

## CARBON MONOXIDE, METHANE AND CARBON DIOXIDE OVER CHINA RETRIEVED FROM SCIAMACHY/ENVISAT BY WFM-DOAS

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### ABSTRACT

The three “carbon gases” carbon monoxide (CO), methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>) are important atmospheric constituents affecting air quality and climate. We present maps of time averaged (yearly, tri-monthly) CO, CH<sub>4</sub> and CO<sub>2</sub> over China for the year 2003. They are derived from the near-infrared nadir observations of the SCIAMACHY spectrometer aboard the European environmental satellite ENVISAT using the scientific WFM-DOAS retrieval algorithm (version 0.4 for CO<sub>2</sub>, version 0.5 for CO and CH<sub>4</sub>). The maps show vertical columns of carbon monoxide and dry-air column averaged mixing ratios of methane (denoted XCH<sub>4</sub>) and CO<sub>2</sub> (XCO<sub>2</sub>). The SCIAMACHY near-infrared nadir observations are nearly equally sensitive to concentration changes at all altitude levels including the boundary layer and therefore enable the detection of surface source regions of carbon monoxide, methane, and CO<sub>2</sub>. The measurement errors for CO and methane are small enough to clearly identify at least moderate to strong source regions. The XCO<sub>2</sub> measurements are more difficult to interpret because the measurement error due to, e.g., aerosol and albedo variability, is currently on the order of the weak source/sink signal to be detected. Our future work will aim at reducing the sensitivity to these and other error sources.

### 1. INTRODUCTION

Air pollution resulting from large-scale biomass burning, fossil fuel combustion, and other fossil fuel related activities has become a problem with increasing importance, especially for countries with an increasing energy demand and fuel consumption such as China. The quantification of concentrations near the sources and the subsequent transport of pollutants is important, for example, for monitoring and forecasting of air pollution. Increasing concentrations of radiatively active so-called greenhouse gases are expected to result in a warmer climate with possible adverse consequences such as rising sea levels and an increase of extreme weather conditions. China emits the greenhouse gas methane in large quantities, e.g., due rice cultivation, ruminants and waste handling (see, e.g., [15] and references given therein). Also large amounts of the air pollutant CO are emitted due to, e.g., fossil fuel combustion and biomass burning.

Carbon monoxide contributes to air pollution because it is toxic in large concentrations, acts as a pre-cursor to tropospheric ozone and - because CO is the leading sink of the hydroxyl radical (OH) - largely determines the self-cleansing efficiency of the troposphere (see, e.g., [2] and references given therein). CO is highly variable in time and space and detailed monitoring of its spatial pattern and time evolution is therefore important.

Carbon dioxide and methane are the two most important anthropogenic greenhouse gases and contribute to global climate change. A prerequisite to predict future climate change resulting from emissions of carbon dioxide and methane is a good understanding of their (surface) sources and sinks. Information on CO<sub>2</sub> and methane sources and sinks on the global scale are currently derived from a highly precise but rather sparse (~100) network of ground stations (e.g., NOAA/CMDL). Satellite measurements have the potential to overcome the limitations of the surface network and help to obtain a better understanding of the methane and CO<sub>2</sub> sources and sinks (see, e.g., [10,11,15] and references given therein). For the near future dedicated satellite missions are planned to globally measure carbon dioxide and methane accurate and precise enough to obtain information on their surface sources and sinks, e.g., OCO (USA) and GOSAT (Japan), both being primarily passive near-infrared nadir missions. SCIAMACHY [3] is not a dedicated carbon mission but due to its near-infrared nadir observation capability is the first satellite instrument that is very sensitive to methane, CO<sub>2</sub>, and CO boundary layer concentration changes as demonstrated by its averaging kernels for CO [9], methane [10] and CO<sub>2</sub> [10]. Therefore, SCIAMACHY plays a pioneering role in this new area of satellite remote sensing.

Here we present new results for CO and methane obtained with version 0.5 of the WFM-DOAS retrieval method. The CO<sub>2</sub> results have been obtained with WFM-DOAS version 0.4 described in detail in previous papers [10,11]. This paper is organized as follows: first, a short overview about the SCIAMACHY instrument is given followed by a description of the retrieval algorithm and, finally, a discussion of the trace gas results focusing on China.

## 2. SCIAMACHY INSTRUMENT

SCIAMACHY is a spectrometer that measures reflected, scattered and transmitted solar radiation in the spectral region 214-2380 nm at moderate spectral resolution (0.2-1.6 nm). On the Earth's day side SCIAMACHY mainly performs a sequence of alternating nadir and limb observations. The horizontal resolution of the nadir measurements depends on orbital position and spectral interval but is typically 60 km (e.g., for methane) or 120 km (e.g., for CO) across track times 30 km along track. These measurements can be inverted to obtain a large number of (primarily) atmospheric data products [3].

Overall, the in-flight optical performance of SCIAMACHY is as expected from the on-ground calibration and characterization activities [4]. One exception is a time dependent optical throughput variation in the SCIAMACHY near-infrared (NIR) channels 7 (the main CO<sub>2</sub> channel) and 8 (the only CO channel and main CH<sub>4</sub> channel) due to ice build-up on the detectors [4] which adversely affects the trace gas retrieval [11,16,19]. This effect is minimized by regular heating of the instrument. The methane results presented in this paper have been derived from channel 6 not affected by an ice layer. CO is retrieved from channel 8 using a correction procedure for ice induced errors (see next section for details).

## 3. RETRIEVAL ALGORITHM AND ITS VALIDATION

The retrieval of a long-lived and therefore relatively well-mixed gas such as methane is extremely challenging as only small variations on top of a large background are of relevance in order to obtain information on its surface sources. Therefore, the retrieval algorithm has to be very accurate. In addition, the algorithm also has to be very fast to process huge amounts of data. We have developed the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) retrieval algorithm to accomplish this task. Basically the same algorithm is also used to retrieve CO and CO<sub>2</sub>. Because accuracy and speed are typically contradicting requirements a good compromise has to be found. In this context it is relevant to point out that other groups are also work on this challenging retrieval task using somewhat different approaches (see, e.g., [14,15,16,17]).

WFM-DOAS [5,7-11] is an unconstrained linear-least squares method based on scaling (or shifting) pre-selected vertical profiles. The fit parameters for the trace gases are directly the desired vertical columns. The logarithm of a linearised radiative transfer model plus a low-order polynomial is fitted to the logarithm of the ratio of a measured nadir radiance and solar irradiance spectrum, i.e., observed sun-normalized radiance. The WFM-DOAS reference spectra are the logarithm of the sun-normalized radiance and its derivatives computed with a radiative transfer model [6]. In order to avoid time-consuming on-line radiative transfer simulations, a fast look-up table scheme has been implemented.

The validation of our data products with independent ground based (FTS) vertical column measurements is an ongoing activity. Available studies [13,18,19,20] report on the validation of previous versions of our data products (e.g., methane v0.4/0.41 and CO v0.4) and/or on a comparison of a small sub-set of the data as for CO<sub>2</sub> [13,20]. A detailed comparison of our new version 0.5 data products has not yet been performed but will be available in the near future.

### 3.1 CO specific aspects

Carbon monoxide is retrieved from a small spectral fitting window located in channel 8 covering several absorption lines of CO. Compared to our initial version 0.4 CO [9,11] the version 0.5 data product presented here has been significantly improved. The v0.5 CO column product is retrieved from an optimised spectral fitting window and generated without the application of a scaling factor (the v0.4 product was scaled with the factor 0.5). In addition, a correction has been applied to reduce time dependent biases caused by the ice-issue (e.g., slit function change), to improve the retrieval for partially cloud covered scenes and to make the retrieval less sensitive to aerosol and albedo variability. The correction is based on the retrieved methane column obtained from the same fitting window. A correction factor, defined as the ratio of an assumed (a-priori) methane column and the retrieved methane column, is applied to the retrieved CO column. The a-priori methane column is computed using a single (scene independent) profile of methane but taking into account the surface elevation of the corresponding ground pixel. This approach makes use of the fact that the variability of the methane column is small compared to the variability of the CO column. A quality flag is set for each pixel to indicate a (potentially) successful measurement. To decide if a measurement is successful, a number of criteria have been defined based on the value of the root-mean-square of the fit residuum, the CO fit error, and the methane correction factor. The methane correction factor, for example, has to be close to 1.0 (within 20%) for a measurement to be classified as successful. Otherwise the disturbances due to clouds, aerosols, surface reflectivity, calibration issues, etc., are considered too large to be corrected for.

A detailed validation by comparison with ground based FTS measurements has not yet been performed but is planned for the future. To assess the quality of this data product we have compared monthly mean v0.5 CO columns with the

operational CO column product of MOPITT/EOS-Terra [12] (L2V3 data product obtained from NASA Langley DAAC). We found good to reasonable agreement. For example, both sensors show a similar seasonal dependence of CO originating from biomass burning in Africa and South America (not shown here). Our quantitative comparison revealed that the SCIAMACHY v0.5 CO columns (over land and water) are on average systematically higher compared to MOPITT by about  $0.2 \times 10^{18}$  molecules/cm<sup>2</sup> (northern hemisphere: ~16%). The inter-hemispheric difference is in good agreement (the mean difference (SCIA-MOPITT) is -3%, the correlation coefficient is 0.9). For a number of reasons (e.g., different altitude sensitivity, spatial resolution, and measurement time), perfect agreement is not to be expected.

### 3.2 Methane specific aspects

For methane we derive dry air column averaged mixing ratios (denoted XCH<sub>4</sub>) by normalizing the retrieved methane column by the simultaneously observed airmass estimated by retrieving the column of a reference gas whose column is less variable than methane. Initially (for v0.4) we used oxygen (O<sub>2</sub>) columns retrieved from the O<sub>2</sub> A band (760 nm) [10,11]. Unfortunately, the sensitivity of the retrieved O<sub>2</sub> column with respect to, e.g. aerosol, is quite different compared to methane mainly because of the large spectral distance between the two fitting windows. For v0.41 XCH<sub>4</sub> [11] we use CO<sub>2</sub> retrieved from the 1.6  $\mu$ m region (channel 6). The new v0.5 XCH<sub>4</sub> data product is derived in a similar way as the v0.41 data product, except that methane is retrieved from channel 6 instead of channel 8. There are two reasons why methane retrieval from channel 6 should give better results compared to channel 8: (i) channel 6 is not affected by the ice issue, and (ii) the channel 6 methane absorption band is located close to the channel 6 CO<sub>2</sub> band thus improving the cancellation of errors when the ratio is computed (see also [15] where a similar approach has been used for the same reasons).

We have performed an initial comparison of v0.5 XCH<sub>4</sub> with TM5 model simulations [1] performed at the EC Joint Research Centre, Ispra, Italy (similar as described in [11] for XCH<sub>4</sub> v0.4/v0.41) and found in general good agreement, typically within a few percent. Under certain conditions the observed XCH<sub>4</sub> is significantly lower than the model XCH<sub>4</sub>: over Antarctica observations less than 1560 ppbv occur and north of 20 deg North values below 1670 ppbv are found, in particular during January to March and around October. More investigations are needed to find out what the reason for this potential underestimation is. Likely candidates are snow/ice covered surfaces due to their low albedo in the NIR. Excluding the low values from the comparison results in a mean difference of the global daily data of less than 2% for nearly all days of the year 2003 (nearly constant low bias of SCIAMACHY of about 1%). The standard deviation of the difference is in the range 1.5-2.5% and the correlation coefficient is typically higher than 0.6. For each ground pixel a quality flag is set using similar criteria as for CO.

### 3.3 Carbon dioxide specific aspects

Absorption bands of CO<sub>2</sub> are covered by the SCIAMACHY channels 7 (~2  $\mu$ m) and 6 (~1.6  $\mu$ m). Because channel 7 is affected by a changing ice-layer on the detectors we currently retrieve CO<sub>2</sub> from channel 6. The WFM-DOAS version 0.4 CO<sub>2</sub> data products are the absolute CO<sub>2</sub> column (in molecules/cm<sup>2</sup>) and the dry air column averaged mixing ratio of carbon dioxide XCO<sub>2</sub> (in ppmv) obtained by normalizing the CO<sub>2</sub> column by the observed airmass obtained from the simultaneously retrieved O<sub>2</sub> column derived from the O<sub>2</sub>-A-band. Details on our version 0.4 CO<sub>2</sub> retrievals are given in [10,11]. Two points should be mentioned here: (i) we have aimed at rejecting as good as possible all even slightly cloud contaminated ground pixels from the data shown here and (ii) that the WFM-DOAS version 0.4 XCO<sub>2</sub> product is scaled by a constant (i.e., time/space/scene independent) factor which has been applied to eliminate an obvious bias. Therefore, the interpretation of the XCO<sub>2</sub> results shown here should focus on space / time variability but not on the absolute XCO<sub>2</sub> level. The single measurement precision of the XCO<sub>2</sub> v0.4 data product is estimated to be on the order of a few percent [10,11,13,20].

## 4. TRACE GAS RESULTS

We have processed all available SCIAMACHY spectra of the year 2003 (details concerning the processed orbits are given in [11]). The results will be shown in the following focusing on China. Yearly averages will be presented as well as tri-monthly averages. Because for January 2003 only a few orbits were available and no data for November and December (for ground processing related reasons) the tri-monthly averaged data cover the time period February to October using an untypical grouping of the months not reflecting the standard definition of seasons. Because of the low reflectivity of water (oceans, great lakes) in the near-infrared the quality of the measurements over water is typically reduced and only measurements over land are discussed here.

#### 4.1 Carbon monoxide (CO)

The left panel of Fig.1 shows yearly averaged vertical columns of CO over China (grid: 0.5x0.5 degrees). Clearly visible are large regions of elevated CO (shown in red) indicating CO source regions. Elevated CO is present over a large area south of Beijing, in the region around Chengdu/Chongqing (Red Valley), around Shenyang, and over Hainan Dao and Zhanjiang. The elevated CO detected with SCIAMACHY clearly correlates with major industrialized areas.

The three panels on the right hand side of Fig.1 show tri-monthly averaged CO columns for (from top to bottom) February-April, May-July, and August-October 2003. The largest CO columns are observed in the May to July 2003 average over large parts of eastern China and around Chongqing/Chengdu. High columns are also observed over these regions during other times of the year, especially around Beijing during February to April.

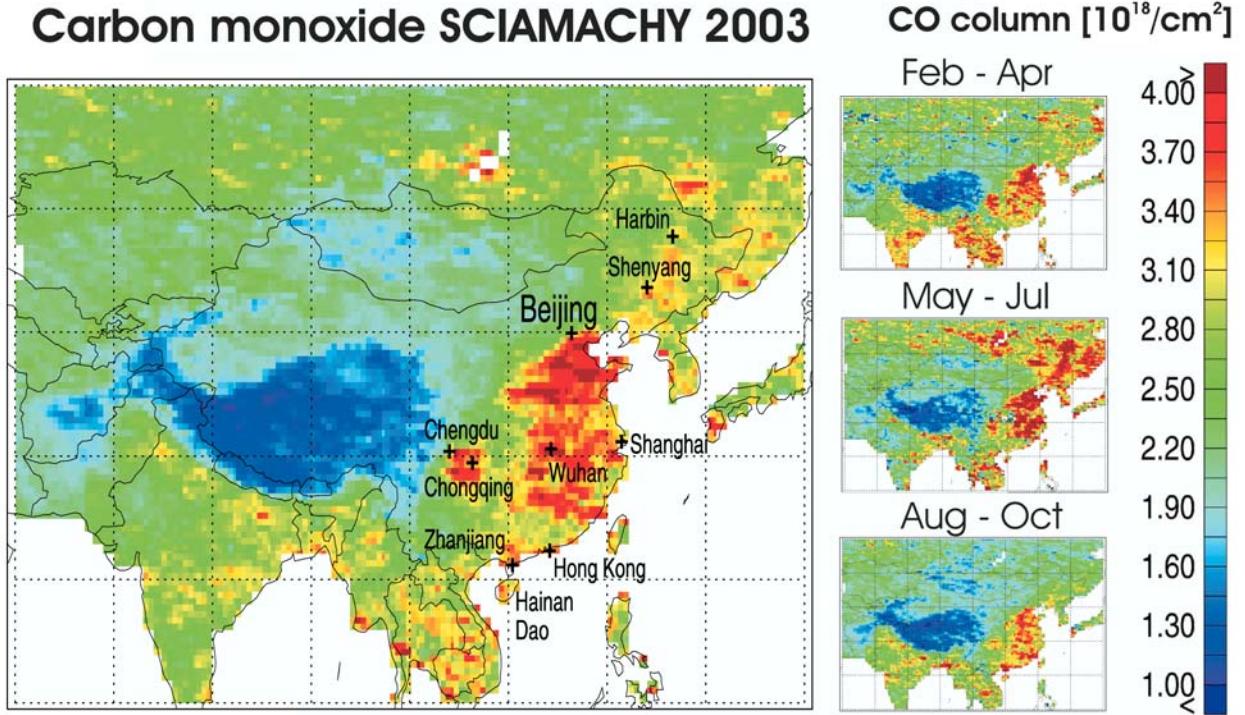


Fig. 1: Carbon monoxide vertical columns over China as measured by SCIAMACHY. The left panel shows the year 2003 average. The three panels on the right hand side show tri-monthly averages for the year 2003. The latitude range covered is 10 °N-60 °N and the longitude range is 60 °E-140 °E.

#### 4.2 Methane (CH<sub>4</sub>)

The left panel of Fig.2 shows the dry air column averaged mixing ratio of methane over China obtained by averaging all SCIAMACHY measurements available for the year 2003. Shown are only measurements over land for which the quality flag determined by the retrieval algorithm indicates a (potentially) successful measurement. Using this approach the measurements over the Himalaya region are filtered out. Clear visible are large regions of elevated methane (shown in red) indicating methane source regions over large parts of south-east China (as well as over India, south-east Asia, and Japan).

The panels on the right hand side of Fig.2 show tri-monthly averages of the year 2003 methane data set. During February to April 2003 (top panel) elevated concentrations are present around Chongqing/Chengdu (see annotation of Fig.1 left panel) and along the Pacific ocean coast line around and south of Shanghai, over Hainan Dao and north of Hainan Dao. During May to July (middle panel) the methane concentration is significantly higher compared to February-April over large parts of south-east China. The highest mixing ratios occur over the Chongqing/Chengdu area. Even larger methane concentrations are visible in the August to October average (bottom panel) were highly elevated mixing ratios are observed over large parts of eastern China (as well as over India and south-east Asia). The August to October 2003 measurements confirm the data shown in [15] also derived from SCIAMACHY but with an independent somewhat different retrieval algorithm. The main methane sources of China are rice cultivation, waste handling, ruminants, fossil-fuel related activities, and wetlands (see [15] and references given therein). The tri-monthly averaged concentrations are highest during August to October in-line with the seasonal dependence of the methane emission from rice-paddies.

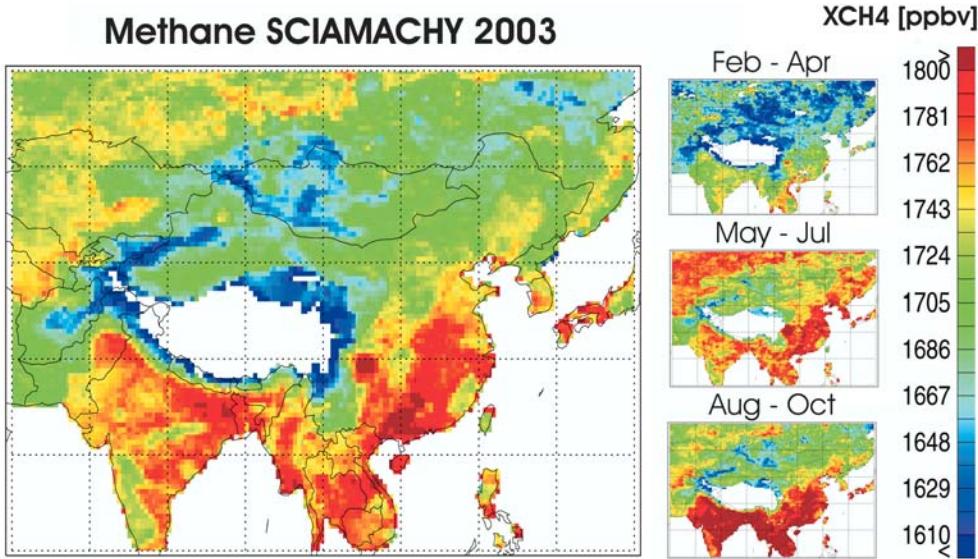


Fig. 2: Methane column averaged mixing ratios over China as measured by SCIAMACHY. The left panel shows the year 2003 average. The three panels on the right hand side show tri-monthly averages for the year 2003. The latitude range covered is 10°N-60°N and the longitude range is 60°E-140°E.

#### 4.3 Carbon dioxide ( $\text{CO}_2$ )

The left panel of Fig. 3 shows the  $\text{XCO}_2$  over and around China obtained by averaging all SCIAMACHY measurements available for the year 2003 [11]. Shown are only measurements over land for which a relatively good fit has been achieved (using this approach the measurements over the Himalaya region are filtered out). The largest mixing ratios are observed in the north-eastern part of China around and north of Harbin. The lowest mixing ratios are observed over the southern part of China along the Mekong river.

The panels on the right hand side of Fig. 3 shows tri-monthly  $\text{XCO}_2$  averages of the year 2003 data set. On average the values are lowest in the May to July average. This minimum around the mid-of the year is consistent with the uptake of atmospheric  $\text{CO}_2$  by vegetation which is in its main growing phase during this time of the year [11]. Superimposed on this large scale behaviour is a considerable regional fine structure. The interpretation of these measurements is difficult because the retrieval errors due to instrument and retrieval noise and biases are estimated to be on the same order as the weak source/sink signals to be detected [8,10,11]. More studies are needed including comparison with land cover maps and model simulations before a clear interpretation of these maps can be made.

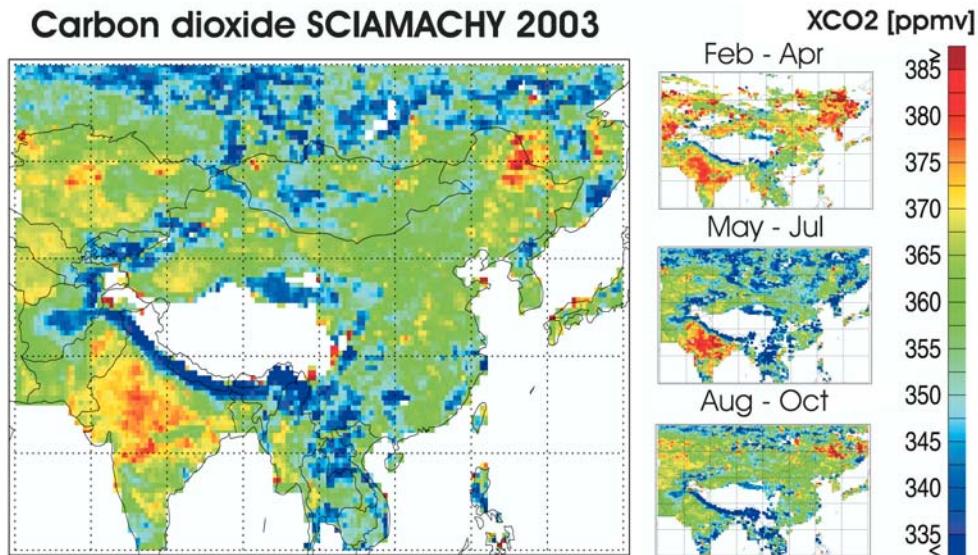


Fig. 3: Carbon dioxide column averaged mixing ratios over China as measured by SCIAMACHY. The left panel shows the year 2003 average. The three panels on the right hand side show tri-monthly averages for the year 2003. The latitude range covered is 10°N-60°N and the longitude range is 60°E-140°E.

## 5. CONCLUSIONS AND OUTLOOK

Satellite derived maps of carbon monoxide, methane and CO<sub>2</sub> over China have been presented. The maps enable the identification of surface source regions. Our future work will focus on further analysing the presented data by performing a detailed comparison with model simulations using the latest emission data bases. Our goal is to generate data products that can provide quantitative information on surface sources and sinks via inverse modelling. So far we have only processed the SCIAMACHY year 2003 spectra. More data will be processed in the near future. Information about the latest status is given on our SCIAMACHY/WFM-DOAS web page:

[http://www.iup.physik.uni-bremen.de/sciamachy/NIR\\_NADIR\\_WFM\\_DOAS/index.html](http://www.iup.physik.uni-bremen.de/sciamachy/NIR_NADIR_WFM_DOAS/index.html).

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