Author's response to comments of Referee #1

AC: We thank the anonymous referee for the detailed review of our manuscript.

RC1: The study aims to assess the relative effect of factors affecting surface spectral radiation, including SZA, total ozone, cloudiness, and albedo. The dataset is from Brewer spectrophotometer measurements at Marambio Base from the Antarctic Peninsula. Spectral measurements at those latitudes are rare and thus the spectral UV time series 2010-2020 is very valuable, and the manuscript gives a good overview of the statistical distribution of the data. The idea of studying the effect of each UV affecting parameter is not new as such, and that has been studied in several publications previously. However, in this study, the use of neural network makes the study interesting and unique, in addition to the Antarctic location. Unfortunately there is one clear mistake in the applied inputs to the model: the surface albedo data can not be taken from the OMUVB product. In the OMUVB product, the albedo is not the actual measured albedo, but it is the albedo climatology which is used to calculate the OMI UV data. I think the paper can not be published before this has been taken into account, either by excluding the part of the study related to albedo effect or by using other data reflecting the actual albedo situation of the site. I have also a concern on the quality and homogeneity of the time series, as there was so few calibrations (see specific comments here below). In addition I don't understand what is the purpose of Figure 11 and text related to that figure (please see specific comments here below).

I am not familiar with ANN modeling, but I have the impression, that some additional information could be given related to the ten different models which were used (Line 140). How did these models differed from each others.

AC: Thank you for valuable comments and suggestions. The surface albedo interpretation part will be taken out of the study. The albedo climatology will only be kept neural network model, as it presents a statistically significant contribution to its accuracy. We have prepared Appendix A (at the end of the Answer sheet, we will also include it in the manuscript) explaining in detail how we built the ten ANN models and showing the differences between their performance.

Please find our answers to the individual specific comments below.

RC1: Specific comments:

Line 28, Reference UNEP2010 could be changed to a more recent one, eg., UNEP2019

EEAP. 2019. Environmental Effects and Interactions of Stratospheric Ozone Depletion, UV Radiation, and Climate Change. 2018 Assessment Report. Nairobi: Environmental Effects Assessment Panel, United Nations Environment Programme (UNEP) 390 pp. https://ozone.unep.org/science/assessment/eeap

AC: Thank you. We will change the reference to Barnes et al. (2022), which is the latest UNEP update, where they also talk about matters covered in the introduction.

Ref.: Barnes, P. W., Robson, T. M., Neale, P. J., Williamson, C. E., Zepp, R. G., Madronich, S., Wilson, S. R., Andrady, A. L., Heikkilä, A. M., Bernhard, G. H., Bais, A. F., Neale, R. E., Bornman, J. F., Jansen, M. A. K., Klekociuk, A. R., Martinez-Abaigar, J., Robinson, S. A., Wang, Q.-W., Banaszak, A. T., Häder, D.-P., Hylander, S., Rose, K. C., Wängberg, S.-Å., Foereid, B., Hou, W.-C., Ossola, R., Paul, N. D., Ukpebor, J. E.,

Andersen, M. P. S., Longstreth, J., Schikowski, T., Solomon, K. R., Sulzberger, B., Bruckman, L. S., Pandey, K. K., White, C. C., Zhu, L., Zhu, M., Aucamp, P. J., Liley, J. B., McKenzie, R. L., Berwick, M., Byrne, S. N., Hollestein, L. M., Lucas, R. M., Olsen, C. M., Rhodes, L. E., Yazar, S., and Young, A. R.: Environmental effects of stratospheric ozone depletion, UV radiation, and interactions with climate change: UNEP Environmental Effects Assessment Panel, Update 2021, Photochem. Photobiol. Sci., 21, 275–301, 2022.

RC1: line 30: incident UV irradiance \rightarrow incident short wavelength UV irradiance. Or change the wording to describe that it prevents the short wavelength UVC and UVB part of the UV spectrum.

AC: Agreed. We will change the wording to "incident short wavelength UV irradiance", because the UV radiation spectrum division is introduced later in the text.

RC1: Line 33: The recent positive stratospheric ozone trends.... Please specify in which part of the globe? Or global mean?

AC: Thank you. This should mean "southern polar regions". It will be changed in the revised version of the manuscript.

RC1: Line 88: Please specify the method for calibration against the reference instrument. Usually a Brewer is calibrated using 1kW or 200W lamps. Please specify also the traceability of the irradiance scale. Any comparison with the PMOD-World reference QASUME spectroradiometer? Either between the IOS and QASUME or between Marambio's Brewer and QASUME? Only two calibration for 11 years of measurements is very little. There is a possibility that the response of the instrument has changed unexpectedly between the calibrations. Do you have any records to check if the instrument has been stable over the years? How did you take into account the change in calibration: linear interpolation between the two calibrations or stepwise? How much was the difference between the two calibrations? Did you perform any final calibration in 2020? Please explain these points in the text.

AC: The instrument was serviced each year in January or February by specialists from CHMI and International Ozone Services, Inc., Canada. Each year, a calibration has been performed using three to five travel 50W lamps, which were calibrated right before the departure to Marambio Base using the B184 Brewer spectrophotometer in the CHMI Solar and Ozone Observatory in Hradec Králové, and three 1000W lamps S1450, S1451, and S1542 calibrated in the World Radiation Center in Davos. Moreover, in 2012 and 2016, the B199 spectrophotometer was calibrated against the world traveling standard B017 and spectrally calibrated with the use of HG and CD spectral lamps directly at Marambio Base. The spectrum was calibrated in 2012, 2016, and also in 2019. A final calibration was performed in 2020 both at Marambio and after the instrument returned to the Solar and Ozone Observatory in Hradec Králové. The yearly calibration results, which were taken in account stepwise, yielded a maximum difference of -7 % in 2014. The mean absolute annual difference in the 2010–2020 period was 4.1 %. The standard lamp ratios R6, R5, and results from Dead time and Run stop test are additional parameters to monitor for checking of the instrument stability. These information are saved in the instrument checklist at the Solar and Ozone Observatory in Hradec Králové.

We will add all this information to the revised version of the manuscript.

RC1: Line 96: Which method did you use for detection of spikes and wavelength shifts? Please add references.

AC: We have used the spike-detection method by Meinander et al. (2003), and the wavelength shift was analyzed based on Fraunhofer lines using the SHICrivm software package (<u>https://www.rivm.nl/en/uv-ozone-layer-and-climate/shicrivm</u>). The information will be added to the text of the manuscript.

Ref.: Meinander, O., Josefsson, W., Kaurola, J., Koskela, T., and Lakkala, K.: Spike detection and correction in Brewer spectroradiometer ultraviolet spectra, Opt. Eng., 42, 1812–1819, 2003.

RC1: Line 120: What did you use as inputs to LibRadtran? Please add the info in the text. And how did you calculated the CMF?

AC: The input parameters for libRadtran were as follows: day of year, solar zenith angle, albedo climatology from OMI, and total ozone column from B199 Brewer at Marambio.

Cloud Modification Factor (CMF) was calculated as the ratio between observed and clear-sky irradiance for each wavelength (like in, for instance, Lindfors et al., 2007), then the weighted mean using the clear-sky intensities as weights was derived for each spectrum. CMF was determined from the ground based spectral UV irradiance data from B199, and the theoretical clear-sky spectral UV irradiance was estimated using the one dimensional DISORT solver of the libRadtran radiative transfer package.

We will add the information on both libRadtran and CMF in the revised manuscript.

Ref.: Lindfors, A., Kaurola, J., Arola, A., Koskela, T., Lakkala, K., Josefsson, W., Olseth, J. A., and Johnsen, B.: A method for reconstruction of past UV radiation based on radiative transfer modelling: Applied to four stations in northern Europe, J. Geophys. Res., 112 (D23201), 1–15, 2007, DOI: doi:10.1029/2007JD008454.

RC1: This is not true. The albedo is the albedo climatology used in calculation of the OMUVB product. Please see my General comments.

AC: Agreed. Relevant changes (correct data product description, omittance of albedo interpretation) will be made in the entire manuscript.

RC1: Line 154: Do you mean the 80% of data was within +-25%?

AC: Yes, instead of "80 % of the data did not exceed approximately ± 25 %" we will change the wording in the revised version of the manuscript as follows "80 % of the modelled data was within approximately ± 25 %".

RC1: Line 156: What is R2?

AC: It is the determination coefficient (R-squared). The correct explanation will be added in the revised version of the manuscript.

RC1: Lines 156-157: What do you mean by shares variability and shared variability? Do you mean shared variance, covariance? Please use other wording.

AC: Determination coefficient represents shared variance between the two datasets (this site provides a very nice and straightforward explanation: <u>https://www.investopedia.com/terms/c/coefficient-of-determination.asp</u>, but it can also be found in McClave and Dietrich, 1991). Based on this statement the sentence will be rephrased in the revised version of the manuscript, and the McClave and Dietrich (1991) citation will be added.

Ref.: McClave, J. T. and Dietrich, F. H.: Statistics, San Francisco, CA: Dellon Publishing Company, 928 p., 1991.

RC1: Line 177: I suggest that you include the info of how many order of magnitude the UV irradiance changes between e.g., 300 nm and 400 nm.

AC: Thank you for the suggestion. Since our measurements only cover the interval between 300–363 nm, we will chose 305 and 340 nm as examples here, as we cover these wavelengths also further in the text (they are contrasting when it comes to absorption by ozone, and they are also frequently used in literature, which allows a comparison of results). The information that between these two wavelengths, the median UV irradiance increased approximately 25 times in summer and over 100 times in early spring and late fall, will be added in the revised version of the manuscript.

RC1: Line 179:it is steeper in low-SZA months...... Explain why (longer atmospheric path – more ozone absorption)

AC: The statement that "it happens because at low SZA, the atmospheric path is shorter, so less UV radiation is absorbed by ozone and other atmospheric gases" will be added to the revised manuscript.

RC1: Line 185: What do you mean with "overall median". Yearly median? Something else?

AC: It is the median value from all available measurements over the entire study period (calculated separately for each wavelength). We will clarify it in the revised version of the manuscript.

RC1: Line 192: The vertical profile of atmospheric ozone affects the absorption in the optical path. This info could be added somewhere, and possibly discussed.

AC: Thank you for the suggestion and the interesting idea, which we may focus on in further research and do a couple of analyses! In the revised version of the manuscript, we will only add a short note to Section 3.2 (explanatory variable effects), to the end of the paragraph about ozone as follows: "However, the shape of vertical ozone profile may play a substantial role in UV radiation absorption in the optical path, as different vertical distributions of ozone may lead to similar TOC values".

RC1: Lines 193-197: I think you should, in a couple of sentences, describe the effect of the Brewer-Dobson circulation and the lack of sunlight in winter \rightarrow ozone accumulates in polar region, but there is no sunlight for the photochemical ozone destruction which then leads to high TOC values at the end of the winter. You should also explain the year to year variation in Antarctic ozone depletion, otherwise it is quite strange that you find within during the same week the highest and the lowest ever measured TOC (6 November 2011, 3 November 2015).

AC: Agreed, thank you for the suggestion. We will add a short explanation, together with relevant citation, to the revised version of the manuscript:

"The yearly cycle, as well as the short and long term fluctuations of TOC in the coastal Antarctic region are conditioned by both chemical and dynamical influences, while the chemical ones (like the catalytic reactions with the contribution of man-made chemicals) are now quite well understood (e.g., Solomon, 1999). The dynamical influences include the Brewer-Dobson circulation, which causes the poleward transport of ozone from the tropic to the poles, and subsequent accumulation of ozone in the polar regions in winter, as no UV radiation is present to induce ozone loss (Weber et al., 2011). Ozone depletion through catalytic reactions starts in early spring and low TOC values are present till the breakdown of the polar vortex, which is caused by the dynamical effect of planetary waves and has much year-to-year variability (e.g., Shepherd, 2008), so it was possible to observe both the absolute ozone minimum and maximum in one month (November), only in different years."

Ref.:

- Shepherd, T. G.: Dynamics, Stratospheric Ozone, and Climate Change, Atmos. Ocean, 46, 117– 138, 2008.
- Solomon, S.: Stratospheric ozone depletion: a review of concepts and history, Rev. Geophys., 37, 275–316, 1999.
- Weber, M., Dikty, S., Burrows, J. P., Garny, H., Dameris, M., Kubin, A., Abalichin, J., and Langematz, U.: The Brewer-Dobson circulation and total ozone from seasonal to decadal time scales, Atmos. Chem. Phys., 11, 11221–11235, 2011.

RC1: Line 199: Where is the site of Lachlan-Cope (2010) located?

AC: Lachlan-Cope (2010) used latitudinal means, because his work was based on satellite data. Corresponding changes (clarification) will be added in the revised version of the manuscript.

RC1: Lines 200-2004: I am not convinced why do you compare with these sites, and about the reasons for the differences. Is there differences in land-sea surroundings, glaciers ...topography, other meteorological reasons, typical synoptic scale phenomena?

AC: All the three mentioned studies used satellite data, which covered, in the case of Lachlan-Cope (2010), the latitude of Marambio, and in the case of the other two mentioned studies, a pixel directly containing the location of Marambio, therefore no big differences in e.g. topography shall be present.

The results in our manuscript, which gave a different annual cycle than the three above-mentioned studies, are not statistically significant (mentioned in the text), and therefore they are non-conclusive, affected likely only by the high cloud cover variability. We will clarify the use of satellite data in the other studies and further stress the non-conclusiveness of our results, which were not statistically significant.

RC1: Lines 228-229: Alongside atmospheric gases, the surface UV irradiance is also affected by the absorption in the troposphere, which is in this study represented by the cloud cover. How about tropospheric O3, SO2? What do you mean by "which is in this study represented by the cloud cover"?

AC: This was only meant to be an introduction sentence. In the revised manuscript, we will change the wording to the simple "Another parameter, important for the UV radiation attenuation, is cloud cover." Unfortunately, it was not within the scope of the study to cover also tropospheric ozone or sulfur dioxide, although it could be an interesting idea for future research. Particular information about this possible further research will be added it to the conclusion paragraph of the revised manuscript.

RC1: Lines 245->end of the section. Please specify what do you mean by "very high UV irradiances". Very high compared to what? Or higher than a certain limit value?

AC: Very high UV irradiances at 305 and 340 nm were defined as the highest 10 % of all recorded values. The statement will be changed in the revised version of the manuscript in order to be more understandable.

RC1: Line 277: ozonosphere This term doesn't exist.

AC: Thank you for the notification. The term will be removed in the revised version of the manuscript, replaced by "ozone layer".

RC1: Line 285: in low-SZA conditions \rightarrow Should this be in high-SZA..?

AC: We don't think so, as the radiation intensity at very short UV wavelengths decreases faster at low SZA, as shown in the figure taken from Kerr and Fioletov (2008), which is attached (x-axis is the cosine of SZA). However, to avoid possible confusion, we will rephrased the statement in the revised version of the manuscript.



Fig. 2 Measurements of spectral irradiance at two wavelengths in the UV spectrum. Measurements were taken at Toronto between January 2002 and June 2003. There is strong absorption by ozone at 300 nm (a) and very little absorption at 324 nm (b). The clustering near the top of the 324 nm data set indicates clear-sky conditions. There is no similar clustering at 300 nm since the effects of ozone variability mask cloud variability. The fall-off of irradiance with decreasing cos(sza) is significantly sharper at 300 nm. than that at 324 nm, where it is nearly linear. Note that the irradiance values are about two orders of magnitude larger at 324 nm than at 300 nm.

Ref.: Kerr, J. B. and Fioletov, V. E.: Surface Ultraviolet Radiation, Atmos. Ocean, 46, 159–184, 2008.

RC1: Lines 309-317 I think this discussion should be excluded if actual albedo values are not used (See general Comments)

AC: Agreed. This part will be taken out from the revised version of the manuscript, alongside any other conclusions drawn from albedo effects (in summary, abstract, etc.).

RC1: From line 318 until the end of the section. I don't understand what has been done. Why didn't you keep in the model the same values than in the observation (Figure 11)?

AC: This part of the study has two purposes: first to show the differences between the high and low values of a given parameter (through the model values, where all other variables are fixed to their monthly medians). The second purpose was to show the dissimilarity between the model outputs and observations, which are meant to express the importance of one (model) vs. all (observation) studied parameters. If we ran the model with the observed values, it would have been a validation, which was already performed before (fig. 3). This way, we could compare the dissimilarities between the model and observations, which were caused by the differences in input variables. Based on the abovementioned statement, we will expand the explanation in the Results section and clarify the purposes and the interpretation of the figure in the revised version of the manuscript. Moreover, the wording will be changed also in the Method section, to make it clear this was not a model validation but actually a modelling experiment, a study of the dissimilarities between the modelled and observed values.

RC1: Figures 7-9 caption: Include the information if the effect is the median effect, and if yes, to which quantity do you make the change (actual value during the measured spectrum?).

AC: Thank you. Please note that we have presented the mean change; the relative values were calculated with regards to the monthly median spectra. This information will be added to the captions of related figures in the revised version of the manuscript.

RC1: Figure 10. I think you should exclude this plot, as the albedo is not the actual one. You can not make any conclusion of it's influence.

AC: Thank you. This plot will be excluded, alongside the albedo panels of Fig. 4, 5 and 11 (now Fig. 10) in the revised version of the manuscript.

Appendix A

Artificial Neural Network model development and validation

Out of the ten ANN models we built, nine (ANN02 to ANN10) behaved in a similar way, while one (ANN01) was different. The differences between the models did not result from the ANN setting, which remained the same, but occurred due to the random initialization of the models and the random split of the dataset to training (70 %), testing (15 %), and validation (15 %) subsets. As seen from Fig. A1, the model ANN01 had the most data within \pm 5, respective \pm 10 % from observations, and it had the largest R-squared and lowest RMSE out of all ten models. However, the model was biased toward underestimation of UV irradiance throughout most of the spectrum.

For the purpose of the study, it was best to choose a model with the best precision, i.e. the lowest variability of results, highest R-squared and lowest RMSE (model ANN01). Also, it was possible to tackle the bias present within the model using a simple median correction described in the manuscript in section 2.4.



Figure A1. Development and validation of the 10 artificial neural network models, while (a) is the sensitivity of individual models to given variables; (b), (c) and (d) shows the amount of modelled data within 1, 5, and 10 % difference from observations, respectively; (e) and (f) are the absolute and relative median differences of the modelled data from the observations; (g) shows the R-squared, and (h) is the root mean square error of the individual models.