



Cloud-height-dependent cloud–aerosol state classification based on MAX-DOAS observations in a coastal region and validation using lidar measurements

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Abstract. Ground-based MAX-DOAS retrievals are strongly influenced by clouds and aerosols. However, existing MAX-DOAS-based cloud classification methods mainly focus on identifying whether observations are affected by clouds, while less attention has been paid to cloud relative height and its different effects on retrievals. We develop a cloud-height-dependent cloud–aerosol state classification method based on MAX-DOAS observations in Guangzhou, a major coastal city in southern China. The method is evaluated using independent 355 nm lidar observations about 3 km from the MAX-DOAS site. It combines the measured zenith signal and its temporal variation, the O₄ differential slant column density (dSCD) at 30° elevation, and the retrieved aerosol optical depth (AOD) to distinguish eight observation states.

Radiative transfer simulations show that low clouds can substantially shorten the effective light path and reduce the O₄ dSCD at 30° elevation, whereas high clouds have a weaker effect because MAX-DOAS measurements are primarily sensitive to the near-surface atmosphere. This behaviour provides the physical basis for distinguishing low clouds from high clouds relative to a site-specific clear-sky reference sensitivity height of about 2 km. In addition, under the clear-sky assumption, cloud-induced light-path changes can be partly misattributed to aerosol extinction, so that low-cloud conditions can lead to overestimated AOD.

Non-clear-sky conditions account for a large fraction of the observations in Guangzhou, with low clouds, cloud holes, and cloud-free high-aerosol conditions occurring frequently. Compared with the lidar-based classification, the proposed method reproduces the main evolution of weather processes reasonably well. After grouping the results into three classes, namely cloud-free, high-cloud, and low-cloud conditions, the overall agreement is 0.658, with fractions of 0.703, 0.666, and 0.524 of the lidar-classified cloud-free, low-cloud, and high-cloud samples, respectively, identified as the same class by MAX-DOAS. Sensitivity analyses further show that the classification framework is generally stable with respect to perturbations in the key thresholds. These results provide information on cloud–aerosol observation states for assessing retrieval applicability under complex cloud conditions.



1 Introduction

The planetary boundary layer (PBL) is the main atmospheric region in which aerosols and pollutants accumulate, and its structure strongly affects near-surface air quality and human health (Li et al., 2017; Dockery, 2009). Observations of the vertical distributions of aerosols and trace gases within the PBL are therefore important for understanding atmospheric transport, dispersion, and related physical and chemical processes. Passive remote sensing relies on scattered solar radiation, and clouds and aerosols can substantially modify photon propagation and radiative transfer, thereby affecting observed signals and increasing retrieval uncertainty (Wagner et al., 2014; Tirpitz et al., 2021; Tian et al., 2021).

Ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) is a widely used passive remote-sensing technique for monitoring tropospheric aerosols and trace gases. By measuring scattered sunlight at multiple elevation angles and combining the observations with radiative transfer simulations, MAX-DOAS can be used to retrieve the vertical distributions of near-surface atmospheric constituents (Hönninger et al., 2004; Wagner et al., 2004; Sinreich et al., 2005; Yombo Phaka et al., 2023; Tian et al., 2023; Schreier et al., 2021; Gratsea et al., 2021). In MAX-DOAS retrievals, the absorption of the oxygen collision complex O_4 is commonly used to constrain aerosol retrievals because its vertical distribution is relatively stable and changes in its observed absorption mainly reflect changes in the effective atmospheric light path. Aerosol profiles are therefore usually retrieved first from O_4 absorption, after which trace-gas profiles are derived using the aerosol information together with trace-gas absorption signals. Consequently, the accuracy of aerosol retrieval directly affects the reliability of subsequent trace-gas retrievals (Bösch et al., 2018; Frieß et al., 2019; Wang et al., 2017).

However, clouds strongly affect MAX-DOAS observations. Under cloudy conditions, the interpretation of MAX-DOAS measurements becomes more difficult because cloud distributions change rapidly and their radiative effects are coupled with those of aerosols. Through scattering and shielding effects, clouds can modify the observed radiance and O_4 absorption and thereby increase the uncertainty of aerosol and trace-gas retrievals. Moreover, because clouds can occur over a wide vertical range in the troposphere, their effects on MAX-DOAS observations depend not only on their presence but also on their altitude. Therefore, cloud screening alone is often insufficient for assessing retrieval applicability under cloudy conditions.

During the past decade, cloud detection and classification methods for MAX-DOAS observations have provided an important basis for interpreting measurement conditions (Wagner et al., 2014; Gielen et al., 2014; Wang et al., 2015). In particular, Wagner et al. (2014) developed a classification scheme using radiance, the colour index (CI), O_4 absorption, and the Ring effect, and applied it during the 2009 CINDI campaign. Related methods have also been used in long-term observations (Wang et al., 2015; Ma et al., 2020). The CINDI-2 retrieval intercomparison examined profile retrievals under clear-sky and cloudy conditions and demonstrated the dependence of retrieval performance on cloud conditions (Tirpitz et al., 2021). Cloud classification therefore provides a basis for examining how cloud height relative to the MAX-DOAS sensitive layer affects measurement and retrieval responses.

Ground-based MAX-DOAS observations are primarily sensitive to the near-surface atmosphere, with sensitivity decreasing rapidly with altitude. When clouds occur within this sensitive layer, they modify photon propagation through scattering and shielding and usually shorten the effective light path. At high elevation angles, this effect is often manifested as a decrease in



O₄ dSCD. Because clouds at different altitudes affect photon paths differently, they can produce distinct signatures in MAX-DOAS observations, indicating that MAX-DOAS has the potential to distinguish cloud-height-related observation responses. Building on previous MAX-DOAS cloud-screening and classification studies (Wagner et al., 2014; Gielen et al., 2014; Wang et al., 2015), we propose a cloud–aerosol state classification method that considers cloud height relative to the instrument’s clear-sky reference sensitivity height. The categories describe cloud–aerosol-related MAX-DOAS observation states. They distinguish cloud-free, high-cloud, and low-cloud observation conditions, with subcategories based on retrieved AOD, as well as optically thick clouds and cloud holes. The classification uses only MAX-DOAS-derived quantities. The aim is to interpret cloud effects on MAX-DOAS measurements and aerosol retrievals.

The method is applied to MAX-DOAS observations obtained in Guangzhou in 2025, with aerosol retrievals at 360 nm. The classification results are evaluated using extinction profiles from a near-co-located 355 nm aerosol lidar.

This paper is organized as follows. Sect. 2 introduces the datasets and retrieval settings. Sect. 3 analyses the effects of clouds and aerosols on MAX-DOAS-derived quantities and aerosol retrieval results. Sect. 4 presents the characteristics of different cloud–aerosol conditions and the classification method. Sect. 5 presents the classification results and their comparison with the lidar results, followed by analyses of classification discrepancies and threshold sensitivity.

2 Datasets

2.1 Observation site

This study was conducted in the University Town area of Guangzhou, China (23.04° N, 113.36° E). A UV MAX-DOAS instrument was deployed on the rooftop of a teaching building at Guangzhou University for continuous measurements, while a 355 nm aerosol lidar was operated about 3.3 km east of the MAX-DOAS site. Together, the two instruments formed a near-co-located observation system (Fig. 1). The observation period covered the whole of 2025. The MAX-DOAS instrument provides aerosol retrieval results in the 360 nm band, whereas the lidar provides aerosol extinction profiles at 355 nm. The lidar profiles provide information on aerosol vertical distributions and cloud-layer structure for evaluating the MAX-DOAS retrievals and classification.

Guangzhou is located in southern coastal China and is strongly influenced by land-sea interaction and monsoon circulation. The region is characterized by abundant water vapour and relatively high humidity, which favour the hygroscopic growth of aerosols (Tan et al., 2013; Liu et al., 2020). As a major urban area, Guangzhou is also affected by local anthropogenic emissions, regional transport, and wet removal, leading to temporal variability in aerosol loading. In addition, cloud amount and cloud processes show marked seasonal variation, with frequent low-cloud and convective-cloud events and complex cloud–aerosol states. These conditions make Guangzhou a representative region for investigating MAX-DOAS observation characteristics and retrieval behaviour under complex cloud–aerosol conditions.

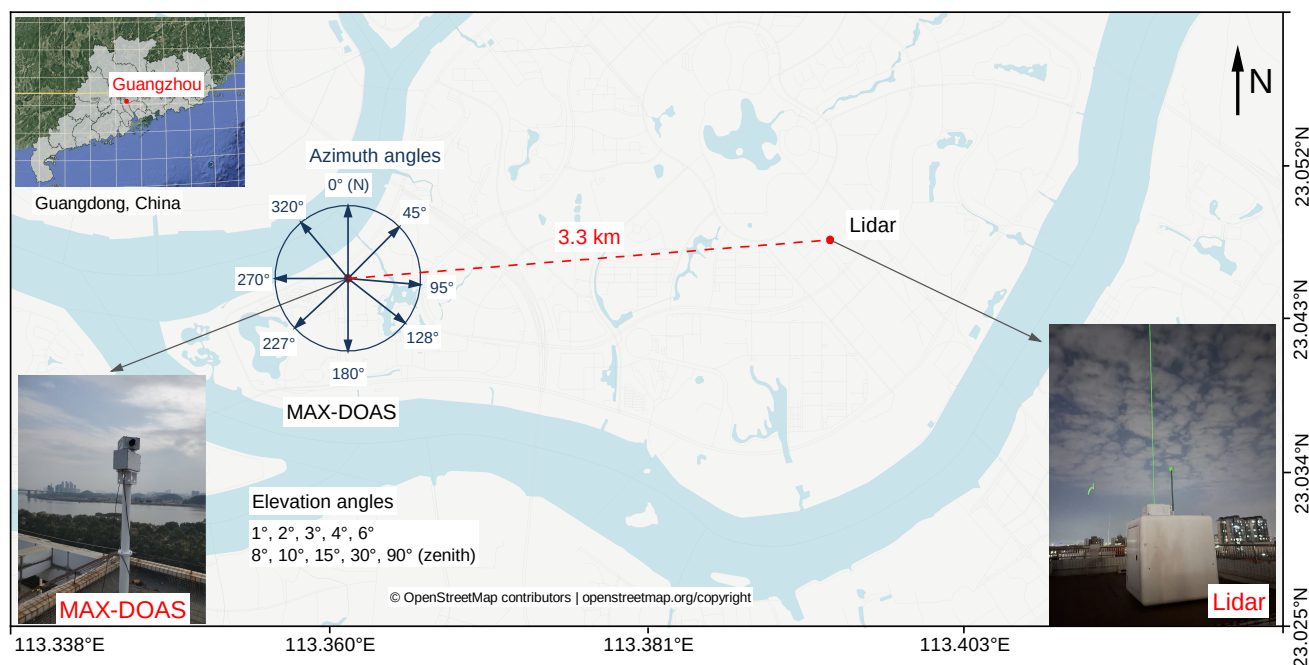


Figure 1. Location of the Guangzhou University Town observation site and schematic illustration of the observing geometry.

2.2 MAX-DOAS observations

2.2.1 MAX-DOAS observational configuration

The MAX-DOAS instrument used in this study operates in the 290–430 nm spectral range with a spectral resolution of about 0.5 nm. The system mainly consists of a telescope, a motor-driven scanning unit, a spectrometer module, a temperature-control unit, and a data acquisition unit. The telescope is installed outdoors and scans multiple azimuth and elevation angles under motor control.

The MAX-DOAS azimuth angles were set to 0°, 45°, 95°, 128°, 180°, 227°, 270°, and 320°, and the elevation angles were set to 90°, 1°, 2°, 3°, 4°, 6°, 8°, 10°, 15°, and 30°. The incoming light was transmitted through an optical fibre to an indoor grating spectrometer for detection, and the measurement process was automatically controlled by a computer. To reduce the influence of dark current and electronic offset, the system was equipped with a shutter for dark measurements, and dark signals were recorded during the elevation scans for correction. The integration time was automatically adjusted according to the scattered solar radiance to maintain the detector signal at about 70% of the saturation level, while the number of spectral acquisitions used for averaging was adjusted to improve the signal-to-noise ratio.

One complete elevation scan typically required about 5 to 15 min, depending on the illumination conditions. Observations were carried out from 08:00 to 18:00 Beijing time (BJT, UTC+8), corresponding to solar zenith angles smaller than 90°. All



times shown in the figures are given in BJT. To ensure spectral data quality, the raw observations were filtered, and data obtained under insufficient illumination conditions were removed. Details of the screening procedure are given in Supplement S1.

2.2.2 MAX-DOAS retrieval configuration

In this study, the O₄ dSCD in the 360 nm spectral region was retrieved using the QDOAS software. For each elevation angle, the corresponding zenith spectrum at 90° within the same scan sequence was used as the reference spectrum to minimize the influence of stratospheric background absorption. The detailed fitting settings for the UV spectral region are listed in Table 1, and an example fit is provided in Supplement S2.

Table 1. DOAS fit settings for the O₄ analysis in the 360 nm spectral region.

Parameter	Setting
Fitting window	338–370 nm
Cross section	NO ₂ at 220 and 294 K (Vandaele et al., 1998) O ₄ at 293 K (Thalman and Volkamer, 2013) BrO at 223 K (Fleischmann et al., 2004) O ₃ at 223 and 243 K (Bogumil et al., 2003) HCHO at 298 K (Meller and Moortgat, 2000) Ring
Polynomial order	5
Offset	Constant

Aerosol extinction profiles were retrieved using the HEIPRO algorithm, which is based on the optimal estimation (OE) method and minimizes the difference between the measured O₄ dSCDs and the corresponding radiative transfer simulations under a priori constraints (Frieß et al., 2006, 2011, 2019). No additional O₄ scaling factor (SF) was applied, as its necessity was not established for the present measurements.

In this work, the HEIPRO algorithm was reimplemented in Python following the retrieval framework and implementation described by Yilmaz (2012). Two modifications were introduced. First, the O₄ dSCDs from a complete elevation sequence were used as the input for each retrieval to achieve higher temporal resolution and better capture rapid changes in cloud conditions. Second, the radiative transfer model was updated to SCIATRAN 4.7 (Rozanov et al., 2014; Institute of Remote Sensing, University of Bremen, 2025) to support simulations under cloudy conditions. To evaluate the reliability of the reimplemented algorithm, cloud-free observations from 9 February 2025 were selected, and the retrieval results from the Python version were compared with those from the original HEIPRO algorithm. The aerosol extinction profiles retrieved by the two versions were generally consistent, indicating that the reimplemented algorithm preserves the main characteristics of the original framework. The remaining differences are mainly related to the temporal resolution of the O₄ dSCD input, the radiative transfer model version, and some model-related a priori settings. A detailed comparison is shown in Fig. 2. The year-round MAX-DOAS aerosol profile retrievals are provided in Supplement S4.



All aerosol retrievals in this study were performed under the cloud-free assumption. In the forward radiative transfer model, the atmosphere was assumed to be horizontally homogeneous, and the instrument was located at the surface. The a priori profiles of temperature, pressure, and trace gases were taken from the climatological database provided with the SCIATRAN package. This database, developed at the University of Bremen, provides monthly mean zonal distributions of the major trace gases and background atmospheric state information for the radiative transfer calculations (Rozanov et al., 2014; Institute of Remote Sensing, University of Bremen, 2025).

The aerosol retrieval settings were as follows. The aerosol a priori profile was assumed to decrease exponentially with altitude:

$$\alpha_{\text{aer},a}(z) = \alpha_{\text{aer},0} \exp\left(-\frac{z}{H}\right), \quad (1)$$

where $\alpha_{\text{aer},a}(z)$ denotes the aerosol a priori extinction coefficient at altitude z , $\alpha_{\text{aer},0}$ is the near-surface aerosol extinction coefficient, and H is the scale height. The total aerosol optical depth (AOD) of the a priori profile was set to 0.1, with a scale height of 1 km. The aerosol single-scattering albedo was set to 0.936. The vertical grid ranged from 0 to 4 km with a resolution of 0.2 km. The relative a priori error was set to 100%, and the measurement error was defined as the fitting error of the O_4 dSCD. The maximum number of iterations was set to 10, and the surface albedo at 360 nm was set to 0.05. A typical retrieval example is shown in Fig. 3.

The MAX-DOAS AOD used for classification and colour coding was calculated by integrating the retrieved aerosol extinction profiles over the fixed altitude range of 0–4 km.

The sensitivity and vertical resolution of the MAX-DOAS aerosol retrievals were characterized using averaging kernels (Rodgers, 2000). The averaging kernel matrix $\mathbf{A}$ is given by

$$\mathbf{A} = \mathbf{G}\mathbf{K}, \quad (2)$$

where $\mathbf{G}$ is the gain matrix and $\mathbf{K}$ is the Jacobian matrix. The diagonal elements of $\mathbf{A}$ describe the sensitivity of the retrieved aerosol extinction at each altitude to the true profile, with larger values indicating a stronger observational constraint and a weaker dependence on the a priori information. The trace of the averaging kernel matrix defines the degrees of freedom for signal (DOFS),

$$\text{DOFS} = \text{tr}(\mathbf{A}), \quad (3)$$

which quantifies the amount of independent information contained in the retrieval. Under the retrieval settings used in this study, the DOFS of the aerosol retrieval is typically about 2.

To quantitatively characterize the effective sensitivity range of the retrieval, an envelope was constructed from the vertical variation of the diagonal elements of the averaging kernel matrix. The altitude at which this envelope decreases to 10% of its maximum value was defined as the upper limit of the sensitivity height, denoted as H_m . Accordingly, the altitude range from 0 to H_m can be regarded as the effective sensitivity range of the MAX-DOAS aerosol retrieval. For the instrument and retrieval configuration used at this site, a reference H_m of about 2 km was obtained from clear-sky observations under favourable



155 observing conditions. These conditions were chosen to characterize the baseline vertical response consistently with the cloud-free forward model, without cloud-induced scattering and shielding. Cloud and aerosol extinction can modify the averaging kernels and retrieval information content (Frieß et al., 2019); the clear-sky sensitivity height therefore provides a reference for interpreting how cloud layers perturb the lower-atmospheric response. In the following sections, the 2 km reference height is used as a fixed cloud-base-height boundary to distinguish low-cloud and high-cloud states relative to this cloud-free sensitive

160 layer.

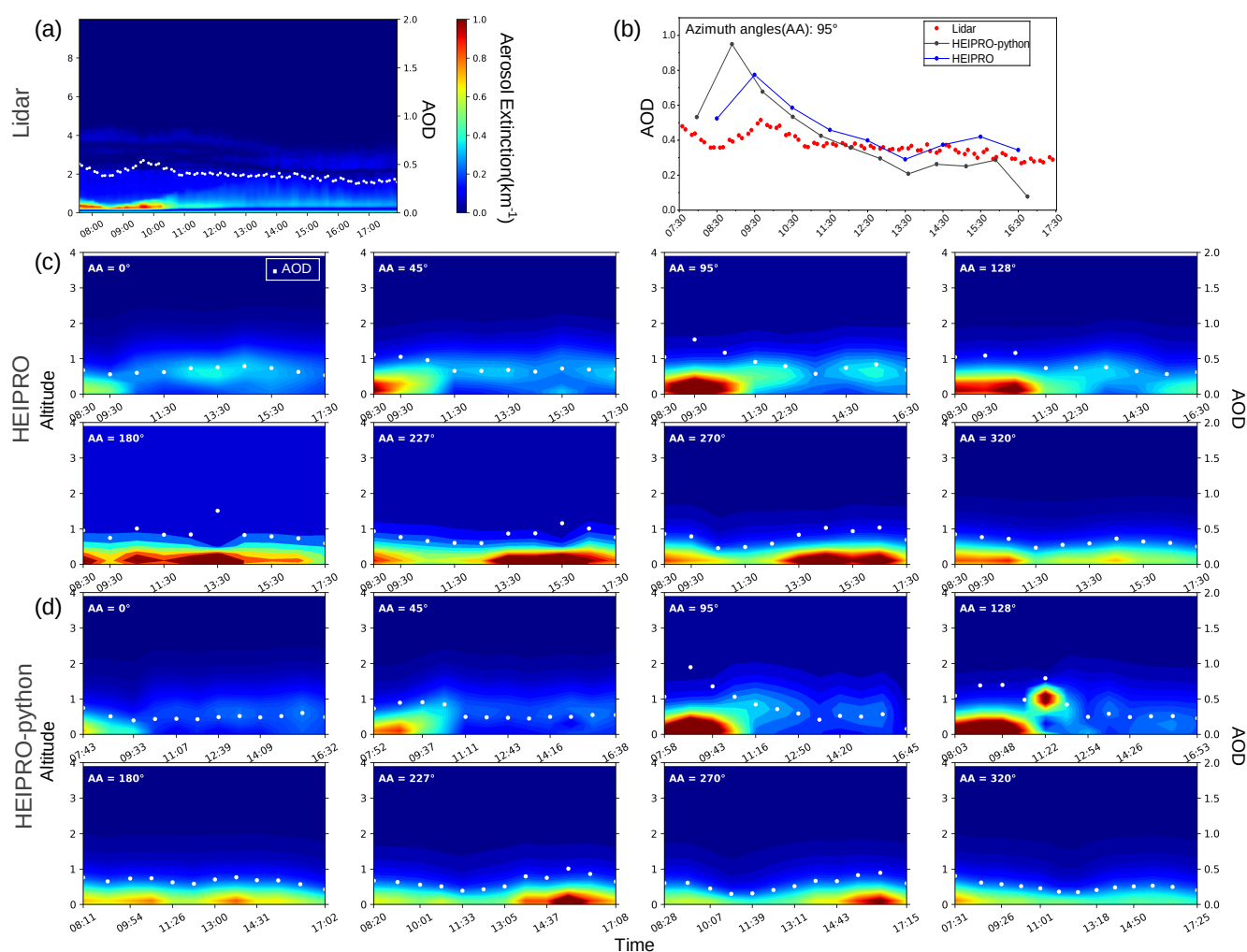


Figure 2. Comparison between lidar observations and aerosol retrievals from the original HEIPRO algorithm and its Python re-implementation. Panel (a) shows the lidar aerosol extinction profiles, with white dots indicating AOD integrated over 0–2 km. Panel (b) compares the AOD(< 2 km) time series at AA = 95°. Panels (c) and (d) show the aerosol extinction profiles retrieved at the eight azimuth angles by the original and Python-implemented HEIPRO algorithms, respectively.

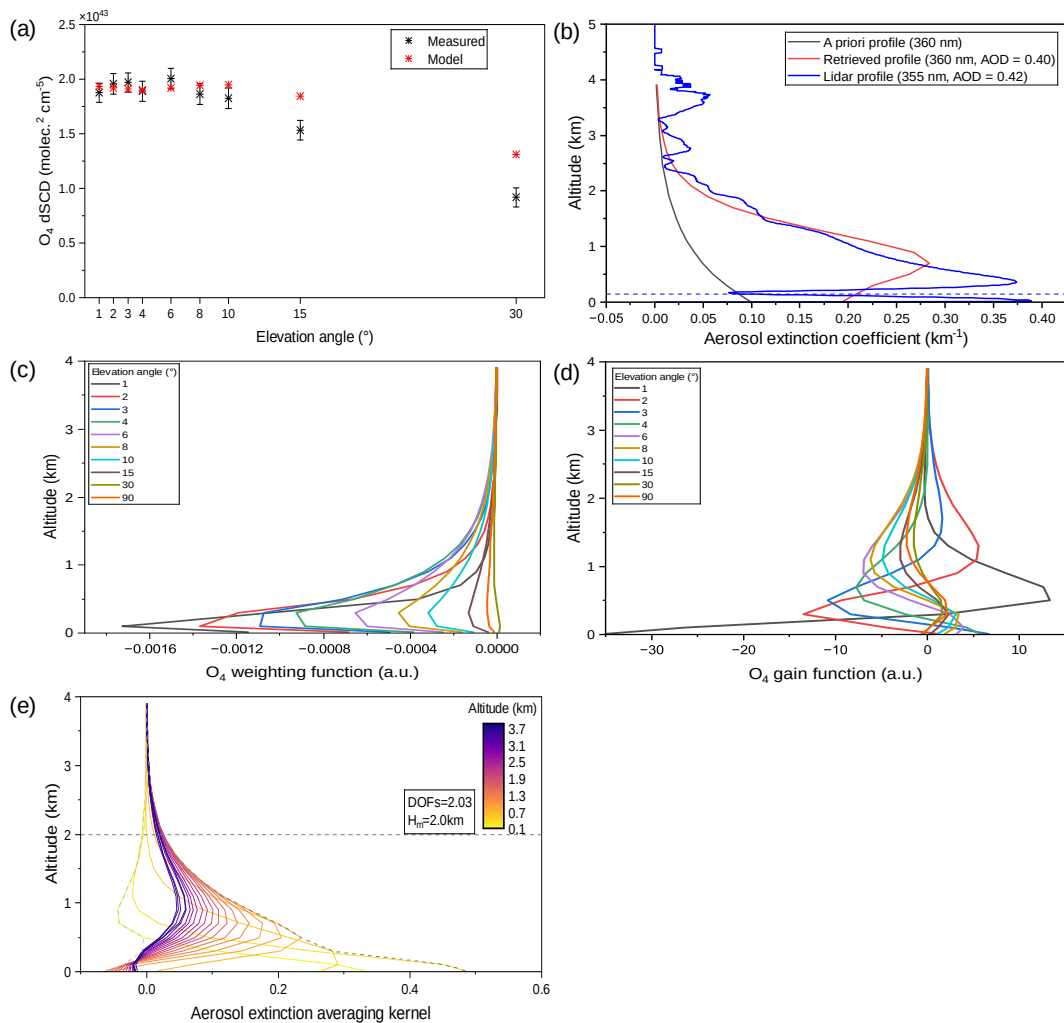


Figure 3. Example of a MAX-DOAS aerosol retrieval for 20 March 2025 at 12:00 local time and AA = 95°. (a) Measured and modelled O_4 dSCDs at different elevation angles. Error bars indicate the spectral-fitting uncertainties reported by QDOAS. (b) Comparison of the a priori profile, the retrieved profile, and the lidar reference profile. (c) Weighting functions of the O_4 absorption with respect to aerosol extinction. (d) Gain functions. (e) Averaging kernels of the extinction retrieval, together with the DOFS and the upper sensitivity height H_m .



2.3 Lidar aerosol extinction profiles

Raman–Mie lidar measurements have been used to derive vertically resolved aerosol optical and microphysical properties (Hu et al., 2019). The 355 nm aerosol extinction profile product used in this study was retrieved from a Raman–Mie scattering lidar system developed by the Anhui Institute of Optics and Fine Mechanics. The lidar system operates at 355 nm with an Nd:YAG laser at a repetition rate of 10 Hz. The aerosol extinction profile product has a temporal resolution of 10 min, a vertical resolution of 7.5 m, a detection range of 0–10 km, and a near-range blind zone of about 30 m. The year-round lidar aerosol extinction profiles are provided in Supplement S5.1.

The averaging kernels describe the vertical sensitivity and resolution of MAX-DOAS retrievals (Rodgers, 2000; Tirpitz et al., 2021). For comparison with the high-vertical-resolution lidar profiles, the lidar extinction profiles were multiplied by the MAX-DOAS averaging kernel matrices to facilitate comparison of profile structures under the different vertical responses of the two instruments.

Previous studies have developed cloud and aerosol layer-detection algorithms for lidar observations (Shen et al., 2024; Vaughan et al., 2009). In this study, cloud layers were identified from the lidar extinction profiles using an extinction threshold. The identified cloud layers were then used to determine cloud-layer presence and boundaries and to derive the cloud optical depth (COD).

3 Effects of clouds and aerosols on MAX-DOAS-derived quantities

This section analyses how clouds and aerosols affect MAX-DOAS-derived quantities through radiative transfer and measurement-response changes. The discussion focuses on zenith radiance, O₄ dSCD, and retrieved AOD under different cloud–aerosol conditions, providing the physical basis for the cloud–aerosol state classification method introduced later.

3.1 Radiance

3.1.1 Zenith radiance

Both aerosols and clouds can modify the radiative transfer process in the atmosphere and enhance particle scattering and multiple-scattering effects, thereby changing the number of photons entering the instrument field of view and affecting the observed radiance. For zenith observations, this radiance response is usually more evident because the light path is relatively short and the contribution of the Rayleigh-scattering background is comparatively limited. At the same time, the radiance change shows a clear wavelength dependence. In the ultraviolet spectral region, Rayleigh scattering is strong and background scattering is dominant, so the relative radiance enhancement caused by particle scattering is somewhat limited. In the visible spectral region, as Rayleigh scattering becomes weaker, the relative contribution of cloud and aerosol scattering increases, and the radiance enhancement becomes more pronounced. This wavelength-dependent response helps explain why the CI derived from zenith radiance is more effective for distinguishing cloudy and aerosol-loaded conditions from clear-sky conditions. In this study, the measured zenith signal, denoted by $I_{\text{obs}}(\lambda)$, is the zenith digital number (DN) normalized by the integration



time in milliseconds (DN ms^{-1}) and is used as a proxy for zenith spectral radiance. The MAX-DOAS instrument was not radiometrically calibrated; therefore, I_{obs} is not expressed in absolute radiance units. Simulated zenith spectral radiance is denoted by $I_{\text{sim}}(\lambda)$ and is expressed in $\text{W m}^{-2} \text{nm}^{-1} \text{sr}^{-1}$. The MAX-DOAS retrieval and the cloud-classification algorithm used here do not rely on absolute radiometric calibration.

The effects of clouds and aerosols on zenith radiance are also closely related to their optical thickness. For aerosols, because their extinction coefficients are relatively low and their vertical distributions are usually more diffuse, their influence on zenith radiance generally increases gradually with increasing AOD. In contrast, clouds have larger extinction coefficients and more vertically concentrated distributions. For optically thin clouds, enhanced multiple scattering can lead to a clear increase in radiance in the viewing direction. However, as cloud optical thickness increases further, photons undergo stronger multiple scattering within the cloud, reducing their probability of reaching the instrument. The shielding and upward reflection of incoming solar radiation by the cloud layer become stronger, leading to a decrease in zenith radiance. Therefore, the effect of clouds on zenith radiance is generally non-monotonic, changing from enhancement to reduction with increasing cloud optical thickness (Chiu et al., 2006). Fig. 4(a) shows simulations of the effects of clouds and aerosols with different optical thicknesses on zenith radiance.

3.1.2 TSI based on the zenith signal

Although variations in zenith radiance and the CI can both indicate the presence of aerosols and clouds, it is usually difficult to determine from a single MAX-DOAS measurement sequence whether these variations are mainly caused by aerosols or by clouds based only on their absolute values (Wagner et al., 2014, 2016). Cloud-edge transitions can produce changes in zenith radiance spectra that are sensitive to cloud properties (Chiu et al., 2010). Compared with cloud-free conditions, clouds usually exhibit stronger spatial and temporal variability as well as larger changes in optical thickness. Therefore, the temporal smoothness indicator (TSI) is introduced to quantify variations in MAX-DOAS-derived quantities associated with changes in cloud properties. We use the TSI formula of Wagner et al. (2014), with the measured zenith signal I_{obs} as the input, to obtain information on cloud variability from its temporal variations.

$$\text{TSI}_{I_{\text{obs}},n} = 2 \left(\frac{\Delta t_1 I_{\text{obs},n+1} + \Delta t_2 I_{\text{obs},n-1}}{\Delta t_1 \Delta t_2 (\Delta t_1 + \Delta t_2)} - \frac{I_{\text{obs},n}}{\Delta t_1 \Delta t_2} \right) \quad (4)$$

where $I_{\text{obs},n}$ denotes the measured zenith signal at 400 nm and n is the measurement-sequence index, and Δt_1 and Δt_2 are the time intervals in seconds between the current sequence and the preceding and following sequences, respectively. This expression approximates the second-order derivative with respect to time. Because I_{obs} is expressed in DN ms^{-1} , the TSI has units of $\text{DN ms}^{-1} \text{s}^{-2}$. When the derived quantity varies smoothly, for example owing to the gradual change in solar zenith angle (SZA), the magnitude of the TSI is usually small. In contrast, rapid changes between adjacent sequences lead to larger TSI magnitudes. The classification thresholds are applied to the absolute value, $|\text{TSI}|$.

Clouds can disturb the typical smooth variation of zenith radiance with SZA. Whether clouds enhance or reduce zenith radiance, the radiance variations are usually not synchronized with the slowly varying SZA-driven changes under cloud-free



conditions. Such deviations mainly reflect the spatial and temporal inhomogeneity of cloud fields, including the alternation
 225 between cloudy and cloud-free conditions. On this basis, the TSI of the measured zenith signal can be used to characterize its
 temporal variability and is particularly sensitive to short-term perturbations caused by cloud layers.

3.1.3 CI based on the zenith signal

Another quantity related to zenith radiance is CI. In this study, CI is defined as the ratio of the zenith spectral signal at 330 nm
 to that at 410 nm:

$$230 \quad CI = \frac{I_{\text{obs}}(330 \text{ nm})}{I_{\text{obs}}(410 \text{ nm})}, \quad (5)$$

where $I_{\text{obs}}(\lambda)$ denotes the measured zenith signal at wavelength λ . For the simulations, CI is calculated from I_{sim} at the same
 two wavelengths.

In existing MAX-DOAS cloud-classification methods, the CI is commonly used as an indicator of sky perturbation. However,
 the CI is influenced not only by clouds, but also by aerosol loading, viewing geometry, and the spectral response of the
 235 instrument. Therefore, changes in the CI cannot be uniquely attributed to cloud effects.

Previous studies proposed simulation-based CI corrections to account for observing conditions and instrumental effects
 (Wagner et al., 2016). CI responds to both clouds and aerosols, which limits its ability to distinguish specific cloud states.

As discussed above, some MAX-DOAS observations obtained under cloudy conditions may still retain retrieval value.
 Therefore, simply classifying observations into clear-sky and disturbed conditions based on the CI may overlook differences
 240 in retrieval applicability under different cloud regimes. In this study, the CI is not used as the main classification parameter, but
 only as an auxiliary quantity for analysing sky-perturbation features.

To further illustrate the response of the CI, radiative transfer simulations and year-round observations were analysed. Fig. 4
 shows the effects of clouds and aerosols on simulated zenith radiance and the CI. The simulations indicate that, relative to cloud-
 free conditions, increases in AOD and COD generally increase zenith radiance, except under optically thick cloud conditions,
 245 while both clouds and aerosols lead to lower CI values. Fig. 5 shows the observed CI-SZA distributions for different retrieved
 AOD ranges and retrieval-failed samples, together with the annual and seasonal frequency distributions of CI. The CI values are
 mainly concentrated around 0.31. Although this range is typically associated with observations affected by clouds or aerosols,
 some cases with relatively low retrieved AOD are also found near this value, as shown in Fig. 5(c) and (d).

3.2 O₄ dSCD

250 O₄ absorption has been widely used in remote-sensing studies of aerosols and clouds. In MAX-DOAS observations, differences
 in O₄ slant column density (SCD) among elevation angles contain information on photon path length and therefore provide
 important constraints for aerosol and boundary-layer retrievals. Because both aerosols and clouds modify photon scattering
 and propagation paths, they also affect the observed O₄ absorption signal. In this study, these effects are analysed using two
 indicators derived from the elevation-angle distribution of O₄ dSCDs. The spread of low-elevation O₄ dSCDs, denoted by
 255 SP_{low} , is defined as the difference between the maximum and minimum O₄ dSCDs across the measured elevation angles from

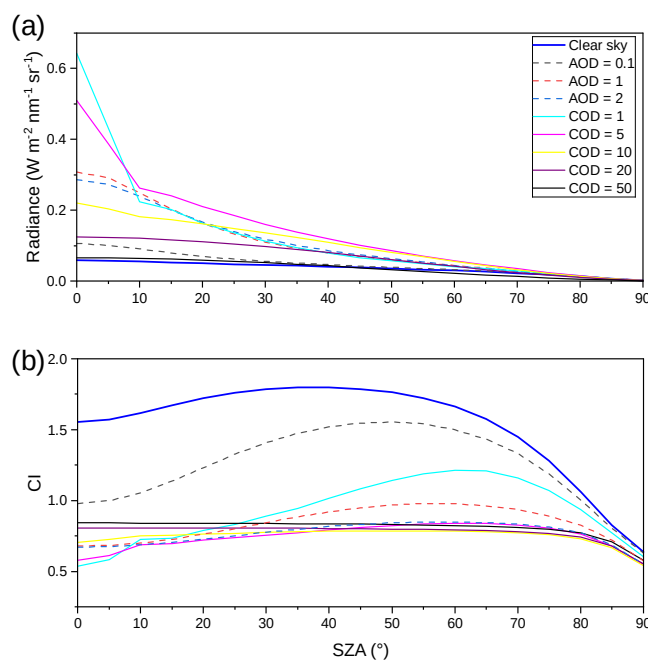


Figure 4. Simulated zenith radiance and colour index under different AOD and COD conditions as a function of SZA. Panel (a) shows the zenith radiance at 360 nm, and panel (b) shows $CI = I_{\text{sim}}(330 \text{ nm})/I_{\text{sim}}(410 \text{ nm})$. The AOD curves represent cloud-free conditions, whereas the COD curves represent cloudy conditions.

1° to 10° within each elevation sequence. In addition, the O_4 dSCD at 30° elevation, hereafter referred to as the 30° O_4 dSCD, is used as a high-elevation indicator to characterize the effect of cloud height on the observation light path.

The low-elevation O_4 dSCD spread, SP_{low} , can be used to characterize aerosol loading. Under low-elevation viewing conditions, O_4 dSCD is relatively sensitive to scattering processes in the near-surface atmosphere. Under clear-sky conditions with low aerosol loading, photon propagation is mainly controlled by Rayleigh scattering, and the light paths corresponding to different elevation angles differ substantially. As a result, the low-elevation O_4 dSCDs are more dispersed, corresponding to a larger SP_{low} . When aerosols are present, multiple scattering in the near-surface atmosphere is enhanced, and the photon propagation paths at different low elevation angles become more similar. The low-elevation O_4 dSCDs therefore become more concentrated, corresponding to a smaller SP_{low} .

By contrast, the 30° O_4 dSCD is more sensitive to the relative height of cloud layers. Clouds modify photon propagation paths through strong scattering and shielding effects, and this influence depends strongly on cloud height. When cloud layers are located within the effective sensitivity range of MAX-DOAS, they can more directly truncate the high-elevation observation paths, shorten the effective light path, and lead to a clear decrease in the 30° O_4 dSCD.

Fig. 6 shows simulations of O_4 SCDs and weighting functions for cloud-free conditions with different aerosol loadings and for cloudy conditions with low aerosol loading. For the cloud-free cases, the AOD values of the aerosol profile were set to 0.01,

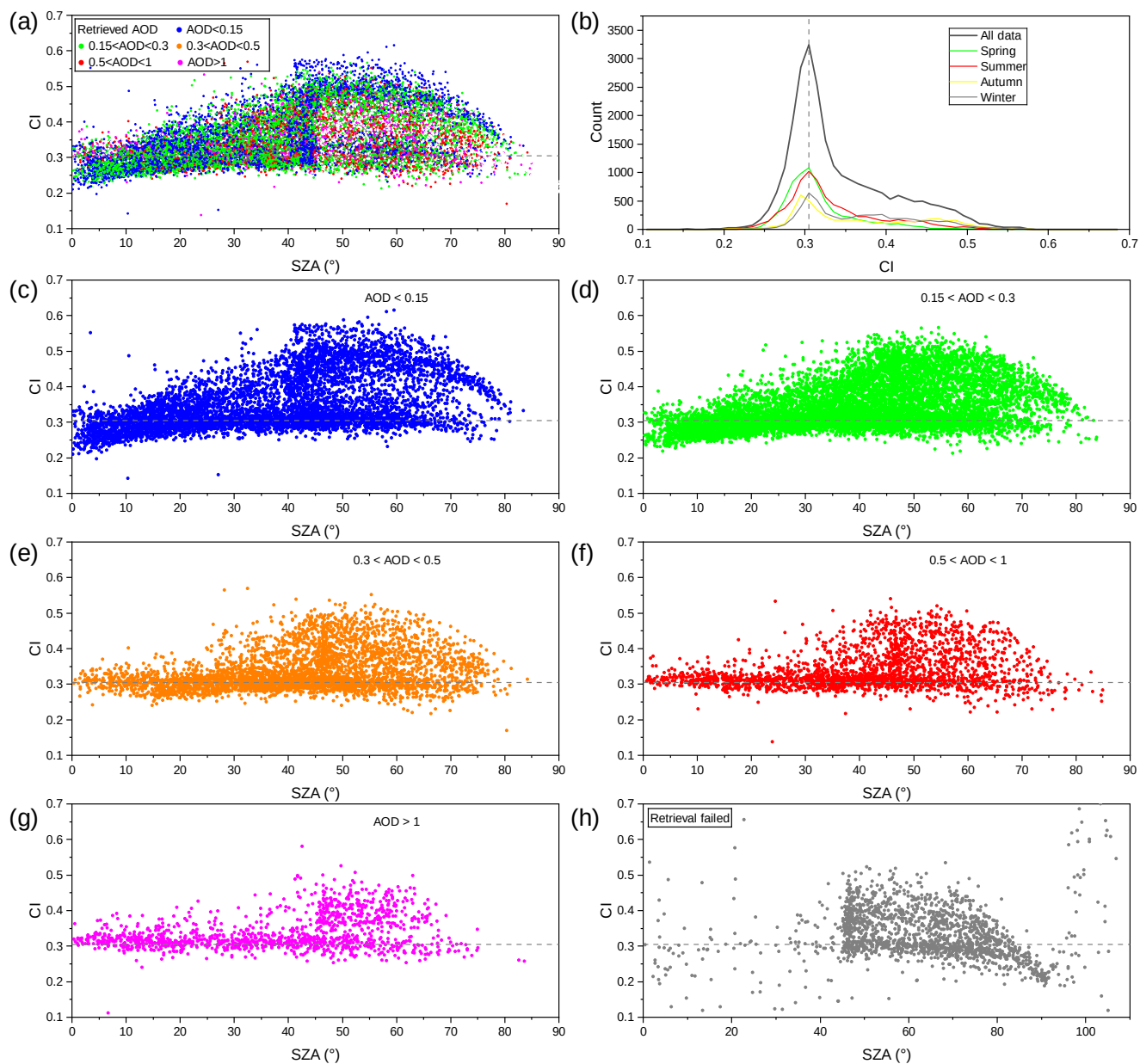


Figure 5. Observed characteristics of the colour index (CI), defined here as $I_{\text{obs}}(330 \text{ nm})/I_{\text{obs}}(410 \text{ nm})$. (a) CI as a function of SZA for all samples, with colours indicating different retrieved AOD ranges. (b) Frequency distributions of CI for the full year and for the four seasons. Panels (c)–(g) show the CI–SZA distributions for different AOD ranges, and panel (h) shows the distribution for retrieval-failed samples. The dashed line indicates CI = 0.31.



0.1, 1, and 2. For the cloudy cases, the cloud-base height (CBH) was set to 0, 0.5, 1, 1.5, 2, 3, and 4 km; the COD was set to 1, 5, 10, 20, and 50; and the cloud geometrical thickness was fixed at 0.2 km. This value is consistent with the typical geometrical thickness of cloud layers observed by the lidar throughout the year. Statistics of cloud geometrical thickness derived from the lidar extinction profiles are provided in Supplement S5.2. The O_4 SCD represents path-integrated O_4 concentration and provides information on effective light-path changes (Wagner et al., 2019; Beirle et al., 2022). The weighting functions describe the vertical sensitivity of observations at different elevation angles to aerosol and cloud extinction in the troposphere.

The simulations show that, under cloud-free conditions, the observation paths at low elevation angles become shorter as AOD increases, and the differences in path length among elevation angles become smaller. For cloudy cases with $CBH = 0$, the simulated situation approximately corresponds to fog or very low clouds and shows features similar to those under very high AOD conditions, with strongly shortened observation paths at all elevation angles. As COD increases, the MAX-DOAS sensitivity to near-surface aerosols decreases further. For low-cloud cases ($CBH < 2$ km), MAX-DOAS mainly responds to aerosols below the cloud layer, while its sensitivity to aerosols above the cloud layer is clearly suppressed, indicating that low clouds partly cut off the observational sensitivity to the atmosphere aloft. At the same time, differences in COD have a relatively limited effect on the low-elevation observation paths and weighting-function distributions under low-cloud conditions. By contrast, when $CBH > 2$ km, changes in cloud height and COD have only a weak influence on MAX-DOAS observations of near-surface aerosols. The corresponding weighting functions remain very similar to those under cloud-free conditions with low aerosol loading ($AOD = 0.01$), indicating that under high-cloud conditions MAX-DOAS retains nearly the same sensitivity to near-surface aerosols as under cloud-free conditions.

Fig. 7 shows simulations of zenith-referenced O_4 dSCDs at different elevation angles. Consistent with the light-path mechanisms revealed in Fig. 6, two main features can be identified. First, under high-AOD conditions, the differences among low-elevation O_4 dSCDs become much smaller, and the distribution becomes more concentrated, corresponding to a smaller SP_{low} . Second, when $CBH < 2$ km, the 30° O_4 dSCD is usually smaller than 5×10^{42} molec. 2 cm $^{-5}$. These results indicate that SP_{low} can be used to characterize aerosol loading, whereas the decrease in the 30° O_4 dSCD serves as an important indicator for identifying low clouds.

3.3 Retrieved AOD

Existing MAX-DOAS aerosol retrievals based on OE are usually developed under the clear-sky assumption. When clouds are present in the actual observations, they strongly modify photon propagation through scattering and shielding, thereby changing the O_4 dSCD. Because cloud effects are not included in the retrieval model, the resulting light-path changes can be misattributed to aerosol extinction, introducing systematic bias into the retrieval. Since MAX-DOAS retrievals are primarily sensitive to the lower atmosphere, this bias is usually amplified in the retrieved lower-layer aerosol extinction, leading to unrealistically enhanced near-surface aerosol extinction and overestimated AOD.

In this study, sensitivity experiments were conducted using radiative transfer simulations together with the OE retrieval framework. First, forward simulations were performed for prescribed aerosol and cloud parameters to generate O_4 dSCDs under different CBH conditions. In these simulations, the aerosol AOD was fixed at 0.1, the COD was fixed at 10, and CBH was



305 varied to represent different cloudy scenarios. The simulated O_4 dSCDs were then used as input to an OE retrieval framework established under the clear-sky assumption, with a cloud-free case at $AOD = 0.1$ used as the reference retrieval setting. The maximum number of iterations was set to 5. By comparing the retrieved aerosol profiles with the prescribed input profiles, the retrieval bias introduced by applying the clear-sky assumption under cloudy conditions was analysed.

Fig. 8 shows the retrieval results under different CBH conditions. For cases corresponding to near-surface fog or very low
 310 clouds, the retrieval terminated after the second iteration and yielded obviously non-physical high values. For $CBH < 2$ km, the lower the cloud base, the larger the overestimation of the retrieved aerosol extinction profile relative to the true profile. When CBH is about 2 km, the retrieval results are relatively close to those under clear-sky conditions. For $CBH > 2$ km, the retrieved aerosol extinction coefficients become slightly smaller than the true profile. This behaviour may be related to the decrease in zenith SCD and the increase in low-elevation SCD under high-cloud conditions, consistent with the simulations for
 315 $COD = 10$ shown in Fig. 6.

These results indicate that the retrieved AOD, as an integrated quantity derived from O_4 absorption, retains to some extent the response of cloud-height-dependent light-path changes. Low clouds can substantially shorten the effective light path, causing the retrieval to compensate by increasing near-surface aerosol extinction and thereby leading to overestimated AOD. By contrast, high clouds exert a weaker influence on observation paths that are mainly sensitive to the lower atmosphere and
 320 therefore have a more limited effect on the retrieval results. This also suggests that, under some conditions with $CBH > 2$ km, MAX-DOAS may still provide relatively reliable aerosol retrievals.

4 Cloud–aerosol classification method

In this study, zenith signal and its temporal variation represented by the TSI, the $30^\circ O_4$ dSCD, and the retrieved AOD are used as the main criteria for cloud–aerosol state classification. Zenith signal and TSI are used to identify cloud occurrence, the 30°
 325 O_4 dSCD is used to characterize relative cloud height, and the retrieved AOD is used to distinguish aerosol loading conditions. This section first summarizes the typical behaviour of these quantities under different weather conditions and then develops the corresponding threshold scheme and classification method.

4.1 Characteristics of derived quantities under different cloud–aerosol states

According to different combinations of cloud height and aerosol loading, sky conditions are classified into eight categories:
 330 clear sky, cloud-free conditions with high aerosol loading, high-cloud conditions with low aerosol loading, high-cloud conditions with high aerosol loading, low-cloud conditions with low aerosol loading, low-cloud conditions with high aerosol loading, optically thick cloud, and cloud holes. Figs. 9–16 present typical observation examples for these eight cloud–aerosol states. In these examples, the lidar aerosol extinction profiles provide independent information on the vertical structure and are used to identify cloud-layer position, aerosol distribution, and the corresponding state. The figures also illustrate the responses of
 335 the MAX-DOAS retrieval results and derived quantities under each condition. The main features of the MAX-DOAS-derived

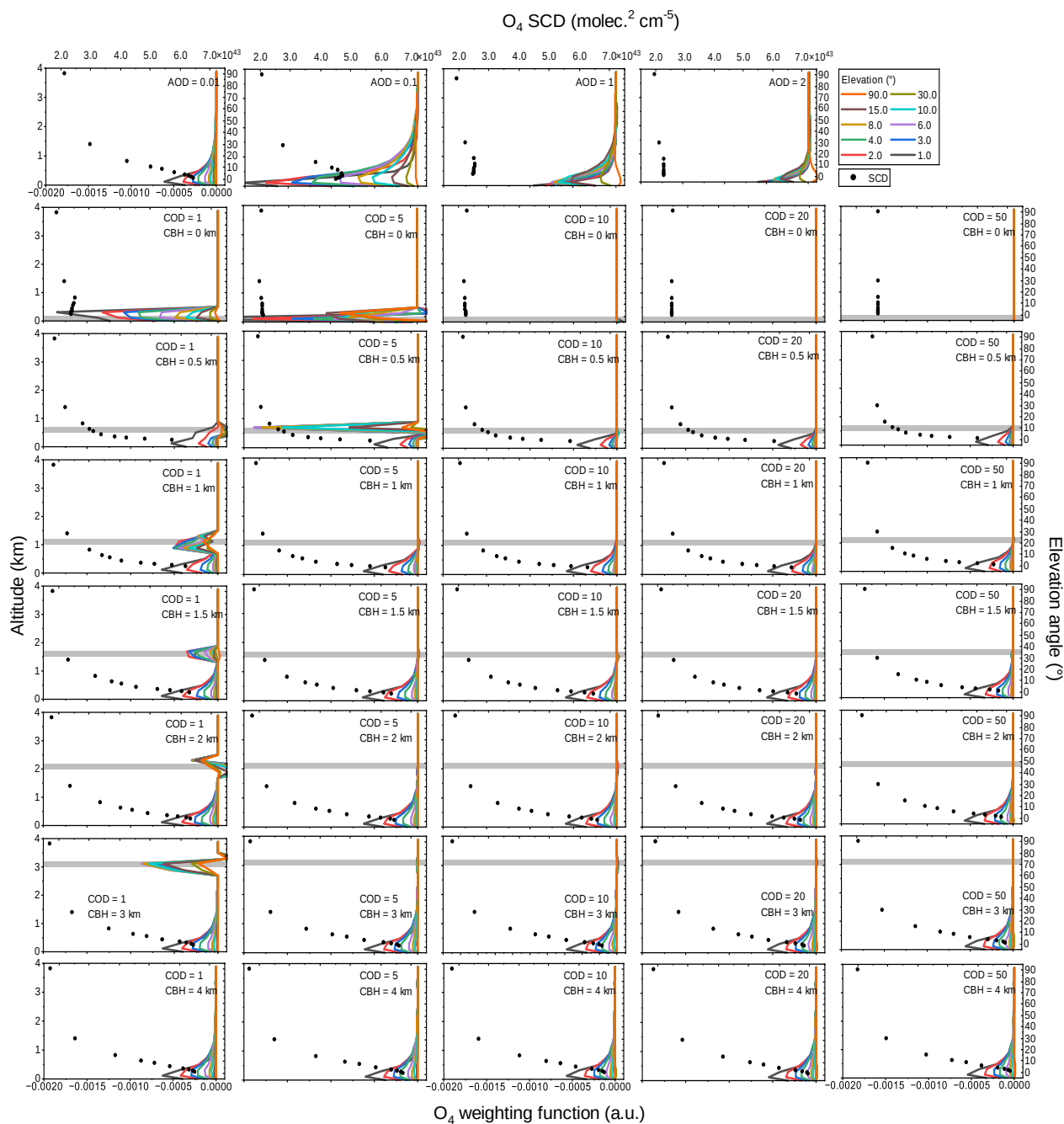


Figure 6. Simulated O_4 slant column densities and weighting functions for different aerosol and cloud scenarios. The first row shows cloud-free cases with different AOD values. The remaining rows show cloudy cases with different cloud-base heights, and the columns correspond to different CODs. The coloured curves indicate the O_4 weighting functions at different elevation angles, the black dots show the corresponding O_4 SCDs, and the grey bands indicate the cloud layer with a fixed geometric thickness of 0.2 km.

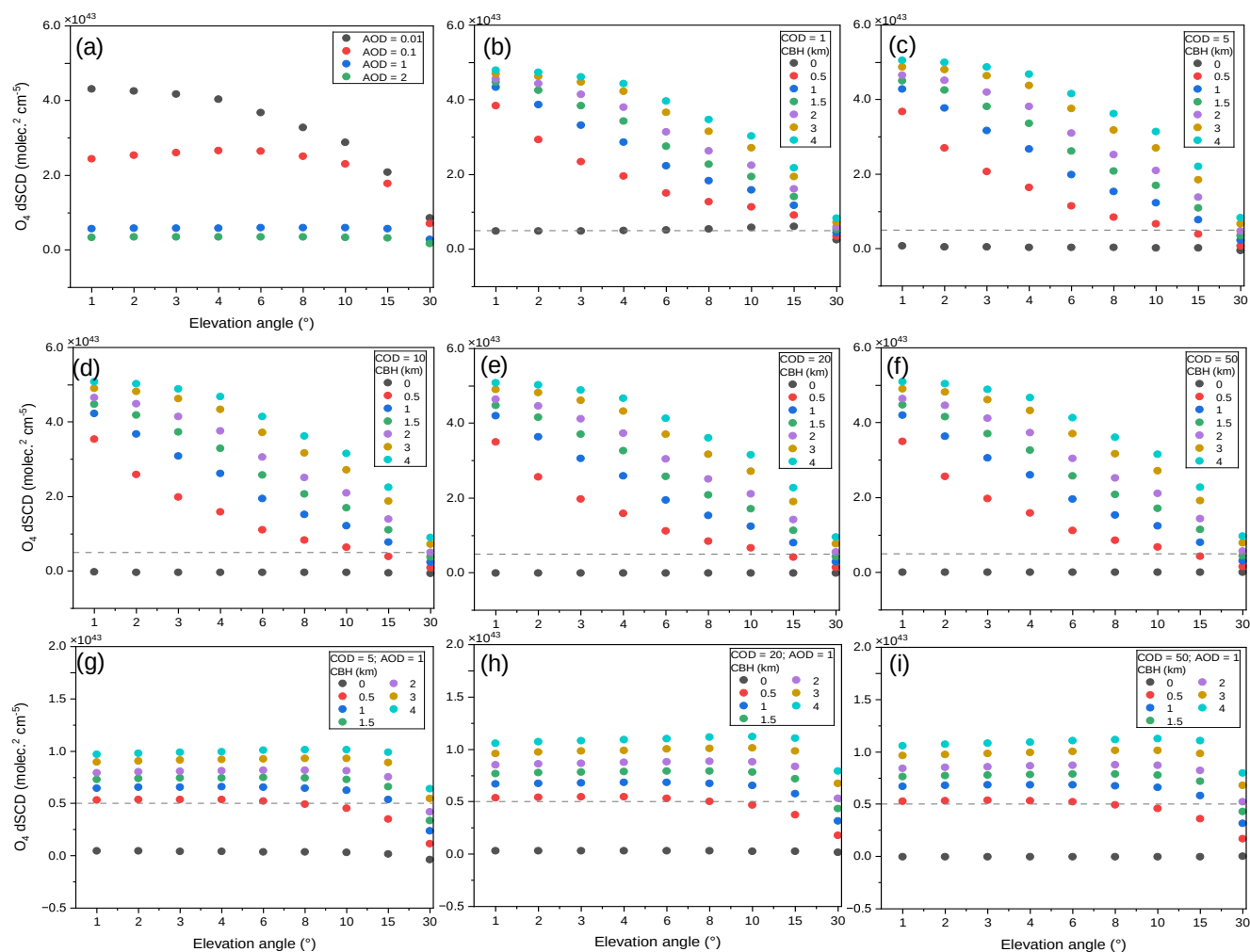


Figure 7. Simulated response of O_4 dSCDs to aerosol loading, CBH, and COD. Panel (a) shows cloud-free conditions with different AODs. Panels (b)–(f) show the dependence of O_4 dSCD on cloud-base height under different CODs. Panels (g)–(i) show the corresponding results for AOD = 1. The grey dashed line indicates the reference threshold of 5×10^{42} molec.² cm⁻⁵ for the 30° O_4 dSCD.

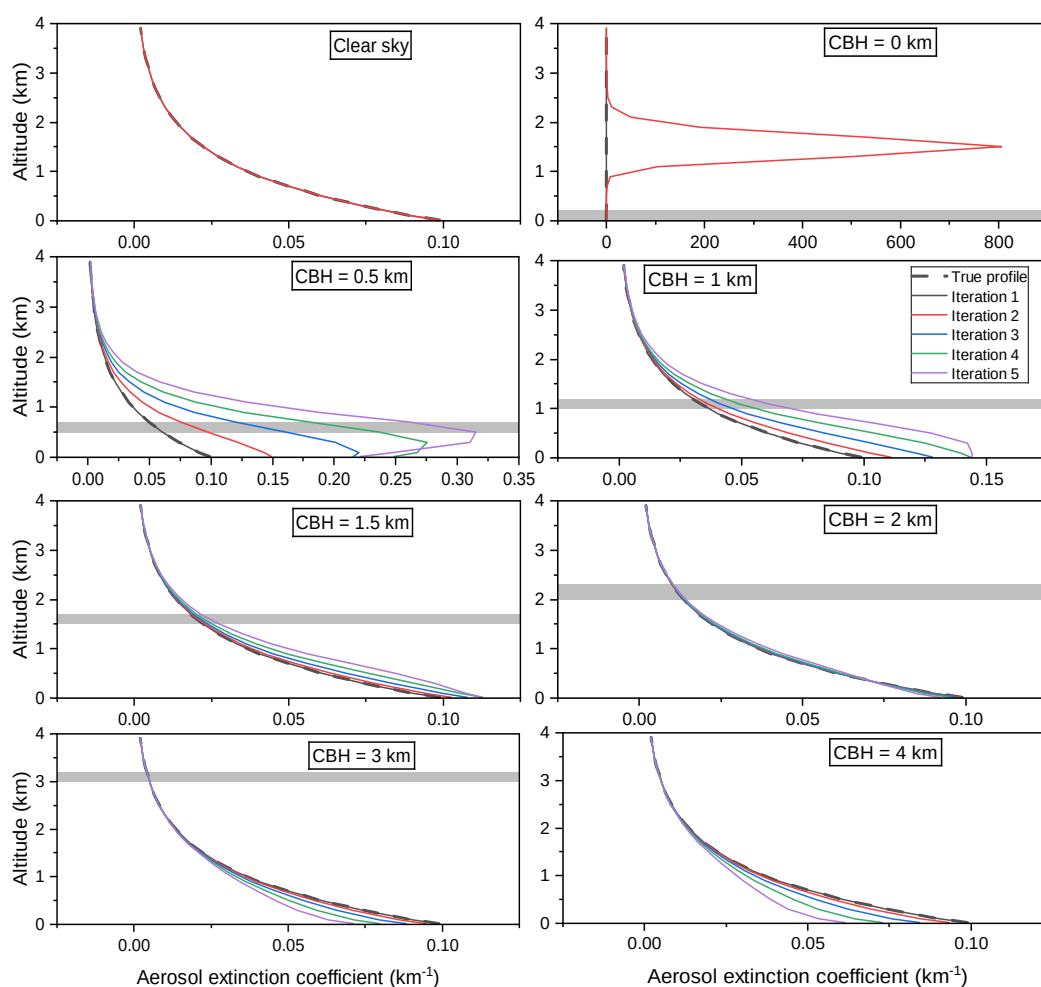


Figure 8. Iterative aerosol retrievals under the clear-sky assumption for COD = 10 and different cloud-base heights. The upper-left panel shows the cloud-free reference case, and the other panels correspond to different CBH values. The black dashed line indicates the true aerosol extinction profile, the coloured curves show the results of iterations 1–5, and the grey band indicates the fixed cloud layer.



quantities are summarized in Table 2. Here, “clear sky” denotes cloud-free conditions with low aerosol loading; the terms “low aerosol” and “high aerosol” in the category labels refer to aerosol loading.

Table 2. Typical characteristics of MAX-DOAS-derived quantities under different cloud–aerosol conditions.

Cloud–aerosol state	Zenith signal I_{obs}	TSI	30° O ₄ dSCD	SP _{low}	Retrieved AOD	CI
Clear sky	Low	Low	High	High	Low	High
Cloud-free (high aerosol)	High	Low	High	Low	High	Low
High cloud (low aerosol)	High	High	High	High	Low, close to clear sky	Low
High cloud (high aerosol)	High	High	High	Low	High	Low
Low cloud (low aerosol)	High	High	Low	High	Low	Low
Low cloud (high aerosol)	High	High	Low	Low	High	Low
Optically thick cloud	Strongly reduced	Low*	Low or negative	Low	Abnormally high	Low
Cloud holes	High and variable	High	High	Variable [†]	Variable [†]	Low, higher than low-cloud cases

* Under optically thick clouds, low zenith signals and long integration times generally lead to small TSI magnitudes. TSI is not used to identify this category. [†] These quantities vary with the aerosol background and do not exhibit a consistent high or low response.

In this study, the optically thick cloud category is identified when both $I_{\text{obs}} < I_{\text{ref},0.01}$ and $\text{AOD} > 1$ are satisfied. Such observation states may occur under very low clouds, near-surface fog, precipitation-related cloud systems, or other weak-signal conditions. Strong attenuation or shortened effective light paths can reduce the zenith signal, while retrievals under the cloud-free assumption may yield abnormally high AOD. The O₄ dSCD may also decrease markedly or become negative, but is not used as a criterion for this category. This label describes an observation state with potentially unreliable aerosol retrievals rather than a quantitative estimate of cloud optical thickness.

Cloud holes represent another special observation state with clear temporal discontinuity. In this study, cloud holes refer mainly to rapid spatial and temporal variations of low clouds below 2 km, namely short periods of open or intermittently transmissive conditions caused by a rapid decrease in cloud optical thickness under low-cloud conditions. Such scenes may correspond either to short-term cloud-free exposure or to very thin cloud conditions. Therefore, the corresponding derived quantities may in some cases resemble those under high-cloud conditions, while their identification uses the temporal context of nearby low-cloud observations. Because the measured zenith signal changes strongly with time, the TSI under cloud-hole conditions is usually high. Compared with low-cloud conditions, the CI is often somewhat larger, although its specific variation still depends on the meteorological background.

Together with the radiative transfer analysis in Sect. 3, these observational responses provide the basis for the classification thresholds and method.

It should be noted that the measured zenith signals shown in Fig. 9(d) are the original measurements and therefore retain the fixed azimuth-dependent bias caused by the non-level installation of the MAX-DOAS instrument. This installation-related structure was accounted for before the TSI-based classification by applying the preprocessing procedure described in Supple-



ment S3.1. In addition, local decreases in the 30° O_4 dSCD associated with the relative observation geometry under cloud-free conditions were treated before applying the O_4 dSCD threshold, as also described in Supplement S3.2.

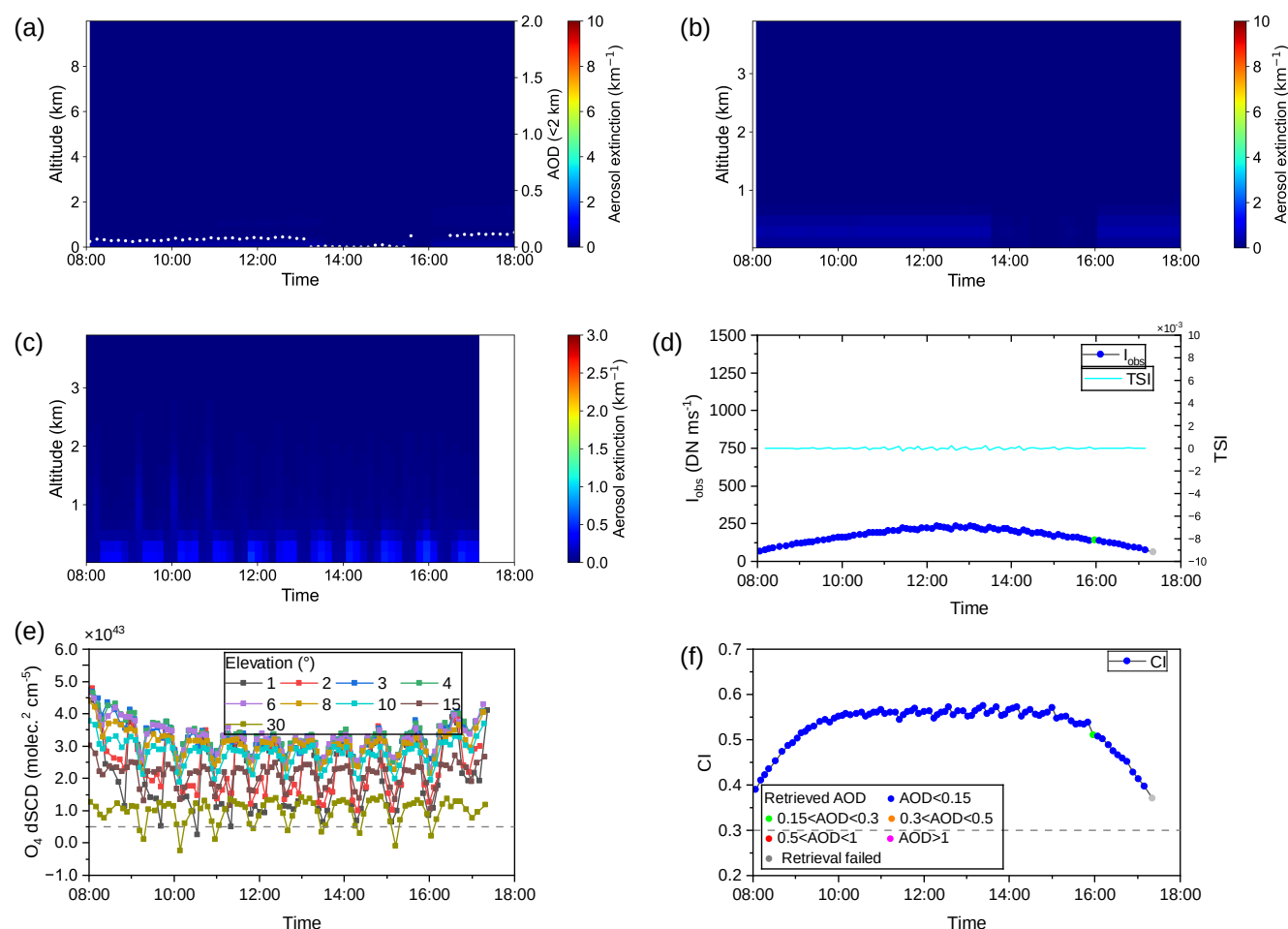


Figure 9. Example of clear-sky conditions on 28 January 2025. (a) Lidar-derived extinction profiles; white crosses indicate cloud-base height, and white dots indicate lidar-derived AOD below 2 km (right axis). (b) Lidar extinction profiles processed with the MAX-DOAS aerosol averaging kernels. (c) MAX-DOAS-retrieved aerosol extinction profiles. (d) MAX-DOAS zenith signal I_{obs} at 400 nm ($DN\ ms^{-1}$; left axis) and its temporal smoothness indicator (TSI; $DN\ ms^{-1}\ s^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS colour index (CI). Symbol colours in (d) and (f) indicate the MAX-DOAS-retrieved AOD ranges; grey indicates failed aerosol retrievals.

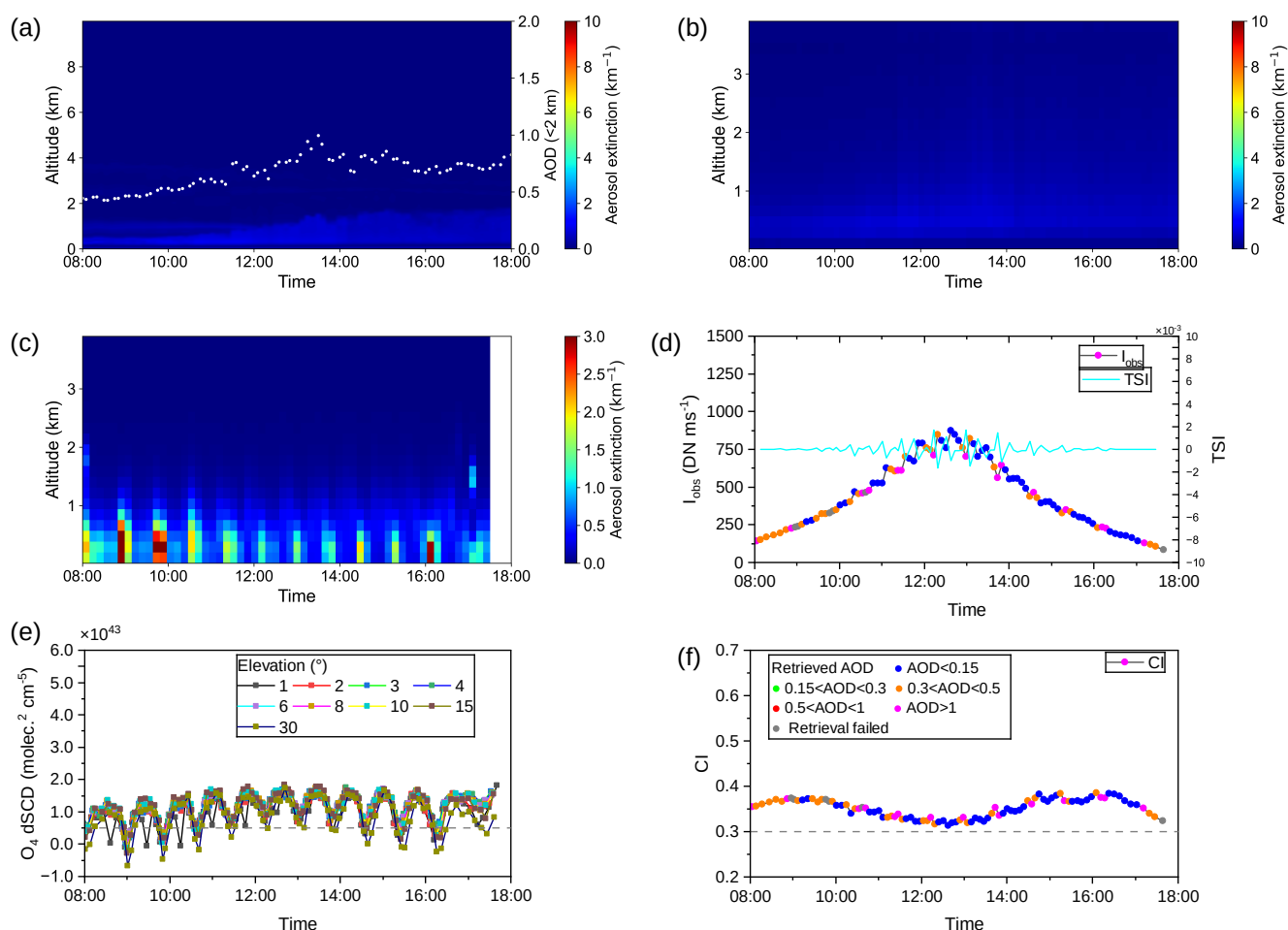


Figure 10. Example of cloud-free conditions with high aerosol loading on 22 March 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI (DN $\text{ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals.

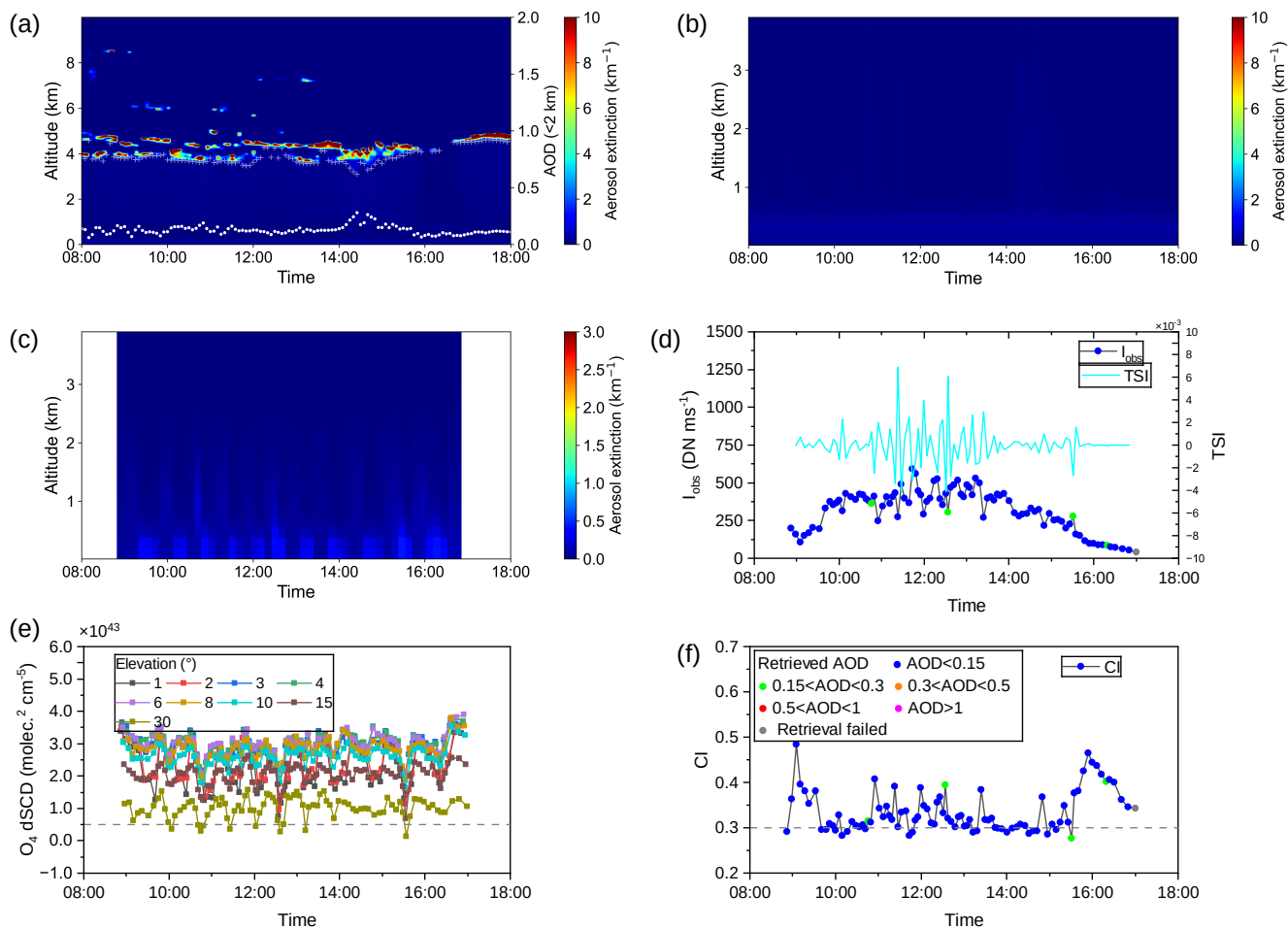


Figure 11. Example of high-cloud conditions with low aerosol loading on 20 November 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI ($\text{DN ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals.

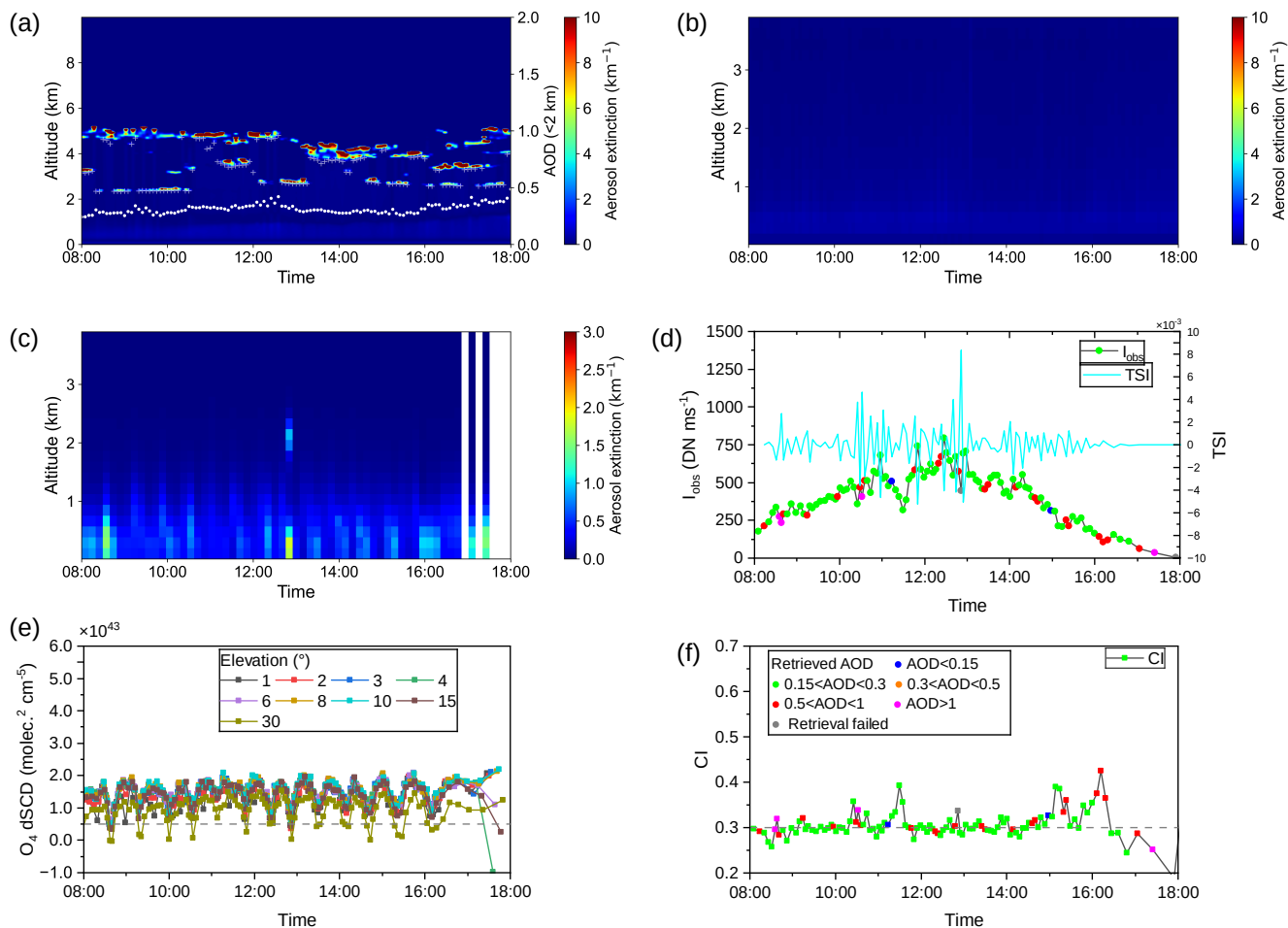


Figure 12. Example of high-cloud conditions with high aerosol loading on 27 October 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI ($\text{DN ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals.

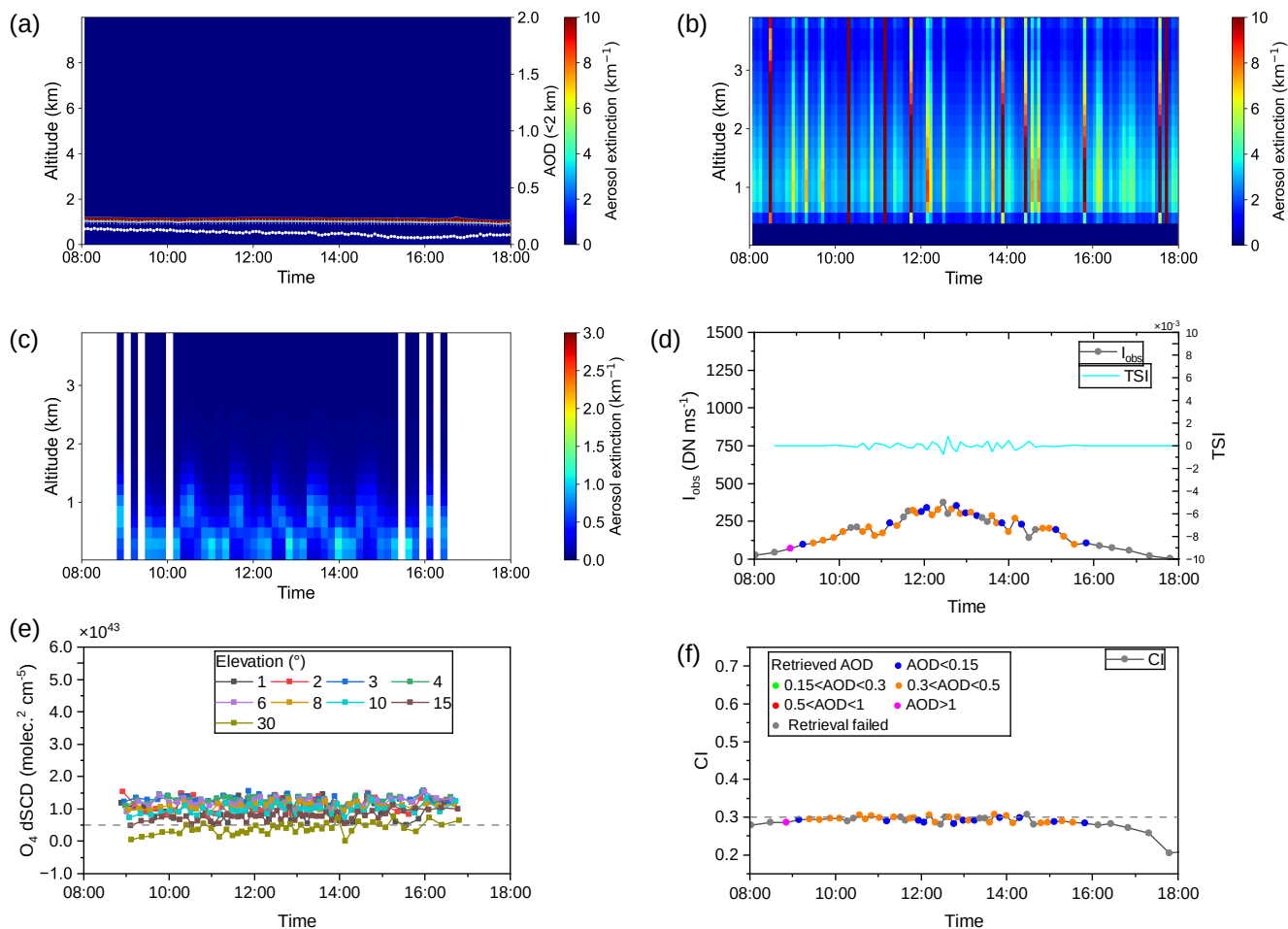


Figure 13. Example of low-cloud conditions with low aerosol loading on 26 December 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI (DN $\text{ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals. The low/high aerosol designation refers to the retrieved-AOD response category.

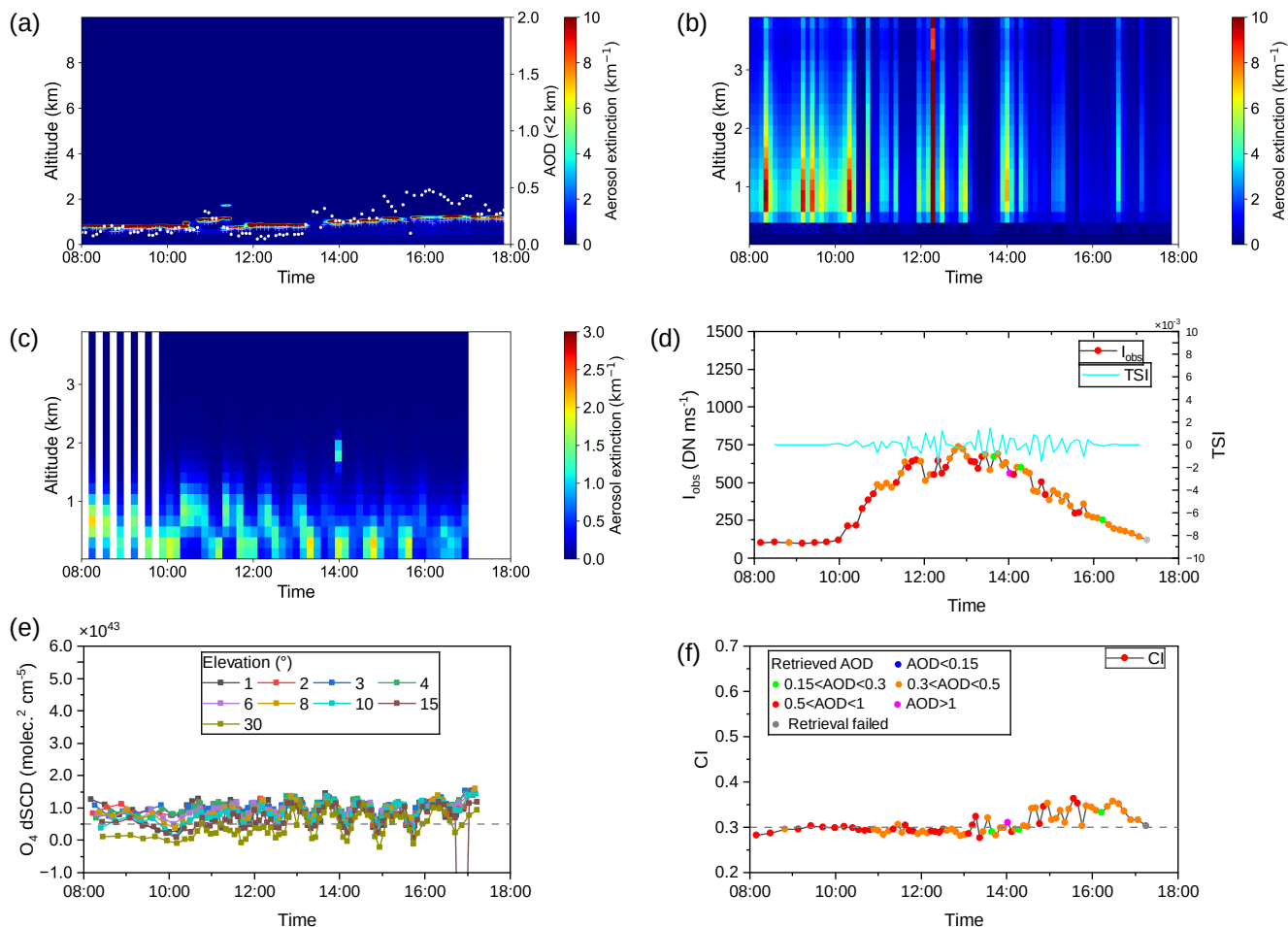


Figure 14. Example of low-cloud conditions with high aerosol loading on 18 February 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI ($\text{DN ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals. The low/high aerosol designation refers to the retrieved-AOD response category.

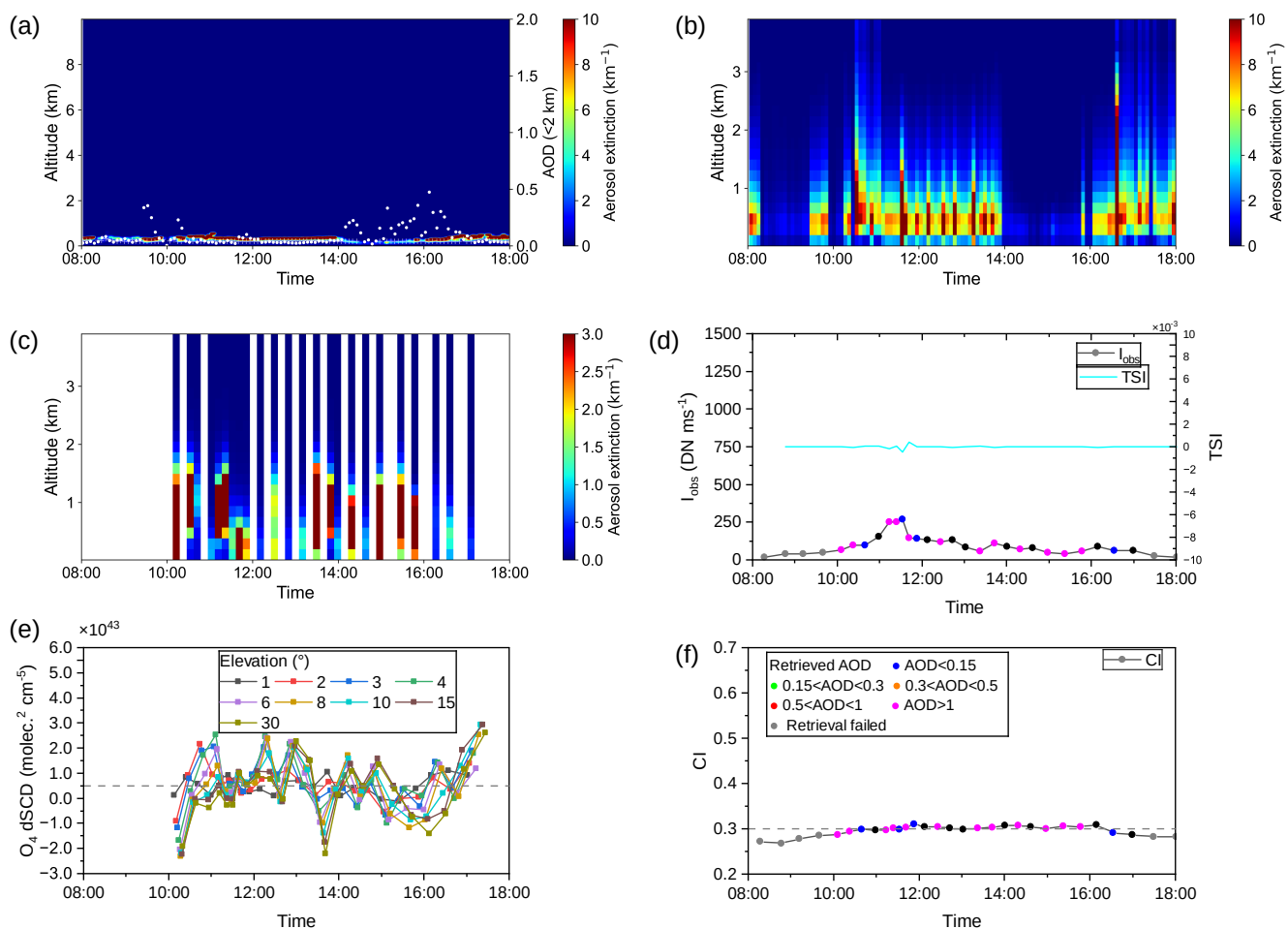


Figure 15. Example of optically thick cloud conditions on 5 March 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI ($\text{DN ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals.

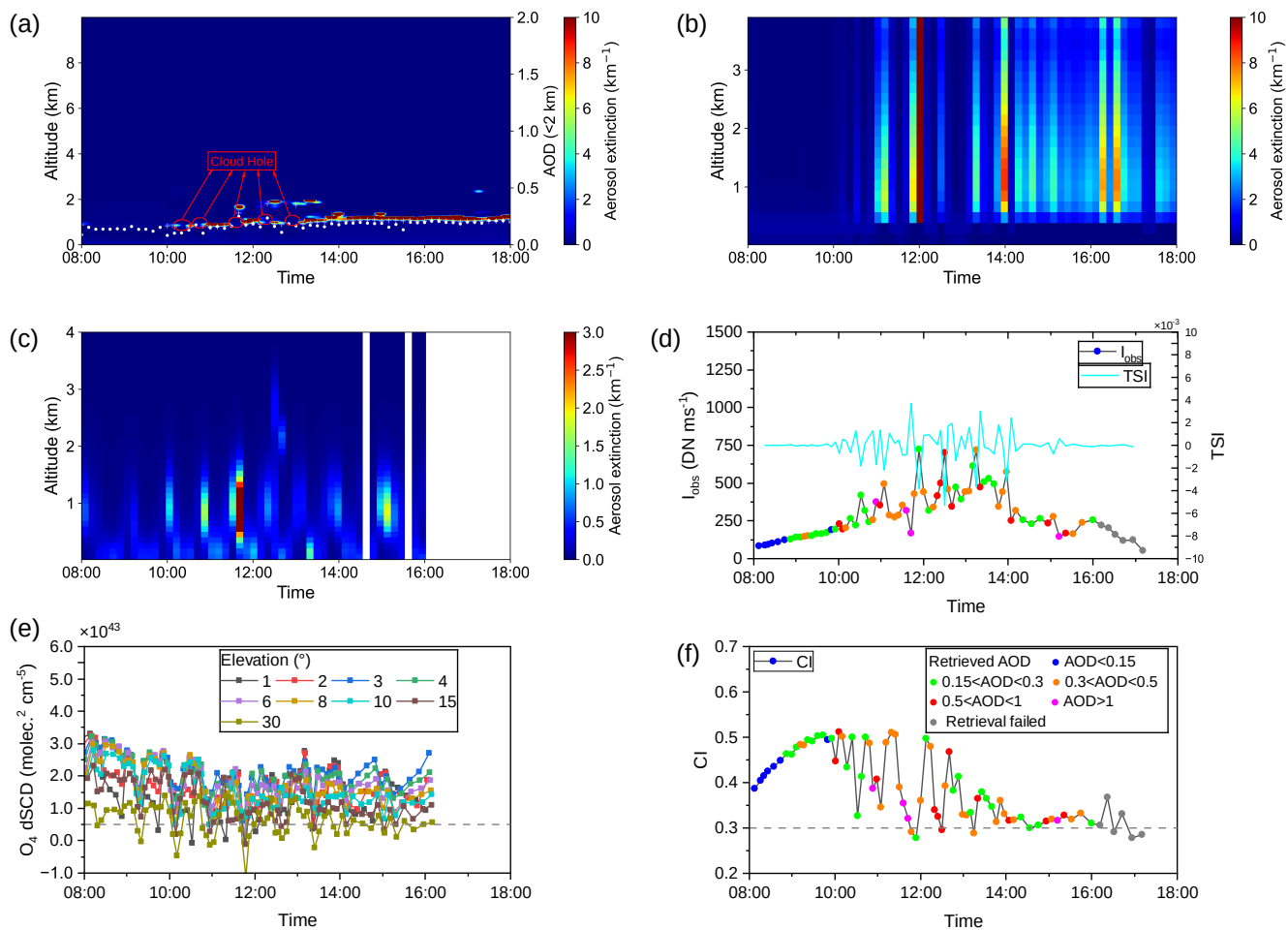


Figure 16. Example of cloud-hole conditions on 31 January 2025. (a) Lidar extinction profiles; white crosses indicate cloud-base height and white dots indicate AOD below 2 km (right axis). (b) Lidar profiles processed with MAX-DOAS averaging kernels. (c) MAX-DOAS-retrieved extinction profiles. (d) MAX-DOAS zenith signal I_{obs} and TSI ($\text{DN ms}^{-1} \text{s}^{-2}$; right axis). (e) MAX-DOAS O_4 dSCDs at different elevation angles. (f) MAX-DOAS CI. Colours in (d) and (f) indicate retrieved-AOD ranges; grey denotes failed retrievals. Red annotations in (a) highlight the cloud-hole intervals discussed in the text.



4.2 Classification thresholds and method

Based on the observational features summarized in Sect. 4.1, zenith signal and its TSI, the 30° O_4 dSCD, and the retrieved AOD were selected as the main criteria for cloud–aerosol state classification. Zenith signal and TSI are used to identify cloud occurrence, the 30° O_4 dSCD is used to separate high-cloud and low-cloud conditions, and the retrieved AOD is used to characterize aerosol loading conditions.

The separation between cloudy and cloud-free conditions is based on the comparison of the measured zenith signal and its TSI with the cloud-free reference behaviour. A relative scaling method based on radiative transfer simulations was used to provide reference signal ranges for different aerosol conditions. In the radiative transfer model, $AOD = 0.01$ and $AOD = 1$ were used to represent clean-background and high-aerosol-loading conditions, respectively. A typical clear-sky day with stable aerosol conditions, 28 January 2025, with a mean AOD of about 0.05, was selected as the reference observation day. The simulated zenith radiance I_{sim} for $AOD = 0.05$ was scaled to match the measured zenith signal I_{obs} , thereby establishing the relationship between model radiance and the observed signal. The corresponding cloud-free reference thresholds, denoted by $I_{ref,0.01}$ and $I_{ref,1}$, are expressed on the same detector-signal scale as I_{obs} and represent the lower and upper cloud-free reference bounds. This scaling does not constitute an absolute radiometric calibration. The scaling results are shown in Fig. 17.

The threshold for the 30° O_4 dSCD was determined from both simulations and observations. The simulation results in Fig. 7 and the relationship between the 30° O_4 dSCD and cloud height shown in Figs. 11– 14 indicate that this quantity responds consistently to low clouds. Therefore, a threshold of $5 \times 10^{42} \text{ molec.}^2 \text{ cm}^{-5}$ was used to distinguish high-cloud from low-cloud conditions.

The retrieved-AOD thresholds used for classification were 0.15, 0.3, and 1. $AOD < 0.15$ identified low-aerosol conditions in cloud-free and high-cloud cases, while 0.3 separated the two low-cloud retrieved-AOD subcategories. Optically thick clouds were identified when $AOD > 1$ and $I_{obs} < I_{ref,0.01}$ were simultaneously satisfied.

The aerosol retrieval assumes cloud-free conditions and does not explicitly include cloud parameters. Cloud-induced changes in the light path can therefore be interpreted by the inversion as changes in aerosol extinction. The simulations in Figs. 6 and 7 show that cloud height and optical depth affect the observation light paths, while Fig. 8 demonstrates the resulting cloud-height-dependent AOD bias at a fixed COD of 10. We therefore empirically increase the AOD threshold to 0.3 for low-cloud cases to distinguish two retrieval-response categories within the classification scheme. This threshold adjustment does not remove cloud-induced bias, and the retrieved AOD under low-cloud conditions cannot be directly interpreted as the actual aerosol loading. The “low aerosol” and “high aerosol” labels consequently describe lower and higher retrieved AOD, respectively.

The main classification thresholds used in this study are listed in Table 3.

Based on these thresholds, an automatic cloud–aerosol state classification method was established using MAX-DOAS-derived quantities. The procedure follows five main steps: (a) optically thick cloud cases are identified when both $I_{obs} < I_{ref,0.01}$ and $AOD > 1$ are satisfied; (b) cloud-free cases are identified using zenith signal and TSI relative to the cloud-free reference behaviour and are further separated into low- and high-aerosol cases using the retrieved AOD; (c) for cloud-affected observations, the 30° O_4 dSCD is used to separate high-cloud and low-cloud conditions; (d) the retrieved AOD is used to

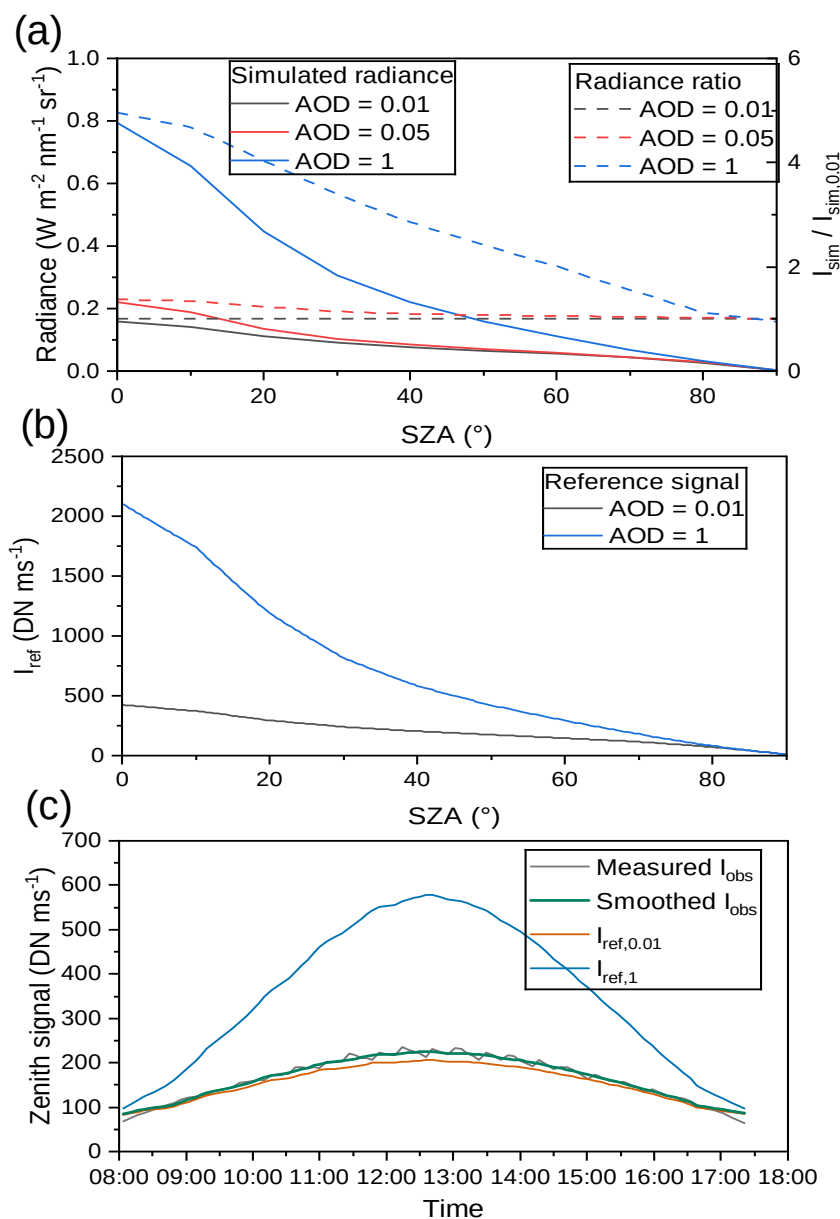


Figure 17. Determination of the zenith-signal reference thresholds. (a) Simulated zenith spectral radiance I_{sim} at 400 nm under cloud-free conditions for different AOD values (solid lines; left axis; $\text{W m}^{-2} \text{nm}^{-1} \text{sr}^{-1}$), and the dimensionless radiance ratio $I_{\text{sim}}/I_{\text{sim},0.01}$ relative to AOD = 0.01 (dashed lines; right axis). (b) Reference thresholds $I_{\text{ref},0.01}$ and $I_{\text{ref},1}$ obtained by scaling the simulated radiance to the measured zenith-signal scale (DN ms^{-1}). (c) Measured and smoothed zenith signals on the clear-sky reference day, 28 January 2025 (mean AOD ≈ 0.05), together with the reference bounds $I_{\text{ref},0.01}$ and $I_{\text{ref},1}$; all signals in (b) and (c) are expressed in DN ms^{-1} .



Table 3. Classification indicators and thresholds.

Indicator	Threshold
Zenith signal I_{obs}	$I_{\text{ref},0.01}$ and $I_{\text{ref},1}$
TSI	$1 \times 10^{-4} \text{ DN ms}^{-1} \text{ s}^{-2}$
$30^\circ \text{ O}_4 \text{ dSCD}$	$5 \times 10^{42} \text{ molec.}^2 \text{ cm}^{-5}$
AOD	0.15, 0.3, 1
Cloud-hole time window	$\pm 30 \text{ min}$

distinguish aerosol loading under high-cloud and low-cloud conditions; and (e) cloud-hole cases are identified using a temporal-window criterion. Specifically, if an observation initially classified as high cloud has low-cloud observations within $\pm 30 \text{ min}$, it is relabelled as a cloud-hole case. The threshold criteria for the eight cloud–aerosol states are summarized in Table 4.

Table 4. Threshold criteria for different cloud–aerosol states. For optically thick clouds, the zenith-signal and AOD conditions must both be satisfied; no $\text{O}_4 \text{ dSCD}$ threshold is applied to this category. TSI thresholds are expressed in $\text{DN ms}^{-1} \text{ s}^{-2}$, and $\text{O}_4 \text{ dSCD}$ thresholds in $\text{molec.}^2 \text{ cm}^{-5}$.

Cloud–aerosol state	Zenith signal I_{obs}	TSI	$30^\circ \text{ O}_4 \text{ dSCD}$	AOD
Clear sky	$< I_{\text{ref},1}$	$< 1 \times 10^{-4}$	$> 5 \times 10^{42}$	< 0.15
Cloud-free (high aerosol)	$< I_{\text{ref},1}$	$< 1 \times 10^{-4}$	$> 5 \times 10^{42}$	> 0.15
High cloud (low aerosol)	$> I_{\text{ref},0.01}$	$> 1 \times 10^{-4}$	$> 5 \times 10^{42}$	< 0.15
High cloud (high aerosol)	$> I_{\text{ref},0.01}$	$> 1 \times 10^{-4}$	$> 5 \times 10^{42}$	> 0.15
Low cloud (low aerosol)	$> I_{\text{ref},0.01}$	$> 1 \times 10^{-4}$	$< 5 \times 10^{42}$	< 0.3
Low cloud (high aerosol)	$> I_{\text{ref},0.01}$	$> 1 \times 10^{-4}$	$< 5 \times 10^{42}$	> 0.3
Optically thick cloud	$< I_{\text{ref},0.01}$	—	—	> 1
Cloud holes	$> I_{\text{ref},0.01}$	$> 1 \times 10^{-4}$	$> 5 \times 10^{42}$	—

A dash indicates that no criterion is applied to that quantity for the corresponding category.

5 Results

5.1 Comparison of classification results from MAX-DOAS and lidar

To evaluate the proposed cloud–aerosol classification method, sky conditions over Guangzhou Panyu in 2025 were classified using MAX-DOAS observations and near-co-located lidar aerosol profile data. The comparison between the two classification results was used to assess the capability of MAX-DOAS to identify cloud–aerosol states under complex cloud conditions.

Fig. 18 shows the year-round cloud–aerosol classification results based on MAX-DOAS observations. The classified states show temporally coherent patterns in the time series. Clear-sky conditions and cloud-free conditions with high aerosol loading usually correspond to relatively stable observation periods, whereas cloudy states are more often characterized by shorter-



duration and more rapidly changing processes, although some cloudy periods also show clear temporal continuity. Non-clear-sky conditions account for a large fraction of the observations, indicating that cloud–aerosol states are common in Guangzhou.

Fig. 19 shows the distribution of lidar-derived below-cloud AOD and CBH. Cloud layers were identified from the lidar extinction profiles using extinction thresholds of 1 km^{-1} below 3 km and 0.5 km^{-1} above 3 km. The results show that both low-AOD clean-background conditions and high-AOD pollution episodes occurred in Guangzhou during the year. Low and high clouds also appeared frequently, resulting in a wide range of cloud–aerosol combinations.

The lidar-derived below-cloud AOD and cloud-layer heights provide a reference for evaluating the MAX-DOAS classification. The frequent occurrence of clouds and high aerosol loading in Guangzhou also indicates that simply discarding all cloud-affected MAX-DOAS observations would lead to a substantial loss of information under complex sky conditions. Therefore, identifying observation scenes that retain retrieval value under different cloud conditions is important for improving the use of long-term MAX-DOAS datasets.

Fig. 20 shows the year-round cloud–aerosol classification results derived from the lidar observations. This classification is based on lidar-derived CBH, below-cloud AOD, and COD, with the corresponding thresholds listed in Table 5. It should be noted that cloud holes are treated as a special type of low-cloud scene in the lidar classification, usually with $\text{CBH} < 2 \text{ km}$ and $\text{COD} < 0.5$. The year-round distribution shows frequent transitions among different cloud–aerosol states in Guangzhou, with low-cloud-related states accounting for a large fraction. Cloud-free conditions show relatively good temporal continuity during some periods. These results indicate that cloud–aerosol conditions over the study region are highly variable, including relatively stable clean or polluted episodes as well as many mixed states under rapidly evolving cloud conditions.

Compared with the MAX-DOAS classification results shown in Fig. 18, the lidar classification shows similar process evolution on the annual scale, indicating general agreement between the two methods in identifying the main cloud–aerosol states. In particular, for clear-sky conditions and some low-cloud processes, the two methods respond consistently to changes in cloud–aerosol state. By contrast, differences remain for scenes such as high clouds and cloud holes, where the boundaries are less distinct or the conditions change rapidly. These differences reflect the intrinsic differences between passive and active remote sensing in observation geometry, sensitivity range, and temporal response.

Fig. 21 shows the annual and monthly statistical distributions of the cloud–aerosol classification results from MAX-DOAS and lidar, based on 22848 and 29159 valid classifications, respectively. These distributions use all valid classifications from each instrument rather than only the time-matched subset used in the agreement analysis. In the annual statistics, cloud-free, high-cloud, and low-cloud-related conditions account for 38.7 %, 20.0 %, and 41.3 %, respectively, in the MAX-DOAS classification, while the corresponding fractions in the lidar classification are 37.5 %, 18.3 %, and 44.3 %. These fractions are calculated from the aggregated class counts before rounding. Both classifications show that low-cloud-related conditions account for the largest fraction in Guangzhou, followed by cloud-free conditions, whereas high-cloud conditions account for a smaller fraction. Both datasets show month-to-month variability, with differences in the fractions of specific categories.

Fig. 22 shows the cross-distribution matrix of the cloud–aerosol classification results from MAX-DOAS and lidar, which is used to quantitatively assess the agreement and misclassification patterns between the two methods. Overall, the concentration of samples along the diagonal indicates good agreement under cloud-free conditions and a relatively strong correspondence



Table 5. Thresholds used for the lidar-based cloud classification.

Cloud–aerosol state	AOD	CBH (km)	COD
Clear sky	< 0.15	—	—
Cloud-free (high aerosol)	> 0.15	—	—
High cloud (low aerosol)	< 0.15	> 2	—
High cloud (high aerosol)	> 0.15	> 2	—
Low cloud (low aerosol)	< 0.15	< 2	> 0.5
Low cloud (high aerosol)	> 0.15	< 2	> 0.5
Cloud holes	—	< 2	< 0.5

A dash indicates that no criterion is applied to that quantity for the corresponding category. For the two cloud-free categories, no cloud layer is detected, so CBH and COD are not applicable.

for some low-cloud cases. By contrast, the off-diagonal elements indicate classification discrepancies, which are more evident among high-cloud, low-cloud, and cloud-hole cases. This suggests that such boundary-type or rapidly changing scenes are more strongly affected by differences in observation geometry and in the physical quantities used for classification.

To evaluate the ability of MAX-DOAS to identify cloud-height categories, the MAX-DOAS and lidar classification results were further grouped into three classes: cloud-free, high-cloud, and low-cloud conditions. The two grouped classifications were then compared using time-matched samples. The overall agreement statistics are summarized in Table 6, and the daily agreement rates are shown in Fig. 23. The overall agreement rate, calculated as the number of pairs assigned to the same class divided by the total number of matched pairs, is 0.658. In Table 6, each entry gives the same-class pair count divided by the class count of the respective instrument, followed by the corresponding percentage in parentheses. Of the samples classified by lidar as cloud-free, low cloud, and high cloud, the fractions also identified as the same class by MAX-DOAS are 0.703, 0.666, and 0.524, respectively. These results indicate that MAX-DOAS has relatively good skill in identifying cloud-free and low-cloud conditions, whereas the agreement for high-cloud conditions is lower.

Table 6. Agreement between MAX-DOAS and lidar cloud classifications.

Cloud class	MAX-DOAS	Lidar
Cloud-free	5842/8168 (71.5 %)	5842/8307 (70.3 %)
High cloud	1754/4184 (41.9 %)	1754/3348 (52.4 %)
Low cloud	5884/8134 (72.3 %)	5884/8831 (66.6 %)

5.2 Analysis of classification discrepancies

Fig. 22 shows that the inconsistencies between the MAX-DOAS and lidar classification results are not randomly distributed, but are mainly concentrated in several specific off-diagonal elements. This indicates that the classification discrepancies mainly

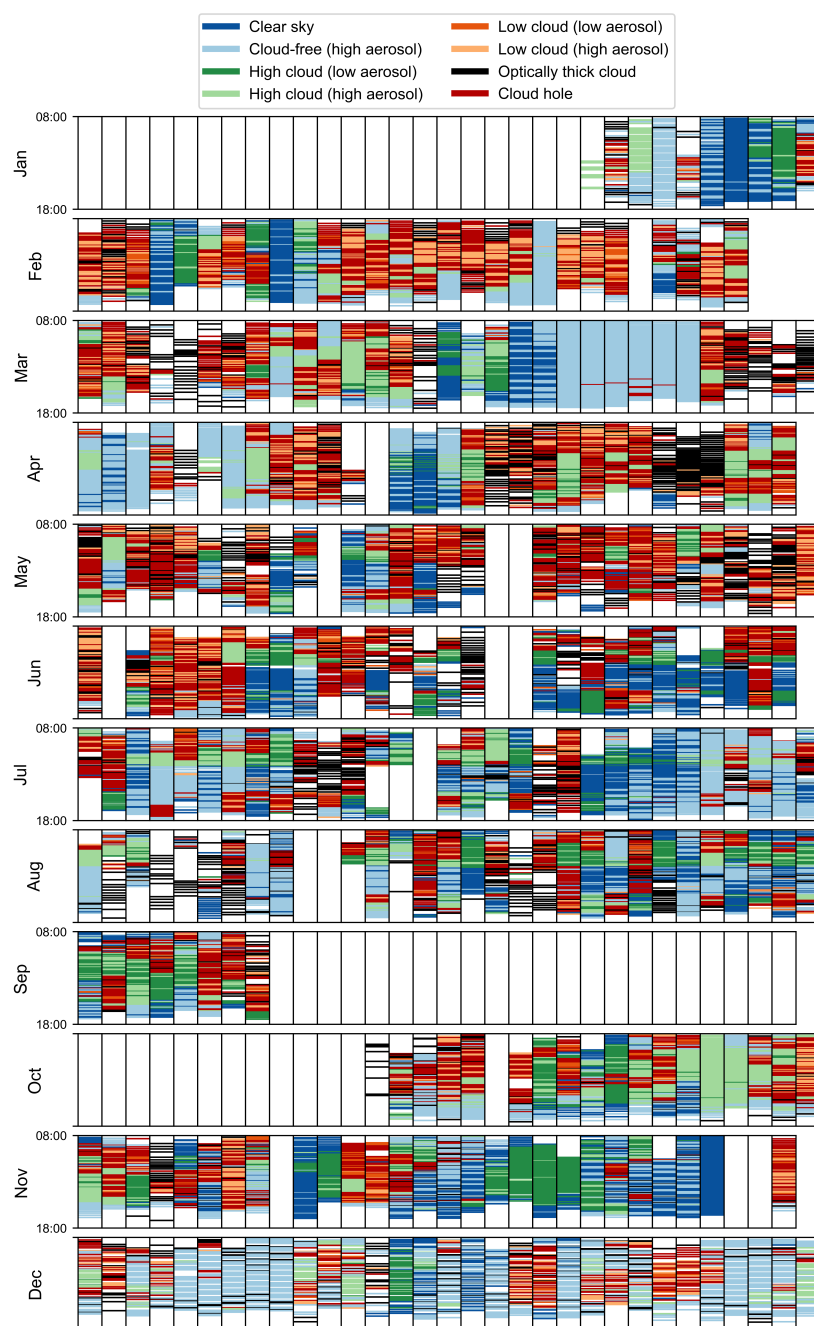


Figure 18. Time series of the MAX-DOAS cloud–aerosol classification results for the year 2025. Different colours indicate clear-sky conditions, cloud-free conditions with high aerosol loading, high-cloud conditions with low or high aerosol loading, low-cloud conditions with low or high aerosol loading, optically thick cloud conditions, and cloud holes. Blank areas indicate missing data or unavailable classifications.

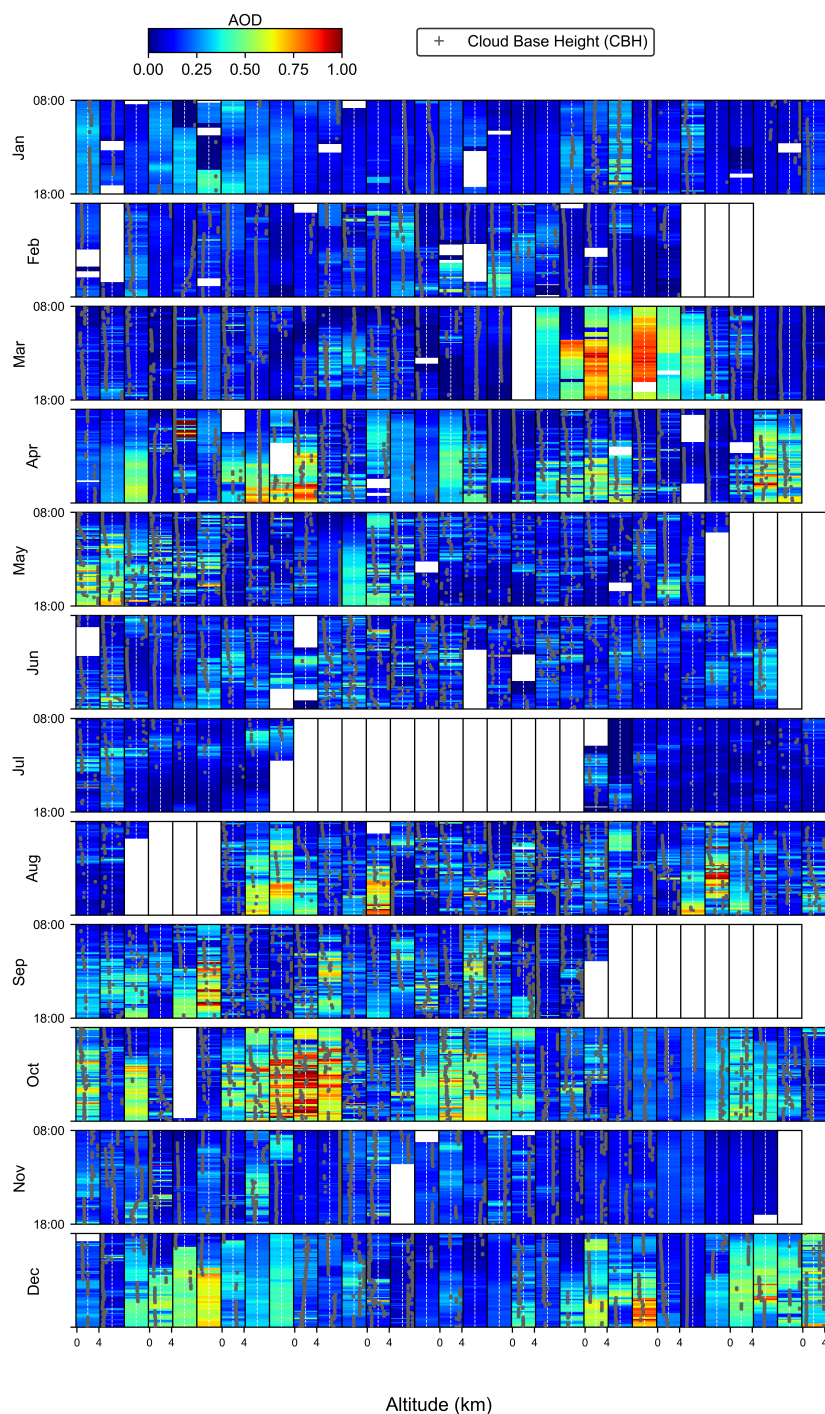


Figure 19. Annual distribution of lidar-derived below-cloud AOD and CBH for 2025. The colour scale indicates below-cloud AOD, and the grey crosses indicate CBH.

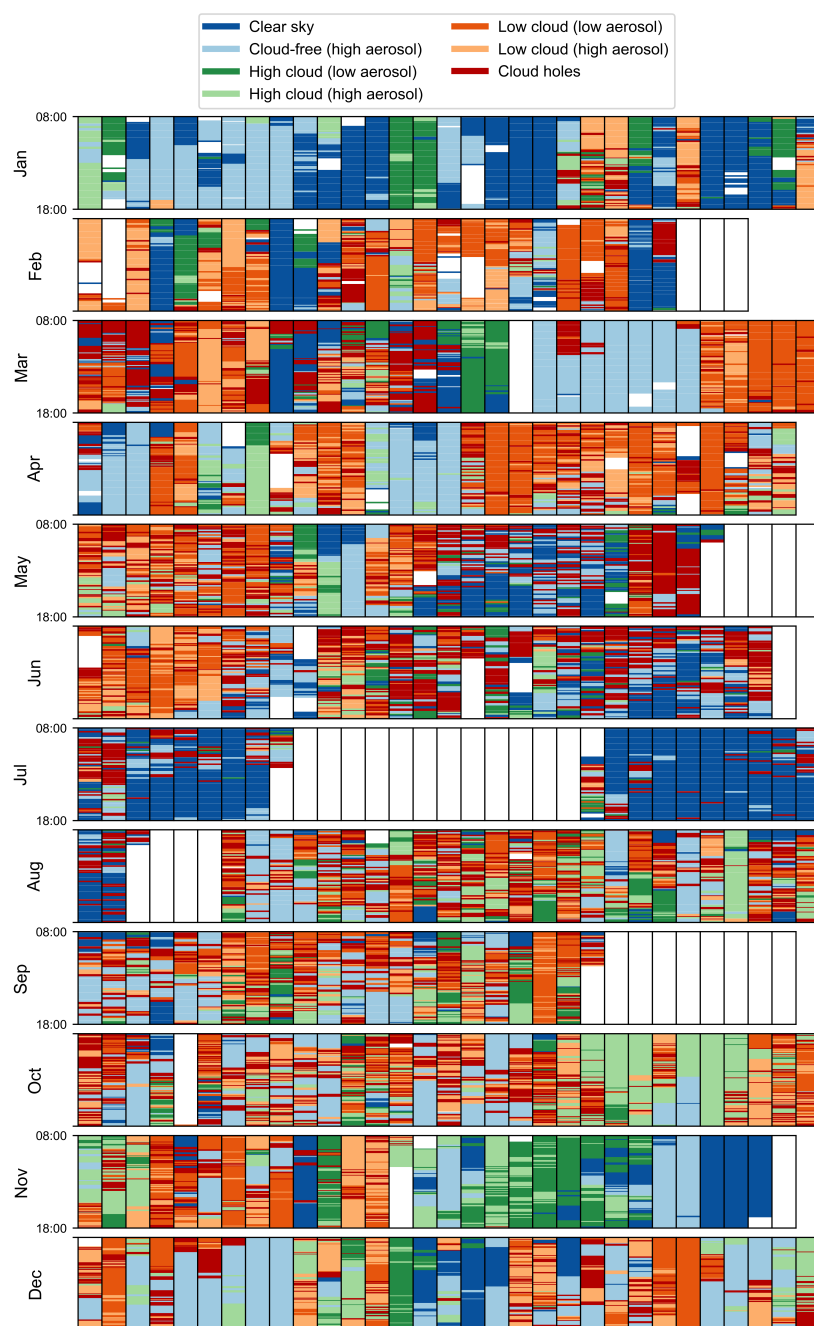


Figure 20. Annual cloud–aerosol classification derived from the lidar observations. Different colours indicate clear-sky conditions, cloud-free conditions with high aerosol loading, high-cloud conditions with low or high aerosol loading, low-cloud conditions with low or high aerosol loading, and cloud holes. Blank areas indicate missing data.

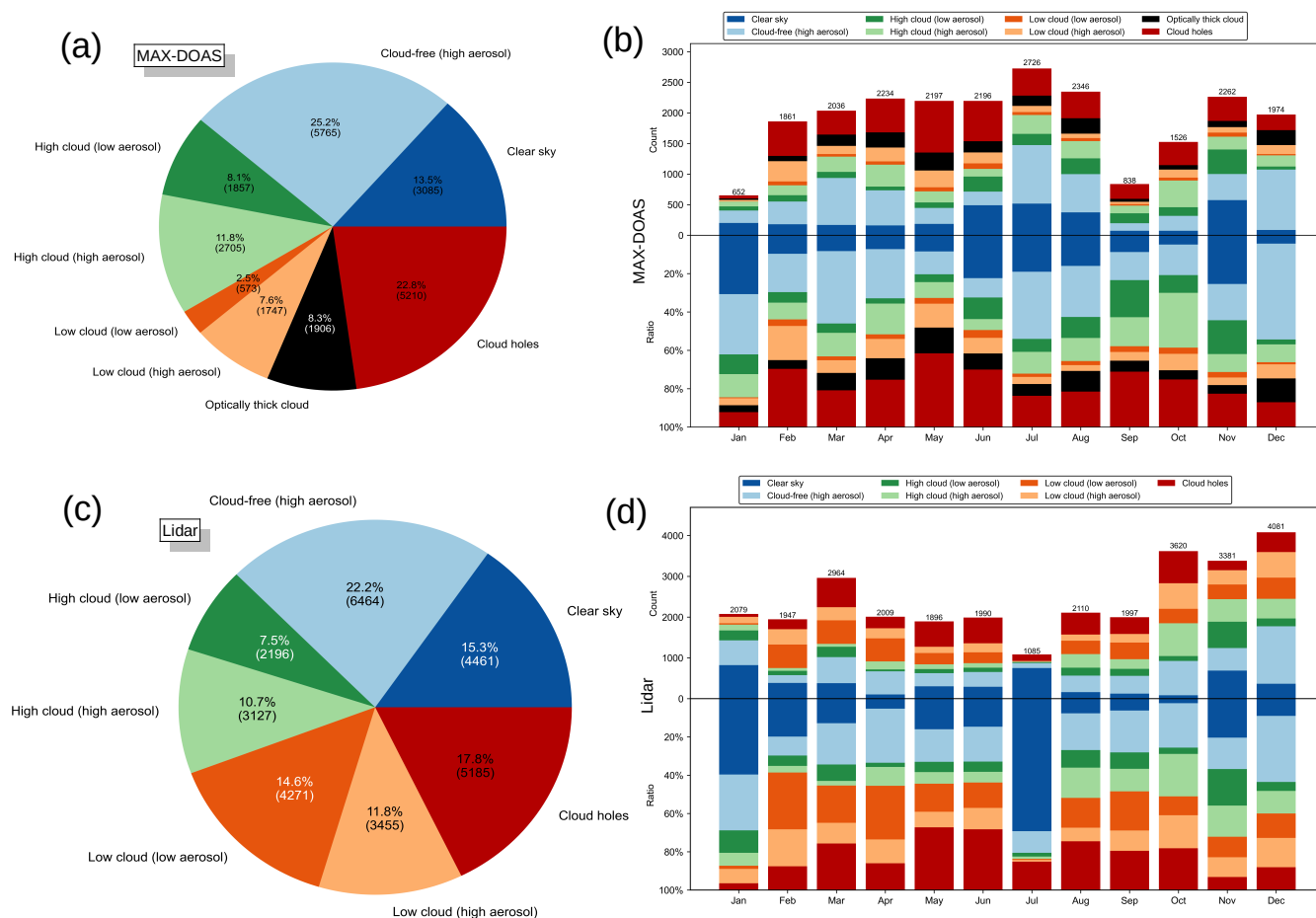


Figure 21. Statistical distributions of the MAX-DOAS and lidar cloud–aerosol classifications. Panels (a) and (c) show the annual fractions and counts for MAX-DOAS (22848 valid classifications) and lidar (29159 valid classifications), respectively, whereas panels (b) and (d) show the monthly counts and fractions. The statistics include all valid classifications from each instrument, not only the time-matched samples.

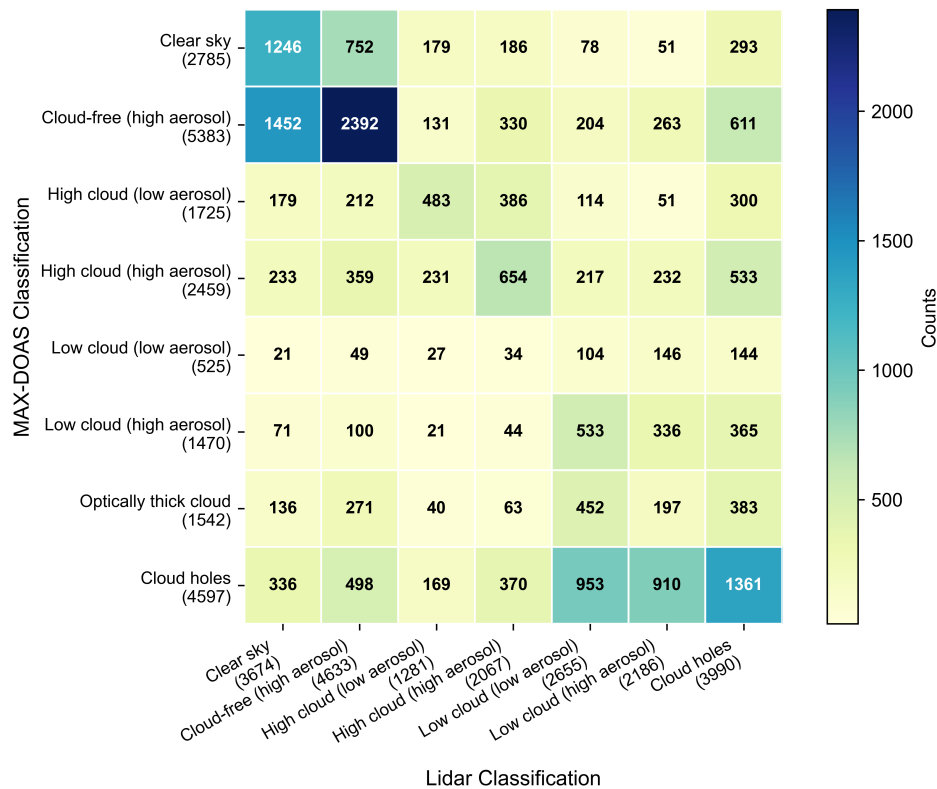


Figure 22. Cross-distribution matrix of the cloud–aerosol classifications derived from MAX-DOAS and lidar observations.

arise from systematic confusion between cloud–aerosol states with similar observational response features, rather than from
455 uniformly distributed random noise. It should be noted that MAX-DOAS observations at eight azimuth directions and the
vertical lidar observations differ substantially in sampling path and representative volume. As a result, the MAX-DOAS-
retrieved AOD, lidar-derived below-cloud AOD, and corresponding aerosol categories are not always strictly consistent. The
lidar comparison does not show a consistent increase in below-cloud AOD for the higher retrieved-AOD category under low-
cloud conditions, further limiting the interpretation of these subcategories in terms of actual aerosol loading. The following
460 discussion therefore focuses mainly on the sources of inconsistency in cloud identification.

Cases classified as cloud holes by MAX-DOAS but as cloud-free by lidar may reflect differences in temporal context and
spatial sampling. The MAX-DOAS cloud-hole label is assigned to an initial high-cloud classification when low-cloud observa-
tions occur within the surrounding time window, whereas the paired lidar classification is based on the detected vertical cloud
structure. These differences can produce a MAX-DOAS cloud-hole label without a corresponding cloud detection by lidar.
465 Conversely, cases classified as cloud holes by lidar but as cloud-free by MAX-DOAS may be associated with the different
operational definitions. The lidar cloud-hole category represents optically thin low clouds with COD < 0.5, which may pro-

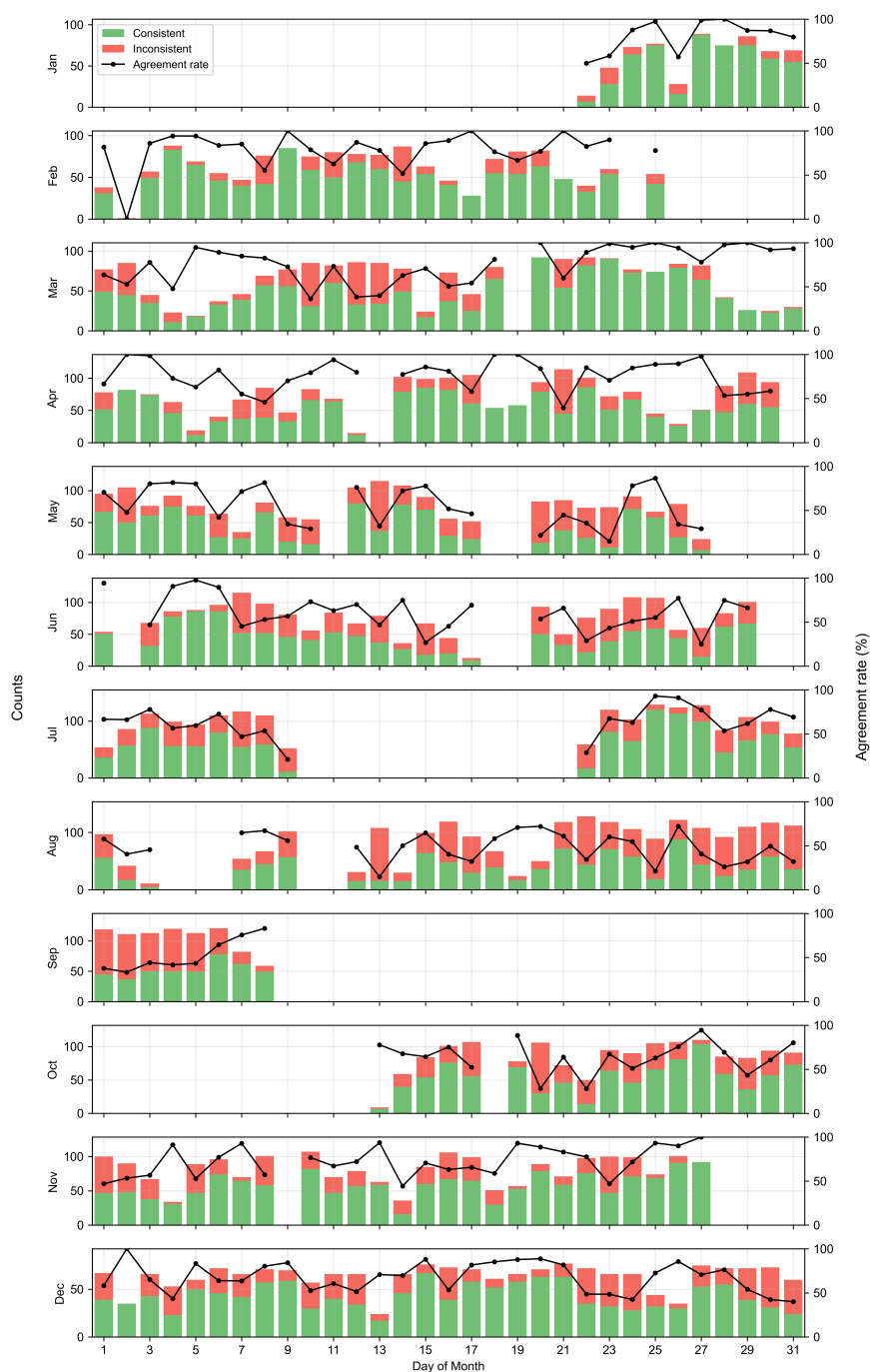


Figure 23. Daily agreement between MAX-DOAS and lidar for the grouped cloud-free, high-cloud, and low-cloud classifications.



duce only weak changes in the MAX-DOAS-derived quantities, leaving these observations within the cloud-free classification criteria.

Discrepancies between the high-cloud and cloud-hole categories may be related to their similar responses in MAX-DOAS-derived quantities. Cases classified as cloud holes by lidar but as high cloud by MAX-DOAS can remain in the initial high-cloud category when the temporal-window criterion is not met, whereas high-cloud candidates can be relabelled as cloud holes when nearby low-cloud observations are present, even if the paired lidar profile is classified as high cloud. The choice of window size therefore affects this distinction.

Cases classified as cloud-free or high cloud by lidar but as optically thick cloud by MAX-DOAS usually occur when the MAX-DOAS signal is weak, for example in the early morning or late afternoon. Under these conditions, the spectral signal-to-noise ratio is low, and the O_4 fitting and aerosol retrieval are more likely to fail. This can result in low zenith signal, abnormally reduced O_4 dSCD, and unrealistically high retrieved AOD, causing such cases to be classified as optically thick cloud.

By contrast, cases classified as cloudy by lidar but as cloud-free by MAX-DOAS mainly occur under large-SZA conditions. In these situations, the overall measured signal is weak, and the cloud-induced signal perturbation may not exceed the prescribed zenith-signal and TSI thresholds. As a result, the cloud perturbation may not be effectively detected by MAX-DOAS.

Some cases classified as high cloud by MAX-DOAS but as cloud-free by lidar are related to the combined effect of a small residual tilt of the MAX-DOAS instrument and small SZA. Because the instrument is not perfectly level, systematic signal differences may occur among zenith observations acquired at different azimuth settings. Under small-SZA conditions, such as around noon in summer when SZA may be smaller than 10° , the zenith signal is strong and this instrumental effect can produce anomalously large TSI values, causing cloud-free cases to be misclassified as high cloud.

Cases classified as high cloud by MAX-DOAS but as low cloud by lidar, or vice versa, are mainly related to the azimuth dependence of the 30° O_4 dSCD. Because MAX-DOAS observations are made at eight azimuth directions, the light-path lengths are not exactly the same in all directions. For some azimuths with shorter light paths, the 30° O_4 dSCD may fall below the threshold, causing cases that should be classified as high cloud to be misidentified as low cloud. Although this effect has been partly corrected in this study, some mismatch remains unavoidable.

In addition to the specific mismatch types described above, the two classification results are affected by more general sources of uncertainty. On the one hand, the MAX-DOAS-derived quantities contain fitting and retrieval errors. On the other hand, additional uncertainty is introduced when the lidar classification is mapped onto the MAX-DOAS time grid. For rapidly evolving cloud-aerosol states such as cloud holes, MAX-DOAS is more sensitive to temporal variability, whereas lidar primarily resolves vertical structure. This difference in representativeness can also lead to classification discrepancies.

5.3 Sensitivity analysis

To evaluate the stability of the classification method under different parameter settings, a sensitivity analysis was performed for the key thresholds used in the classification procedure. The tested parameters include the zenith-signal reference, the TSI threshold, the 30° O_4 dSCD threshold, the AOD thresholds, and the cloud-hole time window. Because these parameters correspond



500 to different classification criteria, their perturbations affect the classification in different ways. In general, each perturbation mainly influences the classification step associated with the corresponding parameter, rather than all classes uniformly.

In this study, perturbations of $\pm 10\%$ were applied separately to the zenith-signal reference, the TSI threshold, the 30° O_4 dSCD threshold, and the AOD thresholds. In addition, the time window for cloud-hole identification was varied to 20 and 40 min. The numbers of classified cases after each perturbation are listed in Table 7, and the agreement between the MAX-DOAS and lidar classifications is shown in Table 8. Each class-specific entry in Table 8 gives the agreement percentage above
 505 the same-class matched count divided by the matched count classified into that state by MAX-DOAS. Signed changes from the baseline are shown in parentheses only when their absolute magnitude exceeds one percentage point (pp), based on the unrounded ratios. Total agreement uses all matched pairs for the corresponding scheme.

Threshold perturbations can move observations into or out of the defined classification conditions: decreasing and in-
 510 creasing the zenith-signal reference by 10 % change the total number of classified samples from the baseline of 22848 to 22922 (+0.32 %) and 22621 (−0.99 %), respectively, as observations near the signal boundaries become classified or unclassified. AOD-threshold perturbations similarly affect coverage through the optically thick-cloud criterion, changing the total by +0.10 % and −0.12 % for factors of 0.9 and 1.1, respectively; TSI perturbations change it by less than 0.02 %, whereas perturbing the O_4 threshold or cloud-hole time window redistributes classes without changing the classified total.

515 Overall, the total agreement rate ranges from 0.645 to 0.667 under different threshold perturbations, with only small variations from the baseline result. This indicates that the proposed classification framework is generally stable with respect to the key threshold settings.

6 Conclusions

Based on year-round near-co-located MAX-DOAS and aerosol lidar observations, this study systematically analysed the effects
 520 of clouds and aerosols on MAX-DOAS-derived quantities and aerosol retrievals. On this basis, a cloud–aerosol state classification method that explicitly incorporates cloud-height information was developed. The main conclusions are as follows.

(a) Although both clouds and aerosols modify the MAX-DOAS light path, their effects are different. Aerosols mainly enhance multiple scattering in the near-surface atmosphere, causing the distribution of low-elevation O_4 dSCDs to become more concentrated. Low clouds, by contrast, shorten the effective light path and lead to a clear decrease in high-elevation O_4
 525 dSCDs, especially the 30° O_4 dSCD. High clouds have a relatively weak influence because MAX-DOAS observation paths are mainly sensitive to the lower atmosphere.

(b) Averaging kernels under clear-sky, favourable observing conditions indicate a reference sensitivity height of about 2 km for the instrument and retrieval configuration used at this site. Together with radiative transfer simulations of cloud-induced light-path changes, this reference provides the physical basis for separating low-cloud and high-cloud states according to
 530 cloud-base height relative to the cloud-free sensitive layer.

(c) When aerosol retrievals are performed under the clear-sky assumption, cloud-induced light-path changes can be partly misattributed to aerosol extinction, leading to retrieval bias. Sensitivity simulations show that this compensation effect is



Table 7. Numbers of MAX-DOAS cloud–aerosol classification results under different parameter perturbation schemes.

Perturbation scheme	Clear sky	Cloud-free (high aerosol)	High cloud (low aerosol)	High cloud (high aerosol)	Low cloud (low aerosol)	Low cloud (high aerosol)	Optically thick cloud	Cloud holes
Baseline	3085	5765	1857	2705	573	1747	1906	5210
Radiance reference $\times 0.9$	3133	5950	1860	2860 (+5.7%)	577	1872 (+7.2%)	1548 (-18.8%)	5122
Radiance reference $\times 1.1$	2966	5526	1812	2583	567	1607 (-8.0%)	2283 (+19.8%)	5277
TSI threshold $\times 0.9$	2949	5568	1895	2750	590	1791	1906	5401
TSI threshold $\times 1.1$	3134	5933	1823	2632	556	1696	1906	5165
30° O ₄ dSCD threshold $\times 0.9$	3162	5840	1916	3042 (+12.5%)	441 (-23.0%)	1563 (-10.5%)	1906	4978
30° O ₄ dSCD threshold $\times 1.1$	3022	5640	1744 (-6.1%)	2292 (-15.3%)	750 (+30.9%)	1986 (+13.7%)	1906	5508 (+5.7%)
AOD threshold $\times 0.9$	2451 (-20.6%)	6334 (+9.9%)	1513 (-18.5%)	2963 (+9.5%)	432 (-24.6%)	1837 (+5.2%)	2097 (+10.0%)	5244
AOD threshold $\times 1.1$	3701 (+20.0%)	5202 (-9.8%)	2189 (+17.9%)	2422 (-10.5%)	723 (+26.2%)	1639 (-6.2%)	1754 (-8.0%)	5191
Cloud-hole window = 20 min	3085	5765	2026 (+9.1%)	3257 (+20.4%)	573	1747	1906	4489 (-13.8%)
Cloud-hole window = 40 min	3085	5765	1734 (-6.6%)	2215 (-18.1%)	573	1747	1906	5823 (+11.8%)

Note: Values are sample counts. Parentheses give the relative change from baseline, $100(N - N_0)/N_0$, only when its absolute magnitude is at least 5 %. This cutoff is used only for display.

strongest under low-cloud conditions and can cause overestimation of near-surface aerosol extinction and retrieved AOD. Under high-cloud conditions, the retrieval bias is relatively small. Therefore, the retrieved AOD reflects not only aerosol loading but also, to some extent, cloud-related light-path effects, and can be used as an auxiliary classification indicator.

(d) The classification combines the measured zenith signal and its TSI, the 30° O₄ dSCD, and retrieved AOD to distinguish eight cloud–aerosol observation states. A temporal-window criterion identifies cloud holes. Under low-cloud conditions, the aerosol subcategories represent lower and higher retrieved AOD rather than actual aerosol loading.

(e) The annual classification results show that low-cloud-related states and cloud-free conditions with high aerosol loading account for relatively large fractions in Guangzhou, reflecting active cloud processes and large variations in urban aerosol loading. The fractions of low-cloud-related states are 41.3 % and 44.3 % in the MAX-DOAS and lidar classifications, respectively, while the fractions of cloud-free conditions with high aerosol loading are 25.2 % and 22.2 %, respectively. This indicates that, in regions such as Guangzhou with high humidity, frequent cloud processes, and high pollution loading, simply excluding all cloud-affected observations would lead to a substantial loss of potentially useful data. It is therefore necessary to further distinguish observation states under different cloud conditions.



Table 8. Agreement between MAX-DOAS and lidar cloud classification under different parameter perturbation schemes.

Perturbation scheme	Cloud-free	High cloud	Low cloud	Total agreement
Baseline	71.5 % 5842/8168	41.9 % 1754/4184	72.3 % 5884/8134	65.8 %
Radiance reference × 0.9	71.6 % 5990/8370	41.3 % 1782/4320	73.9 % (+1.6 pp) 5791/7837	66.1 %
Radiance reference × 1.1	71.9 % 5625/7827	42.3 % 1701/4024	70.8 % (-1.6 pp) 5963/8428	65.5 %
TSI threshold × 0.9	72.7 % (+1.2 pp) 5716/7864	41.7 % 1779/4264	71.7 % 5998/8360	65.9 %
TSI threshold × 1.1	70.8 % 5920/8361	41.9 % 1712/4090	72.7 % 5839/8032	65.8 %
30° O ₄ dSCD threshold × 0.9	70.6 % 5866/8307	40.9 % (-1.0 pp) 1864/4559	73.4 % (+1.0 pp) 5592/7620	65.0 %
30° O ₄ dSCD threshold × 1.1	72.3 % 5794/8013	43.9 % (+1.9 pp) 1633/3724	71.2 % (-1.2 pp) 6227/8749	66.7 %
AOD threshold × 0.9	71.6 % 5802/8107	41.9 % 1731/4127	71.8 % 5932/8267	65.7 %
AOD threshold × 1.1	71.5 % 5872/8217	41.8 % 1760/4213	72.8 % 5846/8034	65.9 %
Cloud-hole window = 20 min	71.5 % 5842/8168	38.7 % (-3.3 pp) 1870/4838	73.5 % (+1.2 pp) 5499/7480	64.5 % (-1.3 pp)
Cloud-hole window = 40 min	71.5 % 5842/8168	44.6 % (+2.7 pp) 1619/3626	71.0 % (-1.4 pp) 6168/8692	66.5 %

Note: Class cells give the agreement rate above the same-class matched count divided by the matched count classified into that state by MAX-DOAS. Total agreement is the sum of same-class matched counts divided by the sum of matched counts. Signed changes from baseline are shown only when their absolute magnitude exceeds 1 percentage point (pp), calculated as $100(r - r_0)$ from unrounded count ratios. Unannotated changes do not exceed this display cutoff.

(f) The MAX-DOAS classification reproduces the main evolution of the lidar-classified states, with an overall three-class agreement of 0.658. Agreement is lower for high-cloud states than for the other grouped states. The cross-distribution highlights differences between cloud-free aerosol subcategories, high clouds and cloud holes, and low clouds and optically thick clouds.

(g) The sensitivity analysis shows that the classification framework is generally stable with respect to the key threshold settings. When perturbations of $\pm 10\%$ are applied to the zenith-signal reference, the TSI threshold, the 30° O₄ dSCD threshold, and the AOD threshold, and when the cloud-hole time window is adjusted, the overall agreement rate for the three cloud classes varies only between 0.645 and 0.667. This indicates that the proposed method is reasonably robust in an overall statistical sense.

The method links cloud-height-dependent observational responses with aerosol retrieval behaviour and provides a physical basis for interpreting MAX-DOAS observation states under complex cloud conditions.

The classification provides observation-state information for subsequent retrieval studies. Future aerosol retrievals could combine O₄ absorption and radiance information to represent cloud and aerosol effects.



Code and data availability. The scripts used for data processing, retrieval, validation, and analysis, including Python-reconstructed HEIPRO retrieval scripts, together with the raw MAX-DOAS and lidar data used in this study, are available from the corresponding author upon reasonable request.

560 *Author contributions.* Luyao Wu conceived the study, designed the experiments, performed data analysis, developed code, prepared the initial manuscript draft, and created the figures. Liang Xi carried out data computations, validated the results, and provided guidance on manuscript writing. Xin Zhao contributed to code development. Yuhao Luo participated in the discussion of results and manuscript proof-reading. Yibin Fu supplied the lidar data. Runze Song was responsible for the preliminary processing of the DOAS data. Minjie Zhao provided essential resources. Fuqi Si organized the MAX-DOAS measurement campaign, contributed to experimental design, and participated in the discussion of results.

Competing interests. The authors declare that they have no conflict of interest.

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