Response to comments of referee 1

Authors would like to express sincere thanks to the referee 1 for valuable comments. We revised a manuscript carefully based on given comments. The comments of the referee 1 are in blue, our replies

5 are in black, and changes made in the revised manuscript are in red. The English in this document has been checked by at least two professional editors, both native speakers of English. Our replies to the comments are as below.

General comments

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The manuscript by Momoi et al. describes a novel method to self-calibrate the POM sun/sky radiometer for water vapour (WV) retrieval using diffuse sky radiance measurements and to estimate precipitable WV from direct irradiance at 940 nm (using a more "physical" approach than a non-linear empirical parametrisation of the Bouguer-Lambert-Beer law). The method is thoroughly and clearly explained, and the description is supported by sensitivity tests using radiative transfer models. The manuscript is skewed in favour of a theoretical/modelling perspective, with only two ending paragraphs focussed on experimental data, which is however justifiable owing to the main purpose of presenting a new method rather than studying the retrieved dataset. I recommend the publication of the manuscript after the authors have addressed some minor remarks, mainly aimed at improving readability by the

20 unexperienced reader.

Specific comments

- The new method seems to provide slightly worse results compared to AERONET, although POM and
 Cimel instruments are similar. It would be good if the authors could elaborate on this, thus enhancing a bit the experimental part of the paper. What is the most likely reason for this result? Is it due to the more physical (less empirical) approach employed in the study, with fewer empirical constraints? Have the authors explored the sensitivity of the retrievals to the accuracy of the instrumental characterisation (e.g., filter response function, field of view, etc.), to the used spectroscopic data (cross sections) or vertical
- 30 profiles? If so, they could mention some of their results. More generally, on the basis of what criteria can the results of the WV retrieval be considered satisfactory? What are the maximum expected/permissible deviations, using such kind of instrument?

2. At least one plot of the time evolution of the retrieved w should be presented, also in order to understand when the maximum deviations from reference instruments occur;

The DSRAD algorithm retrieved the precipitable water vapor by a physical approach although the previous study of the sky-radiometer used an empirical approach (Uchiyma et al., 2014; Campanelli et al., 2014, 2018). The AERONET also uses the empirical method (Holben *et al.*, 1998). Our retrieved value was slightly worse compared with AERONET retrievals and others. The underestimation of

- 40 PWV_{DSRAD+SKYMAP} was due to two factors. The first is the retrieval of PWV by the annual mean calibration constant for the water vapor channel. The calibration constant not only is subject to aging but also undergoes seasonal variation due to temperature dependency (Uchiyama *et al.*, 2018a). Thus, it is possible to underestimate the calibration constant in the wet season. Second, uncertainty regarding the aerosol optical thickness affected PWV retrieval. Figure 18 depicts the differences in PWV and aerosol
- 45 optical thicknesses at 675, 870, and 1020 nm between the DSRAD algorithm and the AERONET retrieval. In the periods from January to May and from October to November, the differences in PWV and aerosol optical thicknesses were less than 0.1 cm and 0.015, respectively. However, the difference in PWV was greater than 0.1 cm from July to September. This corresponds to the difference in aerosol optical thicknesses at 675, 870, and 1020 nm from July to September, which indicates that the
- ⁵⁰ transmittance of water vapor was overestimated by the overestimation of aerosol optical thickness. This led to the underestimation of $PWV_{DSRAD+SKYMAP}$ using the annual mean calibration constant when PWVwas > 3 cm. The above description is added in the revised manuscript (L612-625) and we also added the time series of PWV in the revised manuscript (Figure 18).



Figure 18: The top row shows the time series of PWV in 2017 at Chiba (green and black circles are PWV_{DSRAD+SKYMAP} and PWV_{Cimel}, respectively). The middle row is the difference between PWV_{DSRAD+SKYMAP} and PWV_{Cimel}. The bottom row is the difference in aerosol optical thicknesses at 675 nm (red), 870 nm (blue), and 1020 nm (green) between the DSRAD algorithm and the 60 AERONET retrieval results. Circles and error bars in the middle and bottom rows are means and standard deviations, respectively.

3. It should be stressed that the sensitivity tests using synthetic data do not include measurement noise.If the authors also made some tests with noise, it would be interesting to present those results in thepaper;

We conducted the sensitivity tests using the simulated data with the bias errors in the diffuse radiances. It is added in the revised manuscript (L510-521) as below:

We also conducted sensitivity tests using the simulated data with bias errors to investigate uncertainty in 70 the SKYMAP-derived PWV. The bias errors were $\pm 5\%$ and $\pm 10\%$ for *R*. The value of 5% was given by following reasons. The SVA bias errors of the diffuse radiances for the sky-radiometer observations were estimated to be less than 5% (Uchiyama *et al.*, 2018b). According to Dubovik *et al.* (2000), the uncertainty of the diffuse radiances for the AERONET measurements is $\pm 5\%$. Figures 13 and 14 show the results from the simulated data for the continental average and transported dust aerosols with aerosol

optical thicknesses of 0.02, 0.06 and 0.20 at 940 nm. PWV was overestimated when - 5% bias was applied to *R*. This corresponds to the relationship between *R* and PWV, where *R* decreases with increasing PWV (Section 2.1.2). The bias errors strongly affected the retrieval of PWV at high PWV (> 2 cm), because the sensitivity of high PWV is lower than that of low PWV. The retrieval error of PWV increased with increasing bias errors. The retrieval error of PWV due to ± 5% and ± 10% errors for *R* was within 10% for PWV < 2 cm and up to 200% for PWV > 2 cm.



Figure 13: Comparison of the "true" and retrieval values of PWV from simulated data for continental average aerosol with bias errors. The top, middle, and bottom rows are the retrieval
results at SZA = 30°, 50°, and 70°, respectively. Closed circles are the results with no bias errors. Closed squares and closed triangles are the results with bias errors of plus and minus 5% in *R*,

respectively. Open squares and open triangles are the results with bias errors of plus and minus 10% in *R*, respectively.





Figure 14: Similar to Fig. 13 but for transported dust aerosol.

We also compared our retrievals with more establish methods, such as GNSS/GPS-derived PWV, and AERONET-derived PWV as below:

95 (Section 4.1: L582-587)

Table 5 summarizes the results of comparisons of DSRAD-derived PWV and GNSS/GPS-derived PWV. The magnitude of the bias error and root mean square error were small, less than 0.11 cm and less than 0.226 cm, during 2013 to 2014. Table 6 shows the errors of the retrieved PWV with the annual mean calibration constants for the rank of PWV. The bias error was larger for high PWV than it was for low

100 PWV. The magnitude of the bias errors of PWV was less than 0.163 cm for PWV < 3 cm and less than 0.339 cm for PWV > 3 cm.

(Section 4.2: L602-610)

PWV_{DSRAD+SKYMAP} using the annual mean calibration constant agreed with PWV_{MWR} (Fig. 17c). The

105 error of PWV_{DSRAD+SKYMAP} was – 0.041 < bias < 0.024 cm and RMSE < 0.212 cm for low PWV (<3 cm) and bias < – 0.356 cm and RMSE > 0.465 cm for high PWV (Table 6). Figure 17d shows that PWV_{DSRAD+SKYMAP} using the annual mean calibration constant also agreed with PWV_{Cimel} for low PWV (< 3 cm) but was smaller than PWV_{Cimel} for high PWV (> 3 cm). PWV_{MWR} was larger than PWV_{Cimel} (Fig. 17e). PWV_{DSRAD+SKYMAP} using the annual mean calibration constant was 12% and 9.1% smaller
110 than PWV_{MWR} and PWV_{Cimel}, respectively (Table 5).

Table 5: Comparison of PWV between DSRAD and other instruments.

			Slope C_1	Intercept $C_2[cm]$	γ	Bias [cm]	RMSE [cm]
PS1202091 at Tsukuba, Japan							
	Monthly mean F_0	vs GNSS/GPS receiver (2013)	0.956	0.079	0.938	-0.049	0.138
		vs GNSS/GPS receiver (2014)	0.937	0.161	0.970	-0.110	0.170
	Annual mean F_0	vs GNSS/GPS receiver (2013)	0.919	0.173	0.987	-0.061	0.226
		vs GNSS/GPS receiver (2014)	0.934	0.178	0.987	-0.089	0.223
PS2	501417 at Chiba, Ja	pan					
	Monthly mean F_0	vs MWR (2017)	0.964	0.053	0.961	-0.027	0.091
		vs AERONET (2017)	0.987	0.107	0.976	0.098	0.122
	Annual mean F_0	vs MWR (2017)	0.880	0.132	0.985	0.042	0.231
		vs AERONET (2017)	0.909	0.184	0.991	0.055	0.186

 $C_1, C_2: \text{PWV}_{\text{DSRAD}} = C_1 \times \text{PWV}_{\text{Other}} + C_2$

Bias: $PWV_{DSRAD} - PWV_{Other}$

115 Table 6: Difference in PWV between DSRAD with the annual mean calibration constants and other instruments.

	PWV _{Other}						
	0 - 1 cm	1-2 cm	2-3 cm	3-4 cm	>4 cm		
	Bias [cm]	Bias [cm]	Bias [cm]	Bias [cm]	Bias [cm]		
	(RMSE [cm])	(RMSE [cm])	(RMSE [cm])	(RMSE [cm])	(RMSE [cm])		
PS1202091 at Tsukuba, Japan							
vs GNSS/GPS receiver (2013)	0.083	0.160	0.084	-0.098	-0.339		
	(0.124)	(0.211)	(0.236)	(0.326)	(0.537)		
vs GNSS/GPS receiver (2014)	0.110	0.163	0.107	-0.055	-0.239		
	(0.142)	(0.221)	(0.251)	(0.353)	(0.492)		
PS2501417 at Chiba, Japan							
vs MWR (2017)	0.017	0.024	-0.041	-0.356	-0.594		
	(0.066)	(0.153)	(0.212)	(0.465)	(0.722)		
vs AERONET (2017)	0.088	0.118	0.017	-0.214	-0.264		
	(0.105)	(0.192)	(0.223)	(0.386)	(0.306)		

Bias: PWV_{DSRAD} - PWV_{Other}

- 4. The present algorithm splits the instrumental characterisation (F0, bandpass, FOV) and the atmospheric parameters (WV profiles), while previous approaches use mixed empirical coefficients (a and b) dependent on both the spectral bandpass and the vertical WV profile. If my understanding is correct, this would permit to use the algorithm in different conditions (place/time) compared to the ones when the instrument was calibrated. Could this be an advantage to be underlined in the conclusions?
- 125 Yes, exactly. We add the sentence in the conclusion (L631-636) as below:
- Our DSRAD algorithm retrieves PWV from the direct solar irradiance. This method does not require adjustment parameters used in the empirical methods of previous studies (e.g., Holben *et al.*, 1998; Uchiyama *et al.*, 2014; Campanelli *et al.*, 2014, 2018). Instead, the filter response function and the vertical profiles of water vapor, temperature, and pressure are required as input parameters. Thus, our
- 130 physics-based algorithm has the potential to be applied to sky-radiometers all over the world. This is the greatest advantage of the present study.

Technical corrections

135 - l. 19, "whose aerosol channels": too abrupt beginning, especially for the readers not experienced in measurements with POM photometers. I would argue that aerosols are seldom the only influencing factor at a specific wavelength, therefore "aerosol channel" sounds more like a colloquial shortcut than a technical term;

- l. 28, "aerosol channels": specify the wavelengths;

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We agree with the reviewer. We revised it (L19-26) as below:

The Prede sky-radiometer measures direct solar irradiance and the angular distribution of diffuse radiances at the ultraviolet, visible, and near-infrared wavelengths. These data are utilized for remote sensing of aerosols, water vapor, ozone, and clouds, but the calibration constant which is the sensor

145 output current of the extraterrestrial solar irradiance at the mean distance between the Earth and the sun, is needed. The aerosol channels, which are the weak gas absorption wavelengths of 340, 380, 400, 500, 675, 870, and 1020 nm, can be calibrated by an on-site self-calibration method, the Improved Langley method.

150 - l. 21-22, "by sky-radiometer remain challenge": some articles (a/the) missing;

We agree with the reviewer. We revised it (L28) as below: by the sky-radiometer remains challenging

- 1. 22-23, "generally calibrated by the standard Langley method": only for reference instruments, then 155 calibration is transferred to network instruments (as explained later in the text);
 - 1. 24, "water vapor channel": please mention the channel wavelength here;

We agree with the reviewer. We revised it (L27-31) as below:

However, the continuous long-term observation of precipitable water vapor (PWV) by the sky-160 radiometer remains challenging, because calibrating the water vapor absorption channel of 940 nm generally relies on the standard Langley method (SL) at limited observation sites (e.g., the Mauna Loa Observatory) and the transfer of the calibration constant by side-by-side comparison with the reference sky-radiometer calibrated by the SL method.

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- 1. 53, "columns of water vapor content": do you mean column concentrations? Unclear, since radiosonde also measure vertical profiles;

It means "precipitable water vapor". We revised it (L76).

170

- 1. 60, "these previous studies": even the studies by Fowle («1992)?

We intended "these previous studies" points "SKYNET sky-radiometer (Campanelli et al., 2014, 2018; Uchiyama et al., 2014, 2018a), and AERONET sun-sky photometer (Holben et al., 1998)". Therefore, 175 we revised it (L84-87) as below:

Previous studies of SKYNET and AERONET derived PWV from the observed transmittance of water vapor (\bar{T}_{H20}) , assuming $\bar{T}_{H20} = e^{-a(m \cdot w)^b}$ (Bruegge *et al.*, 1992), where *a* and *b* are adjustment parameters, *m* is the optical air mass, and *w* is PWV.

- 1. 64: do you really mean "radiometric calibration" (as in F0) or, e.g., "spectral sensitivity"? 180

We intend "spectral sensitivity of spectroradiometer". We revised it (L87-89) as below:

However, there is a known noticeable uncertainty in the estimate of PWV because the adjustment parameters depend on the spectral sensitivity of the spectroradiometer as well as the vertical profiles of water vapor and temperature.

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- 1. 75, "Sky-radiometer": article (the) missing?

We revised it (L102) as below:

The sky-radiometer models POM-01 and POM-02 (Prede, Tokyo, Japan), which are ... 190

- 1. 77, "11 wavelengths": maybe a table of the channel wavelengths, together with the main extinction factors, could be useful. See also my first technical comment about the expressions "aerosol channels ... ozone channels" (e.g., even the "ozone channel" is affected by aerosol)

195 - 1. 107-108, "aerosol ... cloud ... water vapor ... ozone channels": cf. previous comments. These approximate expressions could be used only after a short explanation;

We agree with reviewer. We revised it (L102-109) and added the table of sky-radiometer specifications as below:

- 200 The sky-radiometer models POM-01 and POM-02 (Prede, Tokyo, Japan), which are deployed in the international radiation observation network SKYNET, measure solar direct irradiances and diffuse irradiances at the ultraviolet, visible, and near-infrared wavelengths. These measurements are used for the remote sensing of aerosol, cloud, water vapor, and ozone (Table 1; Takamura and Nakajima, 2004; Nakajima *et al.*, 2007). Table 1 shows the relationship between the wavelengths and the main target of
- the remote sensing. The aerosol channels are 340, 380, 400, 500, 675, 870, and 1020 nm; the water vapor channel is 940 nm; the ozone channel is 315 nm; and the cloud channels are 1225, 1627, and 2200 nm.

Wavelength [nm]	Strong gas absorption	Main target substance	POM-01 Standard	POM-02 Standard	POM-02 PS1202091	POM-02 PS2501417
315	O ₃	Ozone	0	0	—	0
340	—	Aerosol	_	0	0	0
380	—	Aerosol	_	0	0	0
400	—	Aerosol	0	0	0	0
500	—	Aerosol	0	0	0	0
675	—	Aerosol	0	0	0	0
870	—	Aerosol	0	0	0	0
940	H_2O	Water vapor	0	0	0	0
1020	—	Aerosol	0	0	0	0
1225	O_2 , CO_2 , H_2O	Cloud	_	—	0	—
1627	CH_4 , CO_2	Cloud	_	0	0	0
2200	CH_4, H_2O	Cloud	_	0	0	0

Table 1: Sky-radiometer specifications. Each sky-radiometer is equipped with a filter indicated by210a circle. "Standard" is the standard specification of sky-radiometer models POM-01 and POM-02.

- 1. 79, "observation ... self-calibration": a bit confusing, please reformulate to avoid mixing of observation and calibration procedures;

215 - 1. 80, "works in turbid atmospheric conditions": "only" in turbid conditions or "also" in turbid conditions?

We agree with the reviewer. We revised it (L109-114) as blow:

Through on-site self-calibration of the aerosol channels by the Improved Langley (IL) method (Tanaka

220 *et al.*, 1986; Nakajima *et al.*, 1996; Campanelli *et al.*, 2004, 2007), the SKYNET system is capable of long-term and continuous aerosol observation. The IL method works not only in clean atmospheric conditions, but also in turbid atmospheric conditions.

- 1. 81-82, "standard ... modified": please, define what a "standard" method and a "modified" one are;

225

The standard and modified Langley methods are developed by Uchiyama et al., (2014) and Campanelli et al. (2014), respectively. We revised it (L115-116) as below:

However, no improved calibration method has replaced the standard (Uchiyama *et al.*, 2014) or modified (Campanelli *et al.*, 2014, 2018) Langley methods for the water vapor channel.

230

250

- 1. 93, "two SKYNET sites": explain why these two sites were selected. Do they have any particular characteristics, or was this choice oriented by the co-located instrumentation?

We chose sites which had been installed not only the sky-radiometer, but also AERONET sun-sky radiometer, GPS/GNSS receiver, and/or MWR. We added the sentence at the end of Section 1 (L129-130) as below:

At these two sites, PWV is observed by the GNSS/GPS receiver, MWR, or AERONET sun-sky radiometer other than the sky-radiometer.

240 - 1. 103, "We explain normalized radiance": article missing?

We agree with the reviewer. We revised it (L142) as below: We explain the normalized radiance...

- Eq. (1): specify earlier in the text that this holds only for a plane-parallel nonrefractive atmosphere (l. 122, now);

We agree with the reviewer. We revised it (L154-162) as below: In the plane-parallel non-refractive atmosphere, *F* at the bottom of the atmosphere (BOA) at the solar zenith angle (SZA) θ_0 and the solar azimuth angle ϕ_0 is derived from

$$F(\lambda) = \frac{F_0}{d^2} \exp\left(-m_0 \tau(\lambda)\right), \quad (1)$$

where F_0 is the calibration constant; *d* is the distance between Earth and the sun (AU); λ is the 255 wavelength; τ is the total optical thickness; and m_0 is optical air mass, represented as $m_0 = 1/\cos\theta_0$.

- Eq. (2): if L is defined as sky radiance (l. 106), then it should be already divided by the solid view angle (omega);

260 We used the sky irradiance instead of the sky radiance in the revised manuscript.

- 1. 150: the sentence is missing its subject. It is also unclear if this limitation (the real atmosphere not being a single layer) will be addressed in the following text;

²⁶⁵ We agree with the reviewer. It is deleted and mentioned in Sect. 2.1.2 in the revised manuscript.

- 1. 152, "sensitivity of R": ... at a wavelength of 940 nm;

Yes, it is at a wavelength of 940 nm. We revised it (L193) as below:

270 We examined the sensitivity of R at 940 nm ...

- 1. 154: I guess that these AOD values refer to 940 nm, too?

Yes exactly. We revised it (L199-201) as below:

Figure 3 shows the dependencies of *R* in the almucantar plane on PWV for continental average aerosol with aerosol optical thicknesses of 0.02 and 0.20 at 940 nm.

- l. 155-156, "the aerosol optical thickness does not affect this relationship": unclear, since R decreases, but the values do depend on AOD;

280

We agree with the reviewer. The relationship that R decreases with an increase of PWV was seen in both low and high AOT cases. We revised it (L201-203) as below: *R* decreases with increasing PWV regardless of the aerosol optical thickness.

- 1. 164: I would not define such a change as "drastic". The variation is only visible in the lower subfigures with a linear y-axis (please, put some letters next to the subplots), and mainly for PWV<=2 cm (as explained later in the text);

It is deleted the word "drastic" in the revised manuscript (L212).

290

- 1. 188: is "SKYMAP" an acronym?

No, it isn't.

295 - 1. 197, "transmittance of the total extinction": isn't just "trasmittance" enough?

We agree with the reviewer. We revised it (L248).

- Eq. (8): please, explain where the 1.65 factor comes from;

300

This factor, which is defined as η in the revised manuscript, was determined in consideration of the smoothness of the VSD. It is written in the Appendix A in the revised manuscript.

Appendix A: Width of the volume size distribution

Because $\frac{dV(r)}{d\ln r}$ is expressed by the superposition of 20-modal lognormal size distributions (Eq. [6]), the 305 width of $\frac{dV(r)}{d\ln r}$ is larger than that of each lognormal size distribution. The width of the lognormal size distribution should be small to deal with the complicated and step variations in $\frac{dV(r)}{d\ln r}$. However, $\frac{dV(r)}{d\ln r}$ cannot represent a natural curve if η is large and *s* is small (Fig. A1). Hence, we have to find the maximum value of η for making $\frac{dV(r)}{d\ln r}$ a natural curve. When C_i is constant, such value of η minimizes the roughness of $\frac{dV(r)}{d\ln r}$, and $\frac{dV(r)}{d\ln r}$ approaches to a flat shape. For a simple formulation, we consider the 310 function A(x) which consists of the multimodal normal distribution function B_i with a constant height. A(x) and B_i are expressed as

$$A(x) = \sum_{i=-\infty}^{\infty} B_i(x) = \sum_{i=-\infty}^{\infty} \exp\left[-\frac{\eta^2}{2} \left(\frac{x-i\xi}{\xi}\right)^2\right], \quad (A1)$$

315 where $i\xi$ and $\frac{\xi}{\eta}$ are the mean and standard deviation, respectively. Its differential is written as

$$\frac{dA}{dx} = \sum_{i=-\infty}^{\infty} \frac{dB_i}{dx} = \sum_{i=-\infty}^{\infty} -\eta^2 \left(\frac{x-i\xi}{\xi}\right) \exp\left[-\frac{\eta^2}{2} \left(\frac{x-i\xi}{\xi}\right)^2\right].$$
(A2)

When the shape of A(x) approaches to be flat, the difference between local maximum and minimum 320 values of A(x) is approximately 0. Because $\frac{dB_i}{dx}$ equals 0 at $x = j\xi$ ($j \in \mathbb{Z}$), A(x) has the local maximum and minimum at $x = j\xi$ and $\left(j + \frac{1}{2}\right)\xi$ in $j \le \frac{x}{\xi} < j + 1$. The difference Δ between the local maximum and minimum values is obtained as

$$\Delta = 1 - \frac{A\left(\frac{2j+1}{2}\xi\right)}{A(j\xi)}.$$
 (A3)

325

Figure A2 shows the relation between η and Δ . The value of Δ increases drastically at around $\eta = 1.5$. in addition, the shape of $\frac{dV(r)}{d\ln r}$ is unnatural when $\eta = 2.0$ (Fig. A1). Therefore, the value of η should be selected from the values around $\eta = 1.5$. In this study, we fixed η at 1.65. This value represents the natural curve of $\frac{dV(r)}{d\ln r}$ and satisfies that the value of Δ is small enough, $\Delta = 3.0 \times 10^{-3}$.

330



Figure A1: Relationship between the volume size distribution and η . The black line is the volume size distribution, which is computed by the integration of 20-modal lognormal distribution functions (red lines). Blue circles are the peak volume of lognormal size distribution.



Figure A2: Relationship between the parameter η and the difference Δ .

340 - 1. 218, "local minimum": does this local minimum change at every retrieval, then?

Yes, exactly. The local minimum of the VSD is calculated at every retrieval.

- 1. 338: specify that the integral of the filter response function was normalised to 1 (not its maximum);

345

The response function is not normalized to 1. We revised it (L386) as below:

$$\bar{T}_{\rm H2O} = \frac{\int_{\Delta\lambda} \Phi(\lambda) T_{\rm H2O}(\lambda) d\lambda}{\int_{\Delta\lambda} \Phi(\lambda) d\lambda} = \frac{\int_{\Delta\lambda} \Phi(\lambda) \exp(-m_{\rm H2O}(\theta) \int_0^z \alpha_{\rm H2O}(g_w(z), K(z), \lambda) dz) d\lambda}{\int_{\Delta\lambda} \Phi(\lambda) d\lambda}$$
(22),

Eq. (25)-(26): it could be useful to use a subscript (j?) for the single F0's. Also, please use another
letter instead of w (in w_H);

We agree with the reviewer. We changed it (L408-417) as below:

The mean value of the calibration constant at the water vapor channel is determined by the robust statistical and iterative method with Huber's M-estimation:

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$$\ln \bar{F}_0 = \sum_i \beta_{\rm H}(t_i) \cdot \ln F_0(t_i), \quad (25)$$

$$\beta_{\rm H}(t_i) = \begin{cases} 1 \quad (|\ln \bar{F}_0 - \ln F_0(t_i)| \le 0.03) \\ \frac{0.03}{|\ln \bar{F}_0 - \ln F_0(t_i)|} \quad (|\ln \bar{F}_0 - \ln F_0(t_i)| > 0.03), \end{cases}$$
(26)

360

where \overline{F}_0 is the mean calibration constant and is calculated at each iterative step, $F_0(t_i)$ is the calibration constant at a specific time *t*, and $\beta_{\rm H}$ is Huber's weight function.

Eq. (28)-(29): perhaps it would be better to identify the indices with other letters than R (already used
for radiances). Also, it should be mentioned that the range of scattering angles for the calculation of index 2 changes during the day;

We agree with the reviewer. We revised it (L430-442) as below:

Next, the running mean of the time series of $\bar{R}_{near}(t)$ with a window of three consecutive data points is 370 calculated as $\langle \bar{R}_{near}(t) \rangle$. Index 1 is defined as the deviation $\tilde{R}_{near}(t)$ of $\bar{R}_{near}(t)$ from $\langle \bar{R}_{near}(t) \rangle$,

$$\tilde{R}_{\text{near}}(t) = |\bar{R}_{\text{near}}(t) - \langle \bar{R}_{\text{near}}(t) \rangle| / \langle \bar{R}_{\text{near}}(t) \rangle.$$
(28)

Index 2 is the deviation \tilde{R}_{far} of normalized angular distributions far from the sun and is defined as 375

$$\tilde{R}_{\text{far}}(t) = \sigma\left(\frac{R(\Theta, t) - \langle R_{\text{far}}(\Theta, t) \rangle}{\langle R_{\text{far}}(\Theta, t) \rangle}\right), \Theta > 10^{\circ}, \quad (29)$$

where $\langle R_{\text{far}}(\Theta, t) \rangle$ is the running mean of $R(\Theta_i, t)$ with a window of three consecutive data points, and $\sigma(\mathbf{X})$ is the standard deviation of data set \mathbf{X} . Note that the data for calculating \tilde{R}_{far} varies depending on SZA, which limits available scattering angles.

- 1. 390, 1. 397 and Table 2, "misjudged"/"incorrect": it is unclear whether the cloud screening criterium correctly works for the portion of the sky seen by the photometer or not. In the first case, the algorithm does its job, and I think that "misjudged"/"incorrect" are misleading terms, since the conditions of whole sky should not be considered as reference;

385

380

"misjudged" and "incorrect" were revised. And "clear-sky" and "cloud affected" were changed to "best condition" and "poor condition" in the revised manuscript (L461-463).

390 - 1. 396-397: are <1 and >2 oktas?

It means cloud cover the range from 0 (no cloud) to 10 (cloud). We used the percentage of cloud cover for whole sky instead of previous "cloud cover the range from 0 (no cloud) to 10 (cloud)". We revised it.

- 1. 407, "line regression": do you mean linear regression using AOD and wavelength (not logs)?

"line regression" means "linear interpolation in the log-log plane". We revised it (L473).

- Sect. 3: were only "aerosol channels" (1. 107) + "water vapor channel" used in the synthetic retrieval?

Yes, Sect. 3 conducted intensive sensitivity tests using aerosol channels and the water vapor channel.

- 1. 464-465: are the -10% and -3% deviations within the expected uncertainty? If not, can you explain these results? Please, use a proper number of significant digits;

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We consider that the difference in the value of the calibration constant between the SKYMAP algorithm and the side-by-side comparison with the reference sky-radiometer was attributable mainly to the calibration period. The calibration constant of the sky-radiometer has seasonal variation due to the temperature dependency of the sensor output (Uchiyama *et al.*, 2018a). Calibration by side-by-side comparison with the reference sky-radiometer was performed only in the winter. However, the calibration constant of the SKYMAP algorithm was the annual mean. The above description is added in the revised manuscript (L551-556).

- 1. 472: maybe it would be more scientifically correct to plot these values anyway (with another
415 colour/marker) even though they will not be considered;

We calculated the monthly mean calibration constant in all of observation periods (Fig. 15a, 16a, 17a). However, we did not retrieve PWV using the monthly mean calibration constants for June and July 2014 because their values were obviously small, and because little data were successfully retrieved due

420

contaminated by clouds. The above description is added in the revised manuscript (L565-568).

- 1. 478: a more natural choice would be to linearly interpolate the monthly calibrations. The authors certainly have good reasons for considering the annual mean value, can they elaborate on this?

to the wet and cloudy conditions in the summer. In addition, it is possible that the measurements were

425

In this study, we used the annual mean calibration constant due to two factors. First, the monthly mean calibration constants were not significant changed in the dry seasons. Second, the accuracy of the calibration constant decreases at the transition between the wet season and the dry season because little data were successfully retrieved due to the wet and cloudy conditions.

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- 1. 518, "much more": "larger"?

Yes. We revised it (L639-640) as below:

Larger retrieval errors occurred in the cases when PWV was >2 cm because PWV became less sensitive to the normalized angular distribution.

- Figs. 1 and 7: the authors should explain the difference between the straight boxes and the rounded ones, and why the latter were not used in Fig. 7;

440 We intend square boxes show the operation of the calculation and input/output parameters and rounded boxes show the operation of the algorithm. We wrote the explanation in the caption of Figures.

- Fig. 2: the label "Principal plane" should be put lower, on the principal place circumference;

445 We agree with the reviewer. We revised it as below:



Figure 2: Observation planes (almucantar and principal planes) of the sky-radiometer.

450 - Fig. 3: mention somewhere that the plots refer to the principal plane;

Figure 3 in the discussion paper show the normalized radiance in the almucantar plane. We revised it.



Figure 3: Normalized angular distributions simulated for continental average aerosol (Table 2) in the almucantar plane with aerosol optical thicknesses of 0.02 and 0.20 at 940 nm. Simulations were conducted for SZA = 70° and PWV (w) = 0, 1, 2, 3, 4, and 5 cm. The top row is the normalized radiance $R(w, \Theta)$, and the bottom row is the ratio of $R(w, \Theta)$ to $R(0, \Theta)$. S-S Approx. is single scattering approximation.

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- Fig. 10: colours in the second row are hardly distinguishable. Explain that they overlap (in the caption);

We agree with the reviewer. We added it in the caption of Figs. 11 and 12 in the revised manuscript as below:

Note that the blue, red, green, and black lines in the middle row overlap.

- Table 3, "Retrieved the PWV": "PWV retrieval"?

470 We agree with the reviewer. We revised it as below:Table 4: References and methodologies of the DSRAD algorithm.

	DSRAD
Solar coordinates	Nagasawa (1999)
Refraction correction	Nagasawa (1999)
Sun-Earth distance	Nagasawa (1999)
Optical mass	Gueymard (2001)
Rayleigh scattering	Fröhlich and Shaw (1980); Young(1981)
Ozone absorption	Sekiguchi and Nakajima (2008)
Water vapor absorption	Sekiguchi and Nakajima (2008)
Filter response function	Stepwise function
Retrieval of PWV	Newton-Raphson method

Development of on-site self-calibration and retrieval methods for sky-radiometer observations of precipitable water vapor

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Abstract.

The Prede sky-radiometer measures direct solar irradiance and the angular distribution of diffuse

- 20 radiances at the ultraviolet, visible, and near-infrared wavelengths. These data are utilized for remote sensing of aerosols, water vapor, ozone, and clouds, but the calibration constant which is the sensor output current of the extraterrestrial solar irradiance at the mean distance between the Earth and the sun, is needed., whose aerosol channels are calibrated by on-site measurements (the Improved Langley method), has The aerosol channels, which are the weak gas absorption wavelengths of 340, 380, 400,
- 25 <u>500, 675, 870, and 1020 nm, can be calibrated by an on-site self-calibration method, the Improved Langley method. This on-site self-calibration method is usefulbeen used for the continuous long-term observation of aerosol properties. However, the continuous long-term observation of precipitable water vapor (PWV) by the sky-radiometer remains challenginge, because the calibrating the water vapor channel of 940 nm is-generally reliescalibrated by on the standard Langley method</u>
- 30 (SL) at limited observation sites (*e.g.*, the Mauna Loa Observatory) and the transfer of the calibration constant by side-by-side comparison with the reference sky-radiometer calibrated by the SL method. In this study, we developed the SKYMAP algorithm, a new on-site self-calibration-method for of selfcalibrating the water vapor channel of the Prede sky-radiometer using diffuse radiances normalized by direct solar irradiance (normalized radiances). Because the sky-radiometer measures direct solar
- 35 <u>irradiance and diffuse radiance using the same sensor, the normalization cancels the calibration constant</u> <u>included in the measurements.</u> The SKYMAP algorithm consists of three steps. First, aerosol optical and microphysical properties are retrieved using direct solar irradiances and the normalized diffuse radiances at aerosol channels. The aerosol optical properties at the water vapor channel are interpolated from those at aerosol channels. Second, the transmittance of PWV is retrieved using the diffuse radiance
- 40 normalized to the direct solar irradiance the angular distribution of the normalized radiances at the water vapor channel, which does not need the calibration constant. Third, the calibration constant at the water vapor channel is estimated from the transmittance of PWV and aerosol optical properties. Intensive sensitivity tests of the SKYMAP algorithm using simulated data of the sky-radiometer showed that the calibration constant is retrieved reasonably well for PWV < 2 cm, indicating which indicates that the
- 45 SKYMAP <u>algorithm</u> can calibrate the water vapor channel on-site in dry conditions. Then <u>Next</u>, the SKYMAP <u>algorithm</u> was applied to actual measurements <u>under the clear-sky and low PWV (< 2 cm)</u> <u>conditionsin the dry season</u> at two sites, (Tsukuba and Chiba, Japan, and the annual mean calibration <u>constants at the two sites were determined.</u>). Because the SKYMAP algorithm is useful for clear-sky and low PWV (< 2 cm) conditions, the water vapor channel was calibrated for the dry season. The
- 50 <u>SKYMAP-derived calibration constants were 10.1% and 3.2% lower, respectively, than those</u> <u>determined by side-by-side comparison with the reference sky-radiometer.</u> After determining the calibration constant, <u>we obtained PWV is able to be retrieved using from the</u> direct solar irradiances for <u>the whole yearin both the dry and wet seasons</u>. The retrieved PWV values corresponded well to those

derived from a Global Navigation Satellite System (GNSS)/Global Positioning System (GPS) receiver,

- 55 a microwave radiometer, and an AERONET sun-sky radiometer at both sites. The (correlation coefficients were greater than? > 0.96. We calculated the bias errors and the root mean square errors by comparing PWV between the DSRAD algorithm and other instruments. The magnitude of the bias error and the root mean square error were < 0.163 cm and < 0.251 cm for PWV < 3 cm, respectively. However, our method tended to underestimate PWV in the wet conditions, and the magnitude of the</p>
- 60 bias error and the root mean square error became large, < 0.594 cm and < 0.722 cm for PWV > 3 cm, respectively.), This problem was mainly due to the overestimation of the aerosol optical thickness before the retrieval of PWV. These results showindicating that the Prede sky-radiometer provides both aerosol andSKYMAP algorithm enables us to observe PWV over the long term, data based on its unique on-site self-calibration methods.

1 Introduction

The highly variable spatiotemporal distributions of aerosols, clouds, and gases (*e.g.*, water vapor and ozone) still include large uncertainties for the quantitative understanding of the Earth's radiation budget at various spatial and temporal scales. Water vapor is specified as an essential climate variable (ECV)

- 70 by the World Meteorological Organization (WMO), a critical key parameter that contributes to characterizing Earth's climate and changes in atmospheric temperature (Schmidt *et al.*, 2010). Water vapor absorbs visible radiation and absorbs and emits infrared radiation to heat and cool the Earth and its atmosphere. Atmospheric heating drives the evaporation of sea water, causing an increase in temperature as positive feedback (IPCC, 2013). In addition, the distribution of water vapor controls precipitation amounts and aerosol-cloud interactions (Twomey, 1990). To understand these effects quantitatively, many previous studies have measured precipitable columns of water vapor content by using a radiosonde, Global Navigation Satellite System (GNSS)/Global Positioning System (GPS) receiver (Bevis *et al.*, 1992), or spectroradiometer (*e.g.*, Fowle, 1912, 1915).
- Precipitable water vapor (PWV), which is the total atmospheric water vapor contained in a vertical column, has been estimated from the measurement of direct solar irradiance at the water vapor 80 channel absorption bands. One of the strong water vapor absorption bands isat around 940 nm and can be measured by sun photometer (Fowle, 1912, 1915; Bruegge et al., 1992; Schmid et al., 1996, 2001; Halthore et al., 1997), SKYNET sky-radiometer (Campanelli et al., 2014, 2018; Uchiyama et al., 2014, 2018a), and AERONET sun-sky photometer (Holben et al., 1998). These pPrevious studies of SKYNET and AERONET –derived PWV from the observed transmittance of water vapor (\bar{T}_{H20}), 85 assuming $\bar{T}_{H20} = e^{-a(m \cdot w)^b}$ (Bruegge *et al.*, 1992), where *a* and *b* are <u>adjustment adjusting</u>-parameters, m is the optical air mass, and w is PWV. However, there is a known noticeable uncertainty in the estimate of PWV because the adjustment parameters depend on radiometric calibration the spectral sensitivity of the spectroradiometer as well as the vertical profiles of water vapor and temperature. Therefore, the adjustment parameters should be determined for each observation site. Therefore, 90 Campanelli et al. (2014, 2018) developed a practical method for determining the adjustment adjusting parameters based on PWV retrieved by a GNSS/GPS receiver or by surface humidity observations.

To estimate PWV using a spectroradiometer, it is necessary to calibrate the water vapor channel. The calibration constant, which is the sensor output current of the extraterrestrial solar irradiance at the mean distance between the Earth and the sun, at the water vapor channel can be determined by the Langley method. For example, Uchiyama *et al.* (2014) calibrated the water vapor channel of a skyradiometer with high accuracy using observations from the Mauna Loa Observatory (3400 m a.s.l.). In the AERONET led by NASA, the field instrument of the AERONET sun-sky radiometer is calibrated every year by lamp calibration and side-by-side comparison with a reference spectroradiometer (Holben 100 *et al.*, 1998). Dedicated effort and <u>cost-expenses</u> are required <u>for maintainingto maintain</u> accurate longterm calibrations using these methods.

The Ssky-radiometer models POM-01 and POM-02 (Prede, Tokyo, Japan), which is are deployed in the international radiation observation network SKYNET, measures solar direct irradiances and angular distributions of diffuse irradiances at the ultraviolet, visible, and near-infrared 105 wavelengths.11 wavelengths including These measurements are used for the remote sensing of aerosol, cloud, water vapor, and ozone channels (Table 1; Takamura and Nakajima, 2004; Nakajima et al., 2007). Table 1 shows the relationship between the wavelengths and the main target of the remote sensing. The aerosol channels are 340, 380, 400, 500, 675, 870, and 1020 nm; the water vapor channel is 940 nm; the ozone channel is 315 nm; and the cloud channels are 1225, 1627, and 2200 nm. Through on-site self-calibration of the aerosol channels by the Improved Langley (IL) method (Tanaka et al., 110 1986; Nakajima et al., 1996; Campanelli et al., 2004, 2007), The SKYNET system is capable of longterm and automatic continuous aerosol observation. through an on-site self-calibration method, called the Improved Langley (The IL) method, which -works not only in clean atmospheric conditions, but also in turbid atmospheric conditions (Tanaka et al., 1986; Nakajima et al., 1996; Campanelli et al., 2004, 2007). However, no improved calibration method has replaced the standard (Uchiyama et al., 115 2014) or modified (Campanelli et al., 2014, 2018) Langley methods for the water vapor channel. In this study, we devised developed a new method of retrieving PWV using the PWV dependency of the normalized radiance, defined as the ratio of diffuse radiance to direct solar irradiance at the water vapor channel. This method enables us to estimate PWV without the calibration constant, and to perform on-120 site self-calibration of the water vapor channel. We developed two algorithms, SKYMAP and DSRAD. The SKYMAP algorithm is a new on-site method for self-calibrating the water vapor channel. It retrieves PWV (PWV_{SKYMAP}) from the angular distribution of the normalized radiance at 940 nmthe

- water vapor channel and calibrates the water vapor channel. The DSRAD algorithm estimates the PWV (PWV_{DSRAD}) from the calibrated direct solar irradiance at 940 nmthe water vapor channel. This method
 does not require adjustment parameters and explicitly uses the filter response function and the vertical profiles of water vapor, temperature, and pressure. The SKYMAP and DSRAD algorithms are described in Section 2. We discuss the results of sensitivity tests of the two-SKYMAP algorithms using simulation data in Section 3 and apply the two algorithms to observational data at two SKYNET sites in Section 4. At these two sites, PWV is observed by the GNSS/GPS receiver, MWR, or AERONET sup-
- 130 <u>sky radiometer other than the sky-radiometer. The retrieval accuracy of our method is evaluated by</u> comparison to these established methods.

2 Methods

In this study, PWV is retrieved using angular distributions of the normalized radiance, which does not require the calibration constant of the sky-radiometer. Section 2.1 shows the normalized radiances and

135 dependencies of the normalized radiance on PWV. Next, we describe two algorithms, the flow and relationships of which are shown in Fig. 1. The SKYMAP algorithm retrieves aerosol optical and microphysical properties and calibrates the water vapor channel by retrieving PWV from the angular distribution of the normalized radiance (Section 2.2). The DSRAD algorithm retrieves PWV from the transmittance derived from the direct solar irradiance at the water vapor channel (Section 2.3).

140 **2.1 Sky-radiometer measurements and <u>the</u> relation<u>ship</u> between normalized radiances and the PWV**

We explain <u>the normalized radiance (Nakajima *et al.*, 1996)</u> in Section 2.1.1 and the theoretical relationship between <u>the normalized radiances</u> and PWV in Section 2.1.2.

2.1.1 Sky-radiometer measurements

The direct solar irradiance (F) and angular distribution of the_diffuse radiance_irradiance (LV) are measured at seven wavelengths by the model POM-01 or 11-eleven wavelengths by the model POM-02, including aerosol (λ = 340, 380, 400, 500, 675, 870, and 1020 nm), cloud (1627 and 2200 nm), water vapor (940 nm), and ozone (315 nm) channels (Table 1). LV is measured in the almucantar and principal planes (Fig. 2). The angular distribution of LV is measured at scattering angles Θ = 2°, 3°, 4°, 5°, 7°, 10°, 15°, 20°, 25°, 30°, 40°, 50°, 60°, 70°, 80°, 90°, 100°, 110°, 120°, 130°, 140°, 150°, and 160° in the almucantar and principal planes, every 10 min. Aerosol-The aerosol channels are calibrated with the IL method using the normalized radiance at Θ < 30°. F and LV(Θ≥4°) at the aerosol and water vapor channels are used in this study.

In the plane-parallel non-refractive atmosphere, FThe direct solar irradiance at the bottom of the atmosphere (BOA) at the solar zenith angle (SZA) θ_0 and the solar azimuth angle ϕ_0 is derived from

$$F(\lambda) = \frac{F_0}{d^2} \exp\left(-m_0 \tau(\lambda)\right), \quad (1)_{\overline{s}}$$

where F_0 is the calibration constant, which is the sensor output current of the direct solar irradiance at the top of the atmosphere (TOA) when the distance between Earth and the sun is 1 AU; *d* is the distance between Earth and the sun (AU); λ is the wavelength; τ is the total optical thickness; and m_0 is optical air mass, represented as $m_0 = 1/\cos\theta_0$ in the plane-parallel nonrefractive atmosphere. In clear-sky conditions, the total optical thickness is the integrated value of _-consists_of_aerosol scattering + /absorption, Rayleigh scattering, and gas absorption coefficients-and integrated from BOA to TOA in the 165 <u>column</u>. Assuming a narrow spectral band filter response function, the normalized radiance (*R*), which is the ratio of \underline{LV} to *F* at the zenith angle (θ) and the azimuth angle (ϕ), is obtained from the radiative transfer equation:

$$R(\Theta,\lambda) = \frac{4V(\Theta,\lambda)}{F(\lambda)m_0\Delta\Omega} = \int_0^{\tau(\lambda)} \exp\left[\left(\tau - \tau'\right)\left(\frac{1}{\mu_0} - \frac{1}{\mu}\right)\right] \omega'(\lambda,\tau')P'(\Theta,\lambda,\tau')d\tau' + Q(\Theta,\lambda), \quad (2)_{\overline{\tau}}$$

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where $P'(\Theta, \lambda, \tau')$ and $\omega'(\lambda, \tau')$ are, the total phase function and the total single scattering albedo, respectively, at the altitude $\tau = \tau'$, $\Delta\Omega$ is the solid view angle (or field of view); Q is the multiple scattering contribution; and

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$$\cos \Theta = \cos \theta \cos \theta_0 + \sin \theta \sin \theta_0 \cos(\phi - \phi_0)_2 \quad (3)_{\overline{2}}$$
$$\mu = \cos \theta; \mu_0 = \cos \theta_0$$

Noted that F₀ is cancelled by the normalization. In the second term of Eq. (2), the solid view angle of each wavelength can be retrieved from the angular distribution around the solar disk (Nakajima *et al.*, 1996; Boi *et al.*, 1999; Uchiyama *et al.*, 2018b). Eq. (2) can be simplified in the almucantar plane due to θ = θ₀:

$$R(\Theta,\lambda) = \int_0^{\tau(\lambda)} \omega'(\lambda,\tau') P'(\Theta,\lambda,\tau') d\tau' + Q(\Theta,\lambda) = \omega(\lambda)\tau(\lambda)P(\Theta,\lambda) + Q(\Theta,\lambda)_{\frac{1}{2}}(4)_{\frac{1}{2}}$$

185 where $P(\Theta, \lambda)$ and ω are the total phase function and the total single scattering albedo, respectively. In contrast, <u>*R*normalized radiances</u> in the principal plane can be described simply, similar to Eq. (4), if we assume that the atmosphere is a single layer:

$$R(\Theta,\lambda) = \frac{\mu_0^2}{\mu_0 - \mu} \omega(\lambda) P(\Theta,\lambda) \left[1 - \exp\left(\frac{\tau(\lambda)}{\mu_0} - \frac{\tau(\lambda)}{\mu}\right) \right] + Q(\Theta,\lambda) \, . \tag{5}$$

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Noted that real atmosphere is not a single layer (Torres et al., 2014).

2.1.2 The relationship between normalized radiances at the water vapor channel and PWV

We examined the sensitivity of *R* at 940 nm in the two observation planes to PWV, aerosol optical properties, and aerosol vertical profiles by simulating *R* using the radiative transfer model RSTAR
(Nakajima and Tanaka, 1986, 1988). The simulation was conducted with two aerosol types based on those used by Kudo *et al.* (2016): the continental average, and the continental average + transported dust in the upper atmosphere (Table 2). The continental average consisted of water-soluble particles,

soot particles, and insoluble particles (Hess et al., 1999). Transported dust was defined as the mineraltransported component from Hess et al. (1999). Figure 3 shows the dependencies of R in the almucantar

- plane on PWV for continental average aerosol (Table 1) with aerosol optical thicknesses of 0.02 and 200 0.20 at 940 nm. The simulations were conducted for at an the SZA of 70° in the almucantar plane. R decreases with increasing PWV regardless of the aerosol optical thickness, and the aerosol optical thickness does not affect this relationship. This suggests that PWV can be estimated from the normalized angular distribution, which is the angular distribution of *R*, without the calibration constant.
- 205 The dependencies of R on PWV cannot be observed in the radiative transfer using single scattering approximation in the almucantar plane. The first term of Eq. (4) is the normalized single scattering contribution and includes only the influences of aerosol and Rayleigh scattering. Noted that this is true only for R, and not for \underline{LV} , because total optical thickness contributes to the single scattering approximation of *LV*. However, the second term for the multiple scattering includes the influence of water vapor absorption and creates the dependencies of R on PWV. Figure 3 shows that the dependency 210 of R on PWV at the forward scattering angles is not strong, but R at the backward scattering angles
 - between 90° and 120° changes drastically with PWV. The range of the scattering angle for R is an important factor.
- Figure 4 illustrates the dependency of R on PWV for different observation planes. The 215 simulation was conducted for transported dust aerosol (Table 24) with an aerosol optical thickness of 0.06 at 940 nm at an SZA of 70° in the almucantar and principal planes. The transported dust aerosol is composed of coarse particles, which have larger impacts on the angular distribution of R at the nearinfrared wavelength than fine particles. The dependency of R in the almucantar plane on PWV is the same as in Fig. 3. The dependency of R on PWV was is also found in both observation the principal
- planes. <u>R</u> increases with increasing PWV at $\theta \ll \theta_0$ and decreases with increasing PWV at $\theta \gg \theta_0$. 220 Although the dependency of R on PWV in the almucantar plane is strong at the backward scattering angles, that in the principal plane is strong at scattering angles between 60° and 90° . R in the principal plane is more sensitive to PWV than R in the almucantar plane because the normalized single scattering contribution in Eq. (5) includes not only Rayleigh and aerosol scattering but also gas absorption.
- In theory, the maximum scattering angle of the principal plane is $\theta_0 + 90^\circ$ and that of the 225 almucantar plane is $2\theta_0$. When the SZA is small, the principal plane has a broader scattering angle range than the almucantar plane. Therefore, the principal plane is more advantageous for PWV retrieval. Figure 5 is the same as Fig. 4 but for an SZA of 30°. Because the maximum scattering angle of the principal plane is obviously larger than that of the almucantar plane, PWV retrieval using the principal plane is more effective compared to that using the almucantar plane. 230

R in the principal plane is affected by the aerosol vertical profile, but this influence can be ignored for R in the almucantar plane (Torres et al., 2014). Figure 6 shows the normalized angular distribution in the two observation planes for the different heights of the transported dust layer. It is obvious that the normalized angular distribution in the principal plane is sensitive to the aerosol vertical

235 profile. Consequently, the principal plane is useful for retrieving PWV when the aerosol vertical profile is known, but the almucantar plane is better when the aerosol vertical profile is not known. In this study, we used the normalized angular distribution in the almucantar plane because the aerosol vertical profile was not known. The influence of SZA on the retrieval of PWV is examined in Section 3.

2.2 SKYMAP algorithm

240 The SKYMAP algorithm consists of three steps (Fig. 7). First, aerosol optical and microphysical properties are retrieved from F and normalized angular distributions at aerosol channels. Second, aerosol optical properties at the water vapor channel are interpolated from those at aerosol channels. PWV is retrieved from the normalized angular distribution at the water vapor channel. Third, the calibration constant at the water vapor channel is estimated from PWV and the aerosol optical 245 properties.

2.2.1 Step 1: Retrieval of aerosol optical and microphysical properties

Aerosol optical and microphysical properties are estimated from sky-radiometer measurements at aerosol channels using normalized angular distributions and transmittance of the total extinction $T = \frac{I_{\text{eff}}Fd^2}{F_0}$ with an optimal estimation method similar to the AERONET and SKYNET retrievals (Dubovik 250 and King, 2000; Dubovik *et al.*, 2006; Kobayashi *et al.*, 2006; Hashimoto *et al.*, 2012; Kudo *et al.*, 2016). Estimated optical and microphysical properties are the real and imaginary parts of the refractive index at aerosol channels (340, 380, 400, 500, 675, 870, 1020 nmTable 1), the volume size distribution, and the volume ratio of non-spherical particles to total particles in coarse mode. Hereafter, these are referred to as aerosol parameters.

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In step 1, we construct the forward model to calculate the sky-radiometer measurements from the aerosol parameters. We assume that the aerosol volume size distribution in the radius range from 0.02 to 20.0 µm consists of 20-modal lognormal volume size spectra distributions as illustrated in Fig. 8:

$$\frac{\mathrm{d}V(r)}{\mathrm{d}\ln r} = \sum_{i=1}^{20} C_i \exp\left[-\frac{1}{2} \left(\frac{\ln r - l}{s}\right)^2\right]_{\star} \tag{6}_{\overline{s}}$$

$$\ln r_i = \ln(0.02\mu m) + \frac{2i-1}{2}\ln\Delta r_2 \qquad (7)$$

$$s \equiv \frac{\ln \Delta r}{\frac{1.65\eta^2}{2}} \qquad (8)_{\overline{2}}$$

$$\ln \Delta r \equiv \frac{1}{20} (\ln(20\mu m) - \ln(0.02\mu m)) = \frac{3}{20} \ln 10, \quad (9)_{3}$$

where C_i, r_i, and s are the volume, radius, and width of each lognormal function, respectively. <u>7</u> is the parameter to determine the width and is given by a fixed value (Appendix A). We can separate the size
distribution into fine and coarse modes by giving the boundary radius r_b, which is obtained as the local minimum. Furthermore, we separate coarse mode into spherical and non-spherical particles:

$$\frac{\mathrm{d}V(r)}{\mathrm{d}\ln r} = \frac{\mathrm{d}V_{\mathrm{f}}(r)}{\mathrm{d}\ln r} + (1-\delta)\frac{\mathrm{d}V_{\mathrm{c}}(r)}{\mathrm{d}\ln r} + \delta\frac{\mathrm{d}V_{\mathrm{c}}(r)}{\mathrm{d}\ln r^{2}}, \quad (10)_{\overline{2}},$$

275 where $\frac{dV_f(r)}{d\ln r}$ is fine mode, $\frac{dV_c(r)}{d\ln r}$ is coarse mode, and δ is the fraction of the non-spherical particles in coarse mode (Fig. 8). The aerosol optical properties are calculated from the size distribution and refractive index, similar to <u>the</u> methods of Kudo *et al.* (2016) and Dubovik *et al.* (2006), as follows:

$$\tau_{\text{ext/sca}}(\lambda) = \sum_{k} \frac{dV_{\text{f}}(r_{k})}{d\ln r} K_{\text{ext/sca}}^{\text{S}}(\lambda, n, k, r_{k}) + \sum_{k} (1 - \delta \gamma) \frac{dV_{\text{c}}(r_{k})}{d\ln r} K_{\text{ext/sca}}^{\text{S}}(\lambda, n, k, r_{k}) + \sum_{k} \gamma \delta \frac{dV_{\text{c}}(r_{k})}{d\ln r} K_{\text{ext/sca}}^{\text{NS}}(\lambda, n, k, r_{k})_{2}$$
(11)

$$\tau_{\rm sca}(\lambda)P_{ii}(\Theta,\lambda) = \sum_{k} \frac{dV_{\rm f}(r_{k})}{d\ln r} K_{ii}^{\rm S}(\Theta,\lambda,n,k,r_{k}) + \sum_{k} (1-\delta\gamma) \frac{dV_{\rm c}(r_{k})}{d\ln r} K_{ii}^{\rm S}(\Theta,\lambda,n,k,r_{k}) + \sum_{k} \frac{\gamma\delta}{d\ln r} \frac{dV_{\rm c}(r_{k})}{d\ln r} K_{ii}^{\rm NS}(\Theta,\lambda,n,k,r_{k})$$
(12)

- where $\tau_{\text{ext/sca}}(\lambda)$ denotes the optical thickness of extinction and scattering, and $\tau_{\text{sca}}(\lambda)P_{ii}(\Theta,\lambda)$ denotes the directional scattering corresponding to the scattering matrix elements $P_{ii}(\Theta,\lambda)$. K^{S} and K^{NS} are the kernels of extinction and scattering properties for spherical and non-spherical particles, respectively. *n* and *k* are the real and imaginary parts of the refractive index, respectively. We use randomly oriented spheroids as non-spherical particles and use the kernels developed by Dubovik *et al.* (2006).
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We compute normalized angular distributions and transmittances of the extinction, using the radiative transfer model RSTAR (Nakajima and Tanaka, 1986, 1988). The model atmosphere is divided by 18 boundary layers altitudes of at 0, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 30, 40, 50, 70, and 120 km. Atmospheric vertical profiles of temperature and pressure are obtained from the NCEP/NCAR Reanalysis 1 data. The absorption coefficients of H₂O, CO₂, O₃, N₂O, CO, CH₄, and O₂ are calculated by the correlated *k*-distribution method from the data table of Sekiguchi and Nakajima (2008).

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The aerosol parameters for the best fit to all measurements (normalized angular distributions and transmittances at aerosol channels) and *a priori* information are obtained by minimizing the following cost function,

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$$f(\mathbf{x}) = \frac{1}{2} (\mathbf{y}^{\text{meas}} - \mathbf{y}(\mathbf{x}))^T (\mathbf{W}^2)^{-1} (\mathbf{y}^{\text{meas}} - \mathbf{y}(\mathbf{x})) + \frac{1}{2} (\mathbf{y}_a(\mathbf{x}))^T (\mathbf{W}_a^2)^{-1} (\mathbf{y}_a(\mathbf{x}))_2$$
(13)

where vector y^{meas} describes the measurements (normalized radiances R^{meas} and transmittances of total extinction T^{meas} at aerosol channels) at the aerosol channels, vector x describes the aforementioned aerosol parameters $-n(\lambda)$, $k(\lambda)$, C_i , and δ — to be estimated, vector y(x) comprises the values corresponding to y^{meas} calculated from x by the forward model (R^{ret} and T^{ret}), and matrix W^2 is the covariance matrix of y and is assumed to be diagonal. The diagonal elements of W are the standard errors in the measurements. We set their values at 0.02 for T^{meas} , and 10% for R^{meas} .

To reduce the effects of observational error on retrieval and to conduct stable analyses, Dubovik and King (2000) considered restricting the spectral variability of the refractive indexvolume size distribution and by limiting the length of the derivatives of the refractive index derivative with respect to the wavelength and the volume size distribution. They considered this *a priori* smoothness constraint as being of the same nature as a measurement and incorporated the smoothness constraint into their retrieval scheme. We also consider the smoothness constraints in this study. The second term of Eq. (13) consists of *a priori* information on the wavelength dependencies of the refractive index, aerosol optical thickness, and smoothness of the volume spectrum, which is described as

$$\mathbf{y}_{a}(x) = \left(\mathbf{y}_{a}^{\text{Re}}, \mathbf{y}_{a}^{\text{Im}}, \mathbf{y}_{a}^{\text{Sca}}, \mathbf{y}_{a}^{\text{Abs}}, \mathbf{y}_{a}^{\text{Vol}}\right)^{T}$$
(14),

where vectors y_a^{Re} , y_a^{Im} , y_a^{Sca} , y_a^{Abs} , and y_a^{Vol} are *a priori* information on the wavelength dependencies of the refractive index (real and imaginary parts), aerosol optical thickness (scattering and absorption parts), and smoothness of the volume spectrum, respectively. The matrix W_a^2 in Eq. (13) is the covariance matrix for determining the strengths of the constrain<u>t</u>s.

We adapt the smoothness constraints of the second derivatives for the real and imaginary parts of the refractive index. The second derivatives are defined as

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$$y_{a}^{\operatorname{Re}(i)}(\boldsymbol{x}) = \left(\frac{\ln n(\lambda_{i}) - \ln n(\lambda_{i+1})}{\ln \lambda_{i} - \ln \lambda_{i+1}} - \frac{\ln n(\lambda_{i+1}) - \ln n(\lambda_{i+2})}{\ln \lambda_{i+1} - \ln \lambda_{i+2}}\right)_{a} \quad (15)$$

$$y_{a}^{\mathrm{Im}(i)}(\mathbf{x}) = \left(\frac{\ln k(\lambda_{i}) - \ln k(\lambda_{i+1})}{\ln i - \ln \lambda_{i+1}} - \frac{\ln k(\lambda_{i+1}) - \ln k(\lambda_{i+2})}{\ln \lambda_{i+1} - \ln \lambda_{i+2}}\right)_{a} (16)_{b}$$
$$(i = 1, \dots, N_{w} - 2),$$

330

where $y_a^{\text{Re}(i)}$ and $y_a^{\text{Im}(i)}$ are the *i*-th elements of the vectors y_a^{Re} and y_a^{Im} , respectively. N_w is the number of wavelengths. The values entered into the weight matrix W_a are 0.2 for the real part and 1.25 for the

imaginary part. These values are adopted from Dubovik and King (2000). Furthermore, we introduce the smoothness constraints to the spectral distributions of the scattering and absorption parts of the 335 aerosol optical thickness by

$$y_{a}^{\text{Sca}(i)}(\mathbf{x}) = \left(\frac{\ln \tau_{sca}(\lambda_{i}) - \ln \tau_{sca}(\lambda_{i+1})}{\ln \lambda_{i} - \ln \lambda_{i+1}} - \frac{\ln \tau_{sca}(\lambda_{i+1}) - \ln sca}{\ln \lambda_{i+1} - \ln sca}\right)_{\pm} \quad (17)_{5}$$

$$y_{a}^{\text{Abs}(i)}(\mathbf{x}) = \left(\frac{\ln \tau_{abs}(\lambda_{i}) - \ln \tau_{abs}(\lambda_{i+1})}{\ln \lambda_{i} - \ln \lambda_{i+1}} - \frac{\ln \tau_{abs}(\lambda_{i+1}) - \ln \tau_{abs}(\lambda_{i+2})}{\ln \lambda_{i+1} - \ln \lambda_{i+2}}\right)_{\pm} \quad (18)_{5}$$

$$(i = 1, \dots, N_{w} - 2),$$

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where $y_a^{\text{Sca}(i)}$ and $y_a^{\text{Abs}(i)}$ are the *i*-th elements of the vectors y_a^{Sca} and y_a^{Abs} , respectively. The value entered in the weight matrix W_a is 2.5 for both the scattering and absorption parts of the aerosol optical thickness. To stabilize the estimation of the volume size distribution, we introduce the smoothness constraint for the adjacent volume size spectrum C_i , as:

$$y_a^{\text{Vol}(i)}(\mathbf{x}) = (\ln C_{i-1} - \ln C_i) - (\ln C_i - \ln C_{i+1}), \quad (19),$$
$$(i = 1, \cdots, 20),$$

$$C_0 = 0.01 \times \min\{C_i | i = 1, \dots, 20\}, C_{21} = 0.01 \times \min\{C_i | r_i > r_b, i = 1, \dots, 20\}$$

where $y_a^{\text{Vol}(i)}$ is the *i*-th element of the vector y_a^{Vol} . The small values of C_0 and C_{21} at r_0 and r_{21} are given to prevent both ends of the size distribution (C_1 and C_{20}) from being abnormal values because <u>*F*</u> the solar direct irradiances and <u>*V*</u>diffuse radiances</u> do not have sufficient information to estimate the size distribution of both small ($r < 0.1 \ \mu\text{m}$) and large particles ($r > 7 \ \mu\text{m}$; Dubovik *et al.*, 2000). Note that r_0 and r_{21} satisfy Eq. (7). The value entered in the weight matrix W_a is 1.6 for the smoothness constraint of

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We minimize f(x) of Eq. (13) using the algorithm developed in Kudo *et al.* (2016), which is based on the Gauss-Newton method and the logarithmic transformations of x and y. Finally, the aerosol optical properties from aerosol channels are obtained from x using Eqs. (11) and (12).

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2.2.2 Step 2: Retrieval of PWV

the size distribution.

We estimate PWV by the following procedure. The aerosol volume size distribution is obtained from step 1, and the refractive index at 940 nm is calculated from those at 870 and 1020 nm by linear interpolation in the log-log plane. Using the size distribution and the interpolated refractive index, we can compute the aerosol optical properties and the normalized angular distribution at the water vapor channel using the forward model described in Section 2.2.1. We retrieve PWV by minimizing the following cost function:

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$$f(\mathbf{x}) = \frac{1}{2} (\mathbf{y}^{\text{meas}} - \mathbf{y}(\mathbf{x}))^T (\mathbf{W}^2)^{-1} (\mathbf{y}^{\text{meas}} - \mathbf{y}(\mathbf{x}))_2 (20)_3$$

where the component of vector x is PWV, vectors y^{meas} and y(x) are the normalized angular distribution in the range of from 4° to 160°, matrix W^2 is assumed to be diagonal, and the values of the diagonal matrix W are assumed to be 10%. The cost function is minimized by the Gauss-Newton method. Note that this process does not require the calibration constants of the sky-radiometer, because we use the normalized angular distribution (Eq. [4]) to obtain PWV instead of using the direct solar irradiance (Eq.

2.2.3 Step 3: Retrieval of the calibration constant of the water vapor channel

 F_0 at the water vapor channel can be obtained from the observed *F* and the band average transmittance 380 \overline{T}_{H2O} converted from PWV in step 2 as follows:

$$F_0 = \frac{Fd^2 e^{m \cdot (\tau_R + \tau_a)}}{\bar{T}_{H2O}}, \quad (21)_{\rm F}$$

where τ_R and τ_a are Rayleigh scattering and aerosol optical thicknesses, respectively. The band average transmittance can be written as

$$\bar{T}_{\rm H2O} = \frac{\int_{\Delta\lambda} \Phi(\lambda) T_{\rm H2O}(\lambda) d\lambda^{\pm}}{\int_{\Delta\lambda} \Phi(\lambda) d\lambda \Delta\lambda} \int_{\Delta\lambda} \Phi(\lambda) T_{\rm H2O}(\lambda) d\lambda =$$

$$\frac{\int_{\Delta\lambda} \Phi(\lambda) \exp\left(-m_{\rm H2O}(\theta) \int_{0}^{z} \alpha_{\rm H2O}(g_{w}(z), K(z), \lambda) dz\right) d\lambda}{\int_{\Delta\lambda} \Phi(\lambda) d\lambda} \frac{4}{\Delta\lambda} \int_{\Delta\lambda}^{-} \Phi(\lambda) \exp\left(-m_{\rm H2O}(\theta) \int_{\theta}^{z} \sigma_{\rm H2O}(g_{w}(z), K(z), \lambda) dz\right) d\lambda} (22)_{5}$$

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[1]).

$$w = \int_0^z g_w(z) dz_2 \qquad (23)_{\overline{z}}$$

where $-\Phi(\lambda)$ is the filter response function, $\Delta \lambda$ is the bandwidth of the filter response function, $T_{\rm H2O}$ is the transmittance of water vapor at wavelength λ , $m_{\rm H2O}(\theta)$ is the optical air mass, g_w is the mass mixing ratio, K is temperature, $\underline{\alpha} \sigma_{\rm H2O}$ is the absorption coefficient at altitude z, and w is PWV. Eq. (22) is discretized by

$$T_{\rm H2O} = \frac{\sum_{i}^{N_{s}} \Phi_{i} \int_{\Delta\lambda_{i}} \exp(-m_{\rm H2O}(\theta) \int_{0}^{z} \alpha_{\rm H2O}(g_{w}(z), K(z), \lambda) dz) d\lambda}{\sum_{i}^{N_{s}} \Phi_{i} \Delta\lambda_{i}} \frac{1}{\Delta\lambda} \sum_{i}^{N_{s}} \Phi_{i} \int_{\Delta\lambda_{i}}^{-} \exp(-m_{\rm H2O}(\theta) \int_{0}^{z} \sigma_{\rm H2O}(g_{w}(z), K(z), \lambda) dz) d\lambda}{(24)}$$

=

where Φ_i is the stepwise filter response function, Δλ_i is the sub-bandwidth of the filter response function, and N_s is the number of sub-bands. We calculate the absorption coefficients at each wavelength by the correlated k-distribution (Sekiguchi and Nakajima, 2008) using the vertical profiles of temperature,
pressure, and specific humidity in the NCEP/NCAR Reanalysis 1 data.

We can calculate a value for F_0 from a data set of the normalized angular distribution. Therefore, for example, the <u>a</u> time series of F_0 in a day is obtained from the daily measurements of the sky-radiometer. The mean value of the calibration constant at the water vapor channel is determined by the robust statistical <u>and iterative</u> method with Huber's M-estimation:

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 $\ln \bar{F}_0 = \sum_i \beta_{\rm H}(t_i) \cdot \ln F_0(t_i) \sum w_{\rm H} \cdot \ln F_0, \quad (25)_{\rm F}$

$$\mathscr{W}\beta_{\rm H}(t_i) = \begin{cases} 1 \quad (|\ln\bar{F}_0 - \ln F_0(t_i)F_{\rm T}| \le 0.03) \\ \frac{0.03}{|\ln\bar{F}_0 - \ln F_0(t_i)F_{\rm T}|} \quad (|\ln\bar{F}_0 - \ln F_0(t_i)F_{\rm T}| > 0.03)^2 \end{cases}$$
(26)

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where \overline{F}_0 is the mean calibration constant and is calculated at each iterative step, $F_0(t_i)$ is the calibration constant at a specific time \underline{t} , and $\underline{w}\beta_{\rm H}$ is Huber's weight function.

2.2.4 Cloud screening using the smoothness criteria of the angular distributions (SCAD method)

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The SKYMAP algorithm can only be applied to measurements under clear-sky conditions. We estimated clear-sky conditions from two indexes calculated from sky-radiometer measurements. Index 1 is a value for diffuse radiances the normalized radiances near the sun. If clouds pass over the sun, index 1 has large temporal variation. Index 2 is a value for the normalized angular distribution. If clouds are detected on the scanning plane of the sky-radiometer, the normalized angular distribution has large variation. Index 1 is defined as follows. First, the mean normalized radiance near the sun \bar{R}_{near} is calculated by

$$\bar{R}_{\text{near}}(t) = \frac{1}{N} \sum_{i=1}^{N} R(\Theta_i, t), \Theta \le 10^{\circ}, \qquad (27)_{\overline{2}}$$

where *N* is the number of measurements, and *R* is the normalized radiance at <u>a</u> time *t*, scattering angle Θ , and wavelength 500 nm. Next, the running mean of the time series of $\bar{R}_{near}R_{near}(t)$ with a window of three consecutive data points is calculated as $<\bar{R}_{near,mean}(t) >$. Index 1 is defined as the deviation $\tilde{R}_{near}(t)$ of $\bar{R}_{near}(t)$ from $<\bar{R}_{near}(t) > R_{near,mean}(t)$,

$$\tilde{R}_{\text{near,dev}}(t) = \left|\bar{R}_{\text{near}}(t) - \langle \bar{R}_{\text{near}}(t) \rangle - \frac{R_{\text{near,mean}}(t)}{R_{\text{near,mean}}(t)}\right| / \langle \bar{R}_{\text{near}}(t) \rangle - \frac{R_{\text{near,mean}}(t)}{R_{\text{near,mean}}(t)}$$
(28).

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Index 2 is the deviation \underline{R}_{far} of normalized angular distributions far from the sun and is defined as

$$\tilde{R}_{\text{far},\text{dev}}(t) = \sigma\left(\frac{R(\Theta,t) - \langle R_{\text{far},\text{mean}}(\Theta,t) \rangle}{\langle R_{\text{far},\text{mean}}(\Theta,t) \rangle}\right), \Theta > 10^{\circ} \text{--degree}, \quad (29)_{\text{far},\text{mean}}(\Theta,t) > 0$$

where $\langle R_{\text{far}}(\Theta, t) \rangle = \frac{R_{\text{far},\text{mean}}(\Theta, t)}{R_{\text{far},\text{mean}}(\Theta, t)}$ is the running mean of $R(\Theta_i, t)$ with a window of three 440 consecutive data points, and $\sigma(\mathbf{X})$ is the standard deviation of data set **X**. Note that the data for <u>calculating</u> \tilde{R}_{far} varies depending on SZA, which limits available scattering angles. We judged clear-sky conditions, when two-indexes 1 and 2 were both below their respective two thresholds (0.1 and 0.2, respectively). We determined the thresholds by comparing the images of the whole-sky camera and the time series of the surface solar radiation observed by the pyranometer. Figure 9 is an example of the 445 results for observations on January 6, 2014, in Tsukuba. Clear-sky conditions continued until 12:30, and then clouds passed over the sky until 15:00. Subsequently, there were clouds near the horizon, but the sky was almost clear. Our algorithm worked well, and cloudy scenes were eliminated. Although the whole-sky camera detected some clouds from 14:00 to 15:00, our algorithm judged the scenes as representative of clear-sky conditions. This may be because there were no clouds in the line of sight of 450 the sky-radiometerHowever, the cloudy conditions from 14:00 to 15:00 were misjudged, because the sky-radiometer observes only a part of the whole sky. The decline in the surface solar radiation around 9:00 was due to wiping of the glass dome of the pyranometer to keep the dome clean.

The method was applied to measurements from 2013 to 2014 at the Meteorological Research
Institute, Japan Meteorological Agency (MRI, JMA), in Tsukuba. The results were validated using visual observation of the amount of clouds in the Aerological Observatory of the JMA. Figure 10a shows the histograms of index 1 for cases in which the sun was and was not covered by clouds. Index 1 had a low value when there were no clouds shading the sun but had a wide range of values when clouds were shading the sun. Fig. 10b shows the histograms of index 2 when cloud cover was and was not <
20%. The peak shifted to the right when cloud cover was ≥ 20%, but the effect was not significant. Table 32 shows the validation results of this method. We defined "clear skybest condition" as cloud cover ≥> 20%2. In less than 17% of cases a

"poor condition" cloudy sky was misjudged as a "best condition" clear sky. The sky-radiometer observes only a part of the whole sky, but our algorithm showed good results.

465 **2.3 Estimation of PWV from direct solar irradiance (DSRAD algorithm)**

The sky-radiometer observes the angular distribution of \underline{LV} every 10 min but observes the direct solar irradiance every 1 min. Once the calibration constant is determined by the SKYMAP algorithm, we can estimate PWV from the direct solar irradiance. The DSRAD algorithm computes the aerosol optical thickness, and PWV from the direct solar irradiances at the aerosol and water vapor channels. Table <u>43</u> 470 shows the references of the DSRAD algorithm. This algorithm consists of two steps. First, aerosol optical thicknesses at aerosol channels are calculated using direct solar irradiances. The aerosol optical thickness at the water vapor channel is interpolated from the aerosol optical thicknesses at 870 and 1020 nm by linear interpolation in the log-log plane by line regression. Second, the band mean transmittance of the water vapor, \bar{T}_{H20}^{meas} , is calculated from the calibrated direct solar irradiance. PWV is retrieved using the formula,

$$\overline{T}_{\text{H2O}}^{\text{meas}} - \frac{\sum_{i}^{N_{s}} \Phi_{i} \int_{\Delta\lambda_{i}} \exp(-m_{\text{H2O}}(\theta) \int_{0}^{z} \alpha_{\text{H2O}}(g_{w}(z), K(z), \lambda) dz) d\lambda_{1}}{\sum_{i}^{N_{s}} \Phi_{i} \int_{\Delta\lambda_{i}}^{-} \exp(-m_{\text{H2O}}(\theta) \int_{\theta}^{z} \sigma_{\text{H2O}}(g_{w}(z), K(z), \lambda) dz) d\lambda} = 0, \quad (30)$$

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where m_{H2O} is the optical air mass calculated by Gueymard (2001). Eq. (30) is solved using the Newton-Raphson method.

To ensure the quality of the data and avoid cloud contamination, we adopt the method of Smirnov *et al.* (2000) with two main differences, similar to Estellés *et al.* (2012). First, an aerosol optical thickness at 500 nm > 2 is considered cloud-affected data. Second, the triplet of the aerosol optical thickness in Smirnov *et al.* (2000) is built from <u>the pre/post</u> 1 min_<u>data</u> instead of 30 s.

3 Sensitivity tests using simulated data

We conducted sensitivity tests using simulated data to evaluate SKYMAP algorithm steps 1 and 2 (Figs. 7a and 7b). The simulation was conducted using the two aerosol types based on those used by Kudo *et al.* (2016): the continental average, and the continental average + transported dust in the upper atmosphere (Table 1). The continental average consisted of water-soluble particles, soot particles, and insoluble particles (Hess *et al.*, 1999).described in Section 2.1.2. Transported dust was defined as the mineral-transported component from Hess *et al.* (1999). The sensitivity test was conducted with sky

- radiances in the almucantar plane for <u>the wavelengths of 340, 380, 400, 500, 675, 870, 940, and 1020</u>
 <u>nm;</u> aerosol optical thicknesses of 0.02, 0.06, and 0.20 at 940 nm; PWV of 0.0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, and 5.0 cm; <u>and SZA of</u>= 30°, 50°, and 70°.
- Figure 101 illustrates the retrieval results from the simulated data for the continental average aerosol with aerosol optical thicknesses of 0.02, 0.06, and 0.20 at 940 nm. The retrievals of the <u>volume</u> size distribution, aerosol optical thickness, and PWV corresponded with their input values <u>("true"</u> <u>values in Fig. 11)</u> when the input of PWV was <2 cm. This was seen regardless of the magnitude of the aerosol optical thickness. When the input of PWV was >2 cm, the <u>volume</u> size distribution, scattering and absorption optical thickness were retrieved well, but PWV was underestimated. When PWV was >2 cm, the normalized angular distribution was insensitive to PWV (Fig. 3). Figure 121 illustrates the retrieval results from the simulated data for the transported dust aerosol with aerosol optical thicknesses of 0.02, 0.06 and 0.20 at 940 nm. The scattering and absorption optical thicknesses were retrieved well. The <u>volume</u> size distribution of fine mode was slightly overestimated. <u>The r</u>Retrieval errors <u>of PWV</u> increased with increasing aerosol optical thickness because the near-infrared wavelength was strongly affected by the retrieval of coarse mode particles.
- 510 We also conducted sensitivity tests using the simulated data with bias errors to investigate uncertainty in the SKYMAP-derived PWV. The bias errors were \pm 5% and \pm 10% for *R*. The value of 5% was given by following reasons. The SVA bias errors of the diffuse radiances for the sky-radiometer observations were estimated to be less than 5% (Uchiyama *et al.*, 2018b). According to Dubovik *et al.* (2000), the uncertainty of the diffuse radiances for the AERONET measurements is \pm 5%. Figures 13
- 515 and 14 show the results from the simulated data for the continental average and transported dust aerosols with aerosol optical thicknesses of 0.02, 0.06 and 0.20 at 940 nm. PWV was overestimated when 5% bias was applied to *R*. This corresponds to the relationship between *R* and PWV, where *R* decreases with increasing PWV (Section 2.1.2). The bias errors strongly affected the retrieval of PWV at high PWV (> 2 cm), because the sensitivity of high PWV is lower than that of low PWV. The
 520 retrieval error of PWV increased with increasing bias errors. The retrieval error of PWV due to ± 5%

and $\pm 10\%$ errors for *R* was within 10% for PWV < 2 cm and up to 200% for PWV > 2 cm.

- When the input of PWV <u>wais</u> $<_2$ cm, the SKYMAP <u>algorithm</u> retrieved PWV very well, within an error of 0.5 cm10%, regardless of the aerosol optical thickness or the aerosol type. <u>This was also</u> <u>observed when the bias errors were added for *R*.</u> The scattering and absorption parts of <u>the</u> aerosol
- 525 optical thickness were also estimated very well within \pm 0.01 in all conditions. Present sensitivity tests suggest the design of a sky-radiometer calibration program as follows: to determine the calibration constant of the water vapor channel in dry days/seasons with PWV <2 cm₇ and to obtain PWV from <u>direct</u> solar irradiance data throughout the year, as illustrated in Fig. 1.

4 Application to observational data

We applied our methods to SKYNET sky-radiometer data in Tsukuba and Chiba. –The results were compared to PWV observed by <u>well-established</u> instruments <u>and methods</u> other than the sky-radiometer. <u>Aerosol The aerosol</u> channels of the sky-radiometer were calibrated by the IL method with SKYRAD.pack version 4.2 (Nakajima *et al.*, 1996; Campanelli *et al.*, 2004, 2007), and the solid view angles of all channels were calibrated by <u>the</u> on-site methods (Nakajima *et al.*, 1996; Boi *et al.*, 1999; Uchiyama *et al.*, 2018b).

4.1 Observation at Tsukuba

In Tsukuba, the sky-radiometer <u>model POM-02</u> (S/N PS1202091) is installed at the MRI (36.05°N, 140.12°E). We used data from 2013 and to 2014. The water vapor channel of PS1202091 was calibrated each winter by side-by-side comparison with the reference sky-radiometer, which was calibrated by the <u>standard</u> Langley method at the NOAA Mauna Loa Observatory (Uchiyama *et al.*, 2014). PWV was also observed using a GNSS/GPS receiver (Shoji, 2013) at Ami station (No. 0584; 36.03°N, 140.20°E), approximately 7.5 km east-southeast of the MRI.

- The calibration constant of the water vapor channel was determined for each month (Figs. 152a and 163a). To obtain the correct value, we used the retrieval results with PWV_{SKYMAP} < 2 cm and sufficiently small cost functions (Eqs. [13]) and [20]). The annual mean values-calibration constants for 2013 and 2014 were 1.886 × 10⁻⁴ A and 2.212 × 10⁻⁴ A, respectively. The annual mean calibration constants changed drastically from 2013 to 2014 (+ 17.2%). This is Bbecause the lens at the visible and near-infrared wavelengths was replaced in December 2013, the calibration constants at these wavelengths changed drastically (annual mean value: + 17.2 % from 2013 to 2014). These results in 2013 and 2014 were less, -10.1% and -3.2-% lower, respectively, than those determined by the side-by-side comparison with the reference sky-radiometer. The difference in the value of the calibration constant between the SKYMAP algorithm and the side-by-side comparison with the reference sky-radiometer. The calibration constant of the sky-radiometer has seasonal variation due to the temperature dependency of the sensor output (Uchiyama *et*
- 555 *al.*, 2018a). Calibration by side-by-side comparison with the reference sky-radiometer was performed only in the winter. However, the calibration constant of the SKYMAP algorithm was the annual mean.

Although the monthly mean calibration constant of the water vapor channel was underestimated every year in the wet season (May to October), it was a good estimate in the dry season (November to April). The number of retrieved results was small in summer because of cloudiness. In summer in Japan, 560 clouds develop every day because it is warm with high relative humidity. Thus, because of higher aerosol optical thickness and as a result of the cloud-affected data, PWV in summer also contained large bias. We rejected the monthly calibration constant, which was calculated from fewer 50 data sets.

Figures 152b and 163b show the DSRAD-retrieved PWV, which is denoted by $PWV_{DSRAD+SKYMAP}$, using the monthly calibration constant. $PWV_{DSRAD+SKYMAP}$ of the sky-radiometer agreed well with that

- 565 of the GNSS/GPS receiver. <u>Note that we did not retrieve PWV using the monthly mean calibration</u> constants for June and July 2014 because their values were obviously small, and because little data were successfully retrieved due to the wet and cloudy conditions in the summer. In addition, it is possible that the measurements were contaminated by clouds.
- Although monthly mean calibration constants values are best, in theory, they could not be 570 obtained during the wet season or during periods of high aerosol optical thickness in-due to the transported dust. Thus, we calculated used the annual mean calibration constant value from all data in a year to estimate PWV. Figures 152c and 163c describe-illustrate PWV using the annual mean value calibration constantsfor the year. The retrieved PWV agreed well with PWV from the GNSS/GPS receiver (correlation coefficient $\gamma = 0.987$ and 0.987, and slope = 0.919 and 0.934 for 2013 and 2014, respectively; Table 5). We estimated PWV, which is denoted by PWV_{DSRAD+LM}, from the DSRAD 575 algorithm using the calibration constant obtained by the side-by-side comparison with the reference skyradiometer. The comparison of PWV_{DSRAD+LM} and the GNSS/GPS-derived PWV in Figs. 12d and 13d shows the good agreement, and the results are similar to those in Figs. 152c and 163c. Then we compared PWV_{DSRAD+LM} and PWV_{DSRAD+SKYMAP} in Figs. 152e and 163e. The difference between PWV_{DSRAD+LM} and PWV_{DSRAD+SKYMAP} was small: 17% in 2013, and 8% in 2014. Our self-calibration 580 method showed comparable results to those based on the standard Langley method (Uchiyama et al., 2014). Table 5 summarizes the results of comparisons of DSRAD-derived PWV and GNSS/GPSderived PWV. The magnitude of the bias error and root mean square error were small, less than 0.11 cm and less than 0.226 cm, during 2013 to 2014. Table 6 shows the errors of the retrieved PWV with the 585 annual mean calibration constants for the rank of PWV. The bias error was larger for high PWV than it was for low PWV. The magnitude of the bias errors of PWV was less than 0.163 cm for PWV < 3 cm and less than 0.339 cm for PWV > 3 cm.

4.2 Observation at Chiba

We used 2017-the_data from the sky-radiometer_model POM-02 (S/N PS2501417) at Chiba University
(35.63°N, 140.10°E) in 2017. The-PWV was also obtained by a Radiometrix MP-1500 microwave radiometer (MWR) and AERONET sun-sky radiometer (Cimel, France) at the same location. The MWR measured in the 22-30 GHz region at 1-min temporal resolution and retrieved PWV_{MWR} using default software. PWV_{Cimel} of the AERONET sun-sky radiometer was retrieved by the_direct solar irradiance at 93640 nm with adjustment parameters (direct sun algorithm version 3; Holben *et al.*, 1998; Giles *et al.*, 2018) and adopted the cloud screening method (AERONET Level 2.0). The AERONET product comprises three types of data: Level 1.0 data are not screened for cloud-affected or low_z-quality

-data, Level 1.5 data are screened but not completely calibrated, and Level 2.0 data are finalized data that have been calibrated and screened. We used PWV for the Level 2.0 data.

- Figure 1<u>7</u>4 shows comparisons of PWV_{DSRAD+SKYMAP} using the monthly and annual means of the calibration constants, PWV_{MWR}, and PWV_{Cimel}. PWV_{DSRAD+SKYMAP} using monthly means calibration constants agreed well (correlation coefficient $\gamma = 0.961$ and slope = 0.964) with those of the MWR (Fig. 1<u>7</u>4b). PWV_{DSRAD+SKYMAP} using the annual mean calibration constant agreed with PWV_{MWR} (Fig. 17c). within ±0.05 cmThe error of PWV_{DSRAD+SKYMAP} was – 0.041 < bias < 0.024 cm and RMSE < 0.212 cm for low PWV (<3 cm), but and was bias < – 0.356 cm and RMSE > 0.465 cm smaller than PWV_{MWR} for
- 605 high PWV (Table 6). (Fig. 14c). Figure 17/4d also shows that PWV_{DSRAD+SKYMAP} using the annual mean calibration constant also agreed with PWV_{Cimel} for low PWV (< 3 cm) but was smaller than PWV_{Cimel} when for high PWV was (> 3 cm). PWV_{MWR} was larger than PWV_{Cimel} (Fig. 17/4e). PWV_{DSRAD+SKYMAP} using the annual mean calibration constant was 12% and 9.1% smaller than PWV_{MWR} and PWV_{Cimel}⁵ but the difference was 10 % compared to the MWR and 16% compared to the AERONET sun-sky
- 610 radiometer, respectively (Table 5). These results suggest an underestimation of PWV_{DSRAD+SKYMAP}, as the uncertainty of PWV_{Cimel} compared to the GNSS/GPS receiver is expected to be less than 10% (Giles *et al.*, 2018). The underestimation of PWV_{DSRAD+SKYMAP} was due to two factors. The first is the retrieval of PWV by the annual mean calibration constant for the water vapor channel. The calibration constant not only is subject to aging but also undergoes seasonal variation due to temperature dependency
- 615 (Uchiyama *et al.*, 2018a). Thus, it is possible to underestimate the calibration constant in the wet season. Second, uncertainty regarding the aerosol optical thickness affected PWV retrieval. Figure 18 depicts the differences in PWV and aerosol optical thicknesses at 675, 870, and 1020 nm between the DSRAD algorithm and the AERONET retrieval. In the periods from January to May and from October to November, the differences in PWV and aerosol optical thicknesses were less than 0.1 cm and 0.015, respectively. However, the difference in PWV was greater than 0.1 cm from July to September. This
- corresponds to the difference in aerosol optical thicknesses at 675, 870, and 1020 nm from July to September, which indicates that the transmittance of water vapor was overestimated by the overestimation of aerosol optical thickness. This led to the underestimation of $PWV_{DSRAD+SKYMAP}$ using the annual mean calibration constant when PWV was > 3 cm. In our error estimation, the error of + 0.03
- 625 <u>for the aerosol optical thickness at 940 nm resulted in the error of -0.214 cm for PWV (Appendix B).</u>

5 Summary

We developed a new on-site self-calibration method, SKYMAP, to retrieve PWV from sky-radiometer data at the water vapor channel. This method first retrieves PWV from the <u>normalized angular</u> distribution of the normalized radiance without the calibration constant. Then the calibration constant is retrieved from the obtained PWV. Once the calibration constant is determined, PWV can be estimated

from the direct solar irradiance. Our DSRAD algorithm retrieves PWV from the direct solar irradiance. This method does not require any of the adjustment parameters used in the empirical methods of previous studies (e.g., Holben *et al.*, 1998; Uchiyama *et al.*, 2014; Campanelli *et al.*, 2014, 2018). Instead, the filter response function and the vertical profiles of water vapor, temperature, and pressure are required as input parameters. Thus, our physics-based algorithm has the potential to be applied to

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sky-radiometers all over the world. This is the greatest advantage of the present study.

Sensitivity tests using simulated data from sky-radiometer measurements showed that <u>the</u> SKYMAP <u>algorithm</u> retrieved PWV within an error of 0.5 cm10% for cases when PWV was <2 cm. <u>Much moreLarger</u> retrieval errors occurred, <u>when in the cases when</u> PWV was >2 cm, because PWV became less sensitive to the <u>normalized</u> angular distribution of the normalized radiance. <u>Therefore, the</u> SKYMAP algorithm can be applied only to dry conditions.

We applied SKYMAP and DSRAD algorithms to <u>the</u>sky-radiometer measurements at two SKYNET sites (Tsukuba and Chiba, Japan). At Tsukuba, the calibration constant estimated by <u>the</u> SKYMAP <u>algorithm</u> was compared to that obtained by side-by-side comparison with <u>a-the reference</u>

- 645 sky-radiometer calibrated by the <u>standard</u> Langley method. Their differences were The calibration constant calculated by the SKYMAP algorithm was -10.1% lower in 2013 and -3.2% lower in 2014 compared with the calibration constant estimated by side-by-side comparison. Our retrieved PWV data were compared to those obtained by a GNSS/GPS receiver, a microwave radiometer, and an AERONET sun-sky radiometer. The correlation coefficients and slopes were as good as >0.96 and 1.00
- \pm 0.12, respectively. The magnitude of the bias error and the root mean square error were < 0.163 cm and < 0.251 cm, respectively, for low PWV (< 3 cm). However, our retrieved PWV was underestimated in the wet conditions, and the magnitude of the bias error and the root mean square error were less than 0.594 cm and less than 0.722 cm for high PWV. This was due to seasonal variation in the calibration constant and the overestimation of aerosol optical thickness at 940 nm interpolated from those at 870
- 655 <u>and 1020 nm.</u>

These results show that our new on-site self-calibration method is practical. In future work, we plan to compare our method with others in the SKYNET framework (Uchiyama *et al.* 2014; Campanelli *et al.*, 2014).

660 6 Data availability

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The SKYMAP and DSRAD algorithms are available on request from the first author. The skyradiometer data are available from the SKYNET website (http://www.skynet-isdc.org/), but the skyradiometer data in Tsukuba, Japan, are available on request from the first author. The MWR data at Chiba University are available from CEReS, Chiba University (http://atmos3.cr.chiba-u.jp/skynet/). The 665 AERONET sun-sky radiometer data are available from the AERONET website (https://aeronet.gsfc.nasa.gov/).

Author contributions

This study was designed by MM, RK, KA, TM, KM, and TN. Sky-radiometer measurements at Tsukuba were conducted by RK. Sky-radiometer and MWR measurements at Chiba were conducted by
HO and HI. Analyses of both sky-radiometers were performed by MM. The calibration constant of the sky-radiometer by the Langley method was provided by AU. Analyses of the GPS receiver were conducted by YS. Visual observations at Tsukuba were conducted by OI and MT. The manuscript was

Competing interests

675 The authors declare that they have no conflict of interest.

written by MM and RK, and all authors contributed to editing and revision.

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Appendix A: Width of the volume size distribution

Because $\frac{dV(r)}{d\ln r}$ is expressed by the superposition of 20-modal lognormal size distributions (Eq. [6]), the width of $\frac{dV(r)}{d\ln r}$ is larger than that of each lognormal size distribution. The width of the lognormal size distribution should be small to deal with the complicated and step variations in $\frac{dV(r)}{d\ln r}$. However, $\frac{dV(r)}{d\ln r}$ 690 cannot represent a natural curve if η is large and s is small (Fig. A1). Hence, we have to find the maximum value of η for making $\frac{dV(r)}{d\ln r}$ a natural curve. When C_i is constant, such value of η minimizes

the roughness of $\frac{dV(r)}{d\ln r}$, and $\frac{dV(r)}{d\ln r}$ approaches to a flat shape. For a simple formulation, we consider the function A(x) which consists of the multimodal normal distribution function B_i with a constant height. A(x) and B_i are expressed as

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$$A(x) = \sum_{i=-\infty}^{\infty} B_i(x) = \sum_{i=-\infty}^{\infty} \exp\left[-\frac{\eta^2}{2} \left(\frac{x-i\xi}{\xi}\right)^2\right], \quad (A1)$$

where $i\xi$ and $\frac{\xi}{n}$ are the mean and standard deviation, respectively. Its differential is written as

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$$\frac{dA}{dx} = \sum_{i=-\infty}^{\infty} \frac{dB_i}{dx} = \sum_{i=-\infty}^{\infty} -\eta^2 \left(\frac{x-i\xi}{\xi}\right) \exp\left[-\frac{\eta^2}{2} \left(\frac{x-i\xi}{\xi}\right)^2\right].$$
 (A2)

When the shape of A(x) approaches to be flat, the difference between local maximum and minimum values of A(x) is approximately 0. Because $\frac{dB_i}{dx}$ equals 0 at $x = j\xi$ ($j \in \mathbb{Z}$), A(x) has the local maximum and minimum at $x = j\xi$ and $\left(j + \frac{1}{2}\right)\xi$ in $j \le \frac{x}{\xi} < j + 1$. The difference Δ between the local maximum and minimum values is obtained as

$$\Delta = 1 - \frac{A\left(\frac{2j+1}{2}\xi\right)}{A(j\xi)}$$
 (A3)

Figure A2 shows the relation between η and Δ . The value of Δ increases drastically at around $\eta = 1.5$. 710 in addition, the shape of $\frac{dV(r)}{d\ln r}$ is unnatural when $\eta = 2.0$ (Fig. A1). Therefore, the value of η should be selected from the values around $\eta = 1.5$. In this study, we fixed η at 1.65. This value represents the natural curve of $\frac{dV(r)}{d\ln r}$ and satisfies that the value of Δ is small enough, $\Delta = 3.0 \times 10^{-3}$.

Appendix B: Error propagation from aerosol optical thickness to PWV

We evaluated the influence of the uncertainty of aerosol optical thickness on PWV using the empirical equation of Bruegge *et al.* (1992). PWV is described using the adjustment parameters as follows

$$w = \frac{1}{m_0} \left(-\frac{\ln \bar{T}_{H20}}{a} \right)^{\frac{1}{b}} [cm].$$
 (B1)

The uncertainty of PWV ϵ_{PWV} is given from the partial differentiation of Eq. (B1) with respect to 1n \overline{T}_{H2O} as follows

$$\epsilon_{\rm PWV} = \frac{\partial w}{\partial \ln \bar{T}_{\rm H20}} \epsilon_{\ln \bar{T}_{\rm H20}} = \frac{w}{b \ln \bar{T}_{\rm H20}} \epsilon_{\ln \bar{T}_{\rm H20}}.$$
 (B2)

where $\epsilon_{\ln \bar{T}_{H20}}$ is the uncertainty of \bar{T}_{H20} . Using Eq. (B1) with the adjusting parameters of the skyradiometer, with a = 0.620 and b = 0.625 as the coefficient values for the trapezoidal spectral response function (Uchiyama *et al.*, 2018a), we write the uncertainty of PWV as

$$\epsilon_{\rm PWV} = -\frac{w}{ab} (m_0 w)^{-b} \epsilon_{\ln \bar{T}_{\rm H20}} = -\frac{w}{0.388} (m_0 w)^{-0.625} \epsilon_{\ln^-_{\rm H20}}.$$
 (B3)

730 If the uncertainty of the calibration constant at the water vapor channel is ignored, the uncertainty of \bar{T}_{H20} is given from Eq. (21) as follows

$$\epsilon_{\ln \bar{T}_{H2O}} = m_0 \epsilon_{AOT}$$
. (B4)

735 where ϵ_{AOT} is the uncertainty of the aerosol optical thickness at 940 nm. The uncertainty of PWV is written by Eqs. (B3) and (B4) as

$$\epsilon_{\rm PWV} = -\frac{1}{0.388} (m_0 w)^{0.375} \epsilon_{\rm AOT} = -0.214 \, [\rm cm].$$
 (B5)

740 where $m_0 = 3.0, w = 5.0 \text{ cm}, \text{ and } \epsilon_{AOT} = 0.03.$

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Figure 1: Diagram of the on-site self-calibration method (SKYMAP) and retrieval of PWV from direct solar irradiances (DSRAD). <u>Square boxes show the operation of the calculation and input/output parameters and rounded boxes show the operation of the algorithm.</u>



Figure 2: Observation planes (almucantar and principal planes) of the sky-radiometer.

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Figure 3: Normalized angular distributions simulated for continental average aerosol (Table 21) in the almucantar plane withat aerosol optical thicknesses of 0.02 and 0.20 at 940 nm. The sSimulations were conducted for SZA = 70° and PWV (w) = 0, 1, 2, 3, 4, and 5 cm. The top rowline is the normalized radiance $R(w, \Theta)$, and the bottom rowline is the ratio of $R(w, \Theta)$ to $R(0, \Theta)$. S-S Approx. is single scattering approximation.



Figure 4: Normalized angular distributions simulated for transported dust aerosol (Table 21) in the almucantar and principal planes withat an aerosol optical thickness of 0.06 at 940 nm. The sSimulations were conducted for SZA = 70° and PWV (w) = 0, 1, 2, 3, 4, and 5 cm. The top rowline is the normalized radiance $R(w, \Theta)$, and the bottom rowline is the ratio of $R(w, \Theta)$ to $R(0, \Theta)$.







Figure 6: Normalized angular distributions simulated for transported dust aerosol (Table 21) in the almucantar and principal planes withat an aerosol optical thickness of 0.06 at 940 nm. The sSimulations were conducted for SZA = 70° and PWV = 2 cm. The height of the dust layer (z_c) is changed to 0.5, 1.5, 2.5, and 3.5 km. The top rowline is the normalized radiance $R(z_c, \Theta)$, and the bottom rowline is the ratio of $R(z_c, \Theta)$ to $R(3.5 \text{ km}, \Theta)$.





Determined $F_0(940 \text{ nm})$ of mean value

900 Figure 7: Schematic diagrams of SKYMAP procedures. (a) Step 1. (b) Step 2. (c) Step 3. <u>Square</u> <u>boxes show the calculation and input/output parameters</u>



Figure 8: Assuming volume size distributions in the SKYMAP algorithm. Fine and coarse mode particles are separated at radius *r*_b. Spheroid particles are assumed only in coarse mode. The black line is the volume size distribution, which is computed by the integration of 20-modal lognormal <u>distribution</u> functions (red, blue, and green lines).



Figure 9: An example result of the SCAD method on January 6, 2014, in Tsukuba. (a) Surface solar radiation observed by the pyranometer. (b) Index 1. (c) Index 2. The closed circles indicate clear-sky conditions and the open circles indicate cloudy conditions in (b) and (c). The lines at 0.1
in (b) and 0.2 in (c) are thresholds for indexes 1 and 2, respectively.



Figure 10: Histograms of indexes 1 and 2 of sky-radiometer observations at Tsukuba. (a) Index 1 when the sun is covered by clouds (blue boxes) and not covered by clouds (red boxes). (b) Index 2 when cloud cover is less than to 20% (red boxes) and greater than or equal to 20% (blue boxes).



Figure <u>1110</u>: Retrieval results from simulated data for continental average aerosol. The top <u>rowline</u> is the volume size distribution, the middle <u>rowline</u> is the scattering and absorption parts of aerosol optical thickness, and the bottom <u>rowline</u> is a comparison of the "true" and retrieval values of PWV. Blue, red, and green lines are the retrieval results at SZA = 30° , 50° , and 70° ,



respectively. The black line is the "true" value. <u>Note that the blue, red, green, and black lines in</u> <u>the middle row overlap.</u>



Figure <u>12</u>11: Similar to Fig. 1<u>1</u>0 but for transported dust aerosol. <u>Note that the blue, red, green, and black lines in the middle row overlap.</u>



Figure 13: Comparison of the "true" and retrieval values of PWV from simulated data for continental average aerosol with bias errors. The top, middle, and bottom rows are the retrieval results at SZA = 30° , 50° , and 70° , respectively. Closed circles are the results with no bias errors. Closed squares and closed triangles are the results with bias errors of plus and minus 5% in *R*, respectively. Open squares and open triangles are the results with bias errors of plus and minus 10% in *R*, respectively.



Figure 14: Similar to Fig. 13 but for transported dust aerosol.



Figure <u>15</u>+2: Application of our methods to observational data from Tsukuba in 2013. (a) Seasonal variation in the calibration constant of the water vapor channel (red circles and error bars are monthly means and standard deviations, respectively; green solid and dotted lines are annual means and standard deviations, respectively; the blue line is the value obtained by a side-by-side comparison with the reference sky-radiometer; boxes indicate the number of data points). (b-d) Comparisons of PWV between the GNSS/GPS receiver and the sky-radiometer with (b) the

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by-side comparison with the reference sky-radiometer; boxes indicate the number of data points). (b-d) Comparisons of PWV between the GNSS/GPS receiver and the sky-radiometer with (b) the monthly mean F_0 , (c) the annual mean F_0 , and (d) the reference F_0 . (e) Comparison of PWV from the sky-radiometer with the reference and annual mean F_0 .



Figure <u>16</u>13: Similar to Fig. 1<u>5</u>2 but in 2014.



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Figure 1714: Application of our methods to observational data from Chiba in 2017. (a) Seasonal variation in the calibration constant of the water vapor channel (red circles and error bars are monthly means and standard deviations, respectively; green solid and dotted lines are annual means and standard deviations, respectively; boxes indicate the number of data points). (b,- c) Comparison of PWV between the MWR and the sky-radiometer with (b) the monthly mean F_0 , and (c) the annual mean $F_{0.}$ (d) Comparison of PWV between the Cimel level 2.0 data and the 970 sky-radiometer with annual mean F_{0} . (e) Comparison of PWV between the Cimel level 2.0 data and the MWR.



Figure 18: The top row shows the time series of PWV in 2017 at Chiba (green and black circles are PWV_{DSRAD+SKYMAP} and PWV_{Cimel}, respectively). The middle row is the difference between PWV_{DSRAD+SKYMAP} and PWV_{Cimel}. The bottom row is the difference in aerosol optical thicknesses at 675 nm (red), 870 nm (blue), and 1020 nm (green) between the DSRAD algorithm and the AERONET retrieval results. Circles and error bars in the middle and bottom rows are means and standard deviations, respectively.



Figure A1: Relationship between the volume size distribution and η . The black line is the volume size distribution, which is computed by the integration of 20-modal lognormal distribution functions (red lines). Blue circles are the peak volume of lognormal size distribution.



Figure A2: Relationship between the parameter η and the difference Δ .

990 <u>Table 1: Sky-radiometer specifications. Each sky-radiometer is equipped with a filter indicated by</u> <u>a circle. "Standard" is the standard specification of sky-radiometer models POM-01 and POM-02.</u>

Wavelength [nm]	Strong gas absorption	Main target substance	POM-01 Standard	POM-02 Standard	<u>POM-02</u> <u>PS1202091</u>	<u>POM-02</u> <u>PS2501417</u>
315	<u>O</u> ₃	<u>Ozone</u>	<u>O</u>	<u>O</u>	=	<u> </u>
<u>340</u>	=	<u>Aerosol</u>	=	\bigcirc	<u> </u>	<u>O</u>
<u>380</u>	=	<u>Aerosol</u>	=	\bigcirc	<u> </u>	\bigcirc
<u>400</u>	=	<u>Aerosol</u>	<u> </u>	\bigcirc	<u> </u>	\bigcirc
<u>500</u>	=	Aerosol	\bigcirc	\bigcirc	<u> </u>	\bigcirc
<u>675</u>	_	<u>Aerosol</u>	<u> </u>	\bigcirc	<u> </u>	\bigcirc
<u>870</u>	=	Aerosol	<u> </u>	\bigcirc	<u> </u>	\bigcirc
<u>940</u>	<u>H</u> ₂ O	Water vapor	<u> </u>	\bigcirc	<u> </u>	\bigcirc
<u>1020</u>	=	Aerosol	<u> </u>	\bigcirc	<u> </u>	\bigcirc
<u>1225</u>	<u>O₂, CO₂, H₂O</u>	<u>Cloud</u>	=	=	<u> </u>	_
<u>1627</u>	<u>CH4, CO2</u>	Cloud	=	\bigcirc	\bigcirc	\bigcirc
<u>2200</u>	<u>CH4, H2O</u>	<u>Cloud</u>	=	<u> </u>	\bigcirc	<u> </u>

Aerosol	Components	Particle shape	Size d	istribution	Refractive index at 940 nm		Re in thi 50	lative weight total optical ckness at 0 nm	Vertical profile	
			Mode radius (µm)	Mode width	Real	Ir	naginary	-		
Continental average	Water-soluble	Sphere	0.18	0.81	1.43	0.	0074	0.9	0	exp(-z/H), H = 8 km
	Soot	Sphere	0.05	0.69	1.75	0.	44	0.0	17	$\exp(-z/H),$ H=4 km
	Insoluble	Spheroid	5.98	0.92	1.52	0.	008	0.0	13	exp(-z/H), H=2 km
Transported dust	Dust	Spheroid	3.23	0.79	1.53	0.	004	0.2	5	$\frac{1}{\sqrt{2\pi\sigma}} \exp\left(-\frac{(z-z_c)}{2\sigma^2}\right),\ z_c = 3.5 \text{ km}$ $\sigma = 0.4 \text{ km}$
	Water-soluble	Sphere	0.18	0.81	1.43	0.	0074	0.6	7	$\exp(-z/H),$ H = 8 km
	Soot	Sphere	0.05	0.69	1.75	0.	44	0.0	5	$\exp(-z/H),$ H = 4 km
	Insoluble	Spheroid	5.98	0.92	1.52	0.	008	0.0	13	exp(-z/H), H=2 km
Aerosol	Componen	Partic	<u>le </u>	Size		Refractive			Relative	Vertical profile
	<u>ts</u>	<u>shape</u>	<u>(</u>	listributi	<u>on</u>	index	at 940 ni	<u>n</u>	weight	
									<u>in total</u>	
									<u>optical</u>	
									thicknes	
									<u>s at</u>	
									<u>500 nm</u>	
			1	<u>Mod</u> <u>1</u>	Mod	Rea	Imagina	ar		
			<u>(</u>	<u>e</u> <u>e</u>	<u>e</u>	<u>1</u>	У			
			<u>1</u>	radiu y	widt					
			<u>s</u>	<u>s 1</u>	<u>1</u>					
			(<u>[µm]</u>						
Continent	Water-	<u>Spher</u>	<u>e (</u>	<u>).18</u> ().81	<u>1.4</u>	<u>0.0074</u>		<u>0.90</u>	exp(<i>-z/H</i>) <u>,</u>
al average	<u>soluble</u>					<u>3</u>				H = 8 km
	<u>Soot</u>	<u>Spher</u>	<u>e (</u>	<u>).05</u>). <u>69</u>	<u>1.7</u>	<u>0.44</u>		<u>0.07</u>	$\exp(-z/H)_{\underline{r}}$
						<u>5</u>				H = 4 km

995 Table 21: Microphysical and optical properties and vertical profiles of aerosol used in sensitivity tests.

	Insoluble	<u>Spheroi</u> <u>d</u>	<u>5.98</u>	<u>0.92</u>	<u>1.5</u> <u>2</u>	<u>0.008</u>	<u>0.03</u>	$\exp(-z/H)_{\underline{i}}$ $\underline{H} = 2 \text{ km}$
<u>Transport</u> <u>ed</u> <u>dust</u>	<u>Dust</u>	<u>Spheroi</u> <u>d</u>	<u>3.23</u>	<u>0.79</u>	<u>1.5</u> <u>3</u>	<u>0.004</u>	<u>0.25</u>	$\frac{1}{\sqrt{2\pi\sigma}} \exp\left(-\frac{(z-z_c)}{2\sigma^2}\right)$ $\frac{z_c}{z_c = 3.5 \text{ km}}$ $\frac{\sigma = 0.4 \text{ km}}{\sigma}$
	<u>Water-</u> soluble	Sphere	<u>0.18</u>	<u>0.81</u>	<u>1.4</u> <u>3</u>	<u>0.0074</u>	<u>0.67</u>	$\exp(-z/H)_{L}$ $\underline{H=8 \text{ km}}$
	<u>Soot</u>	Sphere	<u>0.05</u>	<u>0.69</u>	<u>1.7</u> <u>5</u>	<u>0.44</u>	<u>0.05</u>	$\exp(-z/H)$ <u>H = 4 km</u>
	Insoluble	<u>Spheroi</u> <u>d</u>	<u>5.98</u>	<u>0.92</u>	<u>1.5</u> <u>2</u>	<u>0.008</u>	<u>0.03</u>	$\exp(-z/H)$ <u>H=2 km</u>

visual observation	Sky-radiometer	measuring plane	
Cloud cover	Clear sky	Cloud affect	ed
Clear, less than 1	463 (83.4%)*	68 (9.3%)	
Cloud affected, more than 2	92 (16.6%) [†]	663(90.7%))*
Visual observation	Sky-radion Best condit	neter measuring ion	plane <u>Poor condition</u>
Clear, less than 20%	463 (83.4%	<u>()*</u>	<u>68 (8.7%)</u>

*Obviously correct determination.

1005 [†]Obviously incorrect determination.

Table 43: References and methodologies of the DSRAD algorithm.

	DSRAD
Solar coordinates	Nagasawa (1999)
Refraction correction	Nagasawa (1999)
Sun-Earth distance	Nagasawa (1999)
Optical mass	Gueymard (2001)
Rayleigh scattering	Fröhlich and Shaw (1980); Young(1981)
Ozone absorption	Correlated k-distribution
Water vapor absorption	Correlated k-distribution
Filter response function	Stepwise function
Retrieved the PWV	Newton-Raphson method
	DSRAD

Solar coordinates	Nagasawa (1999)
Refraction correction	Nagasawa (1999)
Sun-Earth distance	Nagasawa (1999)
Optical mass	<u>Gueymard (2001)</u>
Rayleigh scattering	Fröhlich and Shaw (1980); Young(1981)
Ozone absorption	Sekiguchi and Nakajima (2008)
Water vapor absorption	Sekiguchi and Nakajima (2008)
Filter response function	Stepwise function
<u>Retrieval of PWV</u>	Newton-Raphson method

Table 5: Comparison of PWV between DSRAD and other instruments.

		<u>Slope</u>	Intercept	Υ	<u>Bias</u>	<u>RMSE</u>
		<i>C</i> ₁	<i>C</i> ₂ [cm]		[cm]	[cm]
PS1202091 at Tsukub	PS1202091 at Tsukuba, Japan					
Monthly mean	vs GNSS/GPS receiver	<u>0.956</u>	<u>0.079</u>	<u>0.938</u>	<u>-0.049</u>	<u>0.138</u>
F_0	<u>(2013)</u>					
	vs GNSS/GPS receiver	<u>0.937</u>	<u>0.161</u>	<u>0.970</u>	<u>-0.110</u>	<u>0.170</u>
	<u>(2014)</u>					
Annual mean	vs GNSS/GPS receiver	<u>0.919</u>	<u>0.173</u>	<u>0.987</u>	<u>-0.061</u>	<u>0.226</u>
F_0	<u>(2013)</u>					
	vs GNSS/GPS receiver	<u>0.934</u>	<u>0.178</u>	<u>0.987</u>	<u>-0.089</u>	<u>0.223</u>
	(2014)					
PS2501417 at Chiba,	<u>Japan</u>					
Monthly mean	<u>vs MWR (2017)</u>	<u>0.964</u>	<u>0.053</u>	<u>0.961</u>	<u>-0.027</u>	<u>0.091</u>
F_0						
	<u>vs AERONET (2017)</u>	<u>0.987</u>	<u>0.107</u>	<u>0.976</u>	<u>0.098</u>	<u>0.122</u>
Annual mean	<u>vs MWR (2017)</u>	<u>0.880</u>	0.132	<u>0.985</u>	<u>0.042</u>	0.231
F ₀						
	vs AERONET (2017)	<u>0.909</u>	<u>0.184</u>	<u>0.991</u>	<u>0.055</u>	<u>0.186</u>

 C_1 , C_2 : PWV_{DSRAD} = $C_1 \times PWV_{Other} + C_2$

 $\underline{Bias:} PWV_{DSRAD} - PWV_{Other}$

Table 6: Difference in PWV between DSRAD with the annual mean calibration constants and other instruments.

			PWV _{Other}		
	<u>0 – 1 cm</u>	<u>$1 - 2 \text{ cm}$</u>	<u>2 – 3 cm</u>	<u>3 – 4 cm</u>	<u>> 4 cm</u>
	Bias [cm]	Bias [cm]	Bias [cm]	Bias [cm]	Bias [cm]
	(RMSE	(RMSE	(RMSE	(RMSE	(RMSE
	[cm])	<u>[cm])</u>	<u>[cm])</u>	[cm])	[cm])
PS1202091 at Tsukuba,					
<u>Japan</u>					
vs GNSS/GPS receiver	<u>0.083</u>	<u>0.160</u>	<u>0.084</u>	<u>-0.098</u>	<u>-0.339</u>
<u>(2013)</u>	<u>(0. 124)</u>	<u>(0.211)</u>	<u>(0.236)</u>	<u>(0.326)</u>	<u>(0.537)</u>
vs GNSS/GPS receiver	<u>0.110</u>	<u>0.163</u>	<u>0.107</u>	<u>-0.055</u>	<u>-0.239</u>
<u>(2014)</u>	<u>(0.142)</u>	<u>(0.221)</u>	<u>(0.251)</u>	<u>(0.353)</u>	<u>(0.492)</u>
PS2501417 at Chiba, Japan					
<u>vs MWR (2017)</u>	<u>0.017</u>	0.024	<u>-0.041</u>	<u>-0.356</u>	<u>-0.594</u>
	<u>(0.066)</u>	<u>(0.153)</u>	<u>(0.212)</u>	<u>(0.465)</u>	<u>(0.722)</u>
<u>vs AERONET (2017)</u>	<u>0.088</u>	<u>0.118</u>	<u>0.017</u>	<u>-0.214</u>	<u>-0.264</u>
	<u>(0.105)</u>	<u>(0.192)</u>	<u>(0.223)</u>	<u>(0.386)</u>	<u>(0.306)</u>

1020 <u>Bias: PWV_{DSRAD} – PWV_{Other}</u>