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TOLNet validation of satellite ozone profiles in the troposphere: impact of retrieval wavelengths

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Abstract. The Tropospheric Ozone Lidar Network (TOLNet) was applied to validate retrievals of ozone (O_3) profiles in the troposphere from the TROPOspheric Monitoring Instrument (TROPOMI) ultraviolet (UV), Cross-track Infrared Sounder (CrIS) infrared (IR), and a combined UV+IR wavelength retrieval from TROPOMI/CrIS. Observations from six separate ground-based lidar systems and various locations of ozonesondes distributed throughout North America and Europe were applied to quantify systematic bias and random errors for each satellite retrieval. Furthermore, TOLNet data were used to intercompare idealized UV, IR, and UV+IR convolved profiles of O_3 in the troposphere during case studies representative of high O_3 events. This study shows that the combination of wavelengths in satellite retrievals (i.e., UV+IR) increases the sensitivity and vertical resolution of the retrievals of O_3 in the troposphere compared to single-wavelength products. The improved sensitivity and vertical resolution in UV+IR retrievals in the middle- and upper-troposphere resulted in tropospheric degree of freedom (DOF) values ~33% higher compared to UV- and IR-only retrievals. The increased DOFs in the UV+IR retrievals allowed for improved reproduction of mid- and upper-tropospheric O_3 enhancements, and to a lesser degree near-surface pollution enhancements, compared to single wavelength satellite products.

The validation of O_3 profiles in the troposphere retrieved with the UV-only, IR-only, and UV+IR Tikhonov regularised Ozone Profile retrievAl with SCIATRAN (TOPAS) algorithm developed at the Institute for Environmental Physics, University of Bremen demonstrated the utility of using TOLNet as a satellite evaluation data set. TOPAS UV-only, IR-only, and UV+IR wavelength retrievals had systematic biases throughout the troposphere of 11.2 ppb (22.1%), -1.7 ppb (-0.3%), and 3.5 ppb (7.8%), respectively, which meet the tropospheric systematic bias requirements of TROPOMI and CrIS. The primary drivers of systematic bias were determined to be the a priori vertical profile shape, solar zenith angle, and surface albedo. Random errors, representative of uncertainty in the retrievals, were large for all three retrievals with UV-only, IR-only, and UV+IR wavelength retrievals having root mean squared errors (RMSE) throughout the troposphere of 17.4 ppb (19.8% of mean tropospheric column values), 10.5 ppb (12.6% of mean tropospheric column values), and 14.0 ppb (14.6% of mean tropospheric column values), respectively. TOPAS UV-only profiles did not meet the uncertainty requirements defined for TROPOMI for the troposphere; however, CrIS IR-only retrievals did meet the uncertainty requirements defined by this mission. The larger random biases reflect the challenge of retrieving daily O_3 profiles due to the limited sensitivity and vertical resolution of these retrievals in the troposphere. Tropospheric systematic biases and random error were lower in IR-only and combined UV+IR retrievals compared to UV-only products due to the increased sensitivity in the troposphere allowing the retrievals to deviate further from the a priori profiles. Consistent daily observations from TOLNet demonstrated that the performance of the three satellite products varied by season and altitude in the troposphere. TOLNet was shown in this study to be a sufficient source of satellite O_3 profile validation data in the troposphere which is critical as this data source is the primary product identified for the tropospheric O_3 validation of the recently-launched Tropospheric Emissions: Monitoring of Pollution (TEMPO) mission.

1 Introduction

Consistent observations of tropospheric ozone (O_3) are critical for understanding atmospheric chemistry, important societal issues such as air quality and human health (WHO, 2003; US EPA, 2006), and long-term trends in atmospheric chemical composition (Cooper et al., 2014). Monitoring tropospheric O_3 is typically done with ground-based in situ measurement networks, tropospheric O_3 lidars, and ozonesonde launches (Lefohn et al., 2018; Tarasick et al., 2019; Sullivan et al., 2022). These observation types provide high accuracy information; however, surface-level monitoring networks do not detect O_3 vertical profiles throughout the tropospheric column and ozonesonde launches and lidars are spatiotemporally sparse. To fill this time and space void, over the past couple decades satellite sensors have been developed to retrieve O_3 profiles in the stratosphere and troposphere with near global coverage (Hoogen et al., 1999; Liu et al., 2005). However, due to the coarse vertical resolution of nadir-viewing passive satellite retrievals of O_3 profiles in the troposphere (>6 km) the representativeness and accuracy of this data source can be degraded compared to ozonesondes and lidars. Given the benefit from the observational coverage of satellites, it is vital to quantify these sensor's systematic biases and unresolved errors in the troposphere.

The first spaceborne sensor to retrieve tropospheric O_3 vertical profiles was the Global Ozone Monitoring Experiment (GOME) instrument which was launched in 1995 onboard the European Space Agency (ESA) European Remote Sensing Satellite (ERS-2) (Burrows et al., 1999). This ultraviolet (UV) wavelength (between 237–406 nm) retrieval (Hoogen et al., 1999; Liu et al., 2005) from GOME had a vertical resolution of 10–15 km in the troposphere and spatial resolution of 40 km × 320 km (Liu et al., 2005). A follow-on sensor for continued vertical profiling of tropospheric O_3, GOME-2, was launched in 2006 onboard the ESA MetOp-A satellite (Callies et al., 2000). GOME-2 applies an UV wavelength (between 240–403 nm) retrieval and has a ground pixel size of 40 km × 80 km with vertical resolution of 7–15 km in the troposphere (Miles et al., 2015; Kauppi et al., 2016). National Aeronautics and Space Administration (NASA) launched the polar-orbiting Aura satellite in 2004 which is the platform for the Dutch-Finnish nadir viewing spectrometer Ozone Monitoring Instrument (OMI) currently still retrieving tropospheric O_3 profiles (Liu et al., 2010). There are two O_3 profile retrieval algorithms for OMI (NASA - Royal Netherlands Meteorological Institute (KNMI), van Oss et al., 2002; Smithsonian Astrophysical Observatory (SAO), Liu et al., 2010). The SAO algorithm uses UV wavelengths (270–330 nm) to provide data at a spatial resolution of 13 km × 48 km and vertical resolution in the troposphere of 10–14 km (Liu et al., 2010; Bak et al., 2013). The NASA-KNMI OMI algorithm uses the same UV wavelengths resulting in similar spatial and vertical resolution in the troposphere as the SAO product (Kroon et al., 2011). Finally, TROPOspheric Monitoring Instrument (TROPOMI) was launched onboard the ESA's Sentinel-5 Precursor (S5P) satellite in 2017 and retrieves tropospheric O_3 profiles with relatively high spatial resolution (28.8 km × 5.6 km) and vertical resolution of 10-15 km in the troposphere using UV wavelengths (270–330 nm) (Mettig et al., 2021).

Spaceborne sensors using thermal infrared (TIR) wavelengths such as the Infrared Atmospheric Sounding Interferometer (IASI) (Clerbaux et al., 2010), Atmospheric Infrared Sounder (AIRS) (Chahine et al., 2006), Tropospheric Emission Spectrometer (TES) (Beer et al., 2001), and Cross-track Infrared Sounder (CrIS) (Ma et al., 2016) also retrieve tropospheric O_3 vertical profiles. Three IASI sensors have been launched to provide continuous data from 2006 (onboard MetOp-A) to present (onboard MetOp-C) using multiple algorithms applying TIR wavelengths between 975–1100 cm (Keim et al., 2009; Hurtmans et al., 2012). Tropospheric O_3 vertical profiles from IASI sensors have similar spatial resolution as UV-based retrievals (from 12 km × 25 km to 48 km × 50 km) with higher vertical resolution in the troposphere (6–8 km) compared to UV-based sensors (Boynard et al., 2009). TES, also onboard NASA's Aura satellite, uses TIR wavelengths (995–1070 cm) to retrieve O_3 vertical profiles with high spatial resolution (5 km × 8 km) and similar vertical resolution as IASI (6–7 km in the troposphere) (Worden et al., 2007). The NASA Aqua satellite was launched in 2002 which is the platform for the AIRS TIR sensor which retrieves O_3 profiles at ~50 km × 50 km spatial resolution with vertical resolution in the troposphere of 6–8 km using the TIR wavelengths between 985–1318 cm (Fu et al., 2018). The National Oceanic and Atmospheric Administration (NOAA) Suomi National Polar-orbiting Partnership (Suomi-NPP) satellite, which houses CrIS, was launched in 2011 and retrieves O_3 profiles in the TIR wavelengths (650–1095 cm) at 42 km × 42 km spatial resolution and vertical resolution between 4-10 km (Ma et al., 2016).

Given the higher vertical resolution of TIR retrievals compared to UV-only sensors in the troposphere, studies have been conducted to demonstrate the improvements in O_3 vertical profile retrievals when combing both wavelength ranges (e.g., Natraj et al., 2011). This has been demonstrated by combining retrievals from OMI+TES (Fu et al., 2013), GOME-2+IASI (Cuesta et al., 2013), and OMI+AIRS (Fu et al., 2018). In a recent study by Mettig et al. (2022), UV+IR wavelength retrievals from two newer satellite sensors TROPOMI and CrIS were combined to retrieve tropospheric O_3 vertical profiles. The combined UV+IR TROPOMI/CrIS O_3 profile retrievals were evaluated in the troposphere using a small sample (2 of the 8 systems) of ground-based lidar remote-sensing observations from the Tropospheric Ozone Lidar Network (TOLNet) and demonstrated that the combined UV+IR retrievals were more consistent with observations compared to the UV-only product (Mettig et al., 2022).

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One of the primary goals of TOLNet is to validate tropospheric O_3 retrievals from the recently-launched NASA Tropospheric Emissions: Monitoring of Pollution (TEMPO) geostationary satellite mission (Chance et al., 2013; Zoogman et al., 2017). TEMPO will retrieve O_3 profiles, along with partial columns including lowermost tropospheric (0–2 km above ground level (agl)) values, using combined UV (290–345 nm) and visible (VIS, 540–650 nm) wavelengths (Natraj et al., 2011; Chance et al., 2013; Zoogman et al., 2017). UV+VIS retrievals of O_3 profiles have increased sensitivity to O_3 in the lower troposphere when compared to UV-only sensors (Natraj et al., 2011; Zoogman et al., 2017). TEMPO will provide 1-2 hour averaged tropospheric column, 0-2 km partial columns, and O_3 profiles at a high spatial resolution of 8.0 km × 4.5 km.

This study builds upon Mettig et al. (2022) to demonstrate the full capability of TOLNet to validate satellite O_3 retrievals at multiple vertical levels in the troposphere. This study applies all available TOLNet systems to conduct a more robust validation of the UV-only TROPOMI, IR-only CrIS, and UV+TIR TROPOMI/CrIS O_3 profile retrievals. The manuscript is organized in the following manner: Sect. 2 describes the TOLNet data, which serves as the primary validation data set, ozonesondes, and satellite data products applied in this study; Sect. 3 presents the results of the validation; Sect. 4 discusses the overall systematic biases and random errors of the retrievals; and Sect. 5 includes the conclusions of the study.

2 Methods

2.1 TOLNet

TOLNet was established in 2011 and consists of 8 lidar systems distributed throughout North America (Newchurch et al., 2016; https://tolnet.larc.nasa.gov/). Figure 1 shows the geographic locations of the home sites for each of the lidars making up TOLNet. The primary goals of TOLNet are to provide data for: a) understanding physicochemical processes controlling tropospheric O_3 concentrations and morphology, b) evaluation of satellite profile products retrieving tropospheric O_3 (e.g., TROPOMI and TEMPO), and c) chemical transport and air quality model evaluation. TOLNet measurements provide high vertical and temporal resolution data with minimal systematic bias (~5%) and sufficient precision between 0% to 20% depending on specific systems, time of day, altitude, and temporal/vertical averaging (Leblanc et al., 2016, 2018). These high resolution observations with minimal bias/error are a desirable satellite validation data set and have been used to evaluate and better understand O_3 profile retrievals (e.g., Johnson et al., 2018; Sullivan et al., 2022; Mettig et al., 2021, 2022). However, to-date, the full complement of TOLNet lidars have not been used to validate satellite O_3 vertical profiles.

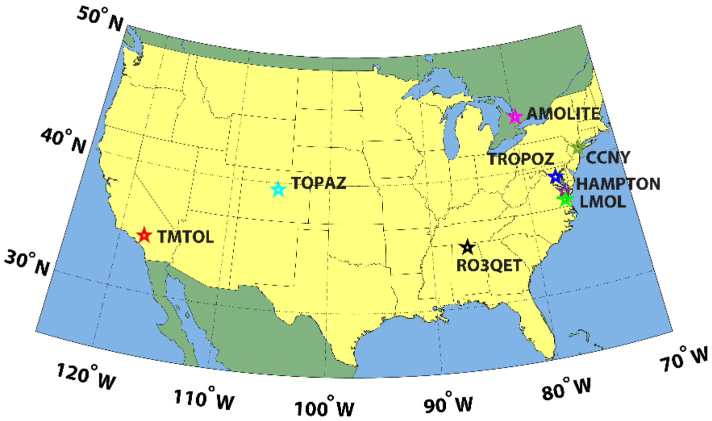

Figure 1. Locations of home stations for the lidar systems of TOLNet (https://tolnet.larc.nasa.gov/).

During the years just after the launch of TROPOMI, TOLNet lidar systems made dedicated observations during the overpass time of this spaceborne sensor (± 1 hour) for validation. For this study, there are a total of 185 TOLNet observations for validation during the time of TROPOMI/CrIS data availability between 2018-2019 (see details in Sect. 2.3). Differential Absorption Lidar (DIAL)-derived vertically resolved O_3 from 6 of the 8 TOLNet lidar systems were applied to validate UV-only TROPOMI, IR-only CrIS, and UV+TIR TROPOMI/CrIS O_3 profile retrievals. Data from the following TOLNet stations were applied: 1) NASA Jet Propulsion Laboratory (JPL) Table Mountain tropospheric ozone lidar (TMTOL) (McDermid et al., 2002), 2) NOAA Tunable Optical Profiler for Aerosol and oZone Lidar (TOPAZ) (Alvarez et al., 2011), 3) University of Alabama in Huntsville Rocket-city O_3 Quality Evaluation in the Troposphere lidar (RO_3QET) (Kuang et al., 2013), 4) Autonomous Mobile Ozone Lidar for Tropospheric Experiments (AMOLITE) (Strawbridge et al., 2018), 5) NASA Langley Mobile Ozone Lidar (LMOL) (De Young et al., 2017; Gronoff et al., 2019; Farris et al., 2019), and 6) NASA Goddard Space Flight Center mobile Tropospheric Ozone Lidar (TROPOZ) (Sullivan et al., 2014). Table 1 provides the basic information about these lidar systems used for validation during 2018-2019.

A portion of TOLNet systems are mobile (e.g., LMOL, TROPOZ, TOPAZ, AMOLITE, and RO_3QET) and were not located solely at their home stations between 2018-2019. For instance, during the summers of 2018 and 2019, LMOL took observations at NASA Langley Research Center, VA (LaRC), Hart Miller Island, MD (HMI), and Sherwood Island, CT (SIC). The majority of the lidar systems applied here were distributed throughout the United States while TROPOZ was in the Netherlands based at the Cabauw Experimental Site for Atmospheric Research (CESAR) for the 2019 TROpomi vaLIdation eXperiment (TROLIX-19) campaign (Sullivan et al., 2022). TOLNet provides observations ideal for satellite validation as the lidars are located at various locations which experience variable atmospheric composition and meteorological conditions and have different viewing conditions (e.g., surface reflectivity, system altitudes, topography, solar zenith (sza) and viewing angles). The lidars also take measurements in all seasons throughout the year providing a robust validation data set. Furthermore, the higher vertical resolution of ground-based lidars, compared to satellite profile products, make these observations ideal for validating satellite O_3 profiles in the troposphere at various altitudes.

Table 1. TOLNet lidar system and ozonesonde observation information between 2018-2019 used for satellite validation.

TOLNet

System Name	Latitude (°N)	Longitude (°E)	Elevation (m)	Observations (number of days)
TMTOL	34.38	-117.68	2285	64
TOPAZ	39.99	-105.26	1674	23
RO$_3$QET	34.73	-86.65	206	25
AMOLITE	57.18	-111.64	266	22
LMOL – LaRC	37.09	-76.38	3	23
LMOL – HMI	39.24	-76.36	6	12
LMOL – SIC	41.11	-73.34	3	6
TROPOZ	51.97	4.93	3	10

Ozonesondes

Location Name	Latitude (°N)	Longitude (°E)	Elevation (m)	Observations (number of days)
HUB	39.06	-76.88	67	7
WCT	41.11	-73.34	3	3
FLP	39.24	-73.14	4	7
HMI	39.24	-76.36	6	6
RU	40.47	-74.43	19	2
UMBC	39.25	-76.71	60	6
UAH	34.73	-86.64	196	4
GML	39.95	-105.20	1743	16

2.2 Ozonesondes

In addition to TOLNet data, ozonesonde observations from the same time period and at similar locations of the lidar systems were applied for validating satellite O_3 profiles. Ozonesonde data from launches at Howard University – Beltsville, MD (HUB), Westport, CT (WCT), Flaxpond, NY (FLP), HMI, Rutgers University, NJ (RU), University of Maryland – Baltimore Country, MD (UMBC), University of Alabama in Huntsville, AL (UAH), and the Global Modeling Laboratory (GML) in Boulder, CO were applied (see Table 1). In all, we apply 51 ozonesonde observations for validation of satellite data between 2018-2019. Similar to TOLNet data, the ozonesondes provide high vertical resolution (effective resolution ~100 m) O_3 information with high accuracy (<15% below ~20 km agl) (e.g., Witte et al., 2018; Sterling et al., 2018; Thompson et al., 2019) from locations distributed throughout the United States in regions which experience variable atmospheric composition, meteorological conditions, and spaceborne sensor viewing conditions.

2.3 Satellite O_3 profile retrievals

This study validates O_3 profiles in the troposphere retrieved with the University of Bremen Tikhonov regularised Ozone Profile retrievAl with SCIATRAN (TOPAS) algorithm which was applied to TROPOMI UV-only, CrIS IR-only, and TROPOMI/CrIS UV+IR data (Mettig et al., 2021, 2022). Mettig et al. (2021, 2022) describe the three-step iterative TOPAS retrieval in detail which is based on the first-order Tikhonov regularization approach (Tikhonov, 1963). Briefly, retrievals of O_3 vertical profiles (x_i) from the surface to 60 km asl, provided in 1 km bins, are done employing Eq. (1):

$$x_{i+1} = x_i + [\boldsymbol{K}^T\boldsymbol{S}_{\boldsymbol{y}}^{-1}\boldsymbol{K} + \boldsymbol{S}_{\boldsymbol{r}}]^{-1} \times [\boldsymbol{K}^T\boldsymbol{S}_{\boldsymbol{y}}^{-1}(y - F(x_i) - \boldsymbol{S}_{\boldsymbol{r}}(x_i - x_a)], \tag{1}$$

where index *i* denotes the iteration number. Climatological a priori O_3 profiles (x_a) are applied in the retrieval and the profile shapes are determined based on total O_3 column abundances (Lamsal et al., 2004). A priori standard deviation is assumed to be 30% and is accounted for in the regularization matrix ($\boldsymbol{S}_{\boldsymbol{r}}$) (Mettig et al., 2022) shown in Eq. (2):

$$\boldsymbol{S}_{\boldsymbol{r}} = (\boldsymbol{S}_{\boldsymbol{a}}^{-1} + \gamma\boldsymbol{S}_{\boldsymbol{t}})^T(\boldsymbol{S}_{\boldsymbol{a}}^{-1} + \gamma\boldsymbol{S}_{\boldsymbol{t}}), \tag{2}$$

where $\boldsymbol{S}_{\boldsymbol{a}}$ is a diagonal matrix containing a priori standard deviations, $\boldsymbol{S}_{\boldsymbol{a}}^{-1}$ is used as the zeroth-order Tikhonov term, γ is a scaling factor, and $\boldsymbol{S}_{\boldsymbol{t}}$ is the first-order derivative matrix (Rodgers, 2002; Rozanov et al., 2011). The forward model Jacobian matrix (K) is needed for the linearization of the ill-posed retrieval problem. $\boldsymbol{S}_{\boldsymbol{y}}$ is the error covariance matrix and is calculated with the fit residuals from the pre-processing step of the retrieval which corrects for effects not accounted for in the radiative transfer model (RTM) such as the rotational Raman scattering, polarisation correction, and secondary calibration (Mettig et al., 2022). Finally, atmospheric pressure and temperature profiles used in the retrieval are taken from ECMWF ERA-5 model simulations (Hersbach et al., 2020). For more detail about the TOPAS retrieval setup see Table 1 of Mettig et al. (2022).

TOPAS results presented in this study are based on TROPOMI Level 1 (L1) version 2.00 radiances from a pre-operational validation dataset and on CrIS Level 2 (L2) CLIMCAPS (Community Long-term Infrared Microwave Coupled Product System) full spectral resolution version 2 radiance data. The specific TOPAS product applied here has retrievals available for 12 weeks total between July 2018 to October 2019 which overlaps with the intensive TROPOMI validation measurements made by TOLNet. The 12 weeks of data include retrievals from 2 weeks every 3 months allowing for seasonal validation of TOPAS. Quality control is performed on each retrieval pixel before application in TOPAS as described in detail in Mettig et al. (2021, 2022).

2.3.1 TROPOMI UV-only retrievals

TROPOMI is a nadir-viewing spectrometer which was launched in October 2017 and has an equatorial overpass time ~13:30 (local time) and a swath width of ~2,600 km providing near daily global coverage. TROPOMI makes retrievals in the UV (270–330 nm), VIS (320–500 nm), near infrared (NIR, 675–775 nm), and shortwave infrared (SWIR, 2305–2385 nm) (Veefkind et al., 2012). The UV spectrometer has 0.5 nm spectral resolution and 0.065 nm sampling. Vertical profiles of O_3 are retrieved using two bands of UV wavelengths [i.e., UV1 (270–300 nm) and UV2 (300–330 nm)] with nadir spatial resolutions of 28.8 × 5.6 km^2 (cross track × along track) and 3.6 × 5.6 km^2, respectively. In order to be combined with the coarser data from CrIS, TROPOMI UV retrievals are degraded to the spatial resolution of 42 × 42 km^2. The TROPOMI TOPAS UV-only wavelength retrieval is described in detail in Mettig et al. (2021). The RTM used to simulate TROPOMI retrievals in the UV1 and UV2 wavelengths is SCIATRAN-V4.5 (Rozanov et al., 2011) with the assumption of a pseudo-spherical atmosphere and O_3 absorption cross sections from Serdyuchenko et al. (2014). For more detail about the TROPOMI retrieval setup see Table 2 of Mettig et al. (2022).

2.3.2 TROPOMI/CrIS UV+TIR retrievals

CrIS retrievals and combined retrievals from TROPOMI and CrIS were produced and described in detail in Mettig et al. (2022). CrIS is a Fourier-transform spectrometer launched in October 2011 and retrieves soundings in the TIR covering the SWIR (3.92–4.64 μm), middle-wave (MWIR, 5.71–8.26 μm), and long-wave infrared (LWIR, 9.14–15.38 μm) (Han et al., 2013; Strow et al., 2013) with a spectral resolution of 0.625 cm^{-1}. Ozone retrievals from the University of Bremen algorithm uses a continuous spectrum from 9350 and 9900 nm with a spectral sampling of 0.05 nm. The same RTM [SCIATRAN-V4.5 (Rozanov et al., 2011)] is applied to model the radiances in both UV and IR ranges. It is possible to combine observations from TROPOMI and CrIS since Suomi-NPP is in the same orbit as S5P and there is only a 3-minute offset in overpass times. For the O_3 profiles, the CrIS field-of-view consisting of 3 × 3 circular pixels, each with 14 km diameter at nadir, which are combined resulting in a spatial resolution of 42 × 42 km^2. For more detail about the CrIS retrieval setup see Table 3 of Mettig et al. (2022).

2.4 Evaluation technique

TOPAS O_3 profile retrievals using TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR data were evaluated using TOLNet and ozonesonde observations. The satellite retrievals were compared to raw observations and when convolved with the averaging kernel (AK) and a priori information from each retrieval using Eq. (3):

$$X_c = X_a + X_a \boldsymbol{AK} X_a^{-1}(X_t - X_a), \quad (3)$$

where X_a is the a priori O_3 profile, X_t is the TOLNet/ozonesonde data interpolated (linear) to the vertical resolution of TOPAS, and $\boldsymbol{AK}$ is the averaging kernel matrix. Statistical comparisons between co-located satellite retrievals and observations were conducted using spatiotemporal thresholds of 2.5 hours and 30 km. Sensitivity studies were conducted using coarser co-location spatiotemporal thresholds of 5 hours and 100 km to maximize the number of co-locations for statistical evaluation and to be consistent with recent TROPOMI O_3 profile validation studies (Mettig et al., 2021, 2022).

To intercompare the performance of UV, IR, and UV+IR TOPAS retrievals in idealized/controlled case studies, TOLNet lidar profiles were convolved using AKs from each of the three retrieval types and a similar calculation as in Eq. (3) except X_t is replaced with a known TOLNet O_3 profile. The TOLNet profiles are shown in Fig. 4 which represent typical clean atmospheric conditions and events of planetary boundary layer (PBL) pollution enhancements and stratospheric intrusions. The same a priori profile was used for each case to isolate the impact of the different wavelength retrieval AKs. To produce the three cases, a clean/background TOLNet lidar observation from RO$_3$QET on September 3, 2019 was applied. To perturb this same O_3 profile to represent a PBL enhancement and stratospheric intrusion, we multiplied the clean/background TOLNet lidar profile by a factor of 1.5 at and below 3 km asl and between 8 and 18 km asl, respectively. The a priori profile used in Eq. (3) was from the TOPAS retrieval for the clean/background case on September 3, 2019.

The statistical evaluation of co-located satellite data and lidar/ozonesonde observations focused on bias, normalized mean bias (NMB, see Eq. (S1)) which is normalized to the magnitude of observational data convolved with retrieval AKs, root mean squared error (RMSE, see Eq. (S2)), and simple ordinary least-squares linear regression (slope, y-intercept, coefficient of determination (R^2)). The evaluation was conducted for the partial column between

0-12 km to represent the troposphere. This vertical extent was chosen as these are the altitudes typically measured by TOLNet. Furthermore, since TOLNet and ozonesonde data provide the unique opportunity to evaluate satellite O_3 profiles in the troposphere at various vertical levels, statistics were calculated for 2 km bins between the surface and 12 km asl.

3 Results

3.1 Intercomparison of the UV, IR, and UV+IR retrievals

The AK is an important aspect of satellite retrievals and illustrates the sensitivity of the satellite measurement at any altitude to the true atmosphere (Rodgers, 2002). Examples of the mean AKs of all three retrievals are shown in Fig. 2. For all the retrievals the UV-only wavelengths have the largest sensitivity to O_3 in the stratosphere above 25 km. Below 20 km, in particular between 10-20 km, IR-only AKs are larger by up to a factor of two compared to UV-only retrievals. While IR retrievals have limited sensitivities above 25 km, it greatly improves the tropospheric sensitivity of UV+IR retrievals compared to UV-only in the troposphere. Total column DOFs (0-60 km asl) in UV+IR retrievals (5.65) are slightly larger compared to UV-only (5.01) with a ~33% increase in tropospheric DOFs (0-12 km asl) in the example shown in Fig. 2. The increases in total column and tropospheric DOFs can be even larger than this at certain times and locations as demonstrated in Mettig et al. (2022). While all retrievals have minimal sensitivity to the lowermost troposphere (0-2 km asl), UV+IR AKs have ~50% higher DOFs in the lower portion of the atmosphere compared to UV-only retrievals.

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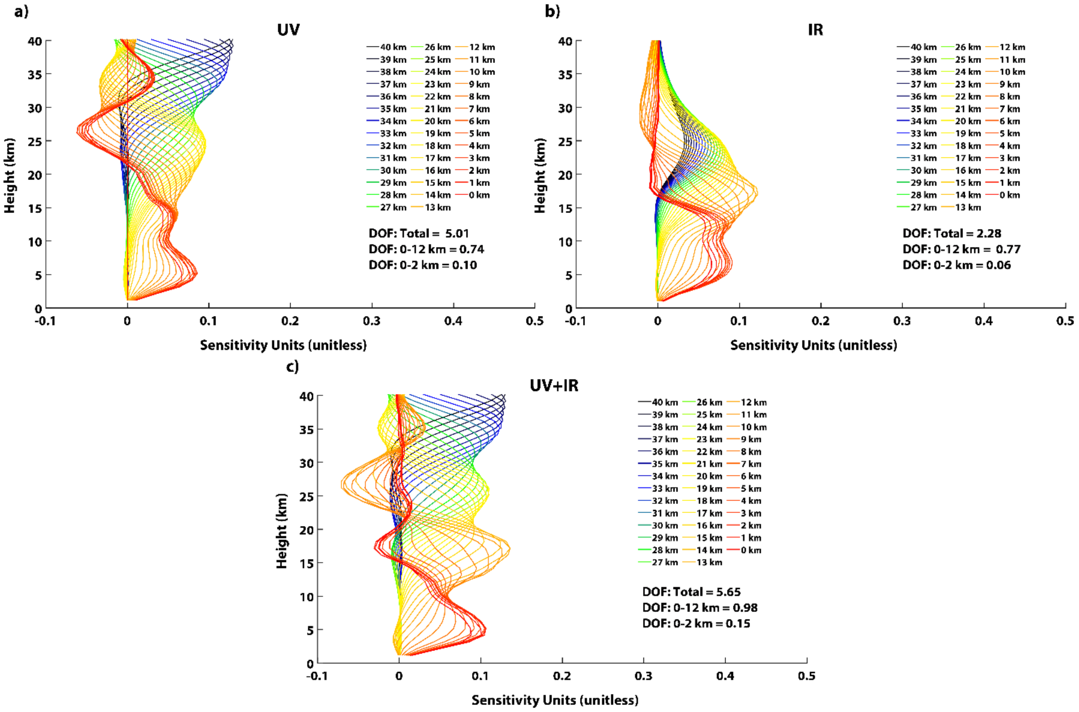

Figure 2. Mean AKs for all a) UV-only, b) IR-only, and c) UV+IR retrievals applied in this study at the location of RO$_3$QET (34.73 °N, 86.65 °W). Each line shows the AK for a particular 1 km vertical bin from the surface to 40 km asl. The figure inset shows the DOF values for the entire atmosphere (Total, 0-60 km), 0-12 km partial tropospheric column, and 0-2 km lowermost tropospheric partial column.

The vertical resolution of the retrieval is calculated by the inverse of the diagonal of the AK matrix (Rodgers, 2002) and an example for each retrieval is shown in Fig. 3. UV-only retrievals have the highest vertical resolution (between 10-12 km) above 20 km in the stratosphere. Below this altitude the UV-based retrievals have decreased vertical resolution (~20 km) with coarsest resolution at altitudes around 15 and 10 km asl. This suggests that UV-only O$_3$ profiles have limited information from the retrieval in the mid- to upper-troposphere. IR-only retrievals have limited information above 30 km asl as vertical resolution and AK values are diminished. However, below 20 km asl, IR-only retrievals have higher vertical resolution compared to UV-only data. Between 5-15 km asl IR-only retrievals have vertical resolutions as low as ~12 km. When combining UV and IR information vertical resolutions of the retrievals are much improved (8-10 km) compared to UV-only profiles below 15 km asl.

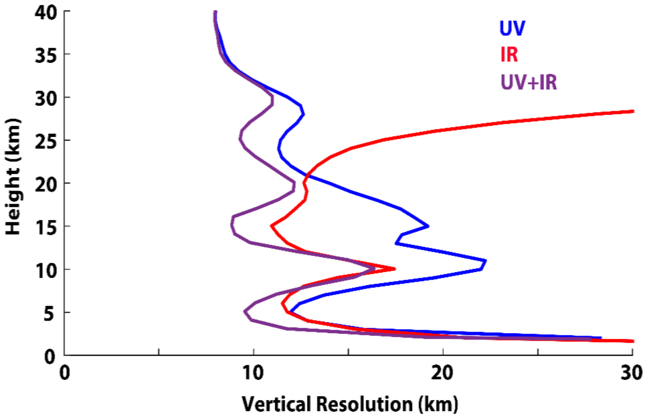

Figure 3. Average vertical resolution of the UV-only (blue), IR-only (red), and UV+IR (purple) retrievals at the location of RO3QET (34.73 °N, 86.65 °W) for each 1 km vertical bin from the surface to 40 km asl.

3.2 Intercomparison of UV, IR, and UV+IR convolved profiles

To evaluate all three retrievals in idealized and controlled case studies, we convolve known lidar profiles with each retrieval's AK and Eq. (3) for three scenarios: a) background/clean conditions, b) PBL pollution enhancement, and c) stratospheric intrusion. The results of this intercomparison for the three case studies are shown in Fig. 4. From Fig. 4a it can be seen that despite the a priori profile having a low bias throughout the troposphere compared to the truth in the clean/background conditions case, the profiles convolved with AKs from all three retrievals are able to accurately reproduce the truth. In the partial column covering the majority of the troposphere (0-12 km asl, hereinafter referred to as the tropospheric column), all convolved profiles have minimal biases <3 ppb (absolute value of NMB ≤3%) where the a priori profile had a low bias of ~-16 ppb (NMB = -27%). This suggests that all three retrievals are able to retrieve tropospheric column O_3 abundance regardless of biases in the a priori profile for typical background/clean conditions. Similar to the tropospheric column, the 0-2 km partial column (hereinafter referred to as the lowermost tropospheric column) was well reproduced by all three convolved profiles with absolute value NMBs ≤6%.

It is most interesting to see how retrievals perform in physicochemical environments which differ from typical clean/background conditions (e.g., pollution events, stratospheric intrusions, wildfires). Figure 4b and 4c demonstrate the capability of the UV-only, IR-only, and UV+IR retrievals to replicate tropospheric and lowermost tropospheric O_3 during a PBL pollution event and a stratospheric intrusion, respectively. During the PBL O_3 enhancement event, results of the convolution of the known lidar profiles with AKs from UV-only, IR-only, and UV+IR wavelength retrievals were similar to those for clean/background conditions with only slightly larger values. This small adjustment is due to these retrievals having minimal sensitivity in the lowermost troposphere. For all retrievals low biases in the lowermost troposphere of ~-40% are seen. Regardless of the inability of the retrievals to fully capture the PBL O_3 enhancement, tropospheric column biases for all profiles representative of the three retrievals had absolute values ≤13%. Overall, in comparison to the a priori profile, convolution with AKs from all three retrievals results in smaller biases throughout the troposphere. This suggests that the retrievals provide some information for studying large pollution events; however, the limited sensitivity to the lowermost troposphere largely limits these retrievals.

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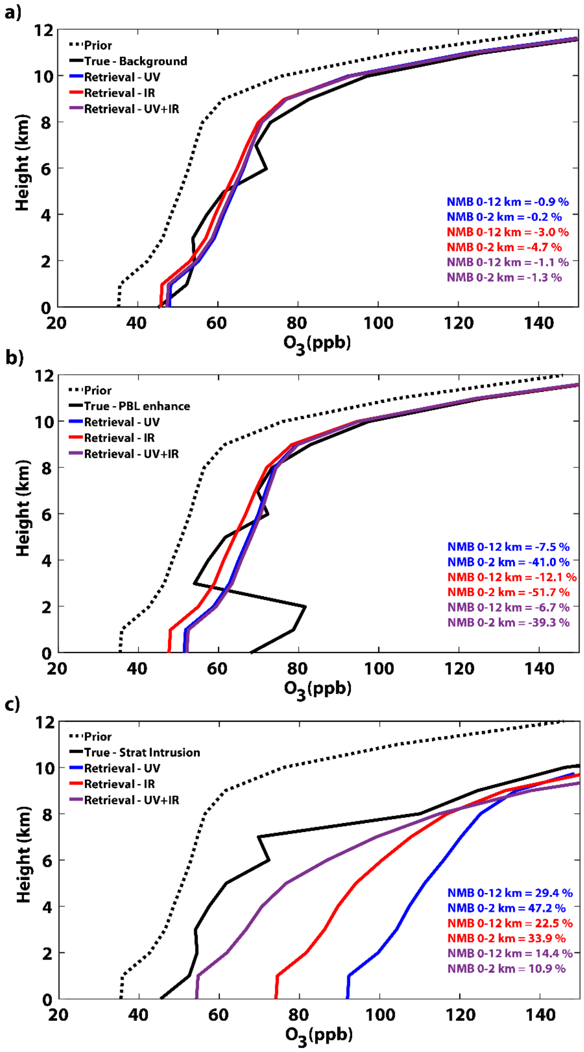

Figure 4. Lidar profiles convolved with AKs from UV-only (blue), IR-only (red), and UV+IR (purple) retrievals at the location of RO$_3$QET (34.73 °N, 86.65 °W) from the surface to 40 km asl for the case studies of: a) clean/background conditions, b) PBL pollution enhancement, and c) stratospheric intrusion. The figure inset shows the normalized mean bias (NMB) percent for the 0-12 tropospheric and 0-2 lowermost tropospheric partial column.

For a stratospheric intrusion, the lidar profiles convolved with retrieval AKs had high biases compared to the truth throughout the troposphere (see Fig. 4c). The large O_3 concentrations in the middle to upper troposphere, where all three retrievals have some sensitivity to the true atmosphere, results in NMBs between 14.4% and 47.2% for tropospheric columns. Regardless of the high biases in the convolved profiles, they still evaluate better for tropospheric column abundances compared to the a priori which had NMB = -51.2%. Compared to the truth, the profiles convolved with AKs from UV+IR retrievals replicate these dynamic O_3 profiles throughout the troposphere with the most skill. Only the UV+IR retrieval was able to replicate lowermost tropospheric O_3 (NMB = 10.9%) better compared to the a priori (NMB = -33.9%). This sensitivity study suggests that retrievals other than UV+IR data may be challenged to accurately observe O_3 profiles throughout the troposphere during times of enhanced middle- and upper-tropospheric O_3 concentrations.

3.3 Validation of TOPAS UV-only, IR-only, and UV+IR retrievals

3.3.1 Mean vertical O_3 profile validation

TOLNet was the primary data set used to validate retrievals of O_3 vertical profiles. Figure 5 shows the comparison of the three vertical O_3 profile satellite retrievals to co-located TOLNet observations at the location of all 6 lidars between 2018-2019 (statistics in Table 2). The validation with TOLNet convolved with TOPAS retrieval AKs (hereinafter TOLNet-AK) demonstrates that the TROPOMI UV-only retrieval meets the defined systematic bias requirement of ±30% (ESA, 2014) throughout the troposphere. The CrIS IR-only retrieval of O_3 profiles meets the lower systematic bias requirement of ±10% defined for this spaceborne sensor (JPSS, 2019) from the surface to ~10 km asl with absolute NMB values <-10% above this altitude. Above 10 km asl the CrIS IR-only retrievals met the higher systematic bias requirement threshold of ±20%. The combined UV+IR retrievals consistently have NMB values lower than ±10% at all altitudes in the troposphere. All three retrievals generally evaluated more consistently to lidar observations compared to the a priori profiles suggesting that the satellite O_3 profiles provide useful information for studying tropospheric composition.

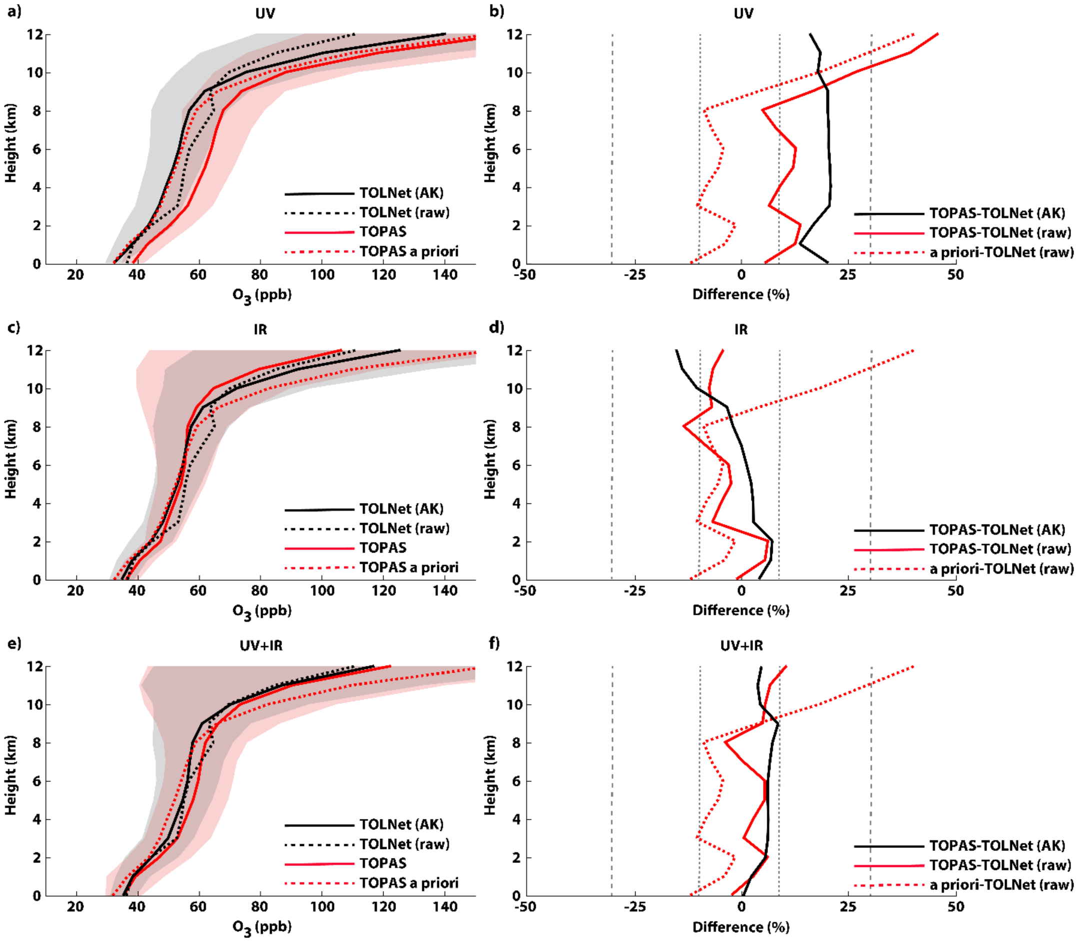

Figure 5. Vertical O_3 profile comparison of TOLNet interpolated to the satellite vertical grid (TOLNet-raw), TOLNet convolved with the TOPAS AK (TOLNet-AK), UV, IR, and UV+IR TOPAS satellite retrievals, and the a priori profile information used in the TOPAS retrieval (total number of colocations (N) = 89). The direct comparison of the profiles and percent difference for UV-only (a, c), IR-only (b, d), and UV+IR (e, f) retrievals are displayed, respectively. The percent difference between TOPAS satellite retrievals and TOLNet-AK and TOLNet-raw are labeled as TOPAS-TOLNet (AK) and TOPAS-TOLNet (raw), respectively. The percent difference between the TOPAS a priori and TOLNet-raw is labeled as a priori-TOLNet (raw). The grey and pink shaded regions illustrate the 1σ standard deviation of TOLNet-AK and satellite O_3 vertical profiles, respectively. NMB values of 30% and 10% are displayed using grey dashed and dotted lines, respectively.

Comparing the three retrievals to TOLNet observations not convolved with the retrieval AKs (hereinafter TOLNet-raw) suggests that the TROPOMI UV-only data has NMB lower than ±15% at all altitudes below 10 km asl with high biases >30% between 10-12 km asl. A general low bias in IR-only profiles compared to TOLNet-raw is determined below 12 km asl (NMB typically between -5 and -15%) except for ~ 2 km asl where a slight high bias is calculated. The combined UV+IR retrievals compare most closely to TOLNet-raw observations with NMB lower than ±10% at all altitudes. Overall, the IR-only and UV+IR satellite retrieval products evaluate more favorably to TOLNet-AK data compared to TOLNet-raw. However, UV-only profiles have higher biases when evaluated with TOLNet-AK data compared to TOLNet-raw below 10 km asl. To have a more consistent validation of the three O_3 profile retrievals we primarily used TOLNet-AK throughout the rest of the study.

For the tropospheric column, the UV-only retrievals are consistently biased high (NMB = 15-20%) compared to TOLNet-AK lidar data (see Fig. 5a, b; Table 2). The systematic high bias determined in this study agrees with the recent TOPAS validation study by Mettig et al. (2022). The UV-only retrievals are consistently higher than observations regardless of the low bias in the a priori below 10 km asl (NMB values ~-5%). Above 10 km asl the a priori has a high bias up to ~35 ppb (NMB = 24%). IR-only O_3 profiles have a small high bias in the lowest 8 km asl and a systematic low bias up to -12% above this altitude. This low bias in the middle to upper troposphere determined in this study agrees with the recent work by Mettig et al. (2022). Finally, the UV+IR retrievals have minimal bias throughout the troposphere with NMB values ranging from 1% to 8% demonstrating minimal dependance on altitude.

Table 2. Statistical validation of TOPAS UV, IR, and UV+IR retrievals with convolved TOLNet-AK observations. All observations and satellite retrievals were co-located using a 2.5 hour and 30 km threshold criteria.

Prior

Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	91	-1.7	-8.0	14.2	0.05
2-4 km	172	-5.1	-6.0	14.5	-0.02
4-6 km	172	-2.8	-7.0	12.5	0.09
6-8 km	159	-5.0	-5.8	18.3	0.16
8-10 km	126	7.3	-2.4	29.9	0.19
10-12 km	84	34.9	23.8	62.8	0.82
Trop. Column	804	1.9	-1.0	26.8	0.47

UV-only

Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	91	6.2	16.3	10.4	0.47
2-4 km	172	9.6	18.0	14.6	0.14
4-6 km	172	10.4	20.1	16.0	0.20
6-8 km	159	10.9	19.7	16.8	0.47
8-10 km	126	12.5	19.5	18.2	0.80
10-12 km	84	19.6	17.5	28.2	1.02
Trop. Column	804	11.2	18.5	17.4	0.96

IR-only

Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	91	2.6	5.4	6.3	0.61
2-4 km	172	1.3	4.9	6.5	0.54
4-6 km	172	0.8	2.4	7.5	0.62
6-8 km	159	-0.5	0.5	8.4	0.76
8-10 km	126	-4.6	-2.7	12.2	0.90
10-12 km	84	-15.9	-12.1	21.2	0.89
Trop. Column	804	-1.7	-0.3	10.5	0.97

UV+IR					
Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	91	1.5	1.3	10.1	0.57
2-4 km	172	3.2	5.8	11.7	0.46
4-6 km	172	3.4	6.2	12.3	0.45
6-8 km	159	4.0	6.4	11.9	0.61
8-10 km	126	4.2	7.8	15.6	1.00
10-12 km	84	4.4	4.1	23.2	1.03
Trop. Column	804	3.5	5.3	14.0	0.92

Ozonesondes were also used to validate TOPAS O_3 retrievals. Figure 6 shows the comparison of the three vertical O_3 profile satellite retrievals to co-located ozonesonde observations at the locations displayed in Table 1 between 2018-2019 (statistics in Table 3). Ozonesondes convolved with retrieval AKs (hereinafter Ozonesondes-AK) when compared to TROPOMI UV-only retrievals suggest these retrievals meet the defined systematic bias requirement of ±30% (ESA, 2014) throughout the troposphere. CrIS IR-only retrievals compared most favorably to ozonesondes meeting the lower systematic bias requirement of ±10% defined for this spaceborne sensor (JPSS, 2019). The combined UV+IR retrievals had NMB values <30% at all altitudes in the troposphere. All three satellite retrieval types performed better than the a priori profile product, further suggesting that the satellite O_3 profiles provide useful information for tropospheric composition. Overall, the comparison of TROPOMI UV-only and CrIS IR-only to both TOLNet and ozonesondes suggests these two satellite sensors meet the systematic bias criteria identified for the O_3 vertical profile products in the troposphere.

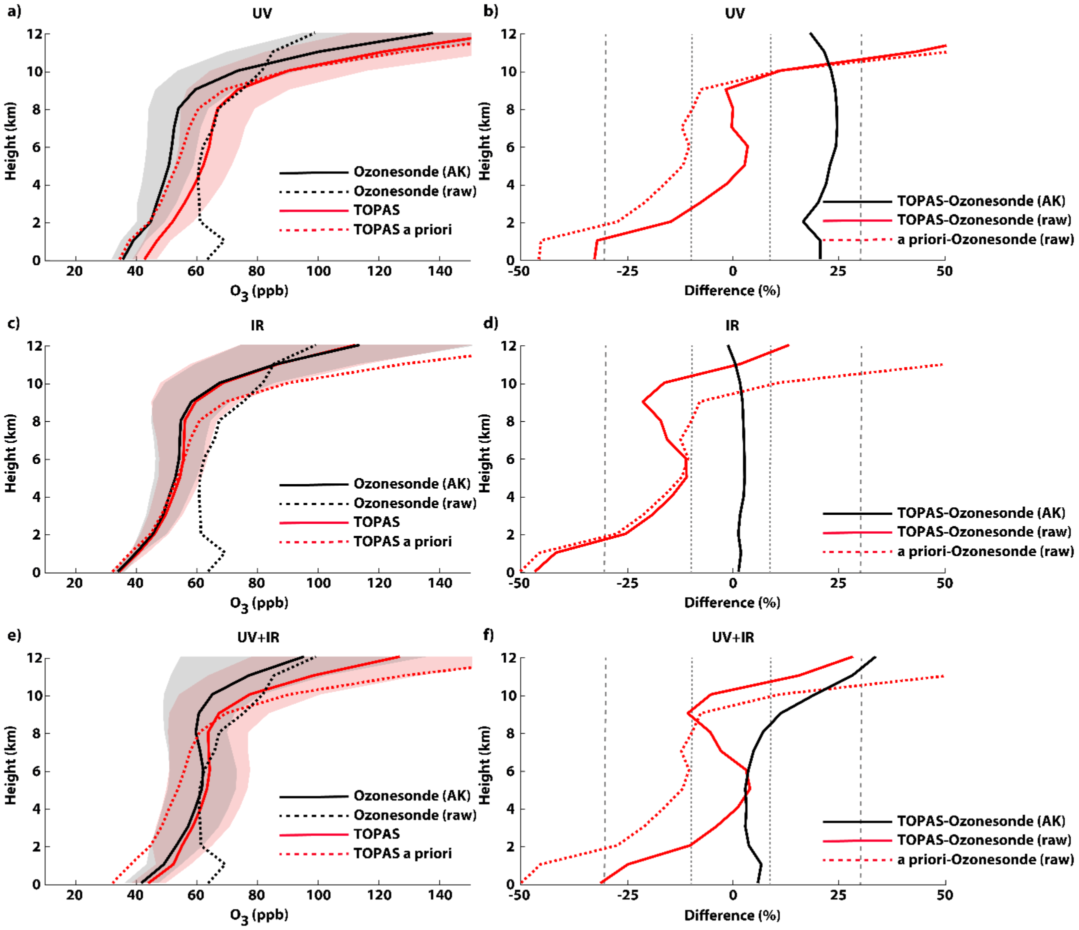

Figure 6. Vertical O_3 profile comparison of ozonesondes interpolated to the satellite vertical grid (Ozonesonde-raw), ozonesondes convolved with the TOPAS AK (Ozonesonde-AK), UV, IR, and UV+IR TOPAS satellite retrievals, and the a priori profile information used in the TOPAS retrieval (total number of colocations (N) = 26). The direct comparison of the profiles and percent difference for UV-only (a, c), IR-only (b, d), and UV+IR (e, f) retrievals are displayed, respectively. The percent difference between TOPAS satellite retrievals and Ozonesonde-AK and Ozonesonde-raw are labeled as TOPAS-Ozonesonde (AK) and TOPAS-Ozonesonde (raw), respectively. The percent difference between the TOPAS a priori and Ozonesonde-raw is labeled as a priori-Ozonesonde (raw). The grey and pink shaded regions illustrate the 1σ standard deviation of Ozonesonde-AK and satellite O_3 vertical profiles, respectively. NMB values of 30% and 10% are displayed using grey dashed and dotted lines, respectively.

Comparing the three satellite profile retrievals to ozonesonde observations not convolved with the retrieval AKs (hereinafter Ozonesonde-raw) suggests TROPOMI UV-only data has NMB lower than ±10% between 3 and 10 km asl; however, much higher biases above and below these altitudes (see Fig. 6). A low bias in IR-only profiles compared to Ozonesonde-raw was determined below 11 km asl (NMB between -10 and -50%) with a small positive bias above this altitude. The combined UV+IR retrievals compare most closely to Ozonesonde-raw observations with NMB lower than ±10% at all altitudes below 11 km and above 2 km asl. The low bias in all three satellite retrievals in the lowermost troposphere is caused by the lack of sensitivity to O_3 at these altitudes not allowing the retrievals to replicate the larger O_3 concentrations observed by Ozonesonde-raw data. In general, the satellite retrievals compare more consistently to Ozonesonde-AK instead of Ozonesonde-raw.

In the tropospheric column the UV-only retrievals are consistently biased high (NMB = 22%) compared to Ozonesonde-AK data (see Fig. 6a, b; Table 3). This systematic high bias is consistent with the validation using TOLNet-AK observations. IR-only O_3 profiles compare very well to Ozonesonde-AK data with NMB values <3% throughout the troposphere. This outperforms the IR-only profiles when compared to TOLNet-AK data. Finally, the UV+IR retrievals have minimal bias below 10 km asl with NMB values <10%; however, with a high bias aloft. The overall validation of the three satellite O_3 profile retrievals using ozonesondes was generally consistent compared to when using TOLNet. Therefore, the rest of this study focuses on the validation using the lidar network observations.

Table 3. Statistical validation of TOPAS UV, IR, and UV+IR retrievals with convolved ozonesonde observations. All observations and satellite retrievals were co-located using a 2.5 hour and 30 km threshold criteria.

Prior

Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	49	-21.8	-45.7	28.2	-0.01
2-4 km	47	-10.4	-24.2	12.5	0.26
4-6 km	50	-8.3	-14.2	12.8	0.25
6-8 km	50	-5.8	-11.4	13.9	0.33
8-10 km	50	4.7	-8.7	18.8	0.71
10-12 km	50	64.4	30.8	75.3	1.28
Trop. Column	296	1.5	-12.2	29.5	0.78

UV-only

Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	49	7.4	20.2	8.6	0.88
2-4 km	47	9.8	18.0	11.5	0.90
4-6 km	50	11.8	22.1	14.1	0.75
6-8 km	50	12.8	24.1	15.1	0.87
8-10 km	50	15.4	24.0	19.0	1.12
10-12 km	50	22.9	22.1	32.1	1.07
Trop. Column	296	12.9	21.8	13.9	1.01

IR-only

Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	49	0.5	1.6	3.8	0.81
2-4 km	47	1.1	1.5	4.8	0.71
4-6 km	50	1.5	2.7	5.7	0.74
6-8 km	50	1.4	2.7	6.8	0.80
8-10 km	50	1.2	2.4	12.0	0.86
10-12 km	50	-0.3	1.2	29.3	0.66
Trop. Column	296	0.8	2.0	6.1	0.85

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UV+IR					
Vertical Level	N (#)	Bias (ppb)	NMB (%)	RMSE (ppb)	Slope
0-2 km	49	2.3	6.1	6.7	0.79
2-4 km	47	1.7	3.2	8.0	0.91
4-6 km	50	1.9	2.9	8.1	0.96
6-8 km	50	3.5	4.1	8.2	1.03
8-10 km	50	9.5	9.0	17.0	1.05
10-12 km	50	26.4	23.5	47.8	0.64
Trop. Column	296	7.2	8.1	11.4	0.93

3.3.2 Impact of co-location criteria on mean vertical O_3 profile validation

To determine the impact of using coarser spatiotemporal co-location criteria (5 hours and 100 km) for satellite O_3 profile validation, more consistent with recent TROPOMI validation studies (Mettig et al., 2021, 2020), we conducted a sensitivity study validation of the mean vertical O_3 profiles using the coarser co-location criteria. Figure S1 shows the comparison of the three vertical O_3 profile satellite retrievals to co-located TOLNet-AK observations at the location of all 6 lidars between 2018-2019 using the coarser spatiotemporal co-location criteria (statistics in Table S1). The coarser co-location criteria resulted in a larger amount of co-location data points for evaluation. While this results in a more reliable statistical evaluation, the validation of all three satellite retrievals is consistent using both the fine and coarse spatiotemporal co-location criteria. Given the consistent validation, and the fact that representation error between ground-based and satellite data is minimized when applying the finer co-location criteria, including the fact that tropospheric O_3 can experience rapid changes during the daylight hours, we feel the finer co-location criteria are more appropriate for this validation.

3.3.3 Validation at different vertical levels in the troposphere using TOLNet

A major advantage of using TOLNet for validation of satellite O_3 profile retrievals is the ability to make observations at different vertical levels of the troposphere over an entire day or more. Figure 7 shows the direct comparison of all co-located satellite and TOLNet-AK O_3 profiles for six separate 2-km vertical layers between the surface and 12 km asl. In the lowest altitudes between the surface and 2 km asl all three retrievals perform similarly; however, the UV-only retrieval has a higher bias and slightly more spread (see bias and RMSE statistics in Table 2). All three retrievals have limited sensitivity to these lower tropospheric regions and are primarily driven by retrieval performance above these altitudes and the shape of the a priori profile. Adding the IR wavelengths, which adds additional sensitivity to the lower- to mid-tropospheric regions, improves performance in the UV+IR retrieval compared to the UV-only.

Above the lowest portions of the troposphere, the three retrievals have more sensitivity to O_3 and differ more in their performance. Between 2-4 km the UV+IR and IR-only retrievals outperform UV-only retrievals (less bias and RMSE) due to the enhanced sensitivity provided by the IR wavelengths. Additionally, the UV+IR and IR-only retrievals have much better linear regression slopes compared to the UV-only product (UV-only results have similar slopes as the a priori profile below 6 km) due to the ability to deviate further from the a priori profile shape. In the vertical layer between 4-6 km, similar to the layer between 2-4 km, the UV+IR and IR-only retrievals outperform UV-only retrievals with less bias and RMSE and better linear regression slopes. Similar comparisons are seen in the vertical layer between 6-8 km. Overall, between 2-8 km asl, IR retrievals have the least bias and spread, along with best linear regression fits. UV+IR retrievals are similar to IR-only data with only slightly worse performance when compared to TOLNet-AK. Both the retrievals using IR wavelengths outperform the UV-only retrievals between 2-8 km asl.

In the upper troposphere (8-12 km asl), UV-only retrievals still display the largest positive bias of all three retrievals; however, the spread in the data between all three profile products are more similar compared to the altitudes below 8 km asl. All three products have linear regression slopes near unity; however, IR-only retrievals have a noticeable low bias above 8 km asl. In general, the IR-only and UV+IR retrievals had the least bias out of all three retrievals between 8-10 km and 10-12 km, respectively, when compared to TOLNet-AK and IR-only data has the least spread in the 8-12 km vertical level when compared to observations.

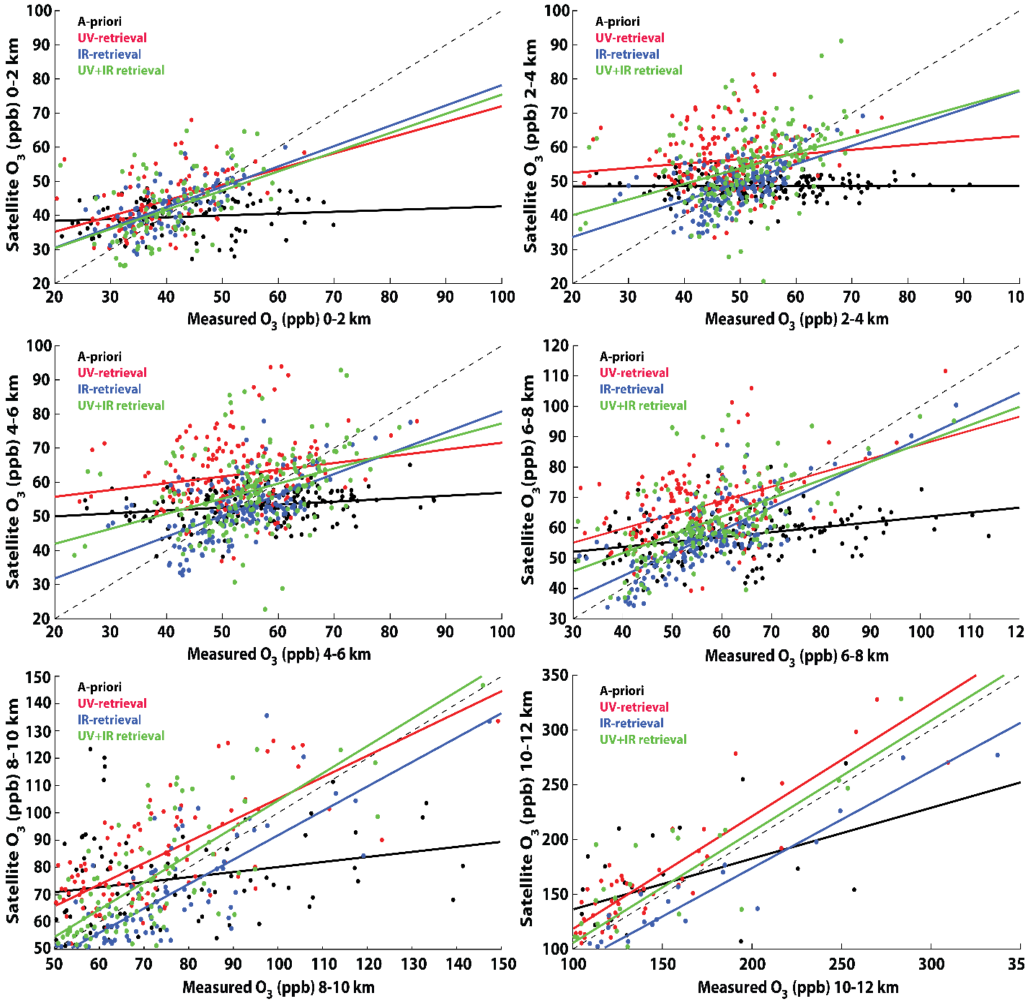

Figure 7. Scatter plot comparison of co-located TOPAS UV-only (red), IR-only (blue), combined UV+IR (green) retrievals, and a priori O_3 vertical profiles to TOLNet observations in 2-km vertical layers between the surface and 12 km asl. The satellite profiles are compared to TOLNet-AK and the a priori data is compared to TOLNet-raw. The solid-

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colored lines illustrate the linear regression fit of each satellite-TOLNet comparison and the dashed line represents the 1:1 fit line. Statistics of the intercomparison at each vertical level are presented in Table 2.

At all altitudes in the troposphere the retrievals typically evaluate better (lower bias and RMSE values) to observations compared to the a priori product. The linear regression slope provides information about the capability of the retrieval to deviate from the prior profile shape and magnitudes. Below 8 km the UV-only retrieval has similar linear regression fits compared to the a priori emphasizing the limited sensitivity of these wavelengths to O_3 in the lower troposphere. In the lower- to mid-troposphere the IR wavelengths provide additional DOFs which allow the IR and UV+IR retrieval to deviate further from the prior profile shape and compare better to observations. Above 8 km all three retrievals have similar linear regression slopes which are able to deviate to some degree from the a priori shape and compare better to observations. While neither of the three retrievals have more than 1.0 DOFs below 12 km asl, the information provided by all retrievals improves upon the prior vertical profile suggesting these satellite data provide useful information for studying tropospheric O_3.

3.3.4 TOLNet validation of seasonal vertical O_3 profiles

A seasonal validation of the three TOPAS O_3 profiles retrievals was performed using TOLNet-AK lidar observations. Figure 8 shows the comparison of satellite retrievals and lidar profiles divided into meteorological season (i.e., winter (DJF), spring (MAM), summer (JJA), and fall (SON)). The number of co-locations are limited during the winter and spring ($N < 10$) outside of the primary O_3 season covering the summer and fall. More robust observational coverage by TOLNet is apparent in the summer ($N = 34$) and fall ($N = 41$). Given the limited number of co-located observations in the winter and spring available for this study, the statistical validation of the satellite retrievals during these months should be viewed as relatively uncertain.

During the winter months, UV+IR retrievals compared most closely to TOLNet-AK observations. This combined retrieval is the only satellite product which validates better to observations compared to the a priori below 8 km asl. The NMB of the UV- and IR-only profiles are >10% throughout the majority of the tropospheric column while UV+IR retrievals have NMB values <7% from the surface to 12 km asl. The prior profile and UV-only retrievals have similar unresolved errors/uncertainties (RMSE) of ~24 ppb throughout the tropospheric column, suggesting the UV-only product was unable to improve upon the a priori information. The random errors in the retrievals including IR wavelengths (e.g., IR-only and UV+IR) had lower RMSE values of ~13 ppb.

In the spring months all three retrievals evaluated more consistently to observations compared to the a priori profiles. At all altitudes in the troposphere the IR-only profiles compared the best to observations with NMB values <10%. The two retrievals which incorporate UV wavelengths had larger positive biases compared to the IR-only data with NMB values between 10-15% and 15-25% for the UV+IR and UV-only vertical profiles, respectively. IR-only retrievals in the spring had the lowest bias and random error (RMSE = 7 ppb). UV-only retrievals also had lower random errors compared to the a priori data source (RMSE = 21 ppb) with unresolved errors of ~14 ppb. UV+IR retrievals in the spring had moderate systematic biases and the largest unresolved errors of all three retrievals (~19 ppb).

During the summer, UV+IR had the lowest biases (within ±10%) above 2 km asl compared to other retrievals and the a priori. The UV-only retrievals had a constant systematic high bias of 10-15% throughout the entire troposphere. The IR-only retrievals had variable biases below 8 km while above this altitude displayed a large negative bias. All three retrievals had smaller RMSE values compared to the a priori of 17 ppb, 11 ppb, and 14 ppb for the UV-only, IR-only, and UV+IR retrievals, respectively. Overall, all three satellite retrievals had smaller bias and uncertainties compared to the a priori profiles for the summer months.

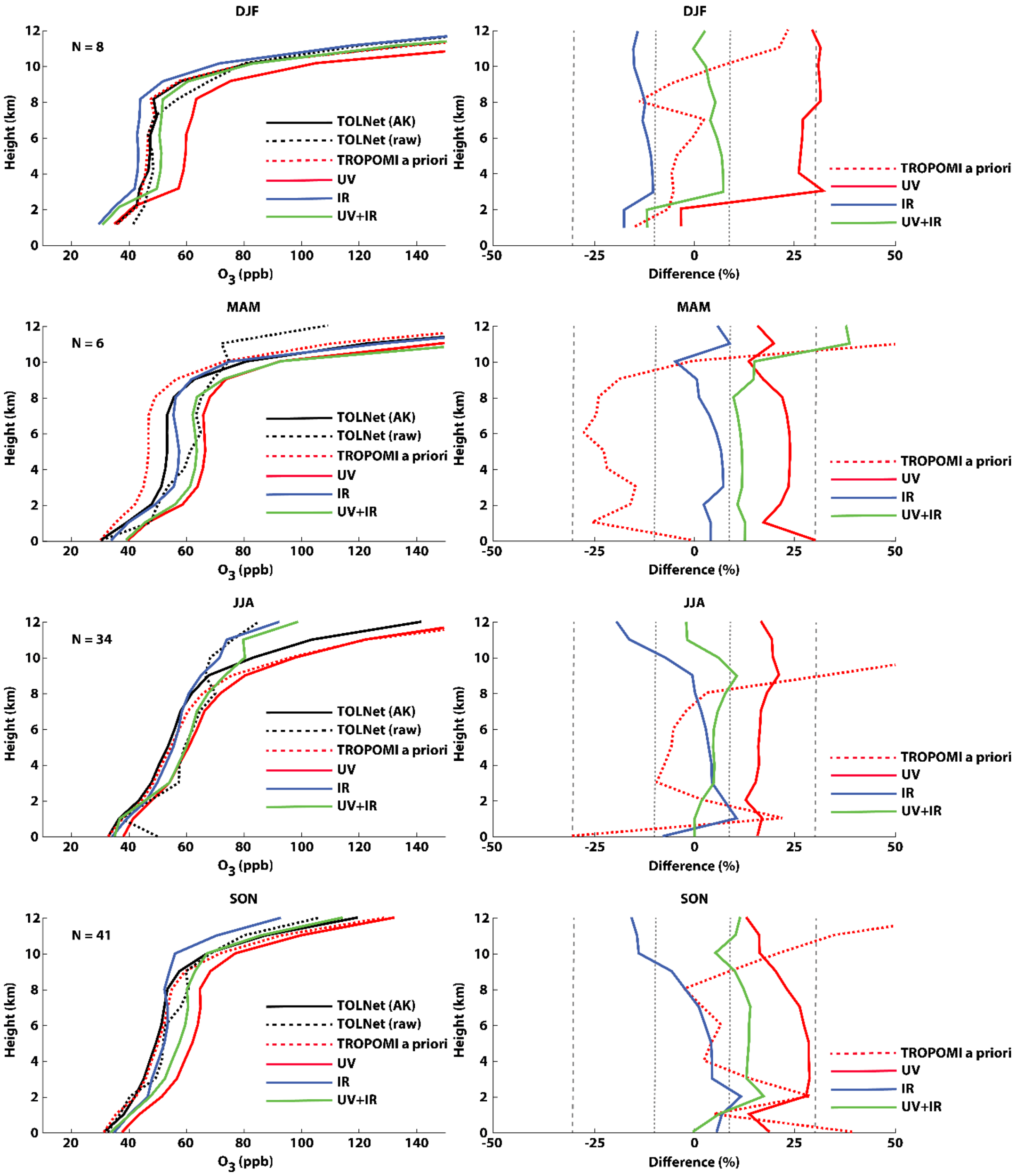

Figure 8. Seasonally-averaged vertical O_3 profile comparison of TOLNet interpolated to the satellite vertical grid (TOLNet-raw), TOLNet convolved with the TOPAS AKs (TOLNet-AK), UV, IR, and UV+IR TOPAS satellite retrievals, and the a priori profile information. The direct comparison of the profiles (left column) and percent difference (right column) for

UV-only, IR-only, and UV+IR retrievals compared to TOLNet-AK are displayed, respectively. NMB values of 30% and 10% are displayed using grey dashed and dotted lines, respectively. The total number (N) of co-located profiles are shown in the figure inset.

At all altitudes in the troposphere during the fall months the UV+IR wavelength profiles compared the best to observations with NMB values <10%. UV-only retrievals had consistent high biases between 10-20% while the IR-only profiles had good performance below 6 km and a large negative bias aloft. All three retrievals had smaller RMSE values compared to the a priori of 16 ppb, 10 ppb, and 13 ppb for the UV-only, IR-only, and UV+IR retrievals, respectively. Similar to the summer months, during the fall all three retrievals had noticeably lower random errors compared to the a priori profiles.

4 Discussion of retrieval bias and uncertainties

To determine a retrieval product's accuracy, it is important to quantify systematic bias and random errors. Figure 5 and Table 2 illustrate each of the three retrieval's systematic biases represented by NMB values when validated with TOLNet-AK observations. TOPAS UV-only retrievals have a consistent positive bias ranging between 16-20% throughout the entire troposphere. The systematic high bias in the UV-only data can partially be explained by biases and shape of the a priori vertical profiles (e.g., Kulawik et al., 2006; Zhang et al., 2010; Johnson et al., 2018). Due to the limited sensitivity of the UV-only retrieval in the upper troposphere, the a priori high bias in the upper troposphere (see Fig. 5) contributes to the UV-only systematic bias throughout the troposphere. This upper tropospheric bias driven by errors in the a priori profile shape and magnitude agrees with results from other recent TROPOMI O_3 profile validation studies (e.g., Sullivan et al., 2022). The addition of the IR-wavelength retrievals to UV-only data was shown to improve the TOPAS retrieval throughout the troposphere. Systematic biases from the IR-only retrievals were minimal (NMB <6%) in the lowest 10 km of the troposphere with the product having a negative bias between 10-12 km (NMB = -12%). A similar low bias in the TOPAS IR-only retrievals when compared to AK-convolved observations was identified in Mettig et al. (2022). When combining the UV and IR wavelengths, the TOPAS retrieval has minimal systematic bias throughout the troposphere with NMB values ranging between 1% and 8% when validated with TOLNet-AK observations. The additional sensitivity in the troposphere in the UV+IR retrieval, compared to the UV-only TOPAS product, resulted in the lower systematic.

The RMSE values in Table 2 represent the random errors in the daily TOPAS O_3 profile retrievals when validated with TOLNet-AK observations. While systematic biases were significantly reduced in the three retrievals compared to the a priori profiles, random errors still remained elevated in most instances. UV-only retrievals had unresolved errors ~35% less compared to the a priori. However, the average RMSE values for this retrieval product still remained large (~17 ppb) throughout the troposphere (compared to ~27 ppb for the a priori). IR-only retrievals displayed the least unresolved errors of all three retrievals with average RMSE values throughout the troposphere of ~10.5 ppb which is ~60% less compared to the a priori. The combined UV+IR profiles had average RMSE values ~14 ppb, ~50% less compared to the a priori, throughout the troposphere. The fact that unresolved errors are reduced in all three retrievals compared to the a priori further emphasizes that satellite O_3 profiles can provide useful information in the troposphere. However, given that unresolved errors of daily profiles on average still remain large (>10 ppb) the accuracy of these satellite products still suffer due to the limited sensitivity of spaceborne sensors to tropospheric O_3.

In addition to the a priori profile shape, sza and surface albedo were determined in this study to be additional controlling factors for systematic bias. All three retrievals had similar bias impacts from sza and surface albedo, so here we discuss the analysis of UV+IR retrievals only. When comparing TOPAS UV+IR retrievals to all co-located raw TOLNet retrievals it was determined that the daily averaged bias was 14.4 ppb. When separating this for high (>60°) and low (<60°) sza it was found that systematic biases were larger (18.3 ppb) for high sza conditions compared to low sza (13.1 ppb). The dependance of satellite O_3 profile retrievals in the troposphere on sza has also been shown in recent TROPOMI and TOPAS validation studies (e.g., Mettig et al., 2021, 2022). As sza values become large the sensitivity of the retrieval in the troposphere are reduced, leading to increased biases in the satellite products. Surface albedo has also been demonstrated to be a controlling factor for the accuracy of tropospheric O_3 retrievals. This validation study using raw TOLNet observations further confirms this. When separating the TOPAS validation for high (>0.2) and low (<0.2) albedo values it was found that systematic biases were larger (16.4 ppb) for low albedo conditions compared to high surface reflectivity (12.8 ppb). RMSE values, representative of unresolved errors in the retrievals, were similar for high and low sza and surface albedo conditions.

5 Conclusions

This study applied the full complement of TOLNet observations (6 out of 8 systems were operational between 2018-2019) to validate UV-only TROPOMI, IR-only CrIS, and UV+TIR TROPOMI/CrIS TOPAS O_3 profile retrievals. TOLNet proved to be a vital validation tool for satellite tropospheric O_3 retrievals. This data source provides: a) highly accurate, high temporal resolution, O_3 observations for multiple continuous hours and/or days, b) retrievals with minimal dependance on a priori information, and c) profiles with higher vertical resolution compared to satellite products. The multi-hour observations provided by TOLNet will be important for validation of tropospheric O_3 profiles and the lowermost tropospheric (0-2 km) partial columns from the recently-launched geostationary TEMPO mission. As a primary validation data source for TEMPO, TOLNet will make dedicated validation observations for this geostationary sensor during all times of the day. These observations will provide hourly tropospheric O_3 observations during all seasons which will greatly increase the amount of data from TOLNet needed for seasonal validation which was not available for this study.

TOLNet was used to intercompare the three retrievals, using idealized case studies by convolving high resolution lidar profiles with retrieval-specific AKs of TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR based on the TOPAS algorithm of the University of Bremen. All three retrievals were determined to be able to reproduce typical/background O_3 profiles. However, the results differed more for physicochemical environments which deviate from typical clean/background conditions. Retrievals using combinations of wavelengths proved to be more capable of capturing conditions with air quality impacts such as pollution events and stratospheric intrusions. UV+IR O_3 profiles most accurately observed O_3 profiles throughout the troposphere during times of enhanced middle- and upper-tropospheric O_3 concentrations such as what occurs during stratospheric intrusions. For near-surface O_3 pollution conditions, all three retrievals were not able to accurately replicate enhancements in the lowermost troposphere due

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to minimal sensitivity to this portion of the atmosphere. However, UV+IR retrievals did have the least bias of all three satellite products throughout the troposphere and lowermost troposphere during times of PBL-level O_3 enhancements. The reason that combined wavelength retrievals (UV+IR) outperform the single wavelength data products (UV, IR) is the increased vertical resolution and sensitivity to O_3 in the troposphere aiding in the ability to deviate further from the a priori profile shape.

TOPAS O_3 profiles from TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR retrievals were validated with TOLNet and ozonesonde observations. The validation results using the two observational data sets were overall consistent. Compared to TOLNet-AK, UV-only TROPOMI retrievals had mean biases which meet the defined systematic bias requirement of ±30% (ESA, 2014) throughout the troposphere. The CrIS IR-only retrieval of O_3 profiles meet the lower systematic bias requirement of ±10% defined for this spaceborne sensor (JPSS, 2019) from the surface to ~10 km asl and above 10 km asl the CrIS IR-only retrievals met the higher systematic bias requirement threshold of ±20%. Finally, the combined UV+IR retrievals consistently had NMB values lower than ±10% at all altitudes in the troposphere. The primary drivers of systematic biases were determined to be the biases in a priori vertical profile shape, sza, and surface albedo. The accuracy of all three retrievals tend to be degraded with increasing sza and lower surface albedo values.

Just as important as systematic bias, this study validates the TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR TOPAS retrievals for daily unresolved errors. Random error (uncertainty) requirements for TROPOMI UV O_3 profile retrievals are ±10% (ESA, 2014) and ±25% for CrIS IR profiles in the troposphere (JPSS, 2019). The validation of UV-only, IR-only, and UV+IR retrievals using the TOLNet-AK observations resulted in troposphere-averaged RMSE values of 19.8%, 12.6%, and 14.6%, respectively. UV-only profiles evaluated here do not meet the uncertainty requirements defined by the ESA. CrIS IR-only retrievals do meet the uncertainty requirements defined by the mission. The ability of the retrievals to deviate from the a priori profile shape assumption is key to lower systematic bias and unresolved error. UV-only retrievals have the least sensitivity to O_3 in the troposphere leading to posterior vertical profiles with nearly identical shape compared to the prior (see Fig. 5). The improved sensitivity of IR wavelengths to O_3 in the troposphere allows IR-only and UV+IR retrievals to deviate further from the shape of the a priori resulting in lower systematic biases and unresolved errors (see Fig. 5 and Table 2).

The results of this validation study can be used to understand the biases and random errors associated with TROPOMI UV, CrIS IR, and TROPOMI/CrIS UV+IR retrievals. While this study is specific to the TOPAS algorithm it reflects the overall accuracy and precision of TROPOMI and CrIS O_3 vertical profile in the troposphere. The satellite retrievals provide useful information for understanding tropospheric O_3; however, the sensitivity of TROPOMI UV-only retrievals is still a limiting factor for accurately assessing variability in tropospheric O_3. IR-retrievals from CrIS provide enhanced sensitivity to tropospheric O_3; however, it is limited by the coarse spatial resolution of the sensor and lack of sensitivity to O_3 in the stratosphere. Combining these retrievals improves the ability to observe tropospheric O_3 to some degree. Future work should consider combination of individual satellite retrievals in order to improve the sensitivity of spaceborne retrievals of O_3 in the troposphere. TOLNet will make dedicated observations for TEMPO validation to evaluate the combination of UV+VIS wavelengths. The UV+VIS retrievals have enhanced lowermost tropospheric sensitivity, and in combination with the sensor's high spatiotemporal resolution, should provide important spaceborne information of tropospheric column and lowermost tropospheric O_3. An important result of this study was showing that TOLNet is a sufficient validation data source for satellite retrievals since TOLNet has been identified as the primary data source for validation of TEMPO O_3 in the troposphere.

Data availability. The TOLNet data used for the satellite data validation is available for download (https://tolnet.larc.nasa.gov/, last access September 6, 2023). The TOPAS satellite retrievals are available upon request to the corresponding author and University of Bremen coauthors. Ozonesonde data can be downloaded for GML (https://gml.noaa.gov/dv/data/index.php?category=Ozone&type=Balloon&site=BLD, last access December 19, 2021), HUB (https://www-air.larc.nasa.gov/cgi-bin/ArcView/owlets.2018?SONDE=1, last access January 22, 2022), HMI (https://www-air.larc.nasa.gov/cgi-bin/ArcView/owlets.2018?SONDE=1, last access January 22, 2022), UMBC (https://www-air.larc.nasa.gov/cgi-bin/ArcView/owlets.2018?SONDE=1, last access January 22, 2022), FLP (https://www-air.larc.nasa.gov/cgi-bin/ArcView/listos?GROUND-FLAX-POND=1, last access January 30, 2022), WCT (https://www-air.larc.nasa.gov/cgi-bin/ArcView/listos?GROUND-WESTPORT=1, last access January 30, 2022), and RU (https://www-air.larc.nasa.gov/cgi-bin/ArcView/listos?GROUND-RUTGERS=1, last access January 30, 2022). Ozonesondes from UAH can be acquired by email to the corresponding author.

Author contributions. MSJ, JS, and MJN were responsible for acquiring the funding for this study. MSJ designed the technical methods and performed the experiments. AR, MW, and NM developed and produced the satellite retrieval data applied in this study. JS, MJN, SK, TL, FC, TAB, GG, RJA, AOL, CJS, GK, BC, and LT were instrumental in obtaining and providing the lidar data used for validation. MSJ prepared the manuscript with contributions from all coauthors.

Competing interests. At least one of the (co-)authors is a member of the editorial board of Atmospheric Measurement Techniques.

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