the absorbing wavelength. Therefore, even when the displacement of SZAmax is small, the sensitivity of the cloud detection may still decrease because of the lower signal available for the retrieval.

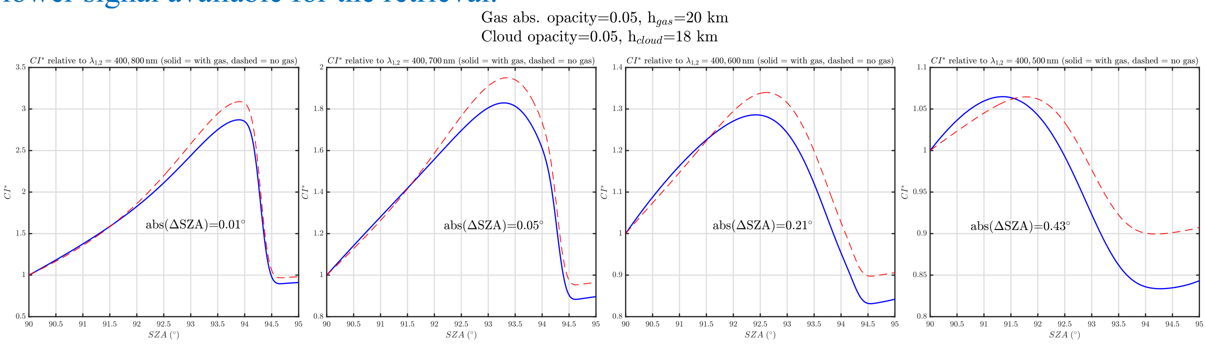

Fig.1 Simulations of the CI.

Figure 2 presents the same set of simulations but for a total gaseous absorption optical depth of 0.02. As expected, the impact of gaseous absorption is reduced. Nevertheless, the same general trend is observed: the smaller the spectral separation between the two wavelengths, the larger the effect on the retrieved SZAmax. Consequently, an inaccurate representation of gaseous absorption may introduce errors in the retrieved cloud altitude. This also provides additional motivation for the proposed single-wavelength methodology, which avoids the need to optimize the selection of two wavelengths while simultaneously reducing the influence of wavelength-dependent gaseous absorption.

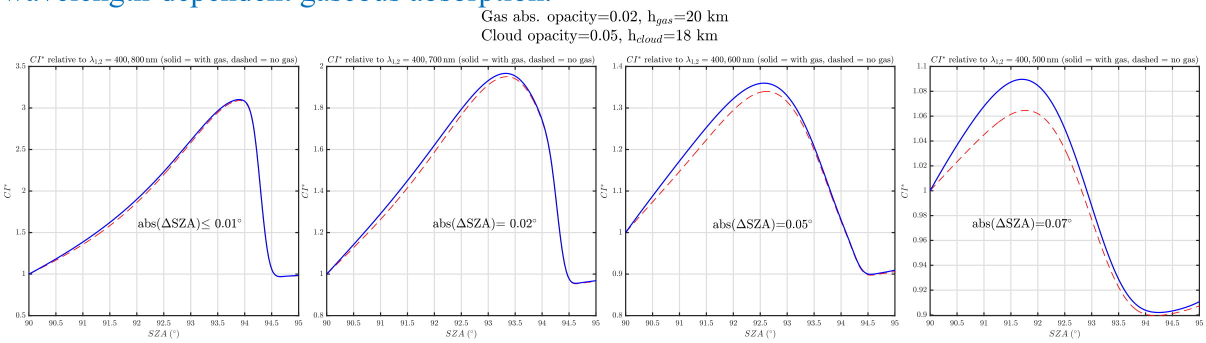

Fig.2 Simulations of the CI.

We have also considered including these additional sensitivity tests in the manuscript. However, since one of the main recommendations from the reviewers was to better emphasize the main contributions, we considered that adding several additional figures and a detailed discussion of the influence of gaseous absorption would unnecessarily increase the length and complexity of the manuscript. Instead, we have added a a text summarizing these results and clarifying the expected impact of gaseous absorption on the retrieval. In particular, the following text has been added in Section 3.4, immediately after "...an inaccurate representation of such temporal cycles may lead to significant errors in the retrieved cloud altitude.":

'To illustrate this point, we performed a series of sensitivity tests considering a Gaussian absorbing layer centred at 20 km, with a geometrical thickness of 10 km, and a total absorption optical depth ranging from 0.01 to 0.1 at λ1 = 400 nm, , while assuming negligible gaseous absorption at λ2. These calculations were not intended to reproduce a particular atmospheric absorber, but rather to provide a generic test case for investigating the influence of gaseous absorption on CI-based cloud-height retrievals. The simulations show that the displacement of SZAmax generally increases as the spectral separation between the two wavelengths decreases. For example, for a cloud located at 18 km and an absorption optical depth of 0.05, the maximum displacement reached approximately 0.43∘for the 400/500 nm wavelength pair, corresponding to an error of about 2 km in the retrieved cloud height. This value should not be regarded as universal, since the impact depends on the selected wavelengths, the cloud altitude, and the optical depth and vertical distribution of the absorbing species. To mitigate these modelling uncertainties, one possible approach is to select λ1 and λ2 outside the main absorption bands of the gases present, thereby simplifying the RT modelling.'

-The manuscript contains a number of valuable contents, but the main achievements – namely, the mechanistic explanation of the CI maximum and the derivation of a new single-wavelength formalism for detecting high-altitude clouds – get a little bit lost in the overall length and technical depth. I recommend overthinking the paper's structure to better highlight these novel findings. Consider shortening the more mathematical sections in the first half of the manuscript or moving the detailed derivations to an appendix.

*Author response: We thank the reviewer for this suggestion. In the revised manuscript, Section 2 has been shortened to improve the overall balance of the paper and place greater emphasis on the new single-wavelength methodology. In particular, Fig.~1 and its associated discussion have been removed, and several introductory explanations have been condensed. Regarding the mathematical derivations, we believe that, given the strong methodological focus of the manuscript, moving part of the derivations to an appendix would interrupt the flow of the presentation and make it more difficult for the reader to follow the development of the new formalism. For this reason, we have considered it more appropriate to retain the main derivations in the body of the manuscript.

-Finally, the conclusions would benefit from a broader outlook. How does the proposed Rayleigh-referenced CI advance our understanding of high-altitude cirrus and polar stratospheric clouds compared to previous approaches?

*Author response: To provide a broader outlook on the implications of the proposed methodology, the following sentence has been added at the end of the second conclusion, where the Rayleigh-referenced CI is introduced:

'Consequently, the proposed Rayleigh-referenced CI provides a more flexible framework for future studies of optically thin high-altitude clouds, including cirrus and polar stratospheric clouds, using a broader range of observing systems.'

Specific comment and technical corrections

-l. 22: "Clouds play a major role in Earth climate system" -> "Clouds play a major role in the Earth's climate system"

*Author response: Corrected.

-l. 35: "heterogeneous reaction that provide" -> "heterogeneous reaction providing"

*Author response: We thank the reviewer for the comment. However, the sentence in the current manuscript reads "which provide surfaces for heterogeneous reactions that release reactive chlorine and bromine species that subsequently participate in catalytic ozone destruction cycles." We believe that this construction is grammatically correct, with the relative clause referring to "heterogeneous reactions". Therefore, we have retained the original wording.

-l. 119: Please rephrase the sentence to avoid misunderstandings regarding "the transition above (?) which the atmosphere no longer receives direct illumination."

*Author response:

-l. 165: Remove parentheses around the citation: (Toledo, 2016) -> Toledo (2016)

*Author response: Done.

-l. 171: It appears that P (not P_total) is shown in the figures. Please revise the corresponding text accordingly.

*Author response: The reviewer is correct. The figures show P, rather than Ptotal, and the corresponding text has been corrected accordingly.

-l. 259: Remove parentheses: (see, e.g., (Gomez-Martin et al., 2021)) -> (see, e.g., Gomez-Martin et al., 2021)

*Author response: Done.

-l. 325: "at lower SZA<95°" -> "at SZA below 95°"

*Author response: Corrected.

-l. 366/367: For improved readability, move the phrase "as will be discussed in the following sections" to the end of the sentence.

*Author response: Done.

-l. 422: Briefly define “droxtals”.
*Author response: A brief definition of droxtals has been added when the term is first introduced in the manuscript.
-Fig. 1: Please specfy the resulting optical thickness of the cloud layer considered.
*Author response: Following the recommendation of Reviewer #1 to simplify Section 2 and improve the focus of the manuscript, Fig. 1 has been removed from the revised version. Consequently, this comment is no longer applicable.
-Fig. 2: The small inset in the top-left corner of panel (a) is not referenced in the caption or main text. Consider removing it for clarity, or integrate the zenith angle directly into the Sun’s ray depiction.
*Author response: The inset has been retained because it provides a clear definition of the solar zenith angle, which is used throughout the manuscript. However, the caption has been revised to explicitly describe the inset.
-Fig. 5: Use a consistent color scheme for the 450 nm and 950 nm results in both panels. Consider using black or a third color for CI* in the right panel.
*Author response: Thank you for the suggestion. In panel (b), the blue and red colours were intentionally chosen to match the left and right vertical axes, respectively, thereby facilitating the association between each curve and its corresponding axis. For this reason, the original colour scheme has been retained.
-Fig. 6: The wavelengths listed in the caption do not match those shown in the left panel. Additionally, no results are displayed for the transition region between 550 and 700 nm. Please either include data for these wavelengths or justify the exclusion of 600 and 650 nm.
*Author response: The figure caption has been corrected to include all wavelengths displayed in panel (a). Regarding the second point, the results at 600 and 650~nm have not been included because they follow the smooth spectral evolution observed between the neighbouring wavelengths (550 and 700~nm) and therefore do not provide additional qualitative information. To clarify this point, the following sentence has been added to the manuscript:
"*The results at 600 and 650nm have been omitted for clarity, as they follow the smooth spectral evolution observed between the neighbouring wavelengths (550 and 700nm) and therefore do not provide additional qualitative information.*"
-Fig. 7: The study by Lauster et al. (2022) has been referenced earlier. For completeness, consider including the corresponding CI ratios from that work.
*Author response: The wavelength pairs used by Lauster et al. (2022) to compute the CI have now been added to Fig.6 of the revised manuscript as dashed vertical lines.
-Fig. 10 + 11: It may be helpful to add the value of SZA_max for better visual orientation, similar to Fig. 9.
*Author response: In the revised manuscript, vertical dashed lines indicating the value of SZAmax have been added to Figures 10 and 11 (9-10 of the revised manuscript). In addition, the figure captions have been revised to explicitly state that the reported values of SZAmax were obtained assuming $\lambda 1$=400 nm in each case.