ISSN 1674-1056
http://cpb.iphy.ac.cn
http://iopscience.iop.org/cpb
中国物理 B
Chinese Physics
中国物理 Chinese Physics B

# Vertical profile of aerosol extinction based on the measurement of $O_4$ of multi-elevation angles with MAX-DOAS

Fusheng Mou(牟福生), Jing Luo(雒静), Suwen Li(李素文), Wei Shan(单巍), Lisha Hu(胡丽莎)

**Citation:**Chin. Phys. B . 2019, 28(8): 084212. **doi:** 10.1088/1674-1056/28/8/084212
Journal homepage: http://cpb.iphy.ac.cn; http://iopscience.iop.org/cpb

**What follows is a list of articles you may be interested in**

---

## Detection performance improvement of photon counting chirped amplitude modulation lidar with response probability correction

Yi-Fei Sun(孙怿飞), Zi-Jing Zhang(张子静), Li-Yuan Zhao(赵丽媛), Wei-Min Sun(孙伟民), Yuan Zhao(赵远)

Chin. Phys. B . 2018, 27(9): 094213. **doi:** 10.1088/1674-1056/27/9/094213

## The intensity detection of single-photon detectors based on photon counting probability density statistics

Zijing Zhang(张子静), Long Wu(吴龙), Jie Song(宋杰), Yuan Zhao(赵远)

Chin. Phys. B . 2017, 26(10): 104207. **doi:** 10.1088/1674-1056/26/10/104207

## A high precision phase reconstruction algorithm for multi-laser guide stars adaptive optics

Bin He(何斌), Li-Fa Hu(胡立发), Da-Yu Li(李大禹), Huan-Yu Xu(徐焕宇), Xing-Yun Zhang(张杏云), Shao-Xin Wang(王少鑫), Yu-Kun Wang(王玉坤), Cheng-Liang Yang(杨程亮), Zhao-Liang Cao(曹召良), Quan-Quan Mu(穆全全), Xing-Hai Lu(鲁兴海), Li Xuan(宣丽)

Chin. Phys. B . 2016, 25(9): 094214. **doi:** 10.1088/1674-1056/25/9/094214

## V-L decomposition of a novel full-waveform lidar system based on virtual instrument technique

Xu Fan, Wang Yuan-Qing

Chin. Phys. B . 2015, 24(10): 104214. **doi:** 10.1088/1674-1056/24/10/104214

## Observation of tropospheric $NO_2$ by airborne multi-axis differential optical absorption spectroscopy in the Pearl River Delta region, south China

Xu Jin, Xie Pin-Hua, Si Fu-Qi, Li Ang, Wu Feng-Cheng, Wang Yang, Liu Jian-Guo, Liu Wen-Qing, Andreas Hartl, Chan Ka Lok

Chin. Phys. B . 2014, 23(9): 094210. **doi:** 10.1088/1674-1056/23/9/094210

---

# Vertical profile of aerosol extinction based on the measurement of $O_4$ of multi-elevation angles with MAX-DOAS*

Fusheng Mou(牟福生)†, Jing Luo(雒静)†, Suwen Li(李素文)‡, Wei Shan(单巍), and Lisha Hu(胡丽莎)

*School of Physics and Electronic Information, Huaibei Normal University, Huaibei* 235000, *China*

(Received 14 March 2019; revised manuscript received 18 May 2019; published online 9 July 2019)

A method for aerosol extinction profile retrieval using ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) is studied, which is based on a look-up table algorithm. The algorithm uses parametric method to represent aerosol extinction profiles and simulate different atmospheric aerosol states through atmospheric radiation transfer model. Based on the method, aerosol extinction profile was obtained during six cloud-free days. The $O_4$ differential air mass factor (dAMF) measured by MAX-DOAS is compared with the corresponding model results under different atmospheric conditions ($R^2 = 0.78$). The aerosol optical thickness, aerosol weight factor in boundary layer, and the height of the boundary layer are obtained after the process of minimization and look-up table method. The retrieved aerosol extinction in boundary layer is compared with PM2.5 data measured by ground point instrument. The diurnal variation trends of the two methods are in good agreement. The correlation coefficients of the two methods are 0.71 when the aerosol optical thickness is smaller than 0.5. The results show that the look-up table method can obtain the aerosol state of the troposphere and provide validation for other instrument data.

**Keywords:** vertical profile of aerosol extinction, look-up table, $O_4$, differential air mass factor, multi-axis differential optical absorption spectroscopy

**PACS:** 42.68.wt, 78.40.–q, 87.64.k–, 95.75.Rs **DOI:** 10.1088/1674-1056/28/8/084212

## 1. Introduction

With the development of industry and the acceleration of urbanization, the concentration of particulate matter and secondary particulate matter in the atmosphere has increased substantially, which has an important impact on atmospheric environmental quality and terrestrial gas system radiation budget.[1,2] Due to the uncertainty of aerosol in morphology, composition, physical and chemical properties, and optical properties, it is difficult to accurately assess the impact of aerosol on the environment and global climate. Therefore, the spatial and temporal distribution, optical properties, and spectral distribution characteristics of aerosol have become a research hotspot in atmospheric environmental monitoring and related climate effects. It is very important to obtain aerosol optical properties for understanding the evolution process of atmospheric pollution, calibration of model, and satellite data.[3]

In recent years, differential optical absorption spectroscopy (DOAS) is used in the field of atmospheric environmental monitoring.[4] The multi-axis DOAS (MAX-DOAS) composed of zenith direction and low observation elevation greatly improves the observation sensitivity of the tropospheric atmosphere. Trace gas profiles in the troposphere can be obtained by combining with the atmospheric radiation transfer model. In 2004, Wagner *et al.* proposed to retrieve the atmospheric scattering light path by using the characteristics of oxygen dimer ($O_4$) vertical profile which is basically unchanged in the atmosphere and evenly distributed horizontally.[5] Since the scattering energy of aerosol strongly affects the photonic optical path of the atmosphere, the aerosol optical thickness and profile information can be obtained by observing the slant column density (SCD) of $O_4$ through ground-based MAX-DOAS. In order to achieve the rapid, accurate, and real-time acquisition of aerosol profile information, a series of studies have been carried out by different research groups.[6–8] At present, two main algorithms of the optimal estimation method and look-up table method are used to obtain aerosol optical properties based on the SCD of $O_4$ through multi-elevation angles with MAX-DOAS. The optimal estimation method can obtain a continuous profile with a high vertical resolution. However, the algorithm relies on prior information and singular values could appear in the retrieval.[9] The look-up table method carries out parameter quantization assumption for aerosol profile, and the retrieval of aerosol profile is relatively fast in look-up table algorithm after the establishment of database. The results are relatively stable because they come from the given situation of database.[10]

*Project supported by the National Natural Science Foundation of China (Grant Nos. 41875040, 41705012, and 1605013).

†These authors contributed equally to this work.

‡Corresponding author. E-mail: swli@chnu.edu.cn

© 2019 Chinese Physical Society and IOP Publishing Ltd 

In 2010, look-up table method was first used to retrieve the extinction profile of aerosol by Li *et al.*[11] The results of different time periods were compared with the ground-based *in situ* data. However, the complete weight factor was not included in the profile hypothesis (0.5–1 proportion of aerosols in the boundary layer) and the effect of extraneous transport on aerosol was not considered. In 2013, Wu *et al.* used look-up table method to retrieve the aerosol extinction profile and compared it with aerosol optical thickness obtained by the Lidar.[12] However, only four hours of measurement results were obtained in the noon period of a day (9:00–13:00).

The look-up table method including complete weight factor (0.1–1) and different time periods (8:00–16:00) is used to retrieve the aerosol profile in this paper. Field experiments are carried out to study the ability of the algorithm to obtain the profile of aerosol extinction under different atmospheric aerosol conditions. The boundary layer aerosol extinction by MAX-DOAS is compared with PM2.5 data obtained by ground-based point instruments.

## 2. Methods and principles

### 2.1. MAX-DOAS system and settings

The one-dimensional MAX-DOAS system was used in the field, which was developed by Anhui Institute of Optics and Fine Mechanics.[13] The experimental site was located in the northern suburb of Huaibei, and continuous observation was carried out in the cloud-free days from September 29, 2018 to October 4, 2018. The telescope pointed downtown (azimuth angle 177°, north is 0°). Figure 1 shows the position of the MAX-DOAS and schematic diagram of the measurement principle. The spectral retrieval band was 338 nm–365.5 nm and the spectrometer had a resolution of 0.5 nm. To reduce the impact of the temperature drift on spectral measurements, the spectrometer was put in the temperature control box and the temperature was set to 27 °C. The measurement spectra in different directions can be obtained by changing the direction of the telescope. The zenith spectrum was used as the reference spectrum, and the retrieval result was the difference of $O_4$ slant column density between the measurement spectrum and reference spectrum, which was called the differential slant column density (dSCD). To facilitate comparison with the model, the concentration divided by $O_4$ vertical column density is converted into the differential air mass factor (dAMF). The point instrument can measure the concentration information of near-surface aerosol particles, which can provide comparative verification for the MAX-DOAS retrieval results in the field campaign.

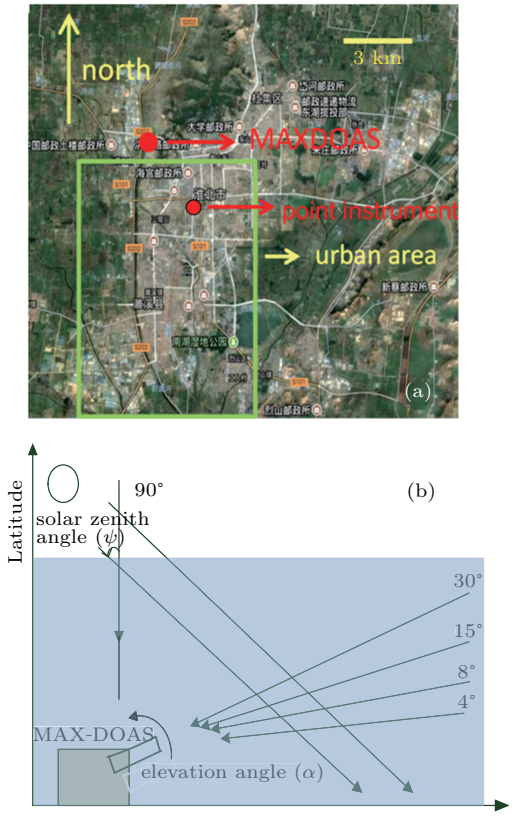



**Fig. 1.** (a) Position of the MAX-DOAS and (b) schematic diagram of the measurement principle.

### 2.2. Algorithm for aerosol profile retrieval based on $O_4$ absorption

For the aerosol profile, a two-layer setup for vertical aerosol distribution is used in the troposphere (0 km–15 km), which can be described by a limited set of parameters. The aerosol is assumed to be homogeneous in a layer near the ground surface (called boundary layer), and the aerosol extinction coefficient (unit: $km^{-1}$) in the layer aloft (*i.e.*, the free troposphere) decreases exponentially with height. This type of aerosol distribution is frequently observed in polluted regions and verified by Lidar observation. The extinction profile $\varepsilon(z)$ can be described as two layers in the range from 0 km to 15 km

$$\varepsilon(z) = \begin{cases} t \cdot w/h, & z \leqslant h, \\ A \cdot \exp(-z/\beta), & z > h, \end{cases} \tag{1}$$

where $z$ is the height above ground, $t$ is the aerosol optical thickness from 0 km to 15 km, and $w$ is the weight factor of total extinction $t$ in the boundary layer. $h$ is aerosol boundary layer height, and $A$ is the normalizing constant for exponentiation function and here, a constant of 5 km is adopted. The norm $A$ is calculated from the integral of $E(z)$ over the entire tropospheric $\mathrm{t} = \int_{0\ \mathrm{km}}^{15\ \mathrm{km}} \varepsilon(z)\mathrm{d}z$, leading to

$$A = \frac{(1-w)\cdot t}{\beta \cdot (\mathrm{e}^{-h/\beta} - \mathrm{e}^{-15\ \mathrm{km}/\beta})}. \tag{2}$$

The parameters are input into the model to obtain the $O_4$ dAMF simulation values of nine elevation angles at different aerosol states (see Table 1). After obtaining the simulation results and measurement results in the measurement cycle, to

optimize the parameters $(t, w, h)$, the cost function $(\chi^2)$ is minimized

$$\chi^2(t,w,h) = \sum_{\alpha=1}^{\alpha=30} \left( \frac{\text{Meas}_\alpha - \text{Sim}_\alpha(t,w,h)}{\sigma(\text{Meas}_\alpha)} \right)^2. \quad (3)$$

The simulated and measured results are $\text{Sim}_\alpha$ and $\text{Meas}_\alpha$, respectively. And $\sigma$ $(\text{Meas}_\alpha)$ is the standard deviation of the measured values. If the minimum result of nine angles in a measurement cycle is the smallest, the corresponding profile parameters can be considered to be more consistent with the real situation. To reduce the uncertainty of a single measurement, the hourly mean of the measured results is carried out at each elevation angle. The output error is bigger when the solar zenith angle (SZA) is greater than 75° and the processing data is selected from 8:00 to 16:00 each day.

The parameters of aerosol optical thickness $t$, aerosol boundary layer height $h$, aerosol extinction weight within the boundary layer $w$, and other atmospheric parameters (such as single scattering albedo) constitute the look-up table.[14] The simulated $O_4$ dAMF of each elevation angle under different atmospheric conditions is calculated through the atmospheric radiation transmission model. The model results and measurement results are compared to obtain the aerosol profile which best conforms to the real atmospheric conditions through the minimization process. The algorithm diagram is shown in Fig. 2.

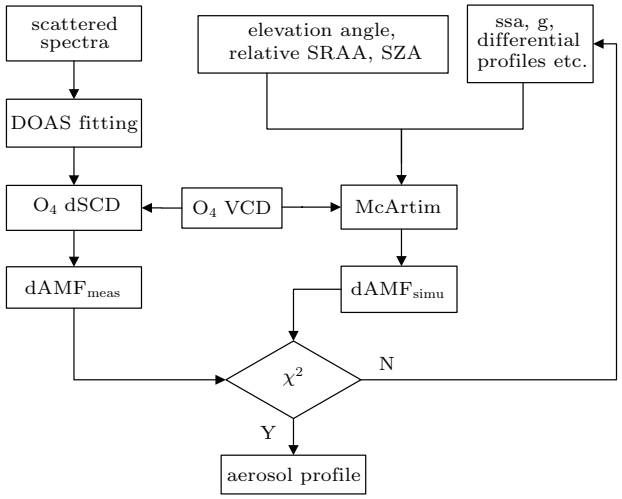



**Fig. 2.** Schematic diagram of aerosol profile retrieved with the absorption of $O_4$ observed by MAX-DOAS.

**Table 1.** Parameters for look-up table. SZA: solar zenith angle. SRAA: solar relative azimuth angle.

| Parameters | Values | Units |
|---|---|---|
| SZA | 62.17, 51.6, 43.03,37.89, 37.68, 42.47, 50.83, 61.29 | degree (°) |
| SRAA | –61.61, –48.9, –32.12, –10.32, 14.24, 36.4, 53.57, 66.54 | degree (°) |
| *g* | 0.72 | |
| *ssa* | 0.92 | |
| *albedo* | 0.05 | |
| Photon number | 200,000 | |
| *t* | 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.5, 2.0 | |
| *w* | 0.1–1, step: 0.1 | |
| *h* | 0.2–2, step: 0.2 | km |
| Free tropospheric height $\beta$ | 5 | km |
| Trace gas profile | $O_4$ US standard atmosphere | |
| Wavelength | 350 | nm |
| Elevation | 1, 2, 3, 4, 5, 6, 8, 15, 30, 90 | degree (°) |

The full spherical Monte-Carlo atmospheric radiative transfer model, Monte Carlo atmospheric radiative transfer and inversion model (McArtim), is used in this study.[15] To improve the operational efficiency, the simulation wavelength is selected as 350 nm, which is the central wavelength of the retrieval band (337 nm–365.5 nm). Sensitivity studies have shown single scattering albedo *ssa*, asymmetric factor *g*, and surface albedo *albedo* have a small influence on air mass factor (AMF). Therefore, fixed values of $ssa = 0.92$, $g = 0.72$, and $albedo = 0.05$ are adopted in the simulation. The specific settings of the model are shown in Table 1.

## 3. Results and analysis

### 3.1. Spectral analysis

The measurement spectrum is retrieved with DOAS method. The fitting structures are $NO_2$ (298 K, vanDaele), $O_4$ (296 K, Hermans), $O_3$ (223 K, Bogumil), and Ring structure. The Ring structure is calculated by DOASIS software.[16]

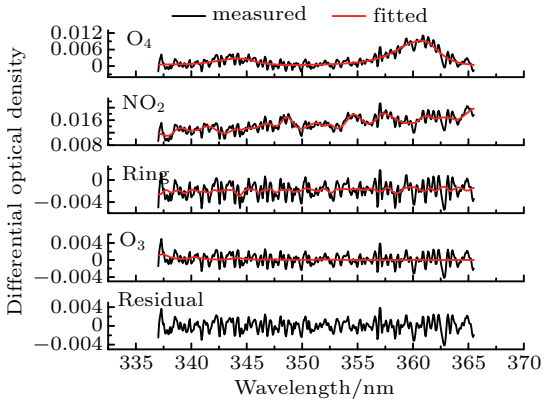



**Fig. 3.** Fitting example of the measured spectrum. The black and red curves indicate the absorption structures and the fitted absorption structures, respectively.

A fitting example of the 2° spectrum is shown in Fig. 3 on October 1, 2018 at 11:56. The zenith spectrum of each measurement cycle is selected as the reference spectrum. The $O_4$ dSCD is $1.84 \times 10^{43}$ molecules$^2$·cm$^{-5}$, and the residual is $1.94 \times 10^{-3}$, which is mainly derived from noise and unknown structure. Differential air mass factor of $O_4$ is obtained according to this method from September 30, 2018 to October 4, 2018 (see Fig. 4). Due to the increase of aerosol content in the atmosphere, the measured results for all elevation angles decrease on October 3 and October 4.

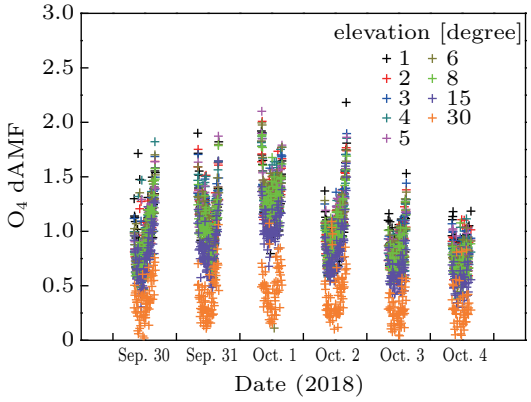



**Fig. 4.** Differential air mass factor of $O_4$ measured on cloud-free days.

### 3.2. Retrieval aerosol profile

Figure 5 shows the $O_4$ dAMF at different elevation angles and the retrieval aerosol profile results in the case of low aerosol (10:00–11:00, October 1, 2018, $t = 0.3$) and high aerosol (12:00–13:00, October 4, 2018, $t = 0.8$). The measurement results and model results are in good agreement under low aerosol and high aerosol. For both the low aerosol (Fig. 5(a)) and high aerosol (Fig. 5(b)) situation, the largest $O_4$ dAMF difference between simulation value and measured value is at 15° and the value is $5.3 \times 10^{-2}$ and $1.2 \times 10^{-1}$, respectively. The aerosol extinction profiles are obtained by look-up table method (see Figs. 5(c) and 5(d)). The dAMF mean values for all elevation angles are 1.07 and 0.65 in low and high aerosol, respectively. The effective absorption path of photons along the direction of the telescope is reduced because the high aerosol concentrations might be close to the ground. The differences of dAMF between different elevations are also decreased.[17] The linear correlation of $O_4$ dAMF between simulation and measurement is shown in Fig. 6. Correlation analysis ($R^2 = 0.78$) shows that the retrieval results and measurement results are very close. The retrieval algorithm minimizes the cost function and the residual error of the inversion is very small.

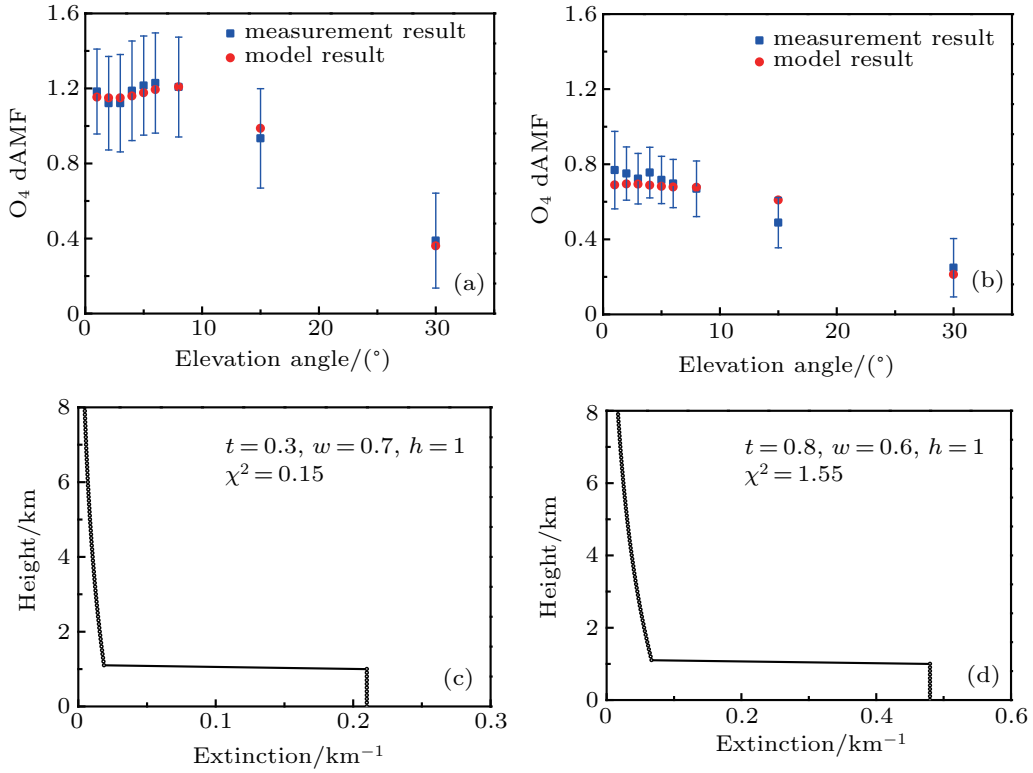



**Fig. 5.** Examples for the retrieval of the aerosol extinction with high and low aerosol loads. Upper panels: comparison of measured and modeled $O_4$ dAMF. Lower panels: retrieved profiles. Left: October 1, 2018, 10:00–11:00, SZA = 43.03°. Right: October 1, 2018, 12:00–13:00, SZA = 37.68°.

From the minimization process described in Subsection 2.1, the aerosol optical parameters ($t$, $w$, $h$) can be obtained from the iterative calculation of the $O_4$ dAMF. These three parameters determine the aerosol extinction profile. Figure 7 shows the retrieval results of aerosol extinction profile for six days. It is found that the aerosol extinction coefficient near the ground on October 4 is larger. The aerosol optical thickness gradually increases from October 3, and the boundary layer height does not change much between 10:00 and 16:00, especially on September 30. From October 1 to

October 4, the boundary layer has changed significantly, and the boundary layer is higher between 12:00 and 14:00. Due to the low height of the boundary layer, the extinction coefficient of aerosols near the ground is larger before 10:00 and after 15:00 every day. The MAX-DOAS system observed towards the urban area. And the point instrument (CEMS system of American thermal power company) was installed in the urban area to monitor the near-ground air quality. After obtaining the extinction coefficient of aerosol by the look-up table method, the extinction coefficient ($E_0 = t \times w/h$) in the boundary layer can be obtained in the field. Figure 8 shows surface aerosol extinction and boundary layer height retrieved by MAX-DOAS for 6 days. PM2.5 data from surface point instrument are also shown in Fig. 8. Surface aerosol extinction $E_0$ by MAX-DOAS is in good agreement with PM2.5 for point instrument. Based on all data ($N = 47$) and $t < 0.5$ ($N = 34$), the correlation coefficient $R^2$ is 0.64 and 0.71, respectively (as shown in Fig. 9).

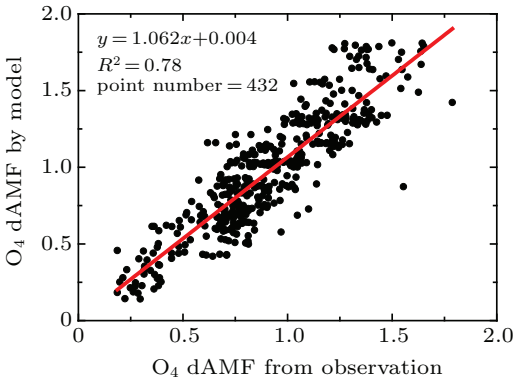



**Fig. 6.** $O_4$ dAMF from MAX-DOAS observation and those derived from the model.

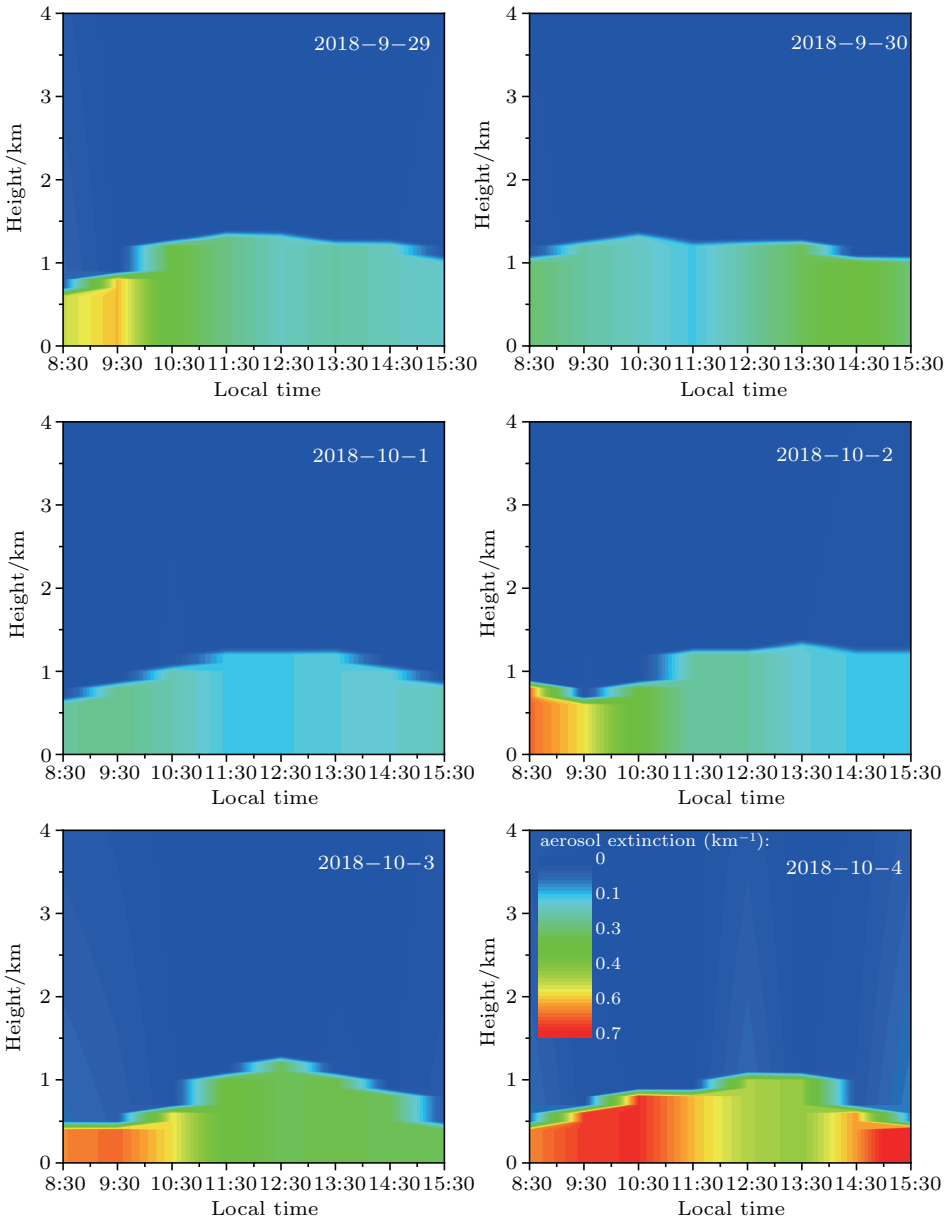



**Fig. 7.** The aerosol profile retrieved by MAX-DOAS.

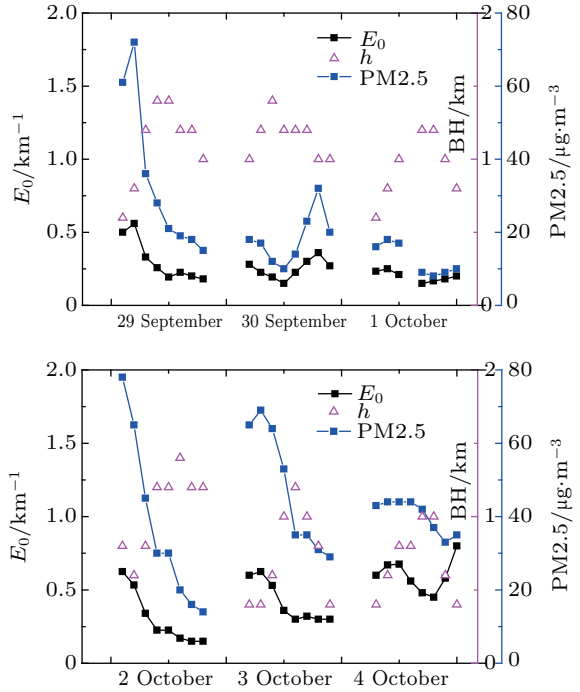



**Fig. 8.** Surface aerosol extinction $E_0$ and boundary layer height BH retrieved by MAX-DOAS for 6 days. Also shown are PM2.5 data from surface point instrument.

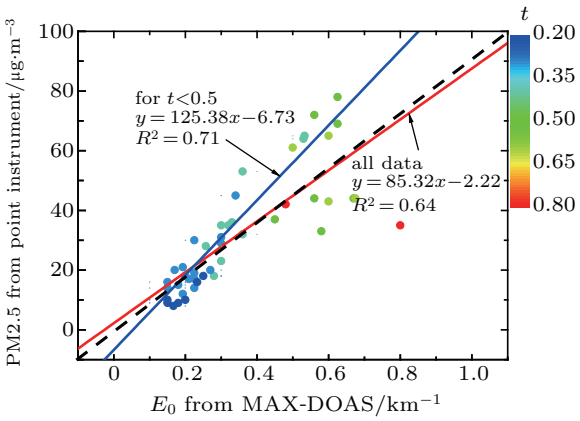



**Fig. 9.** Comparison between surface aerosol extinction measured by MAX-DOAS and PM2.5 data from point instrument. Different colors represent different aerosol optical thicknesses.

The difference between the two instruments is largest in the morning. Considering the fact that MAX-DOAS measures the aerosol extinction averaged over a distance while the point instrument detects the aerosol information near the ground, the results of the linear regression demonstrate good agreement between the measurement results of these two instruments. The latter is more easily influenced by local emission source and could produce high values.[11] In addition, fog and high aerosols in the morning will reduce atmospheric visibility and the effective optical path at all elevation angles of the MAX-DOAS telescope, which restricts the measurement process by MAX-DOAS.[9]

## 4. Conclusion

The retrieval algorithm of aerosol extinction vertical profile is studied based on the $O_4$ absorption of multi-elevation angles by MAX-DOAS. The look-up table method is used to obtain the optimal aerosol extinction profile by minimizing the cost function. By comparing the measured results of $O_4$ dAMF with model results, it is found that this method can accurately obtain the aerosol information for low and high aerosol situation. The retrieved aerosol extinction in boundary layer is compared with PM2.5 data measured by ground-based point instrument. The diurnal variation trend of the two methods is in good agreement. The results show that the look-up table method can obtain the aerosol state of the troposphere and provide validation for other instrument data. However, due to the limitation of the profile shape, it is difficult to retrieve more complex atmospheric pollution state such as the uplift layer. Therefore, different profile shapes will be added to the model and the results will be compared with other algorithms in the future.

## References

[1] Rosenfeld D 2006 *Science* **312** 1323
[2] Che H, Yang Z, Zhang X, Zhu C, Ma Q and Zhou H 2009 *Atmos. Environ.* **43** 1093
[3] Ramanathan V, Crutzen P J, Kiehl J T and Rosenfeld D 2001 *Science* **294** 2119
[4] Wei M H, Tong M M and Li S W 2015 *Chin. J. Chem. Phys.* **28** 688
[5] Wagner T, Dix B and Friedeburg C V 2004 *J. Geophys. Res.* **109** D22205
[6] Frieß U, Baltink H K and Beirle S 2016 *Atmos. Meas. Tech.* **9** 3205
[7] Frieß U, Monks P S and Remedios J J 2006 *J. Geophys. Res.* **111** D14203
[8] Wagner T, Beirle S and Brauers T 2011 *Atmos. Meas. Tech.* **4** 2685
[9] Wang Y, Lampel J and Xie P H 2017 *Atmos. Chem. Phys.* **17** 2189
[10] Li X, Brauers T and Hofzumahaus A 2013 *Atmos. Chem. Phys.* **13** 2133
[11] Li X, Brauers T and Shao M 2010 *Atmos. Chem. Phys.* **10** 2079
[12] Wu F C, Xie P H and Li A 2013 *Acta Opt. Sin.* **33** 0601002
[13] Wang Y, Beirle S and Lampel J 2017 *Atmos. Chem. Phys.* **17** 5007
[14] Mou F S, Luo J, Li S W, Wang J W, Shi R R, Wei M H 2018 *Acta Photon. Sin.* **47** 0701002
[15] Deutschmann T, Beirle S and Frieß U 2011 *J. Quantum Spectrosc. Radiat. Transfer* **112** 1119
[16] Kraus S 2006 Ph. D. Dissertation (Germany: University of Mannheim)
[17] Wang S S, Cuevas C A and Frieß U 2016 *Atmos. Meas. Tech.* **9** 5089