

# Intercomparison of MAX-DOAS vertical profile retrieval algorithms: studies on field data from the CINDI-2 campaign

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**Abstract.** ~~Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) is a well-established ground-based measurement technique for the detection of aerosols and trace gases particularly in the boundary layer and the lower troposphere: ultraviolet and visible radiation spectra of skylight are analysed to obtain information on different atmospheric parameters, integrated over the light path from space to the instrument. An appropriate set of spectra recorded under different viewing geometries ("Multi-Axis") allows retrieval of tropospheric aerosol and trace gas vertical distributions by applying numerical inversion methods.~~

The second Cabauw Intercomparison of Nitrogen Dioxide measuring Instruments (CINDI-2) took place in Cabauw (The Netherlands) in September 2016 with the aim of assessing the consistency of MAX-DOAS measurements of tropospheric species ( $\text{NO}_2$ , HCHO,  $\text{O}_3$ , HONO, CHOCHO and  $\text{O}_4$ ). This was achieved through the coordinated operation of 36 spectrometers operated by 24 groups from all over the world, together with a wide range of supporting reference observations (in situ analysers, balloon sondes, lidars, Long-Path DOAS, direct-sun DOAS, sun photometer and ~~others~~ meteorological instruments).

In the presented study, the retrieved CINDI-2 MAX-DOAS trace gas ( $\text{NO}_2$ , HCHO) and aerosol vertical profiles of 15 participating groups using different inversion algorithms are compared and validated against the colocated supporting observations. ~~The profiles~~, with the focus on aerosol optical thicknesses (AOTs), trace gas vertical column densities (VCDs) and trace gas surface concentrations. The algorithms are based on three different techniques: six use the optimal estimation method, two use a parametrized approach and one algorithm relies on simplified radiative transport assumptions and analytical calculations. To assess the agreement among the inversion algorithms independent of inconsistencies in the trace gas slant column density acquisition, participants applied their inversion to a common set of slant columns. Further, important settings like the retrieval grid, profiles of  $\text{O}_3$ , temperature and pressure as well as aerosol optical properties and a priori assumptions (for optimal estimation algorithms) have been prescribed to reduce possible sources of discrepancies.

The profiling results were found to be in good qualitative agreement: most participants obtained the same features in the retrieved vertical trace gas and aerosol distributions, however sometimes at different altitudes and of different ~~intensity~~ magnitude. Under clear sky conditions, the root-mean-square differences ~~of aerosol optical thicknesses, (RMSDs) among the results of individual participants vary between (0.01 – 0.1) for AOTs,  $(1.5 – 15) \times 10^{14} \text{ molec cm}^{-2}$  for trace gas ( $\text{NO}_2$ , HCHO) vertical columns and surface concentrations among the results of individual participants vary between 0.01 – 0.1,  $(1.5 – 15) \times 10^{14} \text{ molec cm}^{-2}$  and VCDs and  $(0.3 – 8) \times 10^{10} \text{ molec cm}^{-3}$ , respectively.~~ for trace gas surface concentrations. These values compare to approximate average optical thicknesses of 0.3, trace gas vertical columns of  $90 \times 10^{14} \text{ molec cm}^{-2}$  and trace gas surface concentrations of  $11 \times 10^{10} \text{ molec cm}^{-3}$  observed over the campaign period. The discrepancies originate from differences in the applied techniques, the exact implementation of the algorithms and the user defined settings that were not prescribed.

For the comparison against supporting observations, ~~these values increase to the RMSDs increase to (0.02 – 0.2) against AOTs from the sun photometer,  $(11 – 55) \times 10^{14} \text{ molec cm}^{-2}$  against trace gas VCDs from direct-sun DOAS observations and  $(0.8 – 9) \times 10^{10} \text{ molec cm}^{-3}$ .~~ It is likely that a large part of this increase is caused by imperfect against surface concentrations

from the Long-Path DOAS instrument. This increase in RMSDs is most likely caused by uncertainties in the supporting data themselves, spatio-temporal ~~overlap of the different observations~~ mismatch among the observations and simplified assumptions particularly on aerosol optical properties made for the MAX-DOAS retrieval.

~~In contrast to what is often assumed, the~~ As a side investigation, the comparison was repeated with the participants retrieving profiles from their own dSCDs acquired during the campaign. In this case, the consistency among the participants degrades by about 30% for AOTs, by 180% (40%) for HCHO (NO<sub>2</sub>) VCDs and by 90% (20%) for HCHO (NO<sub>2</sub>) surface concentrations.

In former publications and also during this comparison study, it was found that MAX-DOAS vertically integrated ~~extinction profiles and the sun photometer total aerosol optical thickness were found to not necessarily being comparable quantities, unless information on the real aerosol vertical distribution is available to account for the low~~ aerosol extinction coefficient profiles systematically underestimate the AOT observed by the sun photometer. For the first time it is quantitatively shown that for optimal estimation algorithms this can be largely explained and compensated by considering smoothing effects, namely biases arising from the reduced sensitivity of MAX-DOAS observations ~~at higher altitudes~~ to higher altitudes and associated *a priori* assumptions.

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## 15 1 Introduction

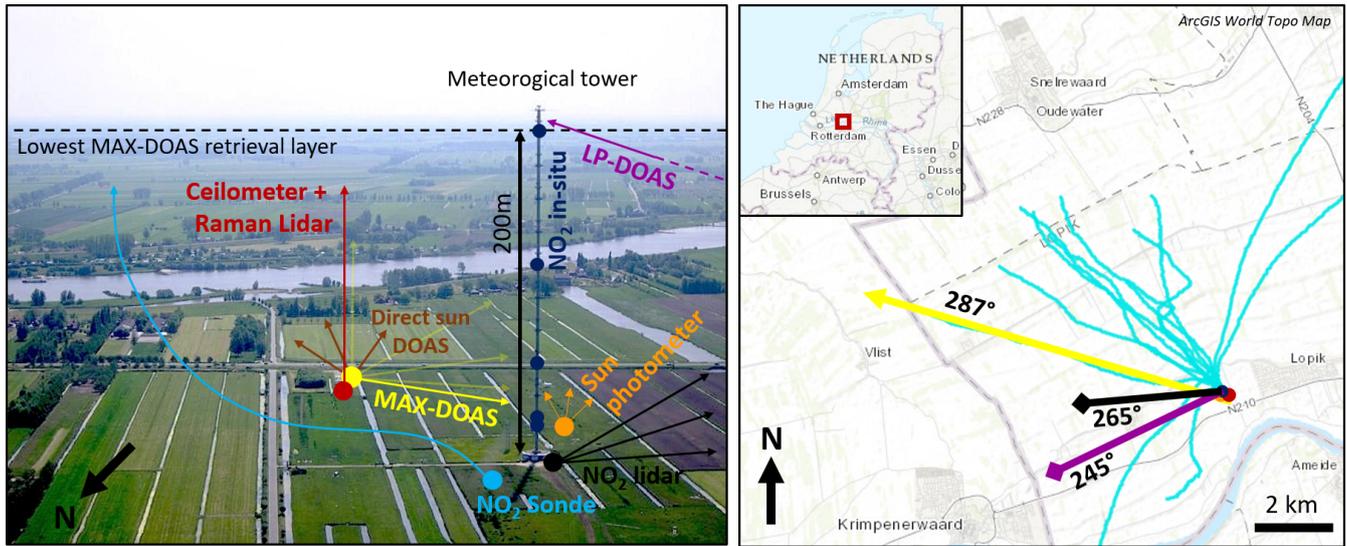
The planetary boundary layer (PBL) is the lowest part of the atmosphere, whose behaviour is directly influenced by its contact with the Earth's surface. Its chemical composition and aerosol load is ~~determined by gas and particulate matter driven by the~~ exchange with the surface ~~and also driven by~~, transport processes and homogeneous and heterogeneous chemical reactions. Monitoring of both, trace gases and aerosols, preferably simultaneous, is crucial for the understanding of the spatio-temporal evolution of the PBL composition and the chemical and physical processes.

Multi-AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) (e.g. Hönninger and Platt, 2002; Hönninger et al., 2004; Wagner et al., 2004; Heckel et al., 2005; Frieß et al., 2006; Platt and Stutz, 2008; Irie et al., 2008; Clémer et al., 2010; Wagner et al., 2011; Vlemmix et al., 2015b) is a ~~well-established widely used~~ ground-based measurement technique for the detection of aerosols and trace gases particularly in the ~~PBL and the lower free lower~~ troposphere: ultraviolet (UV)- and visible (Vis) ~~radiation absorption~~ spectra of skylight are analysed to obtain information on different atmospheric ~~parameters, integrated along absorbers and scatterers, integrated over~~ the light path (in fact a superposition of a multitude of light paths) ~~from the top of the atmosphere (TOA) to the instrument~~. The amount of atmospheric trace gases along the light path is inferred by identifying and analysing their characteristic narrow spectral absorption features, applying differential optical absorption spectroscopy (DOAS, Platt and Stutz, 2008). ~~Detectable gases~~ Gases that have been analysed in the UV and visible spectral range are nitrogen dioxide (NO<sub>2</sub>), formaldehyde (HCHO) nitrogen dioxide (NO<sub>2</sub>), formaldehyde (HCHO), nitrous acid (HONO), water vapour (H<sub>2</sub>O), sulfur dioxide (SO<sub>2</sub>), ozone (O<sub>3</sub>), glyoxal (CHOCHO) and halogen oxides (e.g. BrO, OClO). The oxygen

collision ~~complex-induced absorption (in the following treated as if being an additional trace gas species~~  $O_4$ ) can be used to infer information on aerosols: since the concentration of  $O_4$  concentration is proportional to the square of the  $O_2$  concentration, its vertical distribution is well known. The  $O_4$  absorption signal can therefore be utilized as a proxy for the light path with the latter being strongly dependent on the atmosphere's aerosol content. An appropriate set of spectra recorded under a narrow field of view (FOV, full aperture angle around 10 mrad) and different viewing elevations ("Multi-Axis") provides information on the trace gas and aerosol vertical distributions. Profiles can be retrieved from this information by applying numerical inversion algorithms, typically incorporating radiative ~~transport-transfer~~ models. These profile retrieval algorithms are the subject of this comparison study.

Today, there are numerous ~~such-retrieval~~ algorithms in regular use within the MAX-DOAS community which rely on different mathematical inversion approaches. This study involves nine of these algorithms (listed in Table 2), of which six use the optimal estimation method (OEM), two use a parametrized approach (PAR) and one algorithm relies on simplified radiative transport assumptions and analytical calculations (ANA). The main objective of this study is to assess their consistency ~~with respect to different conditions~~ and to review strengths and weaknesses of the individual algorithms and techniques. Note that this study is strongly linked to the report by Frieß et al. (2019), who performed similar investigations on nearly the same set of profiling algorithms with synthetic data, whereas the underlying data here was recorded during the second "Cabauw Intercomparison for Nitrogen Dioxide measuring Instruments" (CINDI-2, Apituley et al., 2020 in prep.). The CINDI-2 campaign took place from 25 August to 7 October 2016 on the Cabauw Experimental Site for Atmospheric Research (CESAR, 51.9676° N, 4.9295° E) in the Netherlands, which is operated by the Royal Netherlands Meteorological Institute (KNMI). 36 spectrometers of 24 participating groups from all over the world were synchronously measuring together with a wide range of supporting observations (in situ analysers, balloon sondes, lidars, Long-Path DOAS, direct-sun DOAS, sun photometer and ~~others~~meteorological instruments) for validation. This study compares MAX-DOAS profiles of  $NO_2$ , ~~HCHO and aerosol extinction~~ (and HCHO concentrations as well as the aerosol extinction coefficient (derived from  $O_4$  observations)) from 15 of the 24 groups. The results are compared with each other and validated against CINDI-2 supporting observations. For HONO and  $O_3$  profiling results please refer to Wang et al. (2020) and Wang et al. (2018), respectively. ~~The results are compared with each other and validated against CINDI-2 supporting observations.~~ In a recent publication by Bösch et al. (2018), CINDI-2 MAX-DOAS profiles retrieved with the BOREAS algorithm were already compared against supporting observations but regarding a few days only. Finally it shall be mentioned that already in the course of the precedent CINDI-1 campaign in 2009, there were comparisons of MAX-DOAS aerosol extinction coefficient profiles e.g. by Frieß et al. (2016) and Zieger et al. (2011), however also over shorter periods and a smaller group of participants.

The paper is organized as follows: Sect. 2 introduces the campaign setup, the MAX-DOAS dataset with the participating groups and algorithms (Sect. 2.1), the available supporting observations for validation (Sect. 2.2) and the general comparison strategy (Sect. 2.3). The comparison results are shown in Sect. 3. A compact summarizing plot and the conclusions appear in Sect. 4.



**Figure 1.** Left: Image of the CESAR site with position and approximate viewing directions of the MAX-DOAS instruments and supporting observations of relevance for this study. Right: Map (Esri et al., 2018) with instrument locations, viewing geometries and sonde flight paths indicated.

## 2 Instrumentation and methodology

Figure 1 shows an overview of the CINDI-2 campaign setup, including the supporting observations relevant for this study. Instrument locations, pointing (remote sensing instruments) and flight paths (radiosondes) are indicated on the map. Details on the instruments and their data products can be found in the following subsections. For further information refer to Kreher et al.

5 (2019) and Apituley et al. (2020 in prep.).

### 2.1 MAX-DOAS dataset

#### 2.1.1 Underlying dSCD dataset

Deriving vertical gas concentration/aerosol extinction profiles from scattered skylight spectra can be regarded as a two-step process: the 1<sup>st</sup> step is the DOAS spectral analysis, where the magnitude of characteristic absorption patterns of different gas species in the recorded spectra is quantified to derive the so called "differential slant column densities" (dSCDs, definition in the following paragraph). These provide information on integrated gas concentrations along the lines of sight. The 2<sup>nd</sup> step is the actual profile retrieval, where inversion algorithms incorporating atmospheric radiative transfer models (RTM) are applied to retrieve concentration profiles from the dSCDs derived in the 1<sup>st</sup> step.

The very initial data in the MAX-DOAS processing chain are spectra-intensities of scattered skylight  $I_{\lambda}(\alpha)$  at different wavelengths  $\lambda$  (ultra violet and visible spectral range, typical resolutions of 0.5 to 1.5 nm) recorded under different viewing elevation angles  $\alpha$  (ideally the telescope's FOV is usually-negligible compared to the elevation angle resolution). Along the

light path  $l$  from the top of the atmosphere (TOA) to the instrument on the ground, each atmospheric gas species  $i$  imprints its unique spectral absorption pattern (given by the absorption cross section  $\sigma_{i,\lambda}$ ) onto the TOA spectrum  $I_{\lambda,TOA}$  with the optical thickness

$$\tau_{\lambda}(\alpha) = \log\left(\frac{I_{\lambda,TOA}}{I_{\lambda}(\alpha)}\right) = \sum_i \sigma_{i,\lambda} S_i(\alpha) + C \quad (1)$$

5  $S_i(\alpha)$  is the slant column density (SCD), which is the trace gas concentration integrated along  $l$ .  $C$  represents **further** terms accounting for other instrumental and physical effects than trace gas absorption (for instance scattering on molecules and aerosols) that will not be further discussed in this context.  $S_i(\alpha)$  is inferred by spectrally fitting literature values of  $\sigma_{i,\lambda}$  to the observed  $\tau_{\lambda}(\alpha)$ . Since normally  $I_{\lambda,TOA}$  is not available for the respective instrument, optical thicknesses are instead assessed with respect to the spectrum recorded in zenith viewing direction to obtain

$$10 \quad \Delta\tau_{\lambda}(\alpha) = \log\left(\frac{I_{\lambda}(\alpha = 90^\circ)}{I_{\lambda}(\alpha)}\right) \quad (2)$$

Then the spectral fit yields the so called differential slant column densities (dSCDs)

$$\Delta S(\alpha) = S(\alpha) - S(\alpha = 90^\circ) \quad (3)$$

which are the typical output of the DOAS spectral analysis when applied to MAX-DOAS data. For further details on the DOAS method refer to Platt and Stutz (2008).

15 During the CINDI-2 campaign, each participant measured spectra with an own instrument and derived dSCDs applying their preferred DOAS spectral analysis software. The pointings (azimuthal and elevation) of all MAX-DOAS instruments were aligned to a common direction (Donner et al., 2019) and all participants had to comply with a strict measurement protocol, assuring synchronous pointing and spectra acquisition under highly comparable conditions ([Apituley et al., 2009](#)) ([Apituley et al., 2020 in prep.](#)). A detailed comparison and validation of the dSCD results was conducted by Kreher et al.

20 (2019). In the course of their study, Kreher et al. identified the most reliable instruments to derive a "best" median dSCD dataset. This dataset - in the following referred to as the "median dSCDs" - was distributed among the participants. All participants used the median dSCDs as the input data for their retrieval algorithms and retrieved the profiles that are compared in this study. The "median dSCD" approach was chosen for the following reasons: i) it enables to compare the profiling algorithms independently from differences in the input dSCDs, which is necessary to assess the individual algorithm performances. ii) it

25 makes this study directly comparable to the report by Frieß et al. (2019). Among others, this allows to assess to what extent MAX-DOAS profiling studies on synthetic data (with lower effort) can be used to substitute studies on real data. iii) ~~two~~ Two decoupled studies are obtained (Kreher et al. and this study), each confined to a single step in the MAX-DOAS processing chain (the DOAS spectral analysis to obtain dSCDs and the actual profile inversion). A disadvantage of the median dSCD approach is, that the reliability of a typical MAX-DOAS observation undergoing the whole spectra acquisition and processing chain

30 cannot be assessed. Therefore, a comparison of profiles retrieved with the participant's own dSCDs was also conducted, but is not a substantial part of this study. However, these results and a corresponding short discussion can be found in Supplement

S10 and Sect. 3.7, respectively. The median dSCDs cover the campaign core period from 12 to 28 September 2016, considering only data from the first 10 minutes of each hour between 7:00 and 16:00 UT, where the CINDI-2 MAX-DOAS measurement protocol scheduled an elevation scan in the nominal 287° azimuth viewing direction with respect to the north. Hence, the total number of processed elevation scans was 170. An elevation scan consisted of ten successively recorded spectra at viewing elevation angles  $\alpha$  of 1, 2, 3, 4, 5, 6, 8, 15, 30 and 90°, at an acquisition time of 1 minute each. DSCDs were provided for **five** three chemical species, namely O<sub>4</sub> UV, O<sub>4</sub> Vis, HCHO, NO<sub>2</sub> UV and HCHO. O<sub>4</sub> and NO<sub>2</sub> Vis, where “UV” and “Vis” indicate different DOAS were each provided for two different spectral fitting ranges in the ultraviolet, in the ultra-violet (UV) and the visible (Vis) spectral region, respectively resulting in five data products (see Table 1). From the median dSCDs, the participants retrieved profiles for the species listed in Table 1. Not all participants retrieved all species and therefore do not necessarily appear in all plots.

**Table 1.** List of the retrieved species and fitting ranges. For further details on the spectral analysis, please refer to Kreher et al. (2019).

Species	Retrieved quantity	Retrieved from dSCDs of	spectral fitting window [nm]
Aerosol UV	Extinction <u>coefficient</u> [km <sup>-1</sup> ]	O <sub>4</sub> UV	338 - 370
Aerosol Vis	Extinction <u>coefficient</u> [km <sup>-1</sup> ]	O <sub>4</sub> Vis	425 - 490
NO <sub>2</sub> UV	Number concentration [molec cm <sup>-3</sup> ]	NO <sub>2</sub> UV	336.5 - 359
NO <sub>2</sub> Vis	Number concentration [molec cm <sup>-3</sup> ]	NO <sub>2</sub> Vis	425 - 490
HCHO	Number concentration [molec cm <sup>-3</sup> ]	HCHO	336.5 - 359

### 2.1.2 Participating groups and algorithms

Table 2 lists the compared algorithms including the underlying method (OEM, PAR or ANA) and the participating groups with corresponding labels and plotting symbols as they are used throughout the comparison. OEM and PAR algorithms rely on the same idea: a layered horizontally homogeneous atmosphere is set up in a radiative transfer model (RTM) with distinct parameters (aerosol extinction coefficient, trace gas amounts, temperature, pressure, aerosol microphysical properties, ... water vapour and aerosol properties) attributed to each layer. This model atmosphere is then used to simulate MAX-DOAS dSCDs under consideration of the viewing geometries. To retrieve a profile from the measured dSCDs, the model parameters are optimized to obtain maximum agreement minimise the difference between the simulated and measured dSCDs by minimising based on a pre-defined cost function. Typically only  $p = 2$  to 4 Regarding profiles, typically only two to four degrees of freedom for signal (DOFS or  $p$ ) can be retrieved from MAX-DOAS observations, such that general profile retrieval problems with more than  $p$  layers are underconstrained and a priori independent retrieved parameters are ill-posed and prior information has to be assimilated to obtain unambiguous solutions achieve convergence. For OEM algorithms, this is provided in the form of an *a priori* profile (Rodgers, 2000) “filling” the lack of information which is most prominent at higher altitudes (see Sect. 3.1). Parametrized approaches achieve this and associated a priori covariance (Rodgers, 2000), defining the most likely profile and constraining the space of possible solutions according to prior experience.

They constitute a portion of the OEM cost function such that with decreasing information contained in the measurements, layer concentrations are drawn towards their *a priori* values. PAR algorithms implement prior assumptions by only allowing predefined profile shapes which can be described by a few parameters.

For OEM algorithms, the radiative transport simulations are typically performed online in the course of the retrieval whereas the PAR algorithms in this study rely on look-up tables, which are pre-calculated for the parameter ranges of interest. Therefore, PAR algorithms are typically faster than OEM algorithms but also require more memory. The ANA approach by NASA was developed as a quick look algorithm and assumes a simplified radiative transport, based on trigonometric considerations. Since the model equations can be solved analytically for the parameters of interest, neither radiative transport simulation nor the calculation of look-up tables is necessary and an outstanding computational performance is achieved compared to other algorithms (factor of  $\approx 10^3$  in processing time, see Frieß et al., 2019).

For further descriptions of the methods and the individual algorithms, please refer to Frieß et al. (2019). ~~The Besides the algorithms described therein, our study includes results from the~~  $M^3$  algorithm by LMU ~~appears as an additional algorithm in our study.~~ Its description can be found in Supplement S1. For details, refer to the references given in Table 2.

Note that two versions of aerosol results from the MAPA algorithm with different  $O_4$  scaling factors ( $SF$ ) are discussed within this paper, referred to as mp-0.8 (retrieved with  $SF = 0.8$ ) and mp-1.0 ( $SF = 1.0$ ), respectively. The scaling factor is applied to the measured  $O_4$  dSCDs prior to the retrieval and was initially motivated by previous MAX-DOAS studies which reported a significant yet debated mismatch between measured and simulated dSCDs (~~Wagner et al., 2019; Ortega et al., 2016, and references~~ (e. g. Wagner et al., 2009; Clémer et al., 2010; Ortega et al., 2016; Wagner et al., 2019, and references therein)). Also for MAPA during CINDI-2, a scaling factor of 0.8 was found to improve the dSCD agreement, to enhance the number of valid profiles and to significantly improve the agreement with the sun photometer aerosol optical thickness (Beirle et al., 2019). However, in the course of this study it was found that for OEM algorithms the disagreement between sun-photometer and MAX-DOAS can largely be explained by smoothing effects (see Sect. 3.4) and that (at least averaged over campaign) there are no clear indications that a  $SF$  is necessary ~~if smoothing effects, in particular the low sensitivity of MAX-DOAS observations to higher altitudes, are taken into account (see Sect. 2.3.2 and (see Supplement S2).~~

### 2.1.3 Retrieval settings

To reduce possible sources of discrepancies, all profiles shown in this study were retrieved according to predefined settings similar to those of the intercomparison study by Frieß et al. (2019): pressure, temperature, total air density, and  $O_3$  vertical profiles between 0 and 90 km altitude were averaged from  $O_3$  sonde measurements performed in De Bilt by KNMI during September months of the years 2013-2015. ~~The surface albedo was fixed to 0.06, according to ?.~~ A fixed altitude grid was used for the retrieval inversion, consisting of ~~20 layers between 0 and 4 km~~ 20 layers between 0 and 4 km altitude, each with a height of  $\Delta h = 200\text{m}$ . The results of the parametrized approaches and OEM algorithms where the exact grid could not ~~be directly implemented~~ readily be applied during inversion, were interpolated/averaged accordingly afterwards. Note that, for radiative transfer simulations, the atmosphere was represented by finer (25 to 100 m) layers close to the surface, increasing with altitude) and farther extending (up to 40 to this grid to simplify the comparison 90 km altitude) grids, inherently defined by the

[individual retrieval algorithms](#). Surface and instruments' altitudes were fixed to ~~0 m~~0 m, which is close to the real conditions: the CESAR site and most of the surrounding area lie at ~~0.7-0.7~~ metres b.s.l., whereas the instruments were installed at ~~0 to 6 m~~0 to 6 m above sea level. The model wavelengths were fixed according to Table 3. In the case of the HCHO retrieval, the aerosol profiles retrieved at ~~360 nm were interpolated to 343 nm~~[360 nm were extrapolated to 343 nm](#) using the mean Ångström exponent for the ~~440-675 nm~~[440 – 675 nm](#) wavelength range derived from sun photometer measurements (see Sect. 2.2.1) on 14 September 2016 in Cabauw. For the aerosol parameters, the single scattering albedo was fixed to 0.92 and the asymmetry factor to 0.68 for both 360 and 477 nm. These are mean values for 14/09/2016 derived from AERONET measurements at ~~440 nm~~[440 nm](#) in Cabauw. The standard CINDI-2 trace gas absorption cross-sections were applied (see Kreher et al., 2019). A scaling of the measured O<sub>4</sub> dSCDs prior to the retrieval was not applied. An exception is the parametrized MAPA algorithm for which two datasets, one without and one with a scaling ( $SF = 0.8$ ) were included in this study. The OEM *a priori* profiles for both aerosol and trace gas retrievals were exponentially-decreasing profiles with a scale height of 1 km and aerosol optical thicknesses (AOTs) and vertical column densities (VCDs) as given in Table 3. For the AOTs the mean value at 477 nm for the first days of September 2016 derived from AERONET measurements are used. Trace gas VCDs are mean values derived from OMI observations in September 2006-2015. *A priori* variance and correlation length were set to 50% and 200 m, respectively.

#### 15 2.1.4 Requested dataset

All participants were requested to submit the following results of their retrieval: ~~(1i)~~ Profiles and profile errors, optionally with errors separated into contributions from propagated measurement noise and smoothing effects. ~~(2ii)~~ Modelled dSCDs as calculated by the RTM for the retrieved atmospheric state. ~~(3iii)~~ Averaging Kernels (AVKs) for assessment of information content and vertical resolution (only available for OEM approaches). ~~(4iv)~~ Optional flags, giving participants the opportunity to mark profiles as invalid. The flagging must be based on inherent quality indicators, which typically are the root-mean-square difference between measured and modelled dSCDs or the general plausibility of the retrieved profiles. Note that only four institutes submitted flags (INTA/[bePRO](#), BIRA/ bePRO, KNMI/[MARK](#) and MPIC/ MAPA). It is assumed that an accurate aerosol retrieval is necessary to infer light path geometries, thus trace gas profiles are generally considered invalid if the underlying aerosol retrieval is invalid. A detailed description of the flagging criteria and flagging statistics can be found in Supplement S3.

## 2.2 Supporting observations

This section introduces the supporting observations, that were used for comparison and validation of the MAX-DOAS retrieved [profiles results](#). It shall be pointed out that a general challenge here was to find compromises between i) using only accurate and representative data with good spatio-temporal overlap and ii) keeping as many supporting data as possible to have a large comparison dataset. Considerations and investigations on this issue (e.g. comparisons between the supporting observations, spatio-temporal variability and overlap,~~...~~) which lead to the decisions finally taken are mentioned in the following subsections and described in more detail in the supplementary material they refer to.

## 2.2.1 Aerosol optical thickness (AOT)

Independent aerosol optical thickness measurements  $\tau_{aer}$  were performed with a sun photometer (CE318-T by Cimel) located close to the meteorological tower of the CESAR site (see Fig. 1), which is part of the Aerosol Robotic Network (AERONET, see Holben et al., 1998). AOTs were derived from direct-sun radiometric measurements in  $\approx 15$  minute intervals at 1020, 870, 5 675 and 440 nm wavelength. The AERONET level 2.0 data was used, which is cloud screened, recalibrated and quality filtered (according to Smirnov et al., 2000). For the extrapolation of  $\tau_{aer}$  to the DOAS retrieval wavelengths of 360 and 477 nm, a dependency of  $\tau_{aer}$  on the wavelength  $\lambda$  according to

$$\ln \tau_s(\lambda) = \alpha_0 + \alpha_1 \cdot \ln \lambda + \alpha_2 \cdot (\ln \lambda)^2 \quad (4)$$

was assumed, following Kaskaoutis and Kambezidis (2006). The parameters  $\alpha_i$  were retrieved by fitting Eq. (4) to the available 10 data points. Note that  $\alpha_1$  corresponds to the Ångström exponent when only the first two (linear) terms on the right hand side are used. The last quadratic term enables to additionally account for a change of the Ångström exponent with wavelength. For the linear temporal interpolation to the MAX-DOAS profile timestamps, the maximum interpolated data gap was set to 30 min, resulting in a data coverage of about 30%. Smirnov et al. (2000) propose a sun photometer total accuracy in  $\tau_s$  of 0.02. Each AOT is actually an average over three subsequently performed measurements. In this study, ~~an enhanced uncertainty of 0.04 is assumed due to temporal and spectral inter-/extrapolation~~ the proposed accuracy of 0.02 was enhanced by the variability between them (typically on the order of 0.008). 15

## 2.2.2 Aerosol extinction profiles

Information on the ~~true aerosol extinction (AE) profiles~~ aerosol extinction coefficient profiles (in the following referred to by "aerosol profiles") was obtained by combining the sun photometer AOT with data from a ceilometer (Lufft CHM15k Nimbus). 20 The latter continuously provided vertically resolved information on the atmospheric aerosol content by measuring the intensity of elastically backscattered light from a pulsed laser beam (1064 nm) propagating in zenith direction (see e.g. Wiegner and Geiß, 2012). The raw data are attenuated backscatter coefficient profiles over an altitude range from 180 m to 15 km, with a temporal and vertical resolution of 12 s and 10 m, respectively. These were converted to extinction coefficient profiles ~~(in the following referred to by "extinction profiles")~~ by scaling with simultaneously measured sun photometer or MAX-DOAS 25 AOTs. This is described in detail in Supplement S4.1. Note that the approach described there presumes a constant extinction coefficient for altitudes  $\leq 180$  m and that the aerosol properties like size distribution, single scattering albedo and shape remain constant with altitude. To check plausibility, Supplement S4.1 compares the resulting profiles at 360 nm to a few available extinction coefficient profiles, measured by a Raman lidar at 355 nm (the CESAR Water Vapor, Aerosol and Cloud lidar "CAELI", operated within the European Aerosol Research lidar Network (EARLINET, Bösenberg et al., 2003; Pappalardo 30 et al., 2014) and described in detail in Apituley et al., 2009). The average RMSD between scaled ceilometer and Raman lidar profiles is  $\approx 0.03$  up to 4 km altitude is  $\approx 0.03 \text{ km}^{-1}$ . However since there are only few Raman lidar validation profiles available and only for altitudes  $> 1$  km, the ceilometer aerosol ~~extinction~~ profiles should be consulted for qualitative comparison only.

### 2.2.3 NO<sub>2</sub> profiles

NO<sub>2</sub> profiles were recorded sporadically by two measurement systems: radiosondes (described in Sluis et al., 2010) and an NO<sub>2</sub> lidar (Berkhout et al., 2006). Radiosondes were launched at the CESAR measurement site during the campaign. For this study, only data from sonde ascents through the lowest 4 km (which is the MAX-DOAS profiling retrieval altitude range) were used. A sonde profile was considered temporally coincident to a MAX-DOAS profile, when the middle timestamps of MAX-DOAS elevation scan and sonde flight were less than 30 minutes apart. The horizontal sonde flight paths are indicated in Fig. 1. Typical flight times (lowest 4 km) were of the order of 10 - 15 minutes. Data was recorded at a rate of 1 Hz, typically resulting in a vertical resolution of approximately 10 m at an approximate measurement uncertainty in NO<sub>2</sub> concentration of  $5 \times 10^{10}$  molec cm<sup>-3</sup>. The horizontal travel distances varied strongly between 4 and 18 km. A detailed overview on the flights is given in Supplement S4.2.

The NO<sub>2</sub> lidar is a mobile instrument setup inside a lorry which was located close to the CESAR meteorological tower. It combines lidar observations at different viewing elevation angles to enhance vertical resolution and to obtain sensitivity close to the ground, despite the limited range of overlap between sending and receiving telescope (see also Sect. 2.2.2). The instrument is sensitive along its line of sight from 300 to 2500 m distance to the instrument. The azimuthal pointing was 265° with respect to the north and the operational wavelength is 413.5 nm. Typical specified uncertainties in the retrieved concentrations are around  $2.5 \times 10^{10}$  molec cm<sup>-3</sup>. Profiles were provided at a temporal resolution of 28 minutes, each profile consisting of a series of (occasionally overlapping) altitude intervals with constant gas concentration. For an exemplary profile and details on its conversion to the MAX-DOAS retrieval altitude grid, please refer to Supplement S4.3. A lidar profile was considered temporally coincident to a MAX-DOAS profile, when the middle timestamps of MAX-DOAS elevation scan and lidar profile were less than 30 minutes apart. [This resulted into 25 suitable Lidar profiles recorded on six different days during the campaign.](#) Example profiles of both radiosonde and NO<sub>2</sub> lidar are shown in the course of a comparison between the two observations in Supplement S4.5.

### 2.2.4 Trace gas vertical column densities (VCD)

Tropospheric trace gas VCDs were derived from direct-sun DOAS (~~DS-DOAS~~) observations, which were performed between minutes 40 and 45 of each hour. NO<sub>2</sub> VCDs were retrieved from combined datasets of two Pandora DOAS instruments (instrument numbers 31 & 32) and calculated based on the Spinei et al. (2014) approach. The reference spectrum was created from the spectra with lowest radiometric error over the whole campaign and the residual NO<sub>2</sub> signal was determined by applying the so-called Minimum Langley Extrapolation (Herman et al., 2009). The temperature dependence of the NO<sub>2</sub> cross sections was used to separate the tropospheric from the stratospheric column.

HCHO VCDs were retrieved from data of the BIRA DOAS instrument (number 4). A fixed reference spectrum acquired on 18 September 2016 at 9:41 UTC and 55.6° SZA was used. DOAS fitting settings were identical to those used for the CINDI-2 HCHO dSCD intercomparison (Kreher et al., 2019). The residual amount of HCHO in the reference spectrum of  $(8.8 \pm 1.6) \times 10^{15}$  molec cm<sup>-2</sup> was estimated using a MAX-DOAS profile retrieved on the same day and a geometrical AMF

corresponding to  $55.6^\circ$  SZA. Because of that, the HCHO VCDs cannot be considered as a fully independent dataset. VCDs were calculated from total HCHO SCDs using a geometrical AMF including a simple correction for the earth sphericity. Only spectra with DOAS fit residuals  $< 5 \times 10^{-4}$  were considered as valid direct-sun data. As for AOTs, these observations can only be performed when the sun is clearly visible, hence the coverage for cloudy scenarios is scarce.

### 5 2.2.5 Trace gas surface concentrations

Note that in the following, “surface concentration” will not refer to measurements in the very proximity to the ground but to the average concentration in the lowest 200 m of the atmosphere, as retrieved for the MAX-DOAS first profile layer. Trace gas surface concentrations of HCHO and  $\text{NO}_2$  were provided by a long path DOAS system operated by IUP-Heidelberg (LP-DOAS, see Pöhler et al., 2010; Merten et al., 2011; Nasse et al., 2019) (LP-DOAS, see Pikelnaya et al., 2007; Pöhler et al., 2010; Mer

10 . The LP-DOAS system consists of a light-sending and receiving telescope unit located at 3.8 km horizontal distance to a retro reflecting mirror mounted at the top (207 m altitude) of the meteorological tower (see Supplement S4.4). Light from a UV-Vis light source is sent by the telescope to the retroreflector and the reflected light is again received by the telescope unit and spectrally analysed applying the DOAS method. The fundamental difference to the MAX-DOAS instruments is the well-defined light path which enables very accurate determination of trace gas mixing ratios, averaged along the line of sight.  
15 Accordingly, with the retroreflector mounted at 207 m altitude, one obtains average mixing ratios over the lowest MAX-DOAS retrieval layer, as indicated in Fig. 1. Considering DOAS fitting errors and uncertainties in the applied literature cross-sections (Vandaele et al., 1998; Meller and Moortgat, 2000; Pinardi et al., 2013) yields an average accuracy of the LP-DOAS of  $\pm 1.5 \times 10^9 \text{ molec cm}^{-3} \pm 3\%$  ( $\pm 5 \times 10^9 \text{ molec cm}^{-3} \pm 9\%$ ) for  $\text{NO}_2$  (HCHO), respectively. Given the high accuracy, the  
20 most reliable dataset for the validation of CINDI-2 MAX-DOAS trace gas profiling results.

Further observations for qualitative validation are the surface values of the  $\text{NO}_2$  lidar and the radiosondes and also in-situ monitors in the CESAR meteorological tower. Teledyne in situ  $\text{NO}_2$  monitors (Teledyne API, model M200E) were located in the tower basement and were subsequently connected to different inlets located at 20, 60, 120 and 200 m altitude (switching intervals approx. 5 minutes). Further, a CAPS (type AS32M, based on attenuated phase shift spectroscopy, Keabian et al.,  
25 2005) and a CE-DOAS (cavity enhanced DOAS, Platt et al., 2009 and Horbanski et al., 2019) were continuously measuring at 27 m altitude. All the in situ measurements at the tower were combined to obtain another set of surface concentration measurements, more representative for concentrations close to the site. The data were combined by linearly interpolating over altitude between the instruments and subsequently averaging the resulting profile over the retrieval surface layer (0 - 200 m altitude). Note that this method gives a large weight to the uppermost measurements, as they are representative for the majority  
30 of the relevant layer.

### 2.2.6 Meteorology

Meteorological data for the surface layer (pressure, temperature and wind information) routinely measured at the CESAR site were taken from the CESAR database (CESAR, 2018) at a temporal resolution of 10 minutes. Cloud conditions were

retrieved from MAX-DOAS data of instruments 4 and 28 according to the cloud classification algorithm developed by MPIC (Wagner et al., 2014; Wang et al., 2015). Basically only two cloud condition states are distinguished in the statistical evaluation: "clear-sky" (green) and "presence of clouds" (red). Only in the overview- and correlation plots, "presence of clouds" is further subdivided into "optically thin clouds" (orange) and "optically thick clouds" (red). According to this classification 72 (98) of the 170 profiles were measured under clear-sky (cloudy) conditions. Over the whole campaign, there was only one rain event (precipitation > 0.01 mm) coinciding with the measurements on 25 September 2016 between 15:00 and 17:00 h UT. At forenoon on 16 September, a heavy fog event strongly limited the visibility (see also Supplement S5).

## 2.3 Comparison strategy

### 2.3.1 General approach

Different MAX-DOAS retrieval algorithms were extensively compared in Frieß et al. (2019) using synthetic data. The crucial differences of the presented study are: i) The underlying spectra are not synthetic, but were recorded with real instruments, meaning that real noise and instrument artefacts propagate into the results. ii) Independent information on the real profile can only be inferred from supporting observations with their own uncertainties and an imperfect spatio-temporal overlap with the MAX-DOAS measurements. iii) The real conditions encountered can exceed the model's scope because horizontal inhomogeneities or the fact that many of the fixed forward model input parameters (such as aerosol properties, surface albedo, ~~T/P-profiles, ...~~ temperature and pressure profiles) are averaged quantities of former observations which might be inaccurate for specific days and conditions. iv) In some cases, different participants used the same retrieval algorithms; this allows assessment of the impact of different settings in the remaining parameters, which were not prescribed (see Sect. 2.1.3). The approaches chosen here are therefore limited to the examination of i) the consistency among the participants, ii) the consistency of the results with available supporting observations and iii) inherent quality proxies of the retrieval (described in the next paragraph). Table 4 summarizes the quantities which are compared, together with the corresponding supporting observations if available.

In this study, agreement between different observations are statistically assessed by ~~correlation analysis (weighted least-squares regression) and i)~~ weighted root-mean-square differences (RMSD), ii) weighted "Bias" as introduced below and iii) weighted least-squares regression analysis. Discussions and summary are focussed on ~~RMSDs as in contrast to correlation~~ RMSD, being the most fundamental quantity as it represents both, statistical and systematic deviations. The Bias was introduced as a general proxy for systematic deviations. Correlation coefficient, slope and offset from the regression analysis ~~, RMSD is representative for both, statistical and systematic deviations. For~~ are provided and consulted for a more differentiated view.

Consider two time series of length  $N_T$ : the retrieval result  $x_{p,t}$  and of a participant  $p$  at time  $t$  and some reference observation  $x_{ref,t}$  (each consisting of  $N_T$  data points,  $t$  and  $p$  indicating time and participant, respectively either MAX-DOAS median results or data from supporting observations, as further described below) with associated uncertainties  $\sigma_{p,t}$  and  $\sigma_{ref,t}$ . Then the RMSD is given by defined as:

$$\text{RMSD: } \sigma_{rms,p} = \sqrt{\frac{1}{N_T} \cdot \frac{1}{\sum_t w_t} \cdot \sum_t w_t (x_{p,t} - x_{ref,t})^2} \quad (5)$$

For both, RMSD calculation and least square regression, contributing data points are weighted by the reciprocal of the quadratic sum of their uncertainties. The weights  $w_t$  are defined according to

$$w_t = \frac{1}{\sigma_{p,t}^2 + \sigma_{ref,t}^2} \quad (6)$$

and are also applied for the Bias calculation and regression analysis. The Bias is defined as

$$w_{Bias}: \quad \sigma_{bias,p} = \frac{1}{\sigma_{p,t}^2 + \sigma_{ref,t}^2} \frac{1}{N_T} \cdot \frac{1}{\sum_t w_t} \cdot \sum_t w_t (x_{p,t} - x_{ref,t}) \quad (7)$$

Sometimes the term "average RMSD" ("average Bias") is used, which refers to the average over the RMSD (Bias) values of the individual participants, hence:-

Average RMSD:

$$\sigma_{arms,p} = 1/N_P \sum_p \sigma_{rms,p}$$

with  $N_P$  being the number of included participants. We further introduce the "average Bias magnitude", that averages the absolute values of the Bias. When referring to "relative RMSDs" ("relative Bias"), the underlying RMSD (Bias) value was divided by the average of the investigated quantity, hence:-

Relative RMSD:

$$\sigma_{rrms,p} = \frac{N_T \sigma_{rms,p}}{\sum_t x_{ref,t}}$$

The. For the linear regression analysis, the vertical distance between the model and the data points is minimised and also here the weights  $w_t$  are applied.

15 To assess the consistency among the participants is assessed by comparing the results of individual participants with the. the median result over the valid profiles of all participants is inserted as  $x_{ref,t}$ . The median is used instead of the mean value, since it is less sensitive to (sometimes unphysical) outliers. This comparison shows how far the choice of the retrieval algorithm/ technique affects the results but it does not reveal general systematic MAX-DOAS retrieval errors. Outliers observed for distinct participants and algorithms are therefore not necessarily an indicator for poor performance.

20 The To assess the consistency with supporting observations, the latter are inserted as  $x_{ref,t}$ . This comparison is a better indicator for the real retrieval performance. However, uncertainties of supporting instruments (see Supplement S4.5), smoothing effects (see Sect. 2.3.2) and imperfect spatial and temporal overlap of the different observations (see Sect. 2.3.3) complicate the interpretation.

Inherent quality indicators for An inherent quality indicator for the retrieval algorithms are the consistency of modelled and measured dSCDs and the consistency of NO<sub>2</sub> results retrieved in different wavelength ranges. During the inversion, the goal is to minimize the deviation between the RTM simulated dSCDs and the actually measured ones. If strong deviations remain after the final iteration in the minimisation process, this indicates failure of the retrieval. The consistency of retrieval results of NO<sub>2</sub> in the UV and the Vis spectral ranges is another indicator for an algorithm's reliability since they should ideally yield the same results.

In a few cases (e.g. [Section 3.2 Sect. 3.2, where full profiles are compared](#)) the scatter among several participants  $p$  (of number  $N_P$ ) and ~~potentially~~ several retrieval layers  $h$  (of number  $N_H$ ) is of interest. For this purpose, we define the "average standard deviation" (ASDev) which is the standard deviation observed among the participants for individual profiles averaged over retrieval layers and time, hence:

$$5 \text{ ASDev: } \sigma_{asdev} = \frac{1}{N_T} \sum_t \frac{1}{N_H} \sum_h \sqrt{\frac{1}{N_P - 1} \sum_p (x_{p,h,t} - \bar{x}_{h,t})^2} \quad (8)$$

with  $\bar{x}_{t,h}$ ,  $\bar{x}_{h,t}$  being the average (over participants) MAX-DOAS retrieved concentration for a given time  $t$  and layer  $h$ . If not stated otherwise, ASDev values of profiles are calculated considering the lowest five retrieval layers (up to 1 km altitude).

In the statistical evaluations, clear-sky and cloudy conditions as well as unfiltered and filtered data ([according to the flags provided by the participants](#)) are distinguished. The distinction between cloud conditions is of major importance, as particularly in the case of aerosol retrievals under broken clouds, the quality of the results is typically strongly degraded. A consequence of regarding these data subsets is that the number of contributing data points not only depends on the number of submitted profiles and the number of coincident data points from supporting observations but further on the filter settings. Any regression ~~or~~ [RMSD-RMSD or Bias value](#) with less than five contributing data points are considered to be statistically unrepresentative and are omitted. If not stated otherwise, numbers given in the text were calculated considering valid data only.

### 15 2.3.2 Smoothing effects

As shown in Sect. 3.1 below, in particular in the UV range, the sensitivity of ground-based MAX-DOAS observations decreases rapidly with altitude, meaning that species above  $\approx 1 \text{ km} \approx 2 \text{ km}$  typically cannot be reliably ~~detected~~ [quantified](#). At higher altitudes, OEM retrieval results are drawn towards the *a priori* profile ~~,~~ [\(according to the definition of the cost-function, see Rodgers \(2000\)\)](#), while the results of parametrized and analytical approaches are driven by the chosen parametrization and their implementation. Further, the vertical resolution is limited (from 100 to several hundred meters, increasing with altitude), which affects the profile shape and - of most importance in this study - the retrieved surface concentration. Both effects cause deviations from the true profile that are in the following referred to as "smoothing effects".

For a meaningful quantitative comparison, they should be considered. This is possible for OEM retrievals, where the information on the vertical resolution and sensitivity is given by the averaging kernel matrix (AVK, see Sect. 3.1 for details).  
 25 For a meaningful quantitative comparison of an OEM retrieved profile and a validation profile  $\mathbf{x}$  (assumed here to perfectly represent the true state of the atmosphere), the validation profile resolution and information content has to be degraded by "smoothing" it with the corresponding MAX-DOAS AVK matrix  $\mathbf{A}$  according to the following equation (Rodgers and Connor, 2003; Rodgers, 2000):

$$\tilde{\mathbf{x}} = \mathbf{A}\mathbf{x} + (\mathbf{1} - \mathbf{A})\mathbf{x}_a \quad (9)$$

30 Here,  $\mathbf{x}_a$  is the *a priori* profile and  $\tilde{\mathbf{x}}$  represents the profile that a MAX-DOAS OEM retrieval (with the resolution and sensitivity described by  $\mathbf{A}$ ) would yield in the respective scenario. For layers with high (low) gain in information,  $\tilde{\mathbf{x}}$  is drawn

towards  $x$  ( $x_a$ ), while vertical resolution is degraded if  $\mathbf{A}$  has significant off-diagonal entries (compare to Sect. 3.1). In this study, this has implications not only for the comparison of profiles, but also the comparison of the total columns (AOTs and VCDs, which are derived simply by vertical integration of the corresponding profiles) and surface trace gas concentrations. For total columns, the dominant issue is the lack of information at higher altitudes. In contrast, there is reasonable information on the surface concentration, however smoothing can have severe impact here in the case of strong concentration gradients close to the surface. The impact on the individual observations is discussed in the corresponding sections below. A particularly important consequence of smoothing effects is the "partial AOT correction" (PAC), which is introduced and discussed in Sect. 3.4.

10 Finally it shall be pointed out that the sensitivity and spatial resolution is strongly affected by the exact approach that is chosen to solve the ill-posed inversion problem. Frieß et al. (2006) for instance demonstrates, that the sensitivity to higher altitudes can be enhanced by relaxing the prior constraints and by retrieving profiles at several wavelengths simultaneously.

### 2.3.3 Spatio-temporal variability

It is obvious already from Fig. 1 and Sect. 2.2 that the MAX-DOAS instruments and the various supporting observations sample different air volumes at different times. In addition, the MAX-DOAS horizontal viewing distance (derived in Supplement S5) is highly variable, changing between 2 and 30 km during the campaign for the lowest viewing elevation angles. Similar investigations were already performed by Irie et al. (2011) using CINDI-1 data, however using a different definition of the viewing distance. Hence, Table S6 summarizes the spatial and temporal mismatches between MAX-DOAS and supporting observations. Spatial mismatches are of the order of 10 km, temporal mismatches vary between 0 and 20 minutes. Consequently, strong spatio-temporal variations of the observed quantities are expected to induce large discrepancies among the observations, independent of the data quality. Quantitative estimates of the impact on the comparison could only be derived for NO<sub>2</sub> surface concentrations and under strong simplifications (for details see S6) yielding an RMSD of  $3.5 \times 10^{10}$  molec cm<sup>-3</sup>. This is indeed of similar magnitude as the average RMSD observed during the comparison (approx.  $5 \times 10^{10}$  molec cm<sup>-3</sup>). It shall further be noted, that under strong spatial variability the horizontal homogeneity assumed by the retrieval forward models is ~~not given. It was not possible to derive a reliable quantitative estimate of the impact on the comparison, but investigations on~~ the NO<sub>2</sub> surface concentration in Supplement S6 and investigations by ? indicate, that it significantly contributes to the residual RMSD observed between different observations: inaccurate.

## 3 Comparison results

### 3.1 Information content

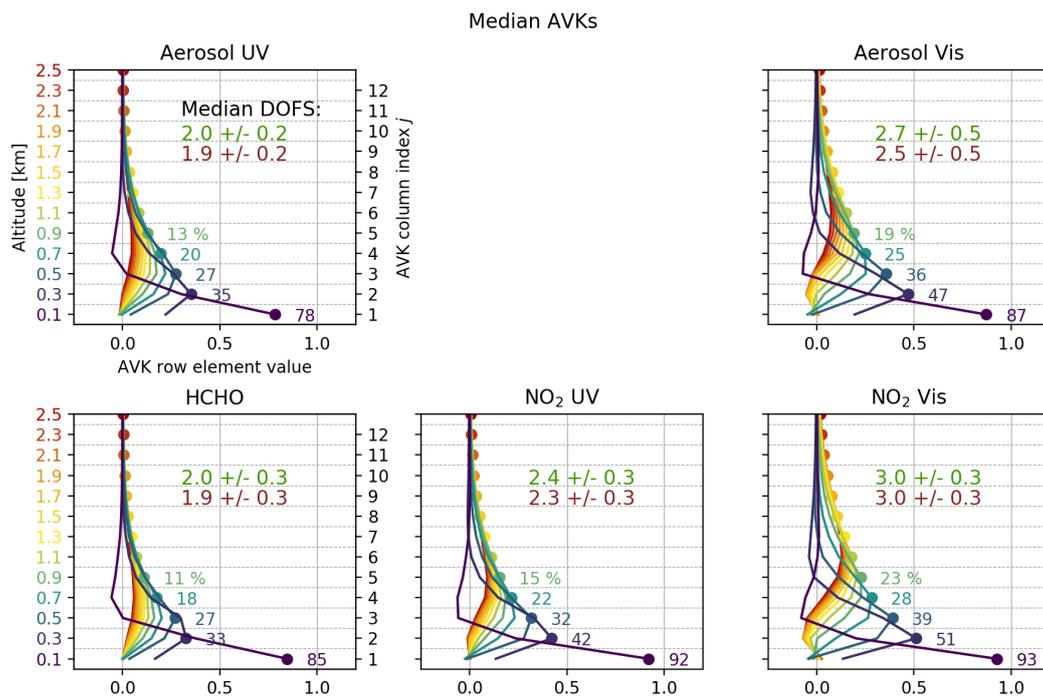
In the case of OEM retrievals, the gain in information on the atmospheric state can be quantified according to Rodgers (2000). Essentially speaking, this is done by comparing the knowledge before (represented by the *a priori* profile and its uncertainties) and after the profile retrieval. The gain in information for each individual vertical profile can be represented by the averaging

kernel matrix (AVK, denoted by  $\mathbf{A}$ ). ~~Each of its elements~~  $A_{ij}$  describes the sensitivity of the measured concentration in the  $i^{\text{th}}$  layer to small changes in the real concentration in the  $j^{\text{th}}$  layer. Each row  $A_i$  can thus be plotted over altitude providing the following information: (1) the value in the layer  $i$  itself (the diagonal element  $A_{ii}$  with a value between 0 and 1) gives the gain in information while  $1 - A_{ii}$  represents the amount of *a priori* knowledge which had to be assimilated to obtain a well defined concentration value. (2) The values in the other layers (off-diagonal elements of  $\mathbf{A}$ ) indicate the cross sensitivity of layer  $i$  to layer  $j$ . Typically, the cross sensitivity decreases with the distance to the layer  $i$ . ~~A reasonably defined characteristic~~ The length of this decay (note that  $i$  can be converted to the corresponding altitude by multiplication with the retrieval layer thickness  $\Delta h$ ) can serve as a measure is an indicator for the vertical resolution of the retrieval. ~~Here, the so-called "spread"  $s(i)$  was chosen as the characteristic length, as defined according to equation 3.23 in Rodgers (2000):~~

$$s(i) = 12 \cdot \Delta h \cdot \frac{\sum_j (i-j)^2 A_{ij}}{\left(\sum_j A_{ij}\right)^2}$$

The trace of  $\mathbf{A}$  equals the degrees of freedom of signal (DOFS), hence the total number of independent pieces of information gained from the measurements compared to the *a priori* knowledge. Figure 2 visualizes the average AVK matrices (median over participants and mean over time) for all five species studied in this work. Note that the AVKs do not necessarily represent the real/ total sensitivity and information content of MAX-DOAS observations as they only consider the gain of information with respect to the *a priori* knowledge. Hence, for stricter *a priori* constraints less gain in information will be indicated by the AVKs.

~~For all species~~ With the *a priori* profiles and covariances used within this study, the sensitivity is limited to about the lowest 1.5 km of the atmosphere for all species. More information is obtained on the Vis species, as the differential light path increases with wavelength resulting in higher sensitivity. The obtained DOFS are generally a bit lower as observed in former studies. This is related to the rather small *a priori* covariance (50%, see Sect. 2.1.3), which implies a good knowledge on the atmospheric state prior to the retrieval and finally leads to less gain in information from the measurements. Figures S35, S36, S37, S38 and S39 in Supplement S8.1 show the average AVKs of the individual participants and reveals, that there are significant differences (up to 1 DOFS) between the participants even when using the same algorithm (up to 0.5 DOFS in the case of PRIAM). This indicates that the information content is not assessed consistently. BOREAS for instance states a very low gain in information especially for Aerosol Vis. This is related to an additional Tikhonov term used as a smoother which was also applied during AVK assessment. Furthermore, all BOREAS results were retrieved on another grid and interpolated onto the submission grid, which leads to a decrease in all AVKs and therefore the DOFS. On average, the dependence of the total amount of information on the cloud conditions is small (typically decrease of 0.1 DOFS). Examination of the AVKs of individual profiles (not shown here), indicated that there are two competing effects: (1) the presence of clouds can increase the sensitivity to higher layers due to multiple scattering and thus light path enhancement in the clouds whereas (2) a decrease in the horizontal viewing distance (e.g. due to fog, rain or high aerosol loads) reduces the information content, since the light paths are shorter and their geometry depends less on the viewing elevation.



**Figure 2.** Mean AVKs for the retrieved species (median over participants, mean over time). Their meaning is described in detail in the text. Each altitude and corresponding AVK line  $A_i$  are associated with a colour, which is defined by the colour of the corresponding altitude-axis label. The dots mark the AVK diagonal elements. The number next to the dots show the exact value in percent, which corresponds to the amount of retrieved information on the respective layer. In each panel, the numbers indicate the DOFS (median among institutes, average over time) for clear-sky (green) and cloudy conditions (red). ~~The vertical bars indicate the vertical resolution (the "spread", defined according to Eq. ??) for the five lowest layers.~~

### 3.2 Overview plots

Figures 3 to 7 show the retrieved profiles of all participants over the whole semi-blind period. They serve as the basis for a general qualitative comparison. For the trace gases, the altitude ranges (full range is 4 km) were reduced to 0–2.5 km for better visibility, considering the MAX-DOAS sensitivity range and the occurrence altitude of the respective species.

**Table 2.** Groups who retrieved and provided profiling results for this study.

Algorithm	Method/Model	Literature	Participants	Acronym	Sym
bePRO	OEM <sup>o</sup> /LIDORT	Clémer et al. (2010)	Aristotle University of Thessaloniki, Thessaloniki, Greece	AUTH	●
		Hendrick et al. (2014)	Royal Belgian Institute for Space Aeronomy, Brussels, Belgium	BIRA	▼
PRIAM	OEM <sup>l</sup> /SCIATRAN	Wang et al. (2013b, 2013a, 2017)	National Institute of Aerospace Technology, Madrid, Spain	INTA	◆
			Anhui Institute Of Optics and Fine Mechanics, <del>Anhui</del> Hefei, China	AIOF	●
			Belarusian State University, Minsk, Belarus	BSU	▼
HEIPRO <sup>x</sup>	OEM <sup>l</sup> /SCIATRAN	Yilmaz (2012)	China Meteorological Administration, Beijing, China	CMA	■
			Max-Planck Institute for Chemistry, Mainz, Germany	MPIC	◆
			Institute of Environmental Physics, University of Heidelberg, Germany	IUPHD	●
BOREAS	OEM <sup>l</sup> /SCIATRAN	Bösch et al. (2018)	Department of Physics, University of Toronto, <del>Toronto</del> -Canada	UTOR	▼
		Chan et al. (2019, 2017)	Institute of Environmental Physics, University of Bremen, Germany	IUPB	●
M <sup>3</sup>	OEM/ LibRadTran		Ludwig-Maximilians-University, Munich, Germany	LMU	●
MMF	OEM <sup>l</sup> /VLIDORT	Friedrich et al. (2019)	Royal Belgian Institute for Space Aeronomy, Brussels, Belgium	BIRA	●
Realtime	ANA <sup>a</sup> /-		NASA-Goddard, Greenbelt, <del>Maryland</del> United States	NASA	●
MARK	PAR <sup>p</sup> /DAK	Vlemmix et al. (2011, 2015a)	Royal Netherlands Meteorological Institute, De Bilt, The Netherlands	KNMI	●
MAPA	PAR/McArtim	Beirle et al. (2019)	Max-Planck Institute for Chemistry, Mainz, Germany	MPIC <sup>y</sup>	●
			Max-Planck Institute for Chemistry, Mainz, Germany	MPIC <sup>y</sup>	▼

<sup>o</sup> OEM: Optimal estimation

<sup>a</sup> ANA: Analytical approach without radiative transfer model

<sup>p</sup> PAR: Parametrized approach

<sup>x</sup> IUPHD and UTOR used different versions of HEIPRO (1.2 and 1.5/1.4, respectively)

<sup>y</sup> Two versions of MAPA (labelled mp-10 and mp08) with different O<sub>4</sub> scaling factors (0.8 and 1.0) are included in the comparison.

<sup>l</sup> Aerosol extinction is retrieved in logarithmic space. This removes negative values and allows larger values.

**Table 3.** Prescribed settings for the radiative ~~transport~~-transfer simulation wavelengths and *a priori* total columns (OEM algorithms only).

Species	RTM wavelength [nm]	<i>A priori</i> VCD/ AOT
Aerosol UV	360	0.18
Aerosol Vis	477	0.18
NO <sub>2</sub> UV	360	$9 \cdot 10^{15}$ molec cm <sup>-2</sup>
NO <sub>2</sub> Vis	460	$9 \cdot 10^{15}$ molec cm <sup>-2</sup>
HCHO	343	$8 \cdot 10^{15}$ molec cm <sup>-2</sup>

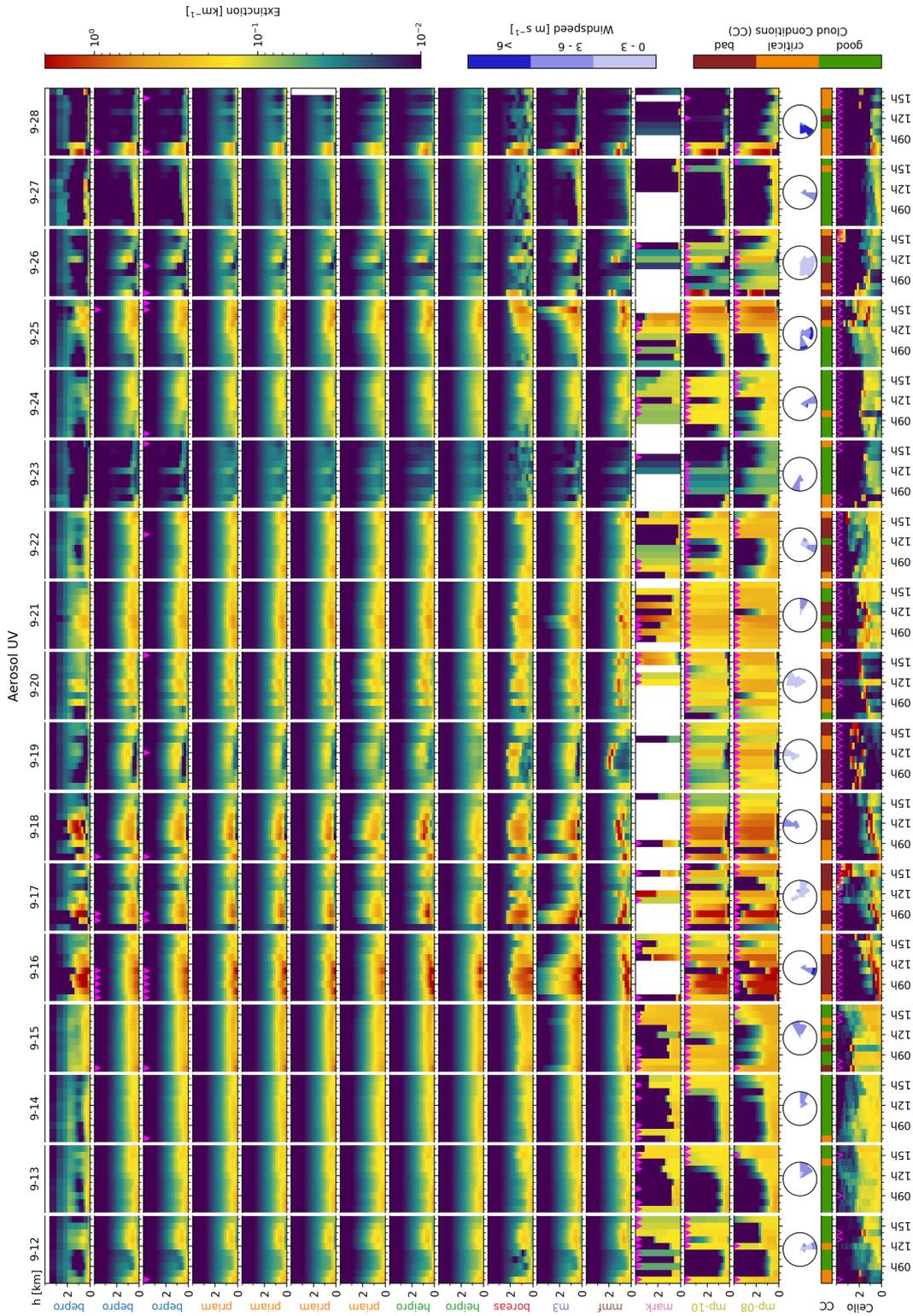
**Table 4.** Overview on compared quantities and available supporting data.

Species	Quantity	Supporting observations	Result section
Aerosol UV	Profiles	Ceilometer <sup>a</sup> (Sec. 2.2.2)	3.2 & Suppl. S8.2
	Aerosol optical thickness (AOT)	Sun photometer (Sec. 2.2.1)	3.4
Aerosol Vis	Profiles	Ceilometer <sup>a</sup>	3.2 & Suppl. S8.2
	Aerosol optical thickness (AOT)	Sun photometer	3.4
HCHO	Profiles	N.A.	3.2 & Suppl. S8.2
	Vertical column (VCD)	Direct-sun DOAS (Sec. 2.2.4)	3.5
	Surface concentration	Long-path DOAS	3.6
NO <sub>2</sub> UV/Vis	Profiles	NO <sub>2</sub> -Lidar & radiosonde <sup>b</sup>	3.2 & Suppl. S8.2
	Vertical column (VCD)	Direct-sun DOAS	3.5
	Surface concentration	Long-path DOAS	3.6
<del>UV vs. Vis retrieval N.A.<sup>c</sup>??</del> All species	Modelled vs. measured dSCDs	N.A. <sup>c</sup>	3.3

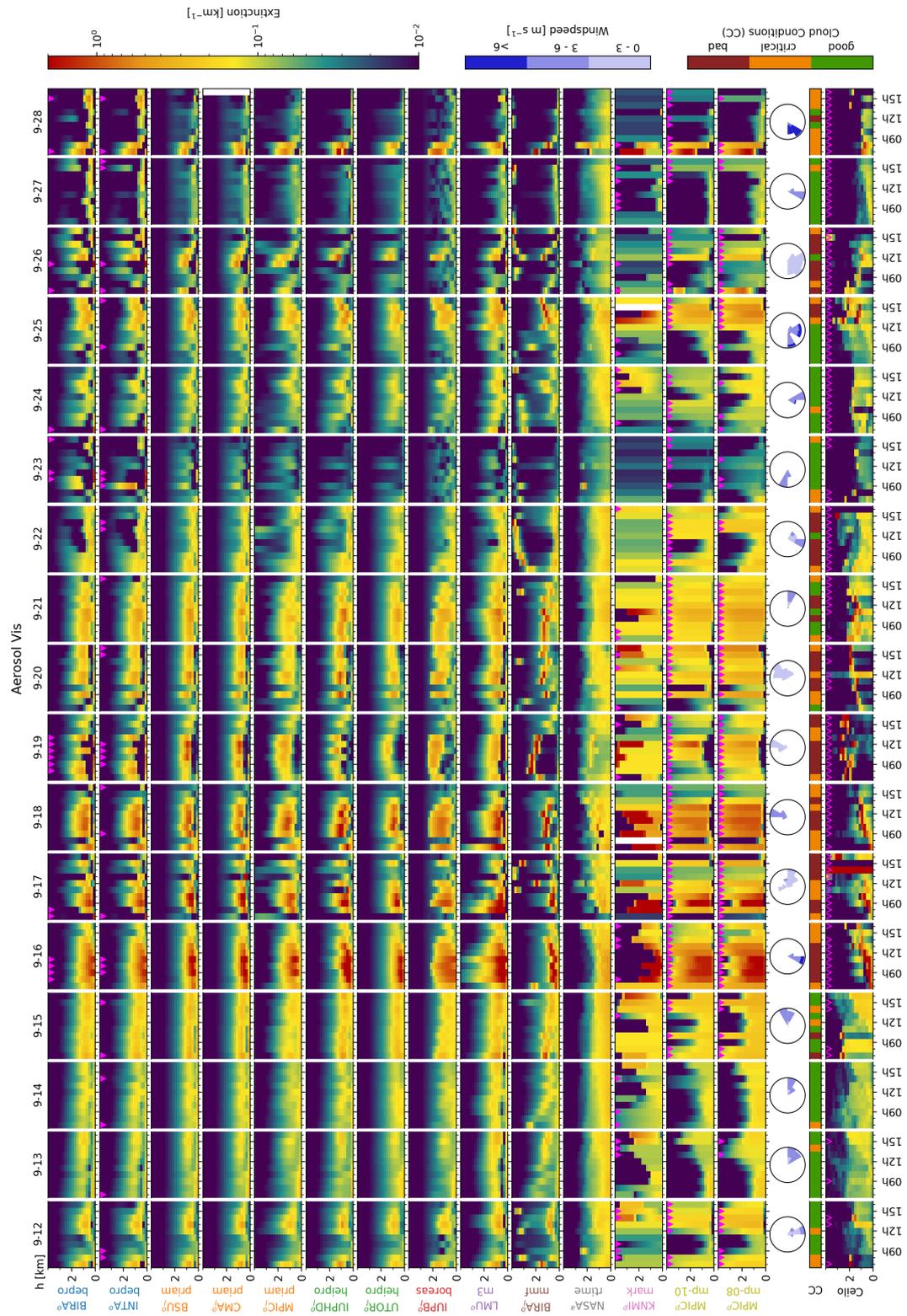
<sup>a</sup> Elastic backscatter profiles scaled with sun photometer or MAX-DOAS AOT.

<sup>b</sup> Scarce data coverage.

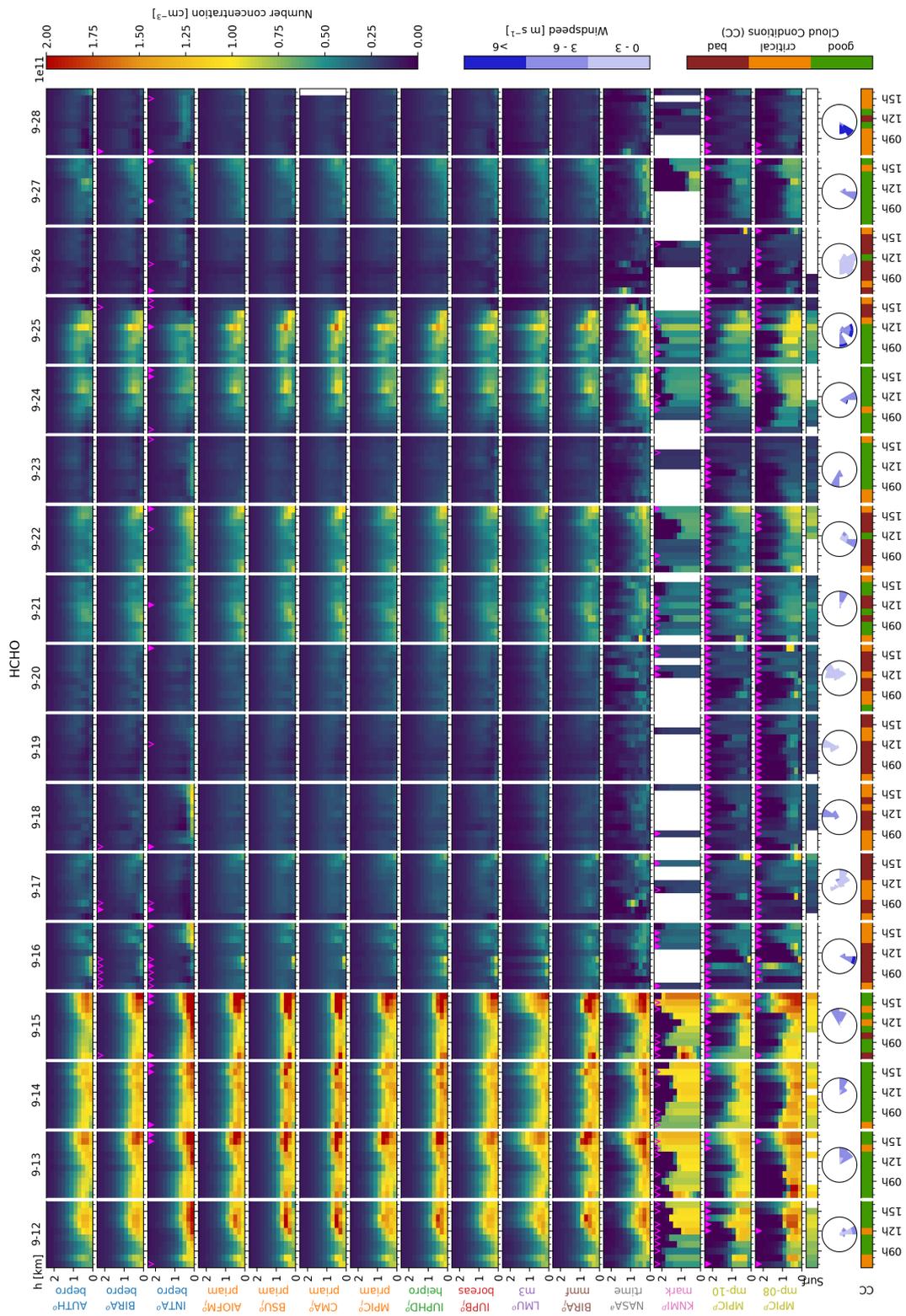
<sup>c</sup> Inherent quality proxy.



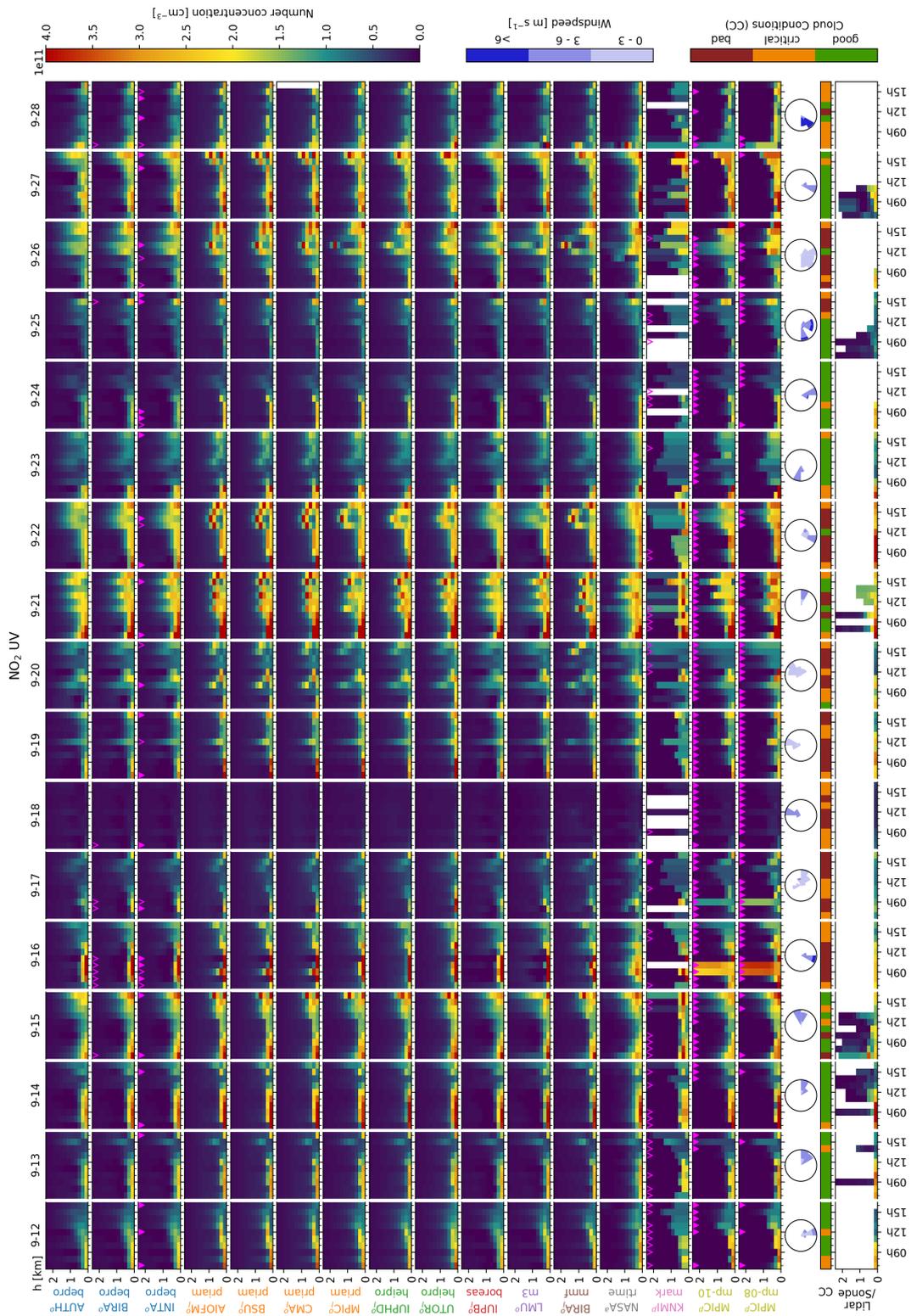
**Figure 3.** Aerosol UV extinction profiles. For MAX-DOAS profiles (plots above the wind roses), **red** pink triangles at the top of the corresponding profile indicate invalid data. The lowest row shows AOT scaled ceilometer backscatter profiles, calculated as described in Sect. 2.2.2 (unsmoothed). Backscatter profiles, which were scaled from MAX-DOAS AOTs (and which are therefore not fully independent) are marked by **red** pink triangles. Maximum extinction values reach  $20 \text{ km}^{-1}$ , exceeding the colour scale. Index letters behind the participant labels indicate whether an OEM (o) or parametrized (p) approach was used and whether aerosol was retrieved in the logarithmic space (l). The wind roses in the lower part of the panel show wind direction (azimuth), wind speed (see colour bar on the right) and occurrences (amplitude). The line close to the panel bottom marked with "CC" indicates the cloud conditions, as described in Sect. 2.2.6.



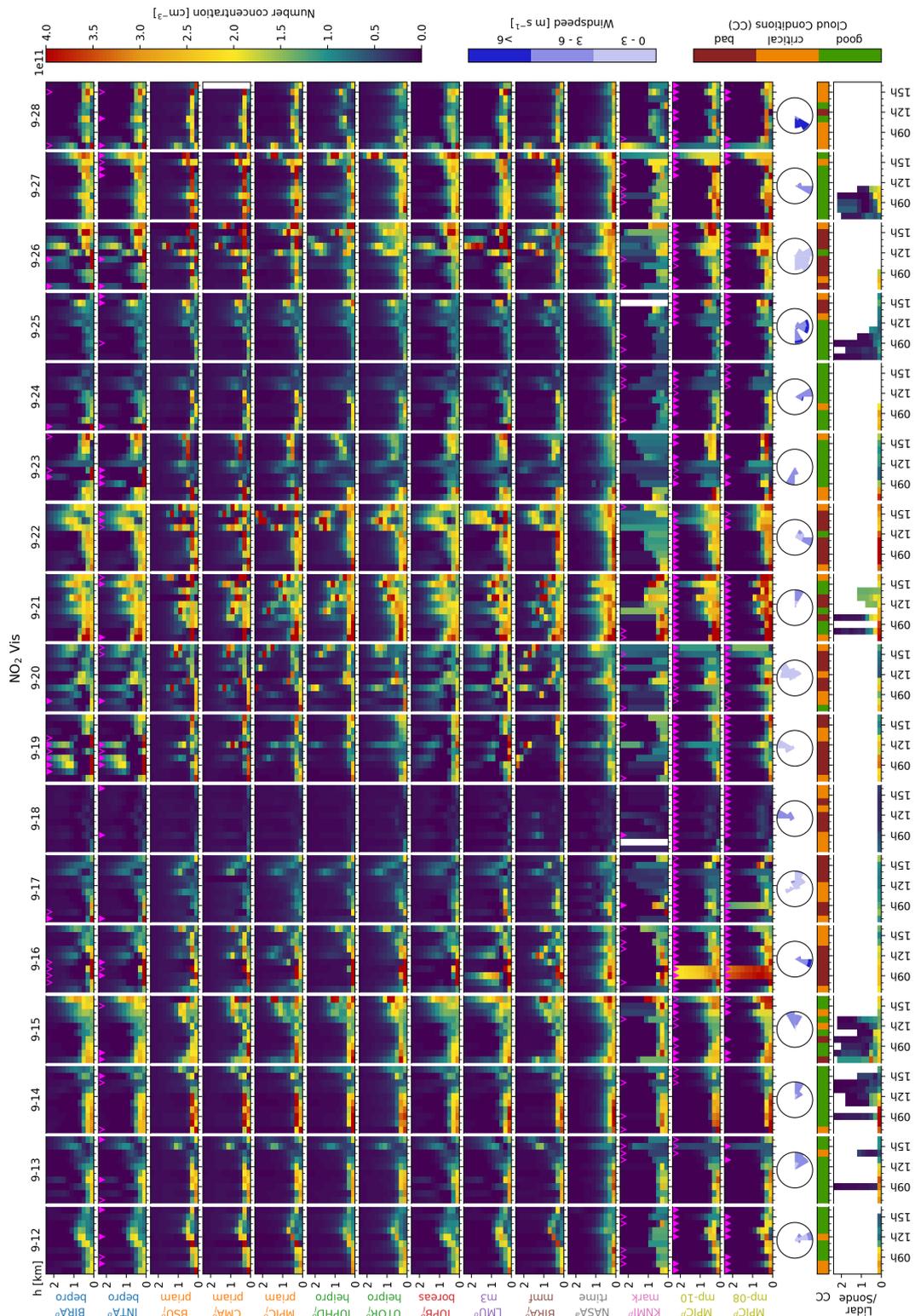
**Figure 4.** Aerosol Vis extinction profiles. Caption of Fig. 3 applies.



**Figure 5.** HCHO concentration profiles. The plot is similar to Fig. 3. Open red-pink triangles at the top of the MAX-DOAS profiles indicate that the underlying aerosol retrieval failed, whereas the trace gas profile retrieval itself was considered successful. The "Surf"-row shows LP-DOAS surface concentrations.



**Figure 6.** NO<sub>2</sub> UV concentration profiles. The lowest row shows a combined dataset of NO<sub>2</sub> lidar, radiosonde, LP-DOAS and tower in-situ data. Redundant surface concentration measurements were averaged.



**Figure 7.** NO<sub>2</sub> Vis concentration profiles. The lowest row shows a combined dataset of NO<sub>2</sub> lidar, radiosonde, LP-DOAS and tower in-situ data. Redundant measurements were averaged here.

Considering valid data only, all algorithms detect similar features in the vertical profiles, but smoothed to different amounts and sometimes detected at different altitudes. For clear sky condition, the observed ASDevs are  $3.5 \times 10^{-2} \text{ km}^{-1}$ ,  ~~$4.0 \times 10^{-2} \text{ km}^{-1}$~~ ,  ~~$1.2 \times 10^{10} \text{ molec cm}^{-3}$~~ ,  ~~$2.4 \times 10^{10} \text{ molec cm}^{-3}$~~  and  ~~$4.4 \times 10^{10} \text{ molec cm}^{-3}$~~  for Aerosol UV,  $4.0 \times 10^{-2} \text{ km}^{-1}$  for Aerosol Vis, ~~HCHO,  $1.2 \times 10^{10} \text{ molec cm}^{-3}$  for HCHO,  $2.4 \times 10^{10} \text{ molec cm}^{-3}$  for NO<sub>2</sub> UV and  $4.4 \times 10^{10} \text{ molec cm}^{-3}$  NO<sub>2</sub> Vis,~~ respectively. When regarding participants using the same algorithm, these values are reduced only by about 50%, indicating that significant discrepancies are caused by differences in the user defined retrieval settings that were not prescribed (e.g. number of iteration-, The latter are for instance the accuracy criteria for the RTMs, the number of iterations in the inversion, accuracy criteria for the RTMs, update interval of the jacobians, ...). Larger the convergence criteria or the decision at which points of the iteration process the forward model jacobians are (re-)calculated. An example are the discrepancies between UTOR/ HEIPRO and IUPHD/ HEIPRO. In this case the number of applied iteration steps in the aerosol inversion was identified as the main reason: UTOR and IUPHD used 5 and 20 iterations here, respectively. The consequences are evident throughout the comparison. Another example is the aerosol UV retrieval of AUTH/ bePro, where in contrast to other bePRO users oscillations seem to appear. We suspect this to originate from similar reasons, which could not yet been identified.

In general, larger discrepancies appear for the species measured in the Vis spectral range than in the UV. For NO<sub>2</sub> (aerosol) the ASDev increases in the Vis by 50 % (90 %). In the case of OEM algorithms, a reason might be that there is lower information content in the UV, meaning that the retrievals are drawn closer to the collectively used *a priori* profile. Further, the larger viewing distance of the Vis retrievals (see Supplement S5) might be problematic, since the exact treatment of the viewing geometries (~~Earth curvature, treatment of~~ like the Earth curvature or the treatment of the instrument field of view, ~~...~~) gain influence. ~~Horizontal inhomogeneities are an unlikely reason because~~ Note that the worse performance in the Vis was also apparent in the study by Frieß et al. (2019) with synthetic data, ~~where horizontal gradients were non-existent~~. The presence of clouds affects ASDevs very differently for different species: for Aerosol UV and Vis it is degraded by a factor of 3 and 4, respectively, which is expected since clouds mostly feature high optical depths > 1 and are detected to very different extent by the individual participants. For HCHO the ASDev decreases by 38% which can be well explained by the systematically lower (-36%) HCHO concentrations observed under cloudy conditions. ASDevs for NO<sub>2</sub> increase by about 20%, while the observed concentrations remain similar (increase < 10%).

Considering valid data only, the parametrized approaches are mostly in good agreement with the other algorithms. For MAPA, unrealistic results are reliably identified and flagged as invalid, whereas in the case of MARK some valid profiles do not look plausible e.g. for Aerosol Vis on 22 September 2016. For both algorithms a large fraction (30 to 70%) of the profiles are discarded as invalid or look unrealistic if the retrieval conditions are not ideal (see also flagging statistics in Sect. 4). Gaps in the MARK data appear where no optimum solution could be found at all.

For aerosol, OEM algorithms often see elevated layers in the Vis even in clear-sky scenarios that cannot be observed in the UV or the ceilometer profiles. On cloudy days, MMF is capable of detecting clouds as very defined features with a good qualitative agreement with the ceilometer data. In the Vis, even high clouds are detected, e.g. on 17 September and 22 September 2016, which indeed coincide with high-altitude clouds above the retrieval altitude range of 4 km. ~~An example for large discrepancies between participants using the same algorithm is AUTH aerosol in the UV, where in contrast to other bePRO~~

~~users oscillations seem to appear. We suspect this to originate from technical problems which could not yet been identified. The discrepancies between IUPHD and UTOR (both using HEIPRO) were found to mainly be caused by differences in the number of applied iteration steps in the Levenberg-Marquardt optimization scheme during aerosol retrieval. IUPHD (UTOR) applied 20 (5) iterations. The consequences are evident throughout the comparison. Compared to the parametrized approaches,~~

5 ~~ØEMs and the Realtime algorithm~~ In contrast to the PAR approaches, OEM and Realtime algorithms yield realistic profiles also under less favourable measurement conditions (e.g. clouds); in particular the OEM results are in qualitative agreement with the ceilometer profiles for many cases.

Regarding HCHO, the agreement of the profiles is exceptionally good considering the particularly low information content of the measurements (due to higher uncertainties in the dSCD data). Probably because observed spatial and temporal concentration  
10 gradients are much smaller than for NO<sub>2</sub>, which might partly be related to enhanced smoothing by the retrieval, but is also well possible to be real, since HCHO sources (mainly the photolysis of volatile organic compounds) are less localized. High HCHO concentrations coincide with clear-sky conditions and with wind from the continent, which is what would be expected from the current knowledge on the origin and chemistry of atmospheric HCHO. As in the case of aerosol, there are significant discrepancies among the bePRO participants, this time with INTA standing out of the group with slight overestimation.

15 For NO<sub>2</sub> very shallow layers and large vertical and horizontal gradients might complicate the retrievals. Nevertheless, good ASDev is achieved in the UV. Week-days and weekends (17, 18, 24 and 25 September) can clearly be distinguished. The lowest concentrations are observed on 18 September, where a Sunday coincides with northerly winds from the sea.

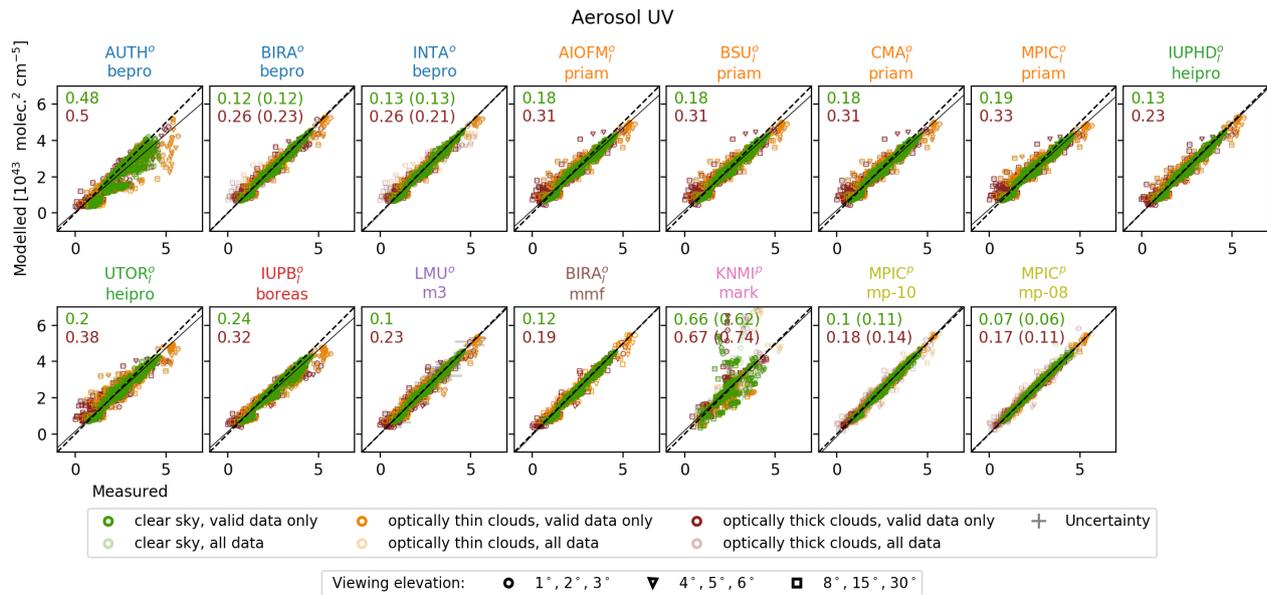
The agreement with the supporting observations will be discussed in detail in the following sections.

### 3.3 Modelled and measured dSCDs

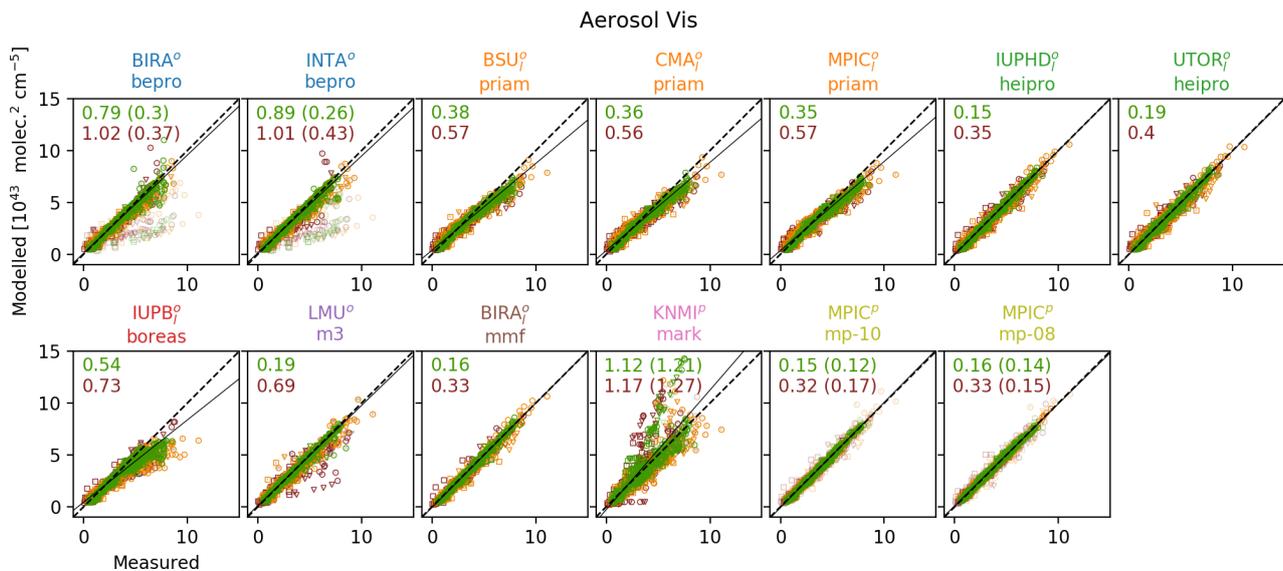
20 An intrinsic indicator for a successful profile retrieval is a good agreement between the measured and the modelled dSCDs, the latter being the dSCDs obtained from the RTM model for the finally retrieved aerosol and trace gas profiles. Poor agreement might indicate that only a local minimum of the cost function was found (OEM approaches), that inappropriate retrieval settings were chosen (e.g. too small number of iterations in the minimisation) or that the RTM is inaccurate for other reasons, for instance because it cannot describe horizontal inhomogeneities. Figures 8 to 12 show the correlation of measured and  
25 modelled dSCDs for all profiles and elevations of each participant. The NASA/ Realtime algorithm is not included since it does not use an RTM and therefore does not provide simulated dSCDs.

For clear-sky conditions, good agreement is achieved by most participants. Only IUPB, ~~AUTH, BSU, KNMI/~~ BOREAS, AUTH/ bePRO, BSU/ PRIAM, and KNMI/ MARK exceed relative RMSDs of 10% and only for O<sub>4</sub> and NO<sub>2</sub> Vis dSCDs. MMF achieves the best overall performance, being the only algorithm with relative RMSDs < 5% for all species. Regarding  
30 HEIPRO, UTOR yields larger RMSD values than IUPHD, which is very likely related to the aforementioned smaller number of iterations applied by UTOR. For the trace gases, small relative RMSD values between 8% and 8% are achieved for all cloud conditions.

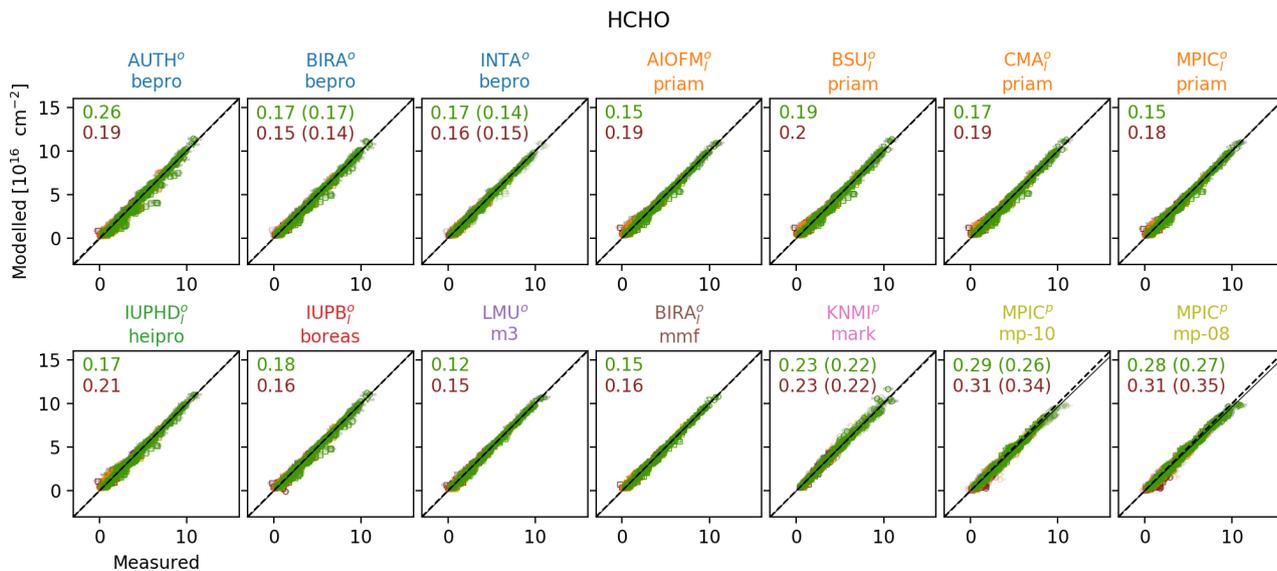
Regarding aerosol, PRIAM and BOREAS feature slightly too low slopes in the UV (approx. 0.9) and more pronounced in the Vis (0.8 to 0.85) interestingly almost exclusively caused by data recorded on the 23 and 27 September where the atmospheric



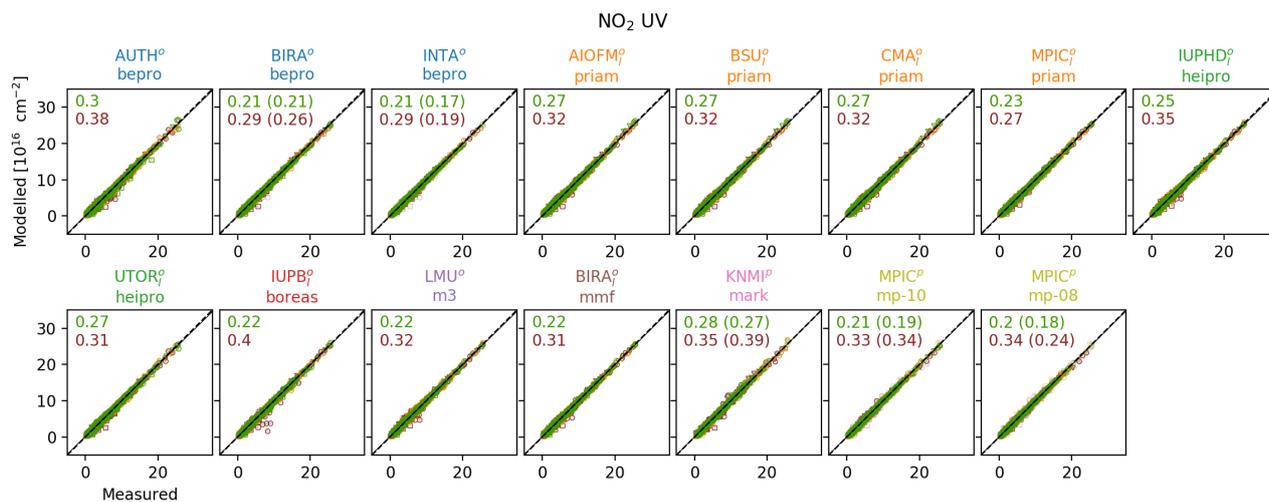
**Figure 8.**  $O_4$  UV dSCD correlation. Marker colours and marker shapes indicate the cloud conditions and viewing elevation angles, respectively, as indicated in the legend. Numbers represent the measurement-error-weighted RMSD between measured and modelled dSCDs in units of  $10^{43} \text{ molec}^2 \text{ cm}^{-5}$  for clear sky (green) and cloudy (red) conditions. Values in brackets were calculated only considering valid data.



**Figure 9.**  $O_4$  Vis dSCD correlation. Legends and description of Fig. 8 apply.

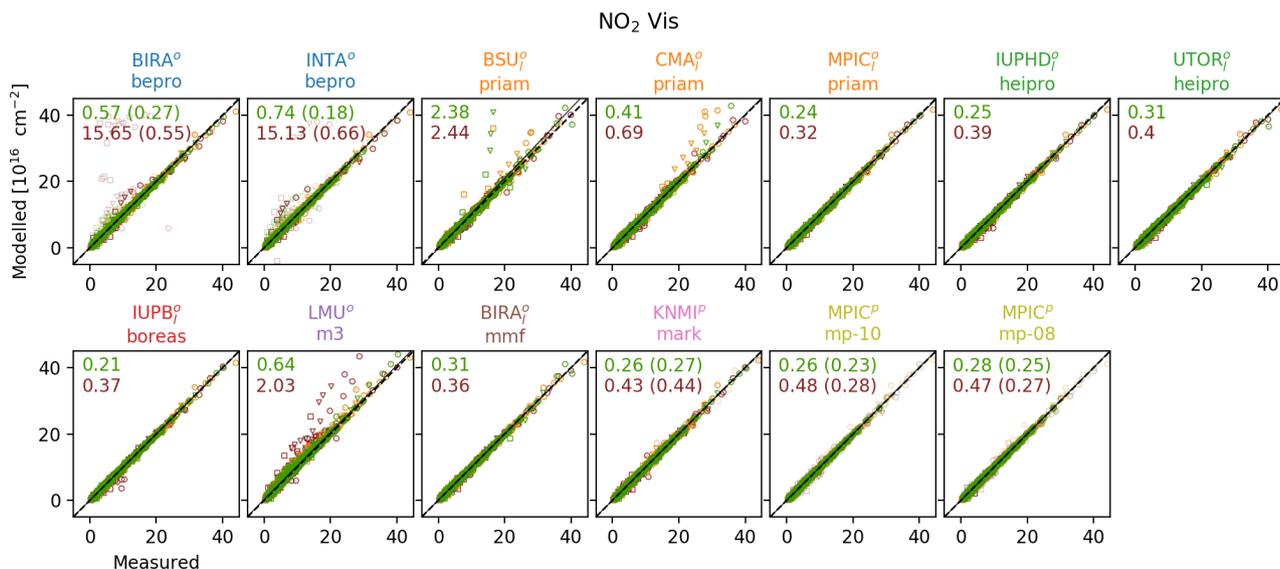


**Figure 10.** HCHO dSCD correlation. RMSD between measured and modelled dSCDs in units of  $10^{16}$  molec  $\text{cm}^{-2}$ . Legends and description of Fig. 8 apply.



**Figure 11.** NO<sub>2</sub> UV dSCD correlation. RMSD between measured and modelled dSCDs in units of  $10^{16}$  molec  $\text{cm}^{-2}$ . Legends and description of Fig. 8 apply.

aerosol load is particularly low. RMSDs increase for cloudy scenarios by 10% (HCHO), 30% (NO<sub>2</sub> UV) and 50% (NO<sub>2</sub> Vis, O<sub>4</sub>), most likely because the horizontal inhomogeneity cannot be adequately reproduced by the 1D models. This is supported by the comparison results from synthetic data by Frieß et al. (2019), where horizontal homogeneity is inherently assured and



**Figure 12.** NO<sub>2</sub> Vis dSCD correlation. RMSD between measured and modelled dSCDs in units of 10<sup>16</sup> molec cm<sup>-2</sup>. Legends and description of Fig. 8 apply.

the scatter remains similar for all cloud scenarios. KNMI/ MARK has problems to reproduce O<sub>4</sub> dSCDs (relative RMSD > 30%), while for trace gases the performance is comparable to the other algorithms. Regarding Vis species, M<sup>3</sup> shows outliers under cloudy conditions (while performing excellently in the UV) and bePRO seems to have convergence problems, which was also evident in the synthetic data (Frieß et al., 2019). This problem is overcome by flagging of approx. 10% of the data, reducing the RMSD by > 50%. PRIAM (except MPIC) shows outliers, in particular for NO<sub>2</sub> Vis. The O<sub>4</sub> scaling factor of 0.8 for MAPA improves O<sub>4</sub> dSCD agreement in the UV by about 35% (for clear sky and valid data), but not in the Vis spectral range (see also Supplement S2).

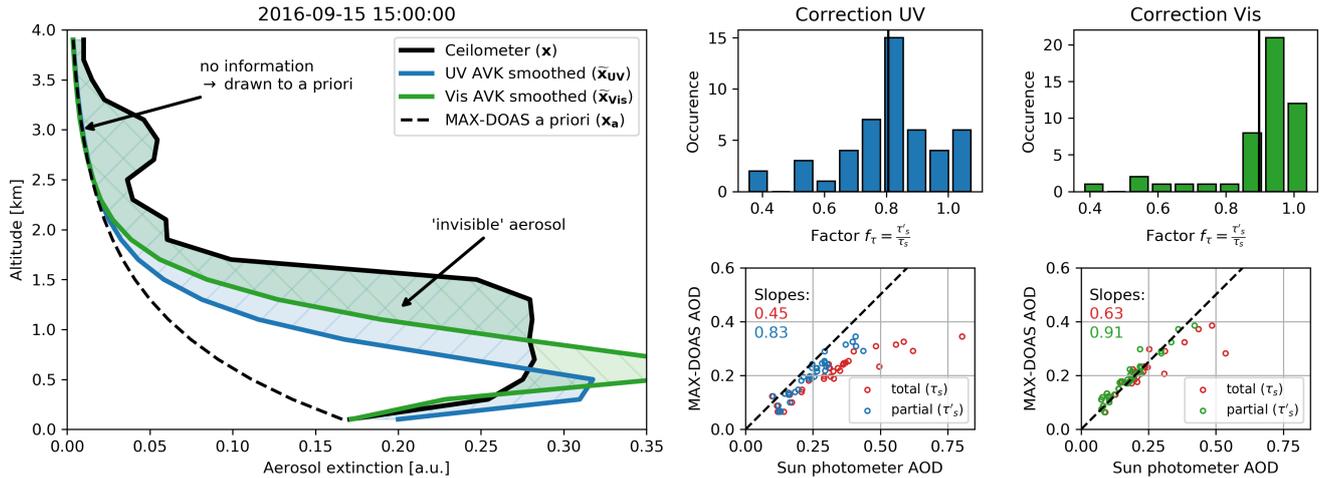
### 3.4 Aerosol optical thickness (AOT)

This section compares vertically integrated MAX-DOAS aerosol extinction profiles with the AOTs observed by the nearby sun photometer. ~~As discussed in Sect. 2.3.2 these two quantities are not necessarily comparable. As shown in Sect. 3.1 the~~ In former publications (e.g. Irie et al., 2008; Clémer et al., 2010; Frieß et al., 2016; Bösch et al., 2018) and also during this comparison study, it was found that MAX-DOAS vertically integrated aerosol profiles systematically underestimate AOTs. It has already been proposed by Irie et al. (2008), Frieß et al. (2016) and Bösch et al. (2018) but not proven that this is related to smoothing effects, namely the reduced sensitivity of MAX-DOAS observations decreases rapidly with altitude to higher altitudes and associated a priori assumptions. Even though the sensitivity to elevated layers was observed to be increased by the presence of optically thick aerosol layers at the ~~corresponding~~ corresponding altitudes (Frieß et al., 2006 and Sect. 3.1 of this study), high-altitude abundances of trace gases and aerosol typically cannot be reliably ~~detected~~ located and quantified by ground-based

MAX-DOAS observations. ~~Thus they can only~~ Integrated profiles rather provide "partial AOTs" which basically only consider low-altitude aerosol and which are additionally biased by *a priori* assumptions on the aerosol extinctions at higher altitudes (for OEM algorithms defined by the *a priori* profile and covariance, for PAR algorithms partly in the form of prescribed profile shapes). Therefore, a comparison between MAX-DOAS ~~vertically-integrated extinction profiles~~ and sun photometer AOTs  $\tau_s$  is not necessarily meaningful. However, for OEM approaches, information on the true aerosol extinction profile  $x$  (which are available from the ceilometer as described in Sect. 2.2.2) and the AVKs  $\mathbf{A}$  can be used to account for this effect: inserting  $x$  and  $\mathbf{A}$  into Eq. (9) yields a smoothed profile  $\tilde{x}$  that can be used to estimate which fraction  $f_\tau$  of the aerosol column is expected to be detected by the OEM retrievals:

$$f_\tau = \frac{\tau'_s}{\tau_s} = \frac{\sum_i \tilde{x}_i}{\sum_j x_j} \quad (10)$$

with  $\tau'_s$  being the actually detectable "partial AOT". ~~Average values over the whole campaign~~ The left panel of Fig. 13 shows an example of an extreme case during the campaign from September 15<sup>th</sup>, 15:00h. Shown are a ceilometer backscatter profile ( $x$ , black) and the same profile smoothed by the MAX-DOAS median OEM averaging kernels for  $f_\tau$  are  $0.81 \pm 0.16$  for Aerosol UV and  $0.90 \pm 0.13$  for Aerosol Vis (using the median AVKs of all OEM retrievals).  $x_{UV}$  and  $x_{Vis}$ , blue and green, respectively. In this particular case it is expected that a large fraction of the aerosol above 1 km altitude will hardly be detected by the MAX-DOAS instruments, resulting in factors  $f_\tau = \frac{\tau'_s}{\tau_s}$  of 0.67 and 0.78, for the UV and the Vis AOT, respectively. Note, however, that corresponding information actually seems to be present in the measurements, since part of the high-altitude aerosol appears to be shifted to lower altitudes which are accessible within the constraints of the *a priori* covariance. Multiplying the AOT observed by the sun photometer with  $f_\tau$  significantly improves the agreement between MAX-DOAS and sun photometer observations in particular in the UV (see Supplement ?? for details). In the following, this correction is referred to as "partial AOT correction" (PAC). The right panels in Fig. 13 show information on  $f_\tau$  and the improvement in the UV and Vis results (2nd and 3rd columns of the figure) over the whole campaign. Average values are  $f_\tau = 0.81 \pm 0.16$  in the UV and  $(0.9 \pm 0.13)$  in the Vis (using the median AVKs of all OEM retrievals). It shall be pointed out that for OEM algorithms the necessity for the PAC can generally be reduced by using improved *a priori* profiles and covariances (e.g. from climatologies, supporting observations and/ or model data). Also the values for  $f_\tau$  will differ, when other *a priori* profiles and covariances than the ones prescribed for this study (see Sect. 2.1.3) are used. Parametrized and analytical approaches typically do not quantify the sensitivity, the effective resolution or the amount of assimilated *a priori* knowledge. For these algorithms, the correction could not be performed and the total sun photometer AOT  $\tau_s$  had to be used for the comparison in this section. However, the comparison results in this section and further investigations in Supplement S2 indicate that a scaling of the measured  $O_4$  dSCDs prior to the retrieval with  $SF \approx f_\tau$  might be used to at least partly account for the PAC for MAPA and probably other PAR and ANA algorithms (see Supplement S2), even though the ~~physical reason for PAC and SF are different~~ motivation for the application of the PAC and the SF are different: the application of the PAC is necessary solely for mathematical reasons related to the concept of OEM and prior constraints applied therein. In contrast, publications that suggest or discuss the application of an SF (e.g. Wagner et al., 2009; Cl mer et al., 2010; Ortega et al., 2016; Wagner et al., 2019) directly compare forward modelled  $O_4$  dSCDs (using an atmosphere derived from supporting observations to reproduce the real conditions to

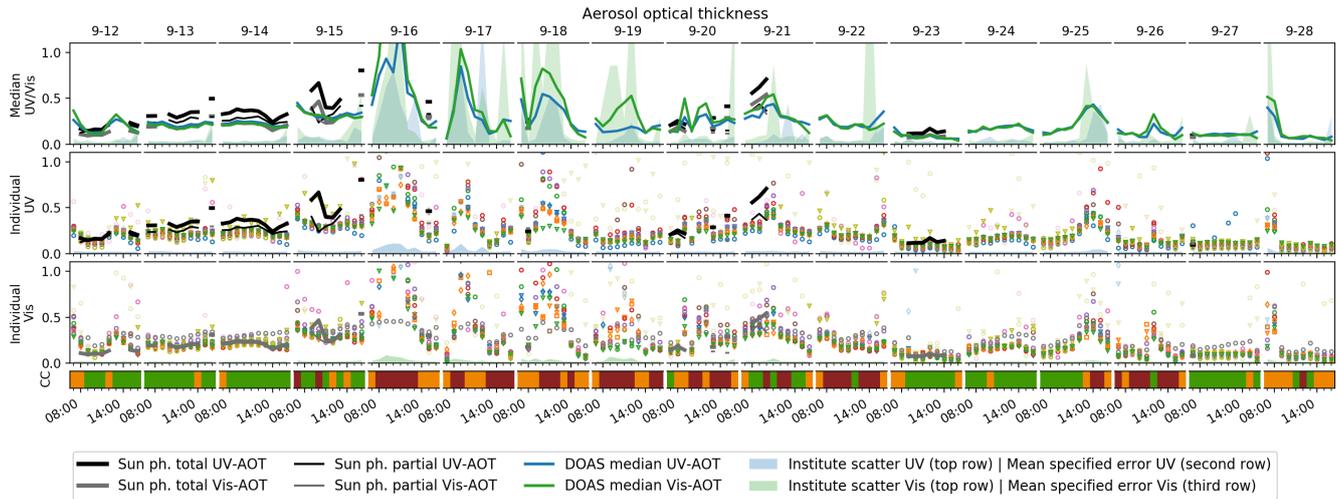


**Figure 13.** Left panel: example for the smoothing of a ceilometer backscatter profile  $x$  (according to Eq. (9)) with particularly heavy aerosol load at high-altitudes retrieved in the UV and Vis, respectively. Right panel: distribution and impact of the correction factor  $f_{\tau} = \tau'_s/\tau_s$  for the UV and the Vis retrieval. Top plots show the distributions of  $f_{\tau}$  with the solid lines indicating the mean values. At the bottom the correlation plots between sun photometer and MAX-DOAS median AOTs are shown. Red circles represent sun photometer total AOTs, other dots represent the partial AOT  $\tau'_s = f_{\tau} \cdot \tau_s$ .

best knowledge) to measured  $O_4$  dSCDs. They do not make use of optimal estimation or prior constraints similar to those used in our study. Thus their findings can be considered independent from any kind of PAC.

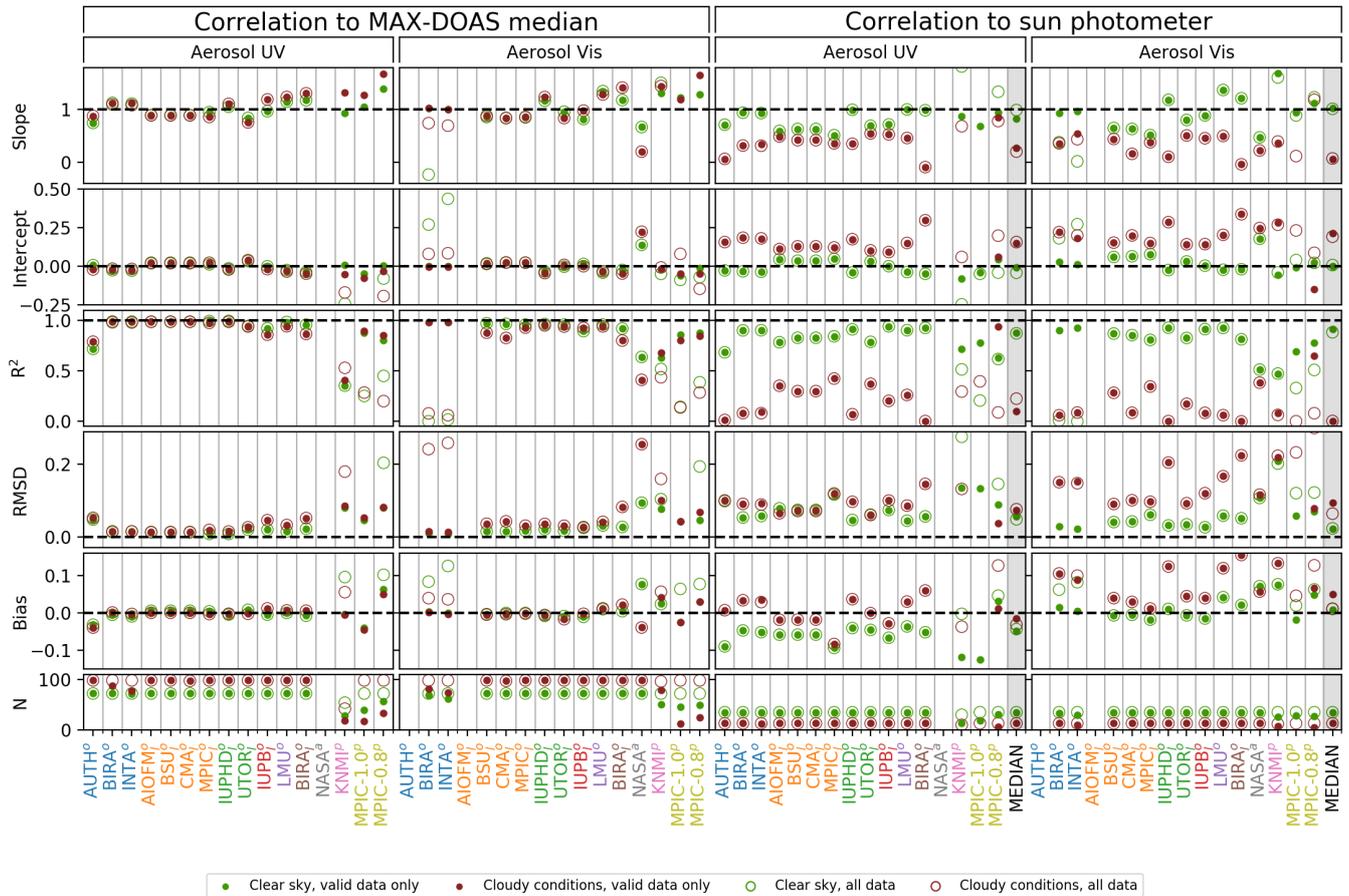
Figure 14 shows time series of the MAX-DOAS retrieved AOTs in comparison to their median and the sun photometer data. For the sun photometer, both the total AOT  $\tau_s$  and the partial AOT  $\tau'_s$  are shown. For the calculation of  $\tau'_s$  in Fig. 14, the median AVKs of all OEM participants were used for the smoothing according to Eq. (9). In the correlation analysis (Fig. 15), AVKs of the individual participants and the individual profiles were applied. Keep in mind that the non-OEM approaches (NASA, ~~KNMI / Realtime, KNMI/ MARK~~ and MPIC/ MAPA) are correlated against  $\tau_s$  and ~~might therefore be underprivileged~~ are therefore expected to generally achieve worse agreement. For correlations of OEM algorithms against  $\tau_s$  please refer to Supplement S8.3. Correlation parameters ~~and RMSD~~, RMSD and Bias values were derived as described in Sect. 2.3.

Under clear sky conditions, average RMSD values against the MAX-DOAS median are 0.028 (in the UV and 0.032 ) ~~for Aerosol UV (Vis) in the Vis.~~ In the presence of clouds they increase by about 30% (and 80%), ~~respectively,~~ which is to a large part caused by mainly due to the periods of particularly large scatter between 16 and 19 September 2016. As already shown in Sect. 3.2, different algorithms detect clouds to very different extent. Especially in the presence of optically thick clouds (AOT > 10), this easily induces discrepancies of several orders of magnitudes. The observed average RMSDs are similar to the specified uncertainties (average is 0.025) that are derived from propagated measurement noise and smoothing effects. Keeping in mind that the retrievals were performed on a common dSCD dataset, this indicates that the choice of the retrieval algorithm and the remaining free settings have severe impact on the results.



**Figure 14.** MAX-DOAS retrieved AOTs in comparison to sun photometer data. Symbol and symbol colours are chosen according to Table 2. Open-Transparent symbols indicate data flagged as invalid. Top row: MAX-DOAS median results vs. the available supporting observations, according to the legend below the plot. The "institute scatter" hatched-areas (sharing the AOT's y-axis scaling but starting at the top of the plot) show the scattering among the participants in terms of standard deviation with valid data considered only. Two lower rows: Comparison of the individual participants for the two spectral retrieval ranges. Here the coloured area is the average retrieval error, as specified by the participants.

For the comparison to the sun photometer, it shall be noted that the PAC induces further uncertainties, as it incorporates the extinction profiles derived from the ceilometer and the algorithms' AVKs, both being error-prone. Further, the comparison to sun photometer data under cloudy conditions might not be very meaningful as (1) there are only 13 measurements available in the presence of clouds and (2) as it is very likely that these measurements were made by looking through very local cloud holes, such that they will not be representative for the MAX-DOAS retrieved AOTs with a typical horizontal sensitivity range of several kilometres (see Supplement S5). The following discussion of the sun photometer comparison therefore refers to clear-sky conditions and valid data only. In general, there is reasonable agreement of the MAX-DOAS retrieved AOT with the sun photometer, with average observed RMSDs of 0.08 (0.06) for Aerosol UV (Vis). Good performance Best performance in the UV is observed for bePRO (except AUTH), IUPHD/ HEIPRO and LMU/ M<sup>3</sup> with RMSDs around 0.05, in the Vis it is the participants using the bePRO (BIRA and INTA), the HEIPRO (IUPHD), M<sup>3</sup> and MMF with RMSDs around 0.05 (0.03). For other OEM algorithms, larger underestimations of the partial AOT ( $0.5 < \text{slope} < 1$ ) are observed and UTOR) and the BOREAS (IUPB) algorithm. For all participants except MPIC-0.8/ MAPA, negative Biases  $< -0.03$  in the UV, which are most evident in the case of PRIAM ( $\approx 0.5$ ). Interestingly, the AVKs at higher layers derived from PRIAM are systematically higher than most other algorithms (see Sect. S8.1), which reduces the impact of the PAC and results in a larger partial AOD  $\tau_s^l$  than for most other datasets. Therefore, the lower slopes of PRIAM might rather be owed to its assessment of information content than to the retrieval algorithm itself. For Aerosol-Vis bePRO remain, even though the PAC has been applied for the OEM algorithms. The



**Figure 15.** Correlation statistics for AOTs. The two left columns give an impression on the agreement among the institutes, as they show the correlation of the individual participant’s retrieved AOT (ordinate of the underlying correlation plot) against the median (abscissa). The two right columns show the correlation against the sun photometer AOT (partial AOT in the case of OEM retrievals) instead of the median. Green and red symbols represent cloud-free and cloudy conditions, respectively. Light symbols-Hollow circles represent values for all submitted data, opaque symbols-the dots only consider data points flagged as valid. The pies indicate, which fraction of  $N$  is the total number of profiles (170) which contributed to the respective valuedata points above. The total number of submitted profiles per participant and species were 170. On the right also the correlation between the MAX-DOAS median results and supporting observations are included (grey shaded columns). The correlation plots are shown in Supplement S8.3.

average Bias in the UV is  $-0.06$ , indicating that the systematic underestimation dominates over random deviations here. Note that the slopes and intercepts vary significantly among the participants, however, in an anti-correlated manner, finally resulting into similar Bias values.

The average Bias in the Vis is only 0.02. Bias magnitudes are much smaller than RMSDs for many participants here, indicating that in these cases Vis AOTs mainly suffer from random discrepancies. BePRO suffers the aforementioned conver-

gence problems during inversion in the Vis (see Sect. 3.3) but the affected results are reliably flagged. KNMI/ MARKand-  
NASA/ Realtime and MPIC-1.0/ MAPA feature the highest RMSDs around 0.1 and strongest Biases below -0.1 in the UV. A  
particular case is KNMI/ Aerosol Vis with  $\text{RMSD} > 0.2$ , with and without flagging being applied.

As described in Supplement S2, the PAC and the application of  $SF \approx f_\tau$  an  $\text{O}_4$  dSCD scaling factor of  $SF \approx f_\tau$  have very  
5 similar impact on the AOT correlation. Consequently, the application of an  $\text{O}_4$  dSCD scaling factor of  $SF = 0.8$  in the case of  
MPIC-0.8/ MAPA significantly improves the agreement to the sun photometer total AOT in the UV ( $f_\tau \approx 0.8$ ) whereas in the  
Vis ( $f_\tau \approx 0.9$ ) it leads to an overcompensation with slope  $> 1$  and intercept  $> 0$ . a Bias of about 0.05.

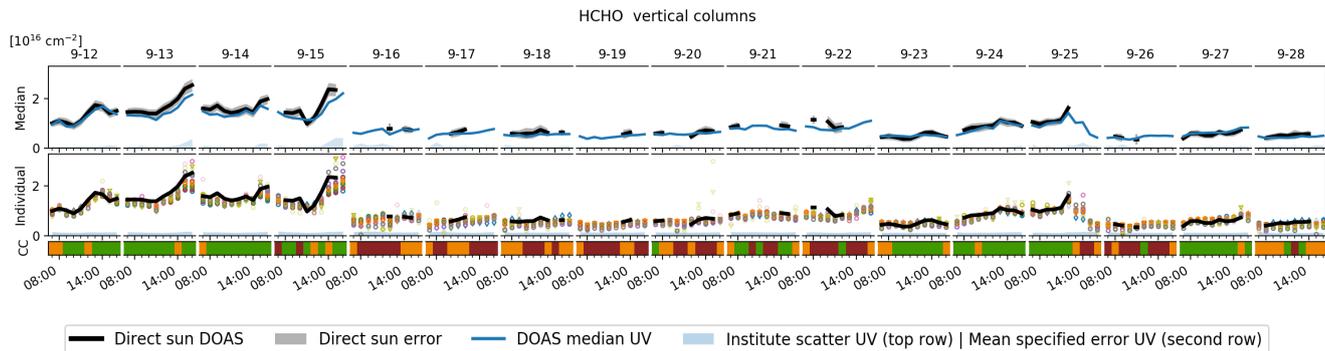
### 3.5 Trace gas vertical column densities

This section compares the VCDs of assesses the consistency of the VCDs for each of the trace gases HCHO and  $\text{NO}_2$ . Independ-  
10 dent observations of VCDs are the direct-sun DOAS observations ( $\text{NO}_2$  and HCHO), but also integrated columns of radiosonde  
and lidar profiles ( $\text{NO}_2$  only). Time series comparisons of all observations are shown in Fig. 16 and 17. For the statistical  
evaluation in Fig. 18, from the supporting observations only direct-sun observations were considered, as they provide the most  
complete dataset.

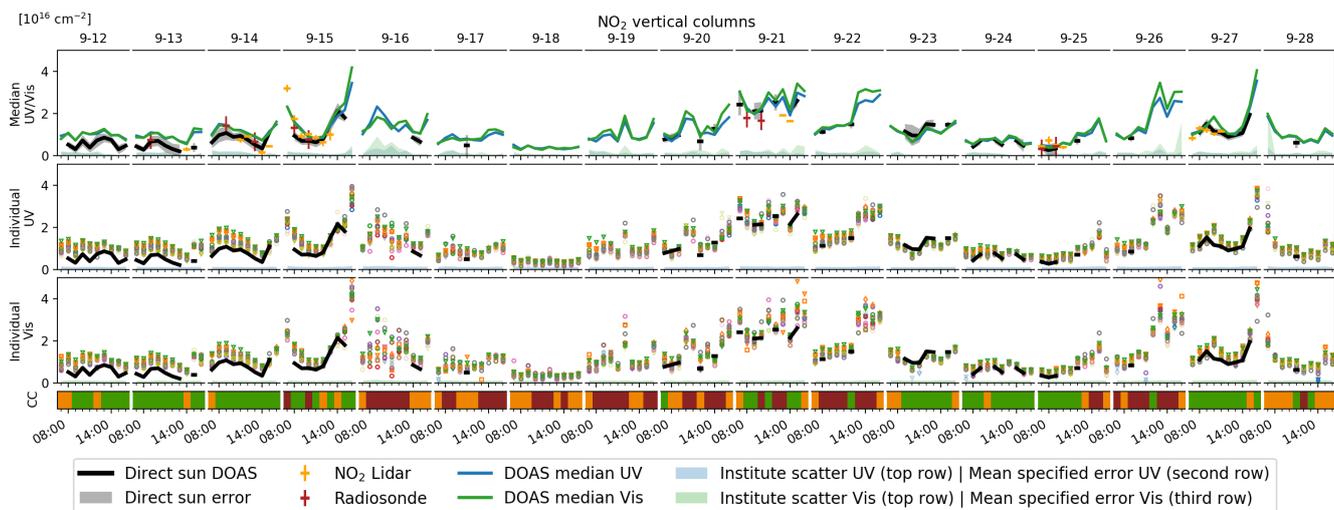
As for AOTs, smoothing effects (in particular the low sensitivity of MAX-DOAS observations for higher altitudes) poten-  
15 tially affects the comparability of MAX-DOAS and direct-sun observations. In contrast to aerosol, only scarce ( $\text{NO}_2$ ) or no  
(HCHO) information on the true profile is available and a correction similar to the PAC cannot be performed. However for  
 $\text{NO}_2$  the available radiosonde profiles could be used for an impact estimate. Ignoring an outlier one problematic radiosonde  
profile on 09-27 07:00:00 -(where  $\text{NO}_2$  concentration was close to the radiosonde detection limit and thus instrumental offsets  
became particularly apparent), correction factors of  $1.06 \pm 0.05$  and  $1.03 \pm 0.03$  in the UV and  $1.03 \pm 0.03$  in the Vis are  
20 obtained, respectively, indicating that the MAX-DOAS retrieved tropospheric  $\text{NO}_2$  VCD is affected by smoothing effects to  
only a few percent. This is expected since  $\text{NO}_2$  mostly appears close to the ground. Also in Fig. 6 and 7,  $\text{NO}_2$  appears to be  
confined to the lowermost retrieval layers with concentrations dropping to around zero already at altitudes where MAX-DOAS  
sensitivity is still significant. Profiles from the  $\text{NO}_2$  lidar were not used in this investigation as they often suffer from artefacts at  
higher altitudes. Regarding HCHO, the MAX-DOAS profiling results on some days show large concentrations over the whole  
25 altitude range where the information content of the measurements is significant (compare Fig. 2 and 5), indicating that there  
might be "invisible" HCHO at even higher altitudes. This is supported by Fig. 16, where MAX-DOAS observations tend to  
yield smaller VCDs than the direct-sun observations in particular in scenarios with high HCHO abundance.

Under clear sky conditions, average RMSD values against the MAX-DOAS median are  $5 \times 10^{14}$  molec  $\text{cm}^{-2}$  for HCHO and  
 $7 \times 10^{14}$  molec  $\text{cm}^{-2}$  for HCHO (for  $\text{NO}_2$  , (both UV and Vis). In contrast to AOTs, these values do not increase significantly  
30 ( $< 15\%$ ) in the presence of clouds. For HCHO it is even reduced by 25% for the same reasons as discussed already in Section  
3.2. Bias values are approximately of half the magnitude of RMSDs for all trace gases.

For HCHO, the comparison against the direct-sun DOAS observations yields an average RMSD of  $1.4 \times 10^{15}$  molec  $\text{cm}^{-2}$ .  
Note however that the two observations are not fully independent, as for the direct-sun data, the residual HCHO amount in



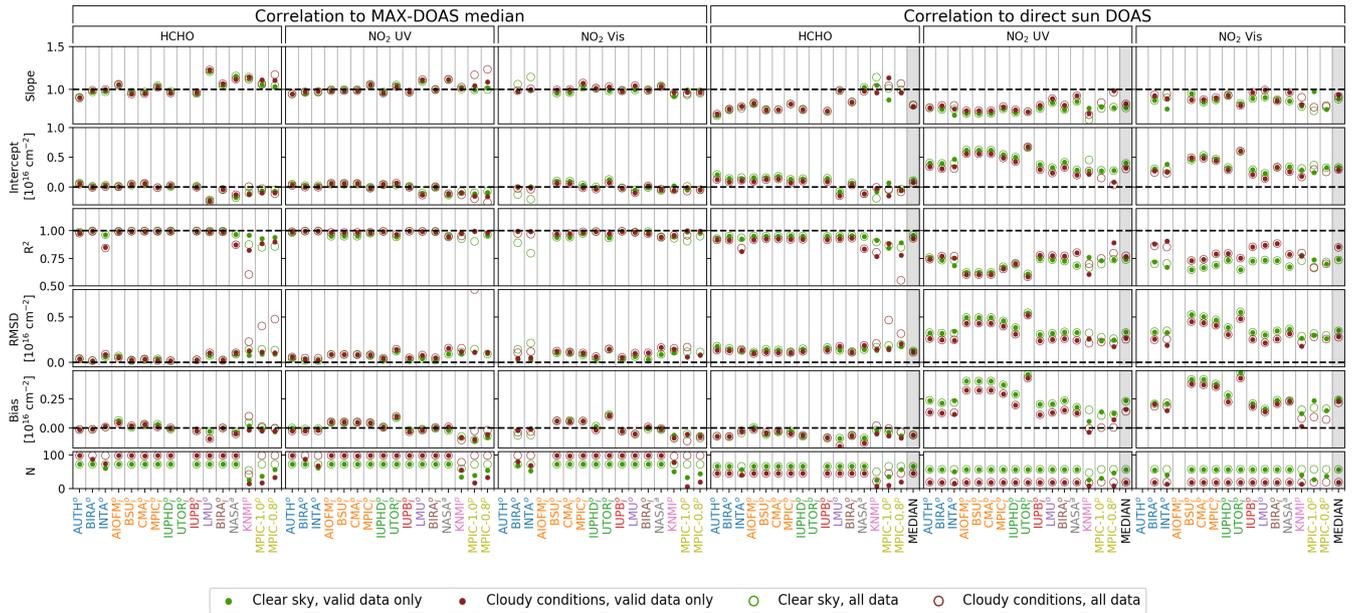
**Figure 16.** Comparison of MAX-DOAS retrieved HCHO VCDs vs. direct-sun DOAS. Basic descriptions of Fig. 14 apply.



**Figure 17.** Comparison of MAX-DOAS retrieved NO<sub>2</sub> VCDs vs. direct-sun DOAS, NO<sub>2</sub> lidar and radiosonde. Basic descriptions of Fig. 14 apply.

the reference spectrum was adapted from the MAX-DOAS VCD (see Sect. 2.2.4). [Bias values are of the order of 35% of the RMSDs, indicating that the deviations are mostly random.](#)

For NO<sub>2</sub> UV (Vis) the comparison to the direct-sun DOAS yields an average RMSD of  $3.7 \times 10^{15}$  molec cm<sup>-2</sup> ( $3.8 \times 10^{15}$  molec cm<sup>-2</sup>), which is about five times the average RMSD of the MAX-DOAS median comparison. Between 12 and 14 September the direct sun VCDs but also most radiosonde and lidar observation are systematically lower than the MAX-DOAS VCDs. [This is also reflected in the correlation statistics: RMSDs and Bias values of different participants appear strongly correlated in Fig. 18 and Bias magnitudes are > 70% of the RMSDs for both UV and Vis.](#) The reason could not yet be identified. [A candidate are the different sampling volumes: while radiosonde, lidar and direct-sun DOAS typically sample air at maximum distances of a few kilometres to the site, the MAX-DOAS instruments have a much larger horizontal sensitivity range](#)



**Figure 18.** Correlation statistics of trace gas VCDs. The plot is similar to Fig. 15. In the underlying correlation plots, ordinates are MAX-DOAS VCDs of individual participants and abscissas are the MAX-DOAS median and direct-sun VCDs, respectively. The correlation plots are shown in Supplement S8.3.

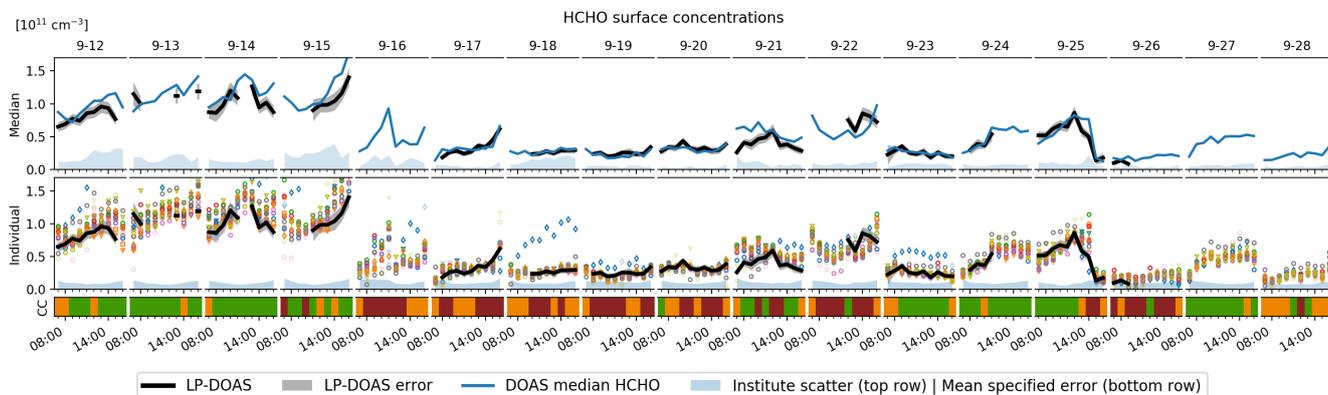
~~(see Supplement S5), even extending to The Hague on some days, which is > 40 km away. Indeed the agreement improves with decreasing visibility. Interestingly, this contrasts with findings on the surface concentration in the following section, where discrepancies to the LP-DOAS are dominated by random deviations.~~

In contrast to the AOTs, the RMSDs against the MAX-DOAS median here are smaller than the specified retrieval errors, which are  $1.3 \times 10^{15} \text{ molec cm}^{-2}$ ,  $1.3 \times 10^{15} \text{ molec cm}^{-2}$  and  $1.2 \times 10^{15} \text{ molec cm}^{-2}$  for HCHO,  $1.3 \times 10^{15} \text{ molec cm}^{-2}$  for NO<sub>2</sub> UV and  $1.2 \times 10^{15} \text{ molec cm}^{-2}$  for NO<sub>2</sub> Vis, respectively. On the other hand NO<sub>2</sub> RMSDs against the direct-sun observations are about three times larger. For the less abundant HCHO, the signal-to-noise ratio (SNR) of the measured in the median dSCDs is smaller than for other species, such that the specified uncertainties derived from the dSCD noise are larger and more representative for the actual retrieval accuracy.

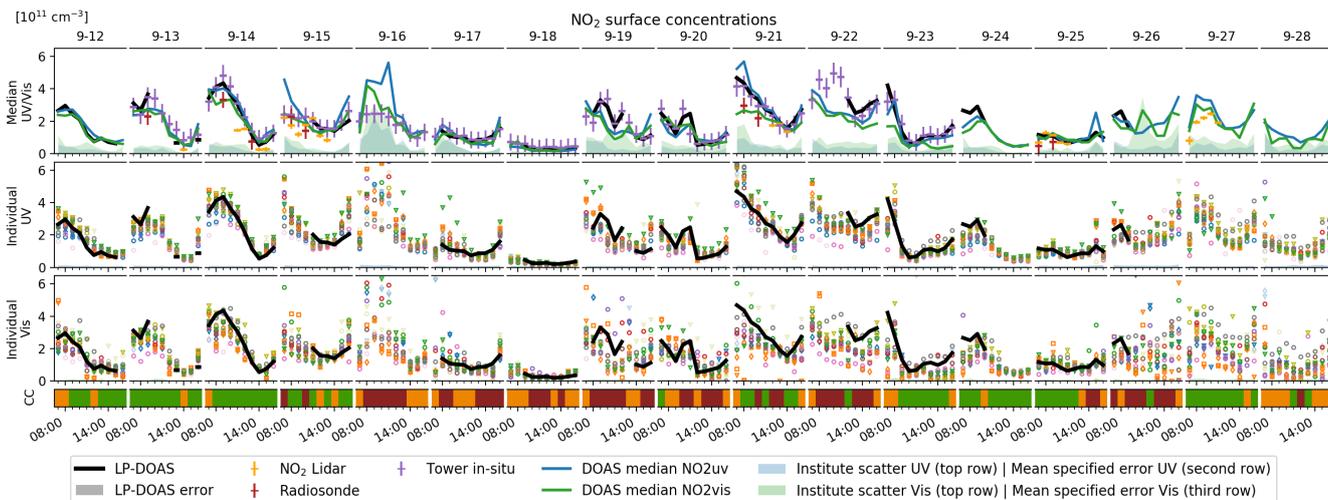
### 3.6 Trace gas surface concentrations

This section compares the number concentration of NO<sub>2</sub> and HCHO observed at the surface. Note that in this paper "surface concentration" refers to the average concentration in the lowest MAX-DOAS retrieval layer extending from 0 to 200 m altitude. Independent observations are the LP-DOAS (NO<sub>2</sub> and HCHO), and the surface values of radiosonde and lidar profiles (NO<sub>2</sub>), as well as integrated values of in situ measurements in the tower (described in Sect. 2.2.5). Comparisons of all observations are shown in Fig. 19 and 20. For the statistical evaluation (Fig. 21) only LP-DOAS data were considered since they provides

provide a very accurate, representative and complete dataset (see Section 2.2.5). The impact of profile smoothing during the retrieval on the retrieved surface concentration was estimated for NO<sub>2</sub> in Supplement S9 from available radiosonde and lidar NO<sub>2</sub> profiles and was found to be around  $5.5 \times 10^9$  molec cm<sup>-3</sup> ( $4 \times 10^9$  molec cm<sup>-3</sup>) in the UV (Vis). Typical RMSD values in the comparison with the LP-DOAS are about one order of magnitude larger, indicating that the impact of smoothing on the NO<sub>2</sub> surface concentration is negligible in this study.

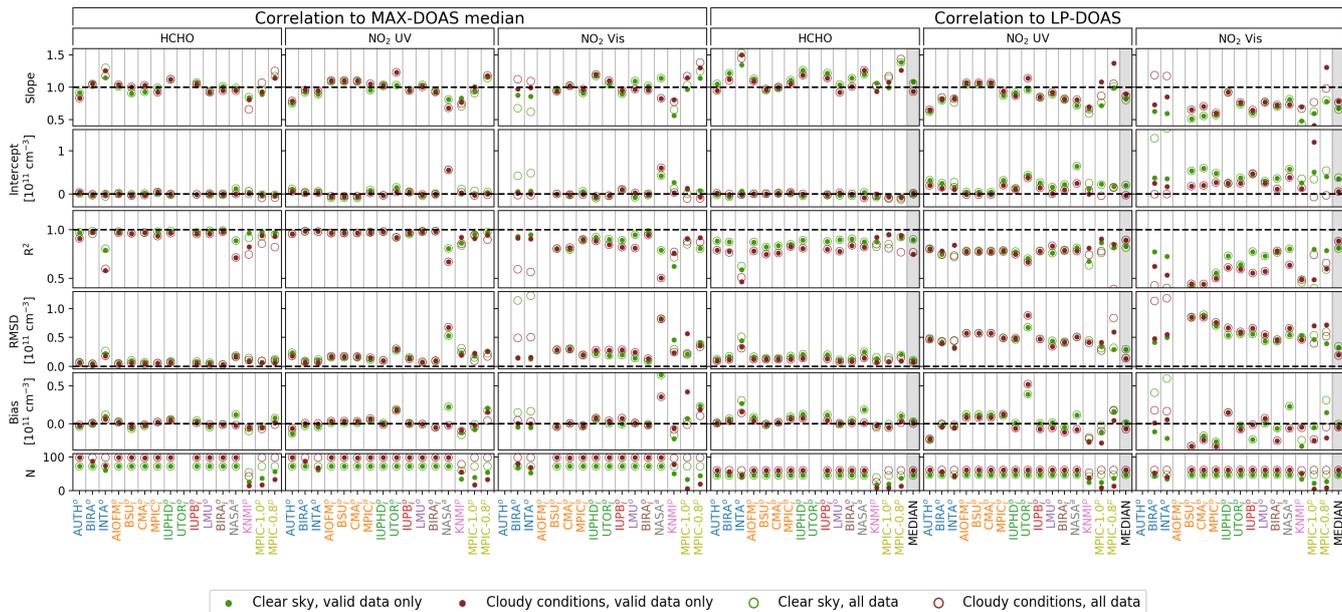


**Figure 19.** Comparison of MAX-DOAS retrieved HCHO surface concentrations. Basic descriptions of Fig. 14 apply. Note that the mean specified uncertainties in the two lower rows of the figure are very small and thus barely visible.



**Figure 20.** Comparison of MAX-DOAS retrieved NO<sub>2</sub> surface concentrations. Basic descriptions of Fig. 14 apply. Note that the mean specified uncertainties in the two lower rows of the figure are very small and thus barely visible.

The comparisons of surface concentrations are particularly useful, because the largest set of validation data is available here and because in contrast to the comparison of AOT and VCDs, the surface concentration comparison also requires an isolation



**Figure 21.** Correlation statistics of trace gas surface concentrations. The plot is similar to Fig. 15. In the underlying correlation plots, ordinates are MAX-DOAS surface concentrations of individual participants and abscissas are the MAX-DOAS median and direct-sun VCDs, respectively. The correlation plots are shown in Supplement S8.3.

of the surface layer from the layers above and therefore reflects the MAX-DOAS' ability to actually resolve vertical profiles ; as it requires an isolation of the surfacelayer from the layers above at least close to the surface.

Figures 19 and 20 show good qualitative agreement between all observations most of the time, even in the presence of clouds. Apparent exceptions for NO<sub>2</sub> are the fog event on 16 September (strong scatter among the participants) and at forenoon on 22 September (MAX-DOAS median shows large deviations compared to the tower measurements probably due to a very local NO<sub>2</sub> emission event close to the tower).

Under clear sky conditions average RMSDs observed for the comparison to the MAX-DOAS median results are  $8.8 \times 10^9 \text{ molec cm}^{-3}$ ,  $1.8 \times 10^{10} \text{ molec cm}^{-3}$  and  $2.7 \times 10^{10} \text{ molec cm}^{-3}$  for HCHO,  $1.8 \times 10^{10} \text{ molec cm}^{-3}$  for NO<sub>2</sub> UV and  $2.7 \times 10^{10} \text{ molec cm}^{-3}$  for NO<sub>2</sub> Vis, respectively. For the comparison to the LP-DOAS, they these values increase to  $1.8 \times 10^{10} \text{ molec cm}^{-3}$ ,  $4.7 \times 10^{10} \text{ molec cm}^{-3}$  and  $5.6 \times 10^{10} \text{ molec cm}^{-3}$ , respectively. For the median comparison, Biases magnitudes are about 40% of the RMSD values. In contrast to the VCDs, deviations to the supporting observations (LP-DOAS) seem to be random to large part, as Bias magnitudes are about three times smaller than RMSDs. Significant Biases are only observed for some participants, e.g. UTOR/ HEIPRO in the UV.

Clouds have very different impact on these the results: the average RMSD to the median increases by 15, 26, 15 for HCHO, 26 for NO<sub>2</sub> UV and 38% for NO<sub>2</sub> Vis, whereas the average RMSD to the LP-DOAS is even reduced by 4, 15, 4, 15 and 17%, respectively. A large fraction of the scatter in the comparison to the LP-DOAS might be related to the spatio-temporal

variability of the gas concentrations, in particular in the Vis spectral range, where the MAX-DOAS viewing distance is large. The good agreement of the surface concentrations with the supporting observations during the first days is opposite to the VCD comparison, which at least for NO<sub>2</sub> points to a problem with the [retrieval results in higher layers or the](#) direct-sun data. For NO<sub>2</sub> Vis, the agreement is generally worse than for NO<sub>2</sub> UV. Convergence problems of bePRO appear again in the form of outliers (see in particular the RMSD values), which are efficiently removed by flagging. INTA shows strong systematic outliers over whole days (e.g. on 18 September), which are not observed for other bePRO users and are very likely produced by technical problems. Again ~~the RMSDs to the MAX-DOAS median even for clear-sky conditions are~~, [as for AOTs and VCDs, the scatter among the participants is](#) similar or larger than the specified errors [even for clear-sky conditions](#) (factors of about ~~1, 2 and 3~~ [one](#) for HCHO, [two for](#) NO<sub>2</sub> UV, ~~NO<sub>2</sub>-Vis, respectively~~).

### 10 3.7 NO<sub>2</sub>-UV-Vis comparison

~~Another intrinsic consistency check for the algorithms, besides the comparison of modelled and measured dSCDs in Sect. 3.3, is the comparison of the~~ [and three for](#) NO<sub>2</sub> retrieval results in the two different spectral ranges (UV and Vis). These should ideally yield equal results at least when assuming a horizontally homogeneous atmosphere. Figures ?? and ?? show the correlation of VCDs and surface concentrations. [Vis, see Fig. 19 and Fig. 20](#).

15 Correlation of MAX-DOAS retrieved NO<sub>2</sub> VCDs in the UV and the Vis spectral ranges. Marker colours and transparency indicate the cloud conditions and flagging, respectively, according to the legend:

Correlation of MAX-DOAS retrieved NO<sub>2</sub> surface concentrations in the UV and the Vis spectral ranges. Legends and description of Fig. ?? apply:

20 For the VCDs, the average RMSD is  $1.5 \times 10^{15} \text{ cm}^{-2}$ , which increase by 70% in the presence of clouds. For clear sky conditions very good agreement (less than 10% relative RMSD) is observed for MAPA, M<sup>3</sup>, MARK, NASA/ Realtme and bePRO/INTA. There is a tendency for Vis VCDs to be larger than UV VCDs (by 6% regarding the campaign averages) which might be caused by the different sensitivity in particular in the retrieval layers between 200 m and 1 km altitude. The extended horizontal viewing distance is an unlikely reason since in contrast to the VCDs, surface concentrations in the Vis are smaller than in the UV.

25 For the surface concentrations, the results are very different for the individual algorithms and participants: The average RMSD is  $6.0 \times 10^{10} \text{ cm}^{-3}$  and increases by 25% in the presence of clouds. Best agreement with  $10\% < \text{relative RMSD} < 20\%$  are achieved by MAPA, Heipro/IUP-HD, MMF and NASA. Both bePRO users show a similar pattern with systematically smaller values in the Vis retrieval. bePRO suffers from a few strong outliers (even exceeding the plotting range), which are however in most cases removed by flagging. For PRIAM, there is large scatter for all the participants. For HEIPRO, there are large discrepancies between the two participants: while IUPHD achieves very good results here, UTOR shows large scatter (approx. factor of 4) similar to PRIAM-users which is once more likely to be explained by the different number of applied iteration steps during the aerosol inversion. The remaining algorithms perform reasonable (relative RMSD  $< 30\%$ ), apart from few outliers that usually occur under cloudy conditions. Particularly good correlation for both, VCDs and surface concentrations, are only achieved by NASA/ Realtme and MPIC/MAPA.

### 3.7 Retrieval from dSCDs of individual participants

As described in Sect. 2.1.1, the results compared so far were retrieved from a common set of median dSCDs. Thus, the results only illustrate the performance of the different retrieval techniques. However, it is also interesting to compare collocated MAX-DOAS measurements which are fully independent, to obtain an estimate of the reliability of a typical MAX-DOAS profile measurement undergoing the whole spectra acquisition and data processing chain. Therefore, the study above was once more conducted with each participant using their own measured dSCDs (see Kreher et al., 2019, for dataset details). ~~The complete results are shown in Supplement S10~~ [Supplement S10 shows further details by means of figures that are equivalent to those shown before in the course of the median dSCD comparison.](#) A summary is given in Table 5 which shows the increase in average RMSD [and average Bias magnitude](#) for the most important comparisons (as described in the precedent subsections for the median dSCDs) when participants use their own instead of the median dSCDs. Only valid data of participants appearing in both studies were considered and BIRA/ bePRO and KNMI were excluded because in contrast to the median dSCD study BIRA/ bePRO and KNMI did not submit flags for the own dSCD study, which heavily impacted the results.

**Table 5.** ~~Increase~~ [Relative increase](#) in average RMSD [\(first value\)](#) and average Bias magnitude [\(values in brackets\)](#) when participants retrieve profiles from their own dSCDs instead ~~from of using~~ the median dSCDs. Values are given for clear sky and cloudy conditions separately. Further the comparisons among the participants (to the MAX-DOAS median) and the comparisons to the supporting observations [\(sun photometer AOTs, direct-sun DOAS VCDs and LP-DOAS surface concentrations\)](#) are distinguished.

Observation	Species	Clear sky		Cloudy	
		To median [%]	To supp. obs. [%]	To median [%]	To supp. obs. [%]
AOT	Aerosol UV	29 (37)	-10 (-16)	32 (48)	45 (58)
	Aerosol Vis	29 (55)	18 (15)	26 (110)	21 (37)
VCD	HCHO	175 (187)	66 (109)	152 (113)	46 (32)
	NO <sub>2</sub> UV	45 (52)	-8 (-18)	45 (31)	-8 (-30)
	NO <sub>2</sub> Vis	43 (8)	6 (13)	27 (-8)	3 (-2)
Surface	HCHO	87 (64)	16 (34)	120 (129)	37 (82)
	NO <sub>2</sub> UV	28 (53)	10 (64)	25 (76)	1 (45)
	NO <sub>2</sub> Vis	13 (11)	6 (37)	-9 (-42)	-13 (-12)

Regarding only the increase in RMSD in the MAX-DOAS median comparison (hence, the degradation of consistency among the participants) is qualitatively consistent with what one would expect from the findings by Kreher et al. (2019) on the CINDI-2 dSCD consistency: for NO<sub>2</sub>, almost all participating instruments were able to deliver good quality dSCDs suitable for profile inversion, while for HCHO the quality was much more variable, resulting in the stronger degradation given in Table 5. Kreher et al. (2019) identified instrumental characterisation (e.g. detector non-linearity and stray-light in the spectrometer) and

pointing issues as the main sources of discrepancy between the participant's own dSCD datasets. The degradation is smaller for the surface concentrations than for the trace gas VCDs and is very similar for different cloud conditions.

For the comparison to the supporting observations, the increase in average RMSD is smaller (second and fourth column of Table 5). This means, that even though using the own dSCDs induces differences among the participants, the average quality of the dSCDs is basically maintained or at least small compared to the discrepancies induced by the retrieval techniques. Interestingly, the RMSD and Bias values for the UV AOT and NO<sub>2</sub> VCD even ~~decreases~~decrease, indicating that the median dSCDs suffer from systematic ~~biases~~errors. Under clear sky conditions, low impact ( $\leq 10\%$ ) was found for Aerosol UV AOTs ~~under clear sky conditions~~ and NO<sub>2</sub> data products. Particularly large impact is observed for HCHO VCDs (66%). Under cloudy conditions, the impact on NO<sub>2</sub> products remains small (again  $< 10\%$ ), whereas for all other products, the increase in average RMSD exceeds 20%.

It is also of interest to explicitly estimate which fractions of the total observed discrepancies among MAX-DOAS observations are caused either by the use of different retrieval algorithms or by inconsistencies in the dSCD acquisition. Note that the RMSD values from the median dSCD comparison represent the error arising solely from using different algorithms while the RMSD values from the own dSCD comparison represent the combined effect of both aspects. For simplicity, we assume that the contributions of both aspects are random and independent so that the effect of using own dSCDs can be isolated by simple RMSD error calculations. In this way, its contribution to the total variance observed among the participants under clear sky conditions can be estimated to 40% (for AOTs), 85% (HCHO VCDs), 70% (HCHO surface concentrations), 50% (NO<sub>2</sub> VCDs), 40% (NO<sub>2</sub> UV surface concentrations) and 20% (NO<sub>2</sub> Vis surface concentrations), respectively. The residual variance can be attributed to the choice and setup of the retrieval algorithm.

## 20 4 Conclusions

Within this study, 15 participants used 9 different profiling algorithms with 3 different technical approaches (optimal estimation (OEM), parametrized (PAR) and analytical (ANA) approach) to retrieve aerosol and trace gas (NO<sub>2</sub>, HCHO) vertical profiles from a common set of dSCDs which was recorded during the CINDI-2 campaign. The results were compared and validated against colocated supporting observations with the ~~aim to assess performance and reliability of individual algorithms but also of the MAX-DOAS profiling technique in general~~ focus on aerosol optical thicknesses (AOTs), trace gas vertical column densities (VCDs) and trace gas surface concentrations. Data from some supporting observations were used for qualitative comparison only (Ceilometer, NO<sub>2</sub> radiosondes, NO<sub>2</sub>-Lidar, NO<sub>2</sub> in-situ instruments) while for a statistical assessment AOTs from the sun photometer, VCDs from direct-sun DOAS observations and surface concentrations from the LP-DOAS were used.

~~Summary of RMSDs from the comparisons in Sect. 3 for clear-sky conditions. The RMSD values of AOT, VCD and surface concentration are calculated with respect to the corresponding supporting observations. Average RMSD values define the colour scale of each column (see colourbar on the top right). White spaces indicate no data. Average observed values (bottom row) are rounded campaign averages of the supporting observations. The column on the far right indicates which fraction of~~

the maximum number (170) of available profiles has been used. Participants who submitted flags are represented by two rows: one considering all data and one using only those flagged as valid ("valid only").

Figure 22 shows an overview of RMSD and Bias values for the ~~inherent quality indicators (correlations correlation~~ between measured and modelled dSCDs ~~as well as between NO<sub>2</sub> UV and Vis results)~~ and the comparisons to ~~available supporting~~ supporting observations (AOT, VCD and surface concentration). General strengths and weaknesses of different algorithms become particularly apparent here. Very good overall performance without the need for validity flagging is achieved by the MMF and the M<sup>3</sup> algorithm. Note ~~;~~ that the results for aerosol are of very similar quality, even though in contrast to M<sup>3</sup>, MMF retrieves aerosol in the logarithmic space. For valid data (about 20% discarded) INTA also shows good overall performance apart from the outliers in the HCHO surface concentration, which are very likely related to technical problems.

Very good performance for aerosol is observed for IUPHD over the full dataset. For NO<sub>2</sub>, best performance is achieved by MAPA. The AOT comparison looks generally worse for parametrized approaches which is expected since no partial AOT correction can be performed and thus - with the MAX-DOAS integrated extinction profile and the sun photometer total AOT - basically two different quantities are compared. Finally, the Realtime algorithm by NASA (being the only ANA algorithm) shall be pointed out: despite its simplified radiative transport and the associated outstanding computational performance it provides reasonable results for trace gases (RMSD/ Average RMSD around unity).

Parametrized approaches appear to be less stable in the sense that for less favourable conditions no convergence is achieved or inconsistent results are returned (30 to 70% of all profiles). For MAPA, these cases are reliably identified and flagged as invalid such that the remaining results achieve very good RMSD and Bias values. In contrast for MARK, even some profiles considered valid do not look plausible. The instability of parametrized algorithms is likely related to the approach: in reality, a vertical profile can be described by an arbitrarily large set of parameters and the information on those contained in a MAX-DOAS measurement depends on the atmospheric conditions, hence the profiles themselves. For parametrized approaches, the number of retrieved parameters is reduced to the number of typically observed DOFs by describing the profile by a few prescribed (not necessarily orthogonal) parameters. Lack of information in those due to particular atmospheric conditions (also if information is available but only on parameters not covered by the chosen parametrization) leads to an under-determined problem with ambiguous solution and the inversion fails. For OEM approaches, the information can be dynamically distributed to a larger number of parameters (20 in this study, namely the species abundances in the retrieval layers) while ~~any lack of information is filled~~ parameters of few or no information are constrained by *a priori* ~~knowledge~~ information. This is why OEM inversions converge under a broader range of atmospheric conditions even when information from the measurement is reduced or shifted between retrieved parameters. On the other hand, this means that OEM algorithms even provide plausibly looking profiles (basically the *a priori* profile) when few/no information is contained in the measurements. Even though such cases can be identified by examining the AVKs, this makes OEM retrievals prone to misinterpretations particularly by inexperienced users.

Regarding full profiles, the overview plots in Sect. 3.2 and figures in Supplement S8.2 show a good qualitative agreement between the algorithms for valid data and clear-sky conditions. In most cases they detect the same features, however sometimes at different altitudes and of different ~~intensity (see also Supplement S8.2)~~ magnitude. Under clear-sky condi-

tions, the RMSDs between individual participants and the MAX-DOAS median results range between (0.01 – 0.1) for AOTs, (1.5 – 15) × 10<sup>14</sup> molec cm<sup>-2</sup> for trace gas VCDs and (0.3 – 8) × 10<sup>10</sup> molec cm<sup>-3</sup> for trace gas surface concentrations~~range between 0.01 – 0.1, (1.5 – 15) × 10<sup>14</sup> molec cm<sup>-2</sup> and (0.3 – 8) × 10<sup>10</sup> molec cm<sup>-3</sup>, respectively.~~ These values compare to approximate average AOTs of 0.3, trace gas VCDs of 90 × 10<sup>14</sup> molec cm<sup>-2</sup> and trace gas surface concentrations of 11 × 10<sup>10</sup> molec cm<sup>-3</sup> observed over the campaign period. Note that profiles were retrieved from a common set of dSCDs and thus these discrepancies solely arise from the choice of the retrieval algorithm and detailed settings, that were not prescribed according to Sect. 2.1.3. Obvious source of discrepancies is the use of different techniques (OEM, PAR and ANA). Further, differences among the two PAR approaches are expected as they use different parametrizations. Discrepancies among the different OEM algorithms are expected as they retrieve aerosol extinction either in logarithmic or linear space and since the exact implementation might differ (consider for instance the Thikonov regularisation approach used by BOREAS). Interestingly, discrepancies among participants using the same OEM algorithm are only about 50% smaller (regarding ASDevs of profiles as defined in Sect. 2.3) than the average discrepancies among all participants. This indicates that user defined retrieval settings that were not prescribed within this study (e.g. number of applied iteration steps in the optimisation process and RTM accuracy options) also have significant impact. An example appearing in this study are the differences between IUPHD and UTOR (both using HEIPRO) that were found to mainly be caused by differences in the number of applied iteration steps in the optimisation process of the aerosol inversions.

As discussed in more detail below and in Sect. 3.7, the discrepancies among the participants are of very similar order of magnitude as discrepancies that are induced when participants retrieve profiles from their own measured dSCDs. It is an important finding that, at least for CINDI-2, the choice of the algorithm/settings has similar impact on the profiling results as the inconsistencies in the dSCD acquisition.

For the comparison against supporting observations ~~, these values increase to~~ (see Fig. 22) RMSDs increase to (0.02 – 0.2) against AOTs from the sun photometer, (11 – 55) × 10<sup>14</sup> molec cm<sup>-2</sup> against trace gas VCDs from the direct-sun DOAS and (0.8 – 9) × 10<sup>10</sup> molec cm<sup>-3</sup> ~~, most likely due to (systematic) errors and~~ against trace gas surface concentrations from the LP-DOAS. For Vis AOTs and trace gas surface concentrations discrepancies are mostly random (average Bias magnitude smaller than half the average RMSD) while for AOT UV and trace gas VCDs systematic deviations are dominant (compare Fig. 22). The average uncertainties of the supporting observations themselves are 0.022, 19 × 10<sup>14</sup> molec cm<sup>-2</sup> and 0.74 × 10<sup>10</sup> molec cm<sup>-3</sup>, respectively, and can therefore be regarded as major RMSD contributors at least in cases where RMSD values are low. Errors in the median dSCDs used as the input for the retrievals are also likely to significantly contribute (see discussion on the own dSCD comparison below). Further, investigations on the spatio-temporal variability (see Sect. 2.3.3 and Supplement S6) indicate that a significant fraction of the RMSD observed between MAX-DOAS and supporting observations is caused by imperfect spatio-temporal overlap ~~of all observations.~~ For NO<sub>2</sub> surface concentrations the RMSD resulting from this could roughly be estimated to be around 3 × 10<sup>10</sup> molec cm<sup>-3</sup> (using strong simplifications though) which is indeed of the order of magnitude of the average RMSDs observed. Finally, simplified assumptions on the fixed RTM atmosphere were made (compare Sect. 2.1.3). While the choice of pressure and temperature profiles has little impact on the overall agreement with supporting

observations (< 5%, see Supplement S7), the assumptions on the aerosol optical properties (Heney-Greenstein approximation with constant single scattering albedo and asymmetry parameter over the whole campaign) are a likely source of error.

The consistency of Aerosol Vis and NO<sub>2</sub> Vis products (in particular the agreement among the participants) is typically worse in comparison to their UV counterparts by up to several ten percent. Only the agreement with the sun photometer AOT improves when going from the UV to the Vis spectral range. This might also be related to the reliability of the sun photometer AOTs  $\tau_s$ : while in the Vis the MAX-DOAS retrieval wavelength (477 nm) is close to the lowest sun photometer wavelength channel (440 nm), in the UV extrapolation of  $\tau_s$  down to 360 nm is required (see Sect. 2.2.1).

The presence of clouds strongly affects the agreement of aerosol retrieval results particularly in the visible spectral range. For AOTs ~~in the UV (Vis) the~~ the increase in average RMSD against the median is around 30% ~~(in the UV and 80% )in the Vis~~ while RMSDs against the sun photometer are degraded by 10% ~~(and 130%), respectively.~~ This is expected as i) high aerosol optical thicknesses at altitudes of low MAX-DOAS sensitivity make the results extremely susceptible to even small changes in the retrieval strategy and ii) the few sun photometer observations under cloudy conditions are likely recorded through local cloud holes and therefore not representative for MAX-DOAS measurements integrating horizontally over several kilometres. In contrast, the impact of clouds on average RMSDs for trace gas VCDs is < 15%. Surface concentration RMSDs against the median are degraded by around 25%, whereas average RMSDs to supporting observations even decrease.

It could be shown that, in the case of CINDI-2, the average impact of smoothing effects on the NO<sub>2</sub> surface concentration is negligible (Supplement S9). In contrast to that, smoothing has a strong impact on the agreement of MAX-DOAS observations with AOTs and probably HCHO VCDs from supporting observations (~~Section Sect. 2.3.2~~). In particular ~~, the low sensitivity at higher altitudes has the effect that~~ it was shown for the first time, that formerly observed systematic discrepancies between MAX-DOAS integrated aerosol ~~extinction profiles~~ and sun photometer ~~total AOTs are not necessarily comparable quantities (Section 3.4 and Supplement ??). Such comparisons can lead to doubtful conclusions if no additional information on the real aerosol distribution is available to perform the necessary corrections. AOTs can be largely explained and compensated by considering biases arising from the reduced sensitivity of MAX-DOAS observations to higher altitudes and associated a priori assumptions (see Sect. 3.4).~~

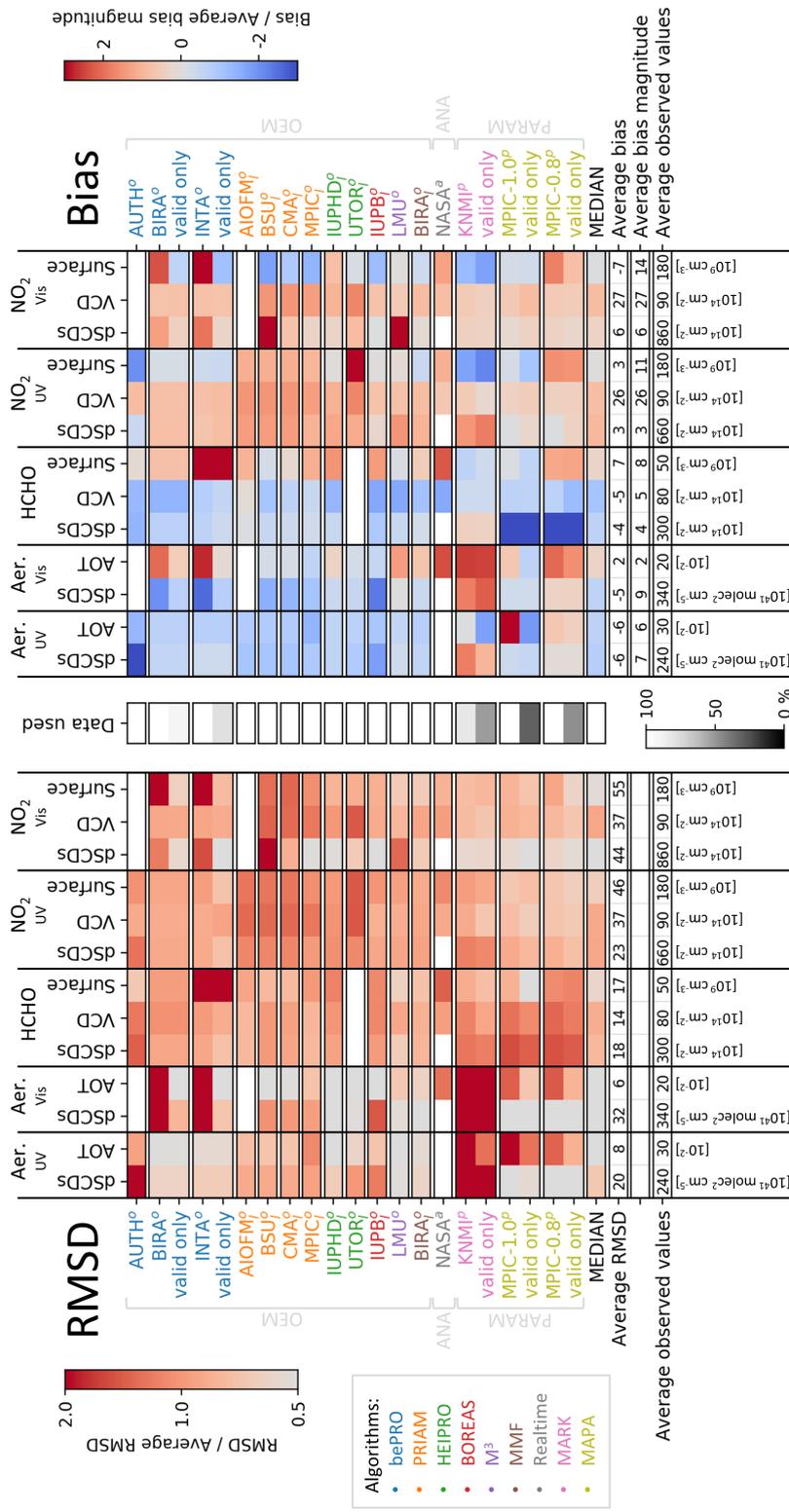
For CINDI-2 data, there is no clear indication that an O<sub>4</sub> dSCD scaling is necessary. On the one hand for OEM algorithms the MAX-DOAS AOT is in good agreement with the sun photometer partial AOT and in contrast to Beirle et al. (2019), we find that a scaling factor of 0.8 is too small (Supplement S2) at least when applied to the whole campaign. On the other hand a less extreme scaling ( $0.8 < SF < 1.0$ ) potentially removes remaining biases (see Fig. S3) and improves the agreement between forward model and reality (see Fig. S4). O<sub>4</sub> scaling and PAC were found to have similar impact on the MAX-DOAS AOT results. Scaling might therefore be used to at least partly replace the PAC in the case of retrieval approaches that do not quantify their sensitivity or the assimilated *a priori* information. At last we think for this study the prescribed SF = 1.0 scaling factor of 1.0 is justified. Even though it might not be ideal, it is the most straightforward approach and yields reasonable and consistent results within the uncertainties introduced by other factors. To draw more concise conclusions, further studies as performed e.g. by Wagner et al. (2019) are necessary.

In most comparisons, RMSDs of individual participants against the MAX-DOAS median results (even when using the same algorithm) was of the order or larger than the uncertainties specified by the algorithms themselves (up to a factor of ~~3~~ three for NO<sub>2</sub> Vis surface concentrations), indicating that the choice of the retrieval algorithm has severe impact on the results. It shows further, that the specified uncertainties (which typically take propagated measurement noise and smoothing errors into account but neglect other effects like model errors) ~~might be~~ are too optimistic as a measure for the MAX-DOAS retrieval accuracy and have to be regarded with care. ~~The discrepancies between the results of the participants using the same algorithm indicate that the retrieval settings that were not prescribed within this study (e.g. number of applied iteration steps in the optimisation process, RTM accuracy options, ...) leave a lot of room for variations. However, technical reasons cannot be fully excluded as the source of the discrepancies. An example appearing in this study are the differences between IUPHD and UTOR (both using HEIPRO) that were found to mainly be caused by differences in the number of applied iteration steps in the optimisation process of the aerosol inversions.~~

If the profiles are retrieved from the participant's individually measured dSCDs instead of using a common median dSCD dataset (see Sect. 3.7), the agreement of MAX-DOAS results with supporting observations (average RMSD) is degraded by very different amounts, depending on species and data product. Low impact ( $\leq 10\%$ ) was found for Aerosol UV AOTs and NO<sub>2</sub> data products. ~~A particularly large impact~~ For Aerosol UV AOTs and NO<sub>2</sub> UV VCDs even improvements were observed, hinting to potential systematic errors in the median dSCDs. A particularly strong degradation was observed for HCHO VCDs (65%).

~~Finally, investigations on the spatio-temporal variability (see Supplement S6) indicate that a significant fraction of the RMSD observed between~~ Further, we estimated what fractions of the observed discrepancies among the MAX-DOAS and supporting observations is caused by imperfect spatio-temporal overlap. Thus for future campaigns participants are caused either by the use of different retrieval algorithms or by inconsistencies in the dSCD acquisition. In average the impact of both aspects is very similar: the effect of using own dSCDs can be estimated to contribute 40% (for AOTs), 85% (HCHO VCDs), 70% (HCHO surface concentrations), 50% (NO<sub>2</sub> VCDs), 40% (NO<sub>2</sub> UV surface concentrations) and 20% (NO<sub>2</sub> Vis surface concentrations) to the total variance introduced by both aspects. The high values for HCHO are expected, since according to Kreher et al. (2019) the acquisition of dSCDs was particular challenging and here and they varied widely among the participants.

For future campaign and comparison exercises, fixed model parameters (particularly aerosol optical properties) and prior constraints might be chosen more carefully. Further we suggest putting enhanced focus on the coordinated operation of all (not only MAX-DOAS) instruments and to incorporate techniques with more appropriate spatial kernels, e.g. limb DOAS observations from unmanned aerial vehicles, to reduce the spatio-temporal mismatch between different observations.



**Figure 22.** Summary of the comparisons in Sect. 3 for clear-sky conditions. Left panel shows RMSD, right panel shows Bias. Average values of RMSD (Bias) define the colour scale of each column of the left (right) panel as indicated by the color bars on the top left (top right) of the figure. Values of AOT, VCD and surface concentration are given with respect to the corresponding supporting observations (sun photometer, direct-sun DOAS and LP-DOAS). White spaces indicate no data. Average observed values (bottom row) are rounded campaign averages of the supporting observations. Average Bias and Average Bias magnitude values (third last and second last row of right panel) represent the averages over the signed and the absolute Bias values, respectively. The "data used"-column in the center indicates which fraction of the maximum number (170) of available profiles has been used. Participants who submitted flags are represented by two rows: one considering all data and one using only those flagged as valid ("valid only").

*Author contributions.* JLT performed the comparison and the associated investigations as described in the paper and wrote the first draft. UF was involved in the planning of the campaign and the profiling activities, operated the IUPHD instrument, evaluated its data, supervised the comparison activities and contributed in scientific discussions and the manuscript revision. FH was involved in the planning of the campaign and the profiling activities, retrieved profiles for BIRA and contributed in scientific discussions and the manuscript revision. FH, GP, MVR, AA, AP, AR, TW, KK, UF, JL designed, planned and organized the CINDI-2 campaign. AL/JX/PX, AP, CF/CH/AM/FT/GP/MVR, CZ/KLC/NH/ZW, EP/FW/TB, ES, IB, JJ/JM, KB/XZ, KLC, MY/OPu, SD and TD/AB prepared and operated the MAX-DOAS instrument(s) of AIOFM, KNMI, BIRA, USTC, IUPB, Pandora, BSU, CMA, UTOR, DLR, INTA, MPIC and AUTH, respectively. AL/JX/PX, AP/TV, CA, CF/MVR, CG/FH, CX/HL/KLC, EP/TB/ FW/ AR, ES, IB, JJ/JM, KB/XZ, KLC, LGM/MY/OPu, MMF, SBei, SD, TD/AB, YW and ZW evaluated the MAX-DOAS data for AIOFM, KNMI/MARK, LMU, BIRA, BIRA/bePRO, USTC, IUPB, NASA/Realtime, BSU, CMA, UTOR, DLR/M3, INTA, BIRA/MMF, MPIC/MAPA, MPIC, AUTH, MPIC/PriAM and DLR/bePRO. AB, AR, CL, KS, MWe, NH and TW supervised the activities of AUTH, Bremen, USTC, UTOR, LMU, DLR and MPIC, respectively. KK as the campaign referee was involved in the actual running of the campaign and the data evaluation up to dSCDs. TW and JK planned and performed the common MAX-DOAS pointing calibration. NH coordinated the cooperation between DLR and USTC. Installation, operation and data evaluation of in-situ NO<sub>x</sub> instrumentation was performed by AF/AH (in-situ profile instrumentation in the tower), AM/FT (CAPS) and JL (ICAD/CE-DOAS). BH calibrated and operated the CIMEL sun photometer that is part of AERONET. DS<sub>w</sub>, LG<sub>a</sub>, RVH and SBe<sub>r</sub> operated the NO<sub>2</sub> lidar and processed its data into NO<sub>2</sub> profiles. SS installed, operated and evaluated the data of the LP-DOAS instrument. DS<sub>z</sub>, MA and MDH operated and evaluated the data of the NO<sub>2</sub> radiosondes. AC and MT provided and installed the Pandora instruments from which NO<sub>2</sub> direct-sun and NASA/Realtime profiling data were deduced. AA, AF, AH, AR, ES, JH, KB, KLC, MMF, MW<sub>i</sub>, SBei, SS, TB, TW, UP and YW contributed to the scientific discussion and interpretation. AM, AR, CF, CH, FT, GP, JH, JV, MMF, MVR, MW<sub>i</sub>, SBei, SS, TB, TW, UP and YW revised and contributed to the manuscript. All authors read and approved the submitted version.

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