IMPROVED TROPOMI HCHO COLUMN VALIDATION USING DUAL-SCAN MAX-DOAS RETRIEVALS

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ABSTRACT

Formaldehyde (HCHO) is the most abundant aldehyde in the atmosphere. HCHO ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements have been performed in Uccle, Belgium during one year and a half from May 2018 to December 2019. The MAX-DOAS instrument was measuring in a dual-scan viewing mode consisting of one scan in elevation in a main azimuthal direction followed by an azimuth scan at a fixed low elevation angle. Aerosols and HCHO vertical profiles are retrieved in the main azimuthal direction by applying a profile inversion algorithm based on the Optimal Estimation Method. With the aid of these vertical profiles and the dual-scan MAX-DOAS retrieval strategy, near-surface HCHO concentrations and columns are retrieved in all directions of the azimuth scan. This ground-based dataset is then used for two main purposes: (1) the characterization of the HCHO horizontal distribution around the station and (2) the validation of HCHO columns measured by the TROPOMI satellite instrument over the Brussels area. This study reveals that (1) the HCHO horizontal distribution differs per season and (2) the use of dual-scan MAX-DOAS measurements improves the TROPOMI HCHO column validation results.

Index Terms— Formaldehyde, Brussels, MAX-DOAS, satellite, TROPOMI, validation

1. INTRODUCTION

Formaldehyde (HCHO) is an important atmospheric constituent mainly produced from the photochemical oxidation of methane (CH_4) and non-methane volatile organic compounds (NMVOCs) emitted in the atmosphere from natural (vegetation), anthropogenic (industrial activities), and pyrogenic (biomass burning) sources. HCHO is therefore often used as an indicator of biogenic and anthropogenic emissions of hydrocarbons [1]. HCHO plays also a critical role in tropospheric chemistry by influencing the formation of tropospheric ozone (O_3) and aerosols. Additionally, even at low concentrations, HCHO can have a direct impact on human health, such as headache, fatigue, and eye and throat irritation. For those reasons, monitoring the regional and global distribution of HCHO is important.

Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) instruments perform spectroscopic observations of scattered sunlight in the visible (Vis) and ultra-violet (UV) spectral ranges at multiple elevation angles towards the horizon and when possible in different azimuthal directions. This allows the retrieval of the vertical and horizontal distribution of aerosols and trace gases in the troposphere [2]. HCHO is one of the many tropospheric species, which can be measured by MAX-DOAS instruments [3].

MAX-DOAS measurements are valuable for the validation of satellite nadir air-quality observations, which provide daily global tropospheric column measurements of various atmospheric species. Since 1995 and the launch of the GOME instrument [4] on the ERS-2 platform, many satellite instruments dedicated to atmospheric composition monitoring have followed, one of the most recent being TROPOMI (TROPOspheric Monitoring Instrument) launched onboard the Sentinel-5p Precursor (S5P) platform. With an unprecedented spatial resolution of 3.5x7.0 km^2 (5.5x3.5 km^2 since 06 August 2019), TROPOMI is ideally suited for air quality measurements at the scale of medium and large cities. This high resolution is a clear advantage, but it also introduces new retrieval challenges that require suitable validation means.

The main motivation of this work is to investigate whether MAX-DOAS HCHO measurements performed in several azimuthal directions can improve the validation of the TROPOMI HCHO product [5-6] in urban conditions. To this aim, we use observations in Uccle (Brussels-Capital Region, Belgium) as a case study.

2. MEASUREMENT SITE AND EXPERIMENTAL SET-UP

Brussels-Capital Region is the most populated area of Belgium and, as a result, pollutants concentrations, such as NO_2, often exceed the European standard upper limit.

A MAX-DOAS instrument has been continuously operated by BIRA-IASB (Koninklijk Belgisch Instituut voor Ruimte-Aeronomie – Institut royal d'Aeronomie Spatiale de

Belgique) in Uccle from January 2017 to February 2020. This measurement site is located in the South of the Brussels city-center and at the East of the Soignes forest. Therefore, it is an ideal measurement site to study moderate to high pollution levels. From March 2018 onward, the MAX-DOAS instrument operated on a dual-scan viewing mode. One complete measurement scan is composed of two different modes: (1) a vertical scan of nine different elevation angles and one fixed azimuthal direction (Northeast direction, i.e., towards the City Centre) and (2) a horizontal scan of nine different azimuthal directions at a fixed elevation angle (2º above the horizon) [7].

3. METHODOLOGY

3.1. DOAS analysis

The radiance spectra measured in the complete dual-scan are analyzed with the QDOAS spectral fitting software [8]. The primary product of this analysis is the HCHO differential slant column density (dSCD), which is the difference between the effective light-path integrated HCHO concentration and the amount of the absorber (HCHO) in a measured reference spectrum. For tropospheric studies, a zenith spectrum (different for each scan) is chosen as a reference in order to remove the contribution of the stratosphere in the so-called off-axis dSCDs, i.e. the dSCDs corresponding to the different elevation angles above the horizon. In the present study, the zenith spectrum of each scan is used as reference.

HCHO dSCDs are retrieved in the 336-359 nm spectral range using the Meller and Moortgat (2000) HCHO cross section [9]. This wavelength region is chosen because it contains three strong HCHO absorption bands.

3.2. Aerosol and HCHO vertical profile retrievals

For each MAX-DOAS elevation scan in the main azimuthal direction, the aerosol extinction coefficient and HCHO vertical profiles are retrieved by applying the Mexican MAX-DOAS Fit (MMF) inversion algorithm to the measured oxygen dimer (O_4) and HCHO dSCDs [10]. The aerosol extinction profile of each vertical scan derived from the O_4 dSCDs is used as input to derive the HCHO vertical profile, as the aerosol content and vertical distribution provide information about the effective light-path in the probed atmosphere. Each aerosol and HCHO retrieval is quality checked based on the degrees of freedom (DOFs), the difference between the measured and simulated O_4 and HCHO dSCDs and the aerosol optical density (AOD) value [7].

As it will be presented in the next sections, the HCHO vertical profiles are also used to estimate the Mixing Layer Height (MLH) of HCHO, which is an effective height defining the atmospheric layer in which HCHO is expected to be uniformly distributed.

3.3. Dual-scan HCHO MAX-DOAS retrieval in Uccle

The dual-scan HCHO MAX-DOAS retrieval is an adaptation of the dual-scan MAX-DOAS retrieval, which was successfully applied to NO_2 retrieval in Uccle, Belgium [7]. It consists in the conversion of the measured trace gas dSCDs at 2° elevation into (1) near-surface trace gas box-averaged Volume Mixing Ratios (VMRs) from the surface to the MLH and (2) trace gas vertical column densities (VCDs) [11].

The measured HCHO dSCDs in one low elevation angle provides information on the near-surface concentration. As mentioned before, the measured dSCDs is the light-path integrated concentration of HCHO. Consequently, the knowledge of the light-path's length is crucial to derive the near-surface HCHO VMRs. O_4 can be used as a tracer for the effective light-path in the atmosphere: as its concentration is well-known (it varies with the square of O_2 concentration), its variation can be attributed to changes in the light-path due to the presence of particles like aerosols and clouds.

However, the direct use of the O_4 light-path's length in the HCHO retrieval is not possible under moderate to high pollution conditions, such as those in Brussels, because the profile shapes of O_4 and HCHO are not the same. For this reason, radiative transfer model (RTM) simulations are necessary in order to estimate a unitless correction factor, which will account for these profile shape differences (see [7]). Those simulations are performed for eight different MLH values of aerosols and HCHO in the range of 500-2000 m and different measurement viewing geometries (Solar Zenith Angle (SZA), Relative Azimuth Angle (RAA) and the corresponding elevation angle of 2º). For every MAX-DOAS measurement, one value of the correction factor is given according to its viewing geometry and MLH value during the measurement. The MLH is derived from the HCHO vertical profiles in the main azimuthal direction and is defined as the ratio of the HCHO VCD to the near-surface concentration of HCHO [7].

The verification of the dual-scan HCHO MAX-DOAS retrieval is conducted by comparing the retrieved dual-scan HCHO VMR and VCD to the same quantities as retrieved by the MMF inversion algorithm in the main azimuthal direction. Both methods are highly correlated with a correlation coefficient equal to 0.98 for both HCHO VMRs and VCDs.

4. RESULTS

4.1. Dual-scan HCHO seasonal variation

The seasonal variation of the MAX-DOAS HCHO VCDs for the period May 2018-December 2019 is presented in Figure 1. Those VCDs correspond to seasonal mean HCHO values in every azimuth direction. It should be noted that only HCHO VCDs measured around TROPOMI overpass time (±1 hour) have been considered. The length of each line corresponds to the seasonally mean HCHO representative horizontal distance, as estimated by the dual-scan HCHO MAX-DOAS retrieval (see [7]). Note that the range of the color-coding differs per season for better visualization purpose.

The HCHO VCDs have a clear seasonal cycle, with maximum values during summer and a minimum during winter. To investigate the temperature dependence of HCHO VCDs, HCHO VCDs are plotted against surface temperature data, as measured by the BIRA-IASB meteorological station in Uccle (Figure 2). The increase of temperature during summer causes HCHO formation from isoprene, which is mostly emitted at forested areas under high temperature and radiation conditions [12]. As presented in Figure 2, higher temperature values are associated with higher HCHO VCDs. The maximum HCHO VCDs during summer is attributed to the temperature-dependent production of HCHO from biogenic sources, which is consistent with the location of the VCD peak towards the Soignes forest, a large forested area in the East/South-East of Brussels (Figure 1).

In order to investigate the role of the different wind regimes in the observed HCHO VCDs per season, the retrieved HCHO VCDs in the main azimuthal direction are classified in different groups according to the wind direction. During summer, the maximum HCHO VCDs are retrieved when the wind is blowing from the Southeast direction (only forested area; free from important anthropogenic sources), which indicates that biogenic sources dominate the HCHO production. During winter, the wind direction related to the maximum HCHO VCDs is N/NE, which suggests that anthropogenic sources provide a significant contribution to the HCHO production given the location of anthropogenic sources (city center, industrial area, and airport) in this direction.

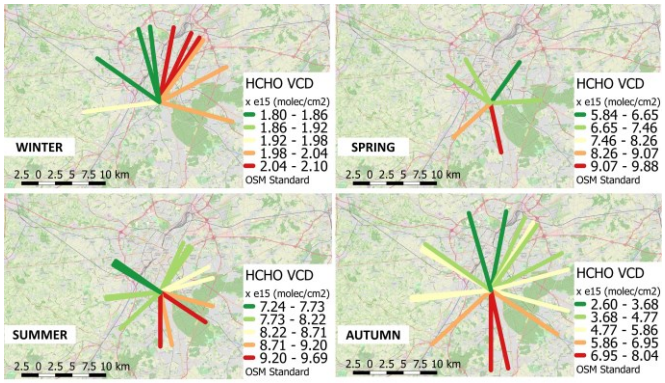

Figure 1. Seasonally mean HCHO VCDs in each azimuthal viewing direction. © OpenStreetMap contributors 2021. Distributed under a Creative Commons BY-SA License.

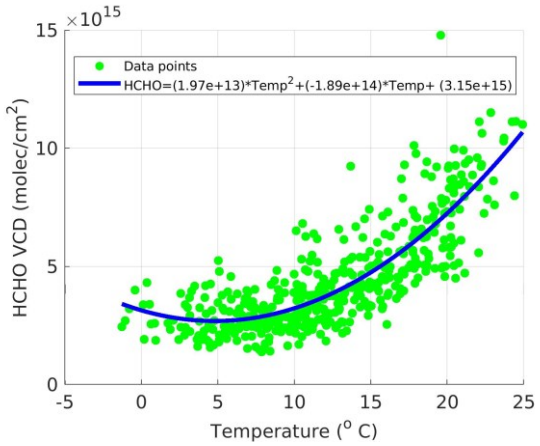

Figure 2. Scatter plot of surface temperature data and MAX-DOAS HCHO VCDs**.** The color bar represents the temperature data, which is the same as in x axis.

4.2. Validation of TROPOMI HCHO columns

4.2.1. Use of one MAX-DOAS azimuthal direction measurements

To investigate if the dual-scan MAX-DOAS HCHO measurements improve the TROPOMI HCHO column validation results in urban conditions, a first comparison between the MAX-DOAS HCHO VCDs derived in the main azimuthal direction by applying the MMF inversion algorithm, and daily-mean TROPOMI HCHO columns is done. Pixels in a radius of 20km around the measurement site are averaged and compared to mean MAX-DOAS HCHO VCDs extracted within ± 1 hour of the TROPOMI overpass time (~13h30).

Figure 3a presents the comparison results between satellite and 1-D ground-based data during one year and a half. The dataset is highly correlated (R ± probable error = 0.72 ± 0.02) and the slope value (± its uncertainty) is equal to 0.76 (± 0.05). The median relative bias between satellite and 1-D ground-based data is –6 %.

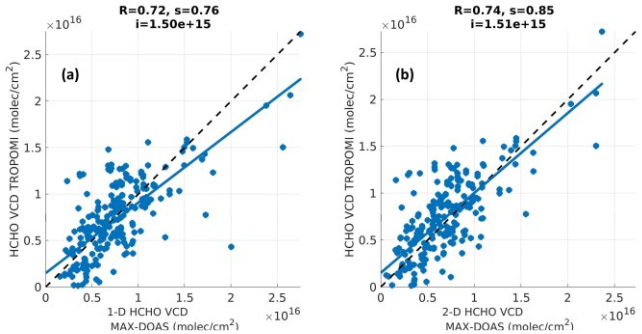

Figure 3. Scatter plots between TROPOMI HCHO columns and (a) 1-D MAX-DOAS observations and (b) dual-scan MAX-DOAS observations during one year and a half.

4.2.2. Use of dual-scan MAX-DOAS measurements

Here, the dual-scan MAX-DOAS HCHO VCDs are compared with TROPOMI HCHO columns. HCHO VCDs in every MAX-DOAS azimuthal direction around TROPOMI overpass time (±1 hour) are averaged and compared to daily-mean TROPOMI HCHO columns (inside a radius of 20km as in Subsection 4.2.1).

Figure 3b shows that the agreement between satellite and MAX-DOAS improved when using the dual-scan dataset instead of the MAX-DOAS observations in the main azimuthal direction, with a slope value (± its uncertainty) of 0.85 (± 0.05) instead of 0.76 (± 0.05). The correlation coefficient value (0.74 ± 0.02 instead of 0.72 ± 0.02) is slightly improved. The median relative bias between satellite and 2-D ground-based data is +7 %, which is slightly higher than the median relative bias in Section 4.2.1 (in absolute value: 7% instead of 6%). This finding indicates that the amelioration of the spatial coincidence between ground-based and TROPOMI observations improves the agreement between both HCHO VCD column data sets in terms of correlation coefficient and slope values, but increases the bias of the comparison.

5. CONCLUDING REMARKS

In the present study, one year and a half (May 2018-December 2019) of MAX-DOAS measurements is used to validate TROPOMI HCHO columns above Brussels.

MAX-DOAS HCHO VCDs are retrieved in several azimuthal directions by applying the dual-scan MAX-DOAS retrieval strategy. The adopted experimental set-up allows to better characterize the HCHO horizontal distribution and its variability. Higher HCHO VCDs are observed during summer, because of the high temperature dependency of HCHO. Anthropogenic sources are the main contributors of HCHO emissions during cold months, while during warmer months HCHO is released in the troposphere from both anthropogenic and biogenic sources, which leads to higher HCHO VCDs.

The 1-D and dual-scan MAX-DOAS HCHO VCDs are compared with TROPOMI HCHO columns above the Brussels-Capital Region during the period May 2018-December 2019. The two comparison exercises reveal that the use of the dual-scan MAX-DOAS HCHO measurements improves the TROPOMI HCHO column validation results under urban conditions.

Future steps include a detailed investigation of the remaining TROPOMI HCHO columns underestimation compared to ground-based MAX-DOAS measurements and more precisely, the role of the *a priori* HCHO profile shape used in the TROPOMI HCHO column retrieval.

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