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B. Dils, M. de Mazière, T. Blumenstock, M. Buchwitz, R. de Beek, et al.. Comparisons between SCIAMACHY and ground-based FTIR data for total columns of CO, CH₄, CO₂ and N₂O. Atmospheric Chemistry and Physics Discussions, 2005, 5 (3), pp.2677-2717. hal-00303909

HAL Id: hal-00303909

<https://hal.science/hal-00303909>

Submitted on 18 Jun 2008

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Comparisons between SCIAMACHY and ground-based FTIR data for total columns of CO, CH₄, CO₂ and N₂O

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Received: 7 March 2005 – Accepted: 23 March 2005 – Published: 3 May 2005

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Abstract

Total column amounts of CO, CH₄, CO₂ and N₂O retrieved from SCIAMACHY nadir observations in its near-infrared channels have been compared to data from a ground-based quasi-global network of Fourier-transform infrared (FTIR) spectrometers. The SCIAMACHY data considered here have been produced by three different retrieval algorithms, WFM-DOAS (version 0.4, 0.41 for CH₄), IMAP-DOAS (version 0.9) and IMLM (version 5.5) and cover the January to December 2003 time period. Comparisons have been made for individual data, as well as for monthly averages. To maximize the number of reliable coincidences that satisfy the temporal and spatial collocation criteria, the SCIAMACHY data have been compared with a temporal 3rd order polynomial interpolation of the ground-based data. Particular attention has been given to the question whether SCIAMACHY observes correctly the seasonal and latitudinal variability of the target species. The ensemble of comparisons, discussed in this paper, demonstrate the capability of SCIAMACHY, using any of the three algorithms, to deliver products for the target species under consideration, which are already useful for qualitative geophysical studies on a global scale. It is expected that the remaining uncertainties in the data products will decrease in future versions of the algorithm to also allow more quantitative investigations on a regional scale.

1. Introduction

The SCIAMACHY instrument (Burrows et al., 1995; Bovensmann et al., 1999, 2004) onboard ENVISAT has the potential to make nadir observations in the near-infrared (NIR; 0.8–2.38 μm) of the most important greenhouse gases such as water vapour (H₂O), carbon dioxide (CO₂), methane (CH₄), and nitrous oxide (N₂O), and of the ozone precursor gas carbon monoxide (CO), which also acts as an important indirect greenhouse gas as it significantly impacts the OH budget. SCIAMACHY is among the first satellite instruments that can measure greenhouse gases in the troposphere

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on a global scale. Its predecessor instrument GOME (Global Ozone Monitoring Experiment) does not include the channels in the NIR (Burrows et al., 1999). IMG (Interferometric Monitor of Greenhouse Gases) flew onboard ADEOS in 1997 to make nadir measurements in the thermal infrared (TIR), but failed after a few months of operation (Kobayashi et al., 1999). At present, MOPITT (Measurements Of Pollution In The Troposphere) is delivering only CO profile data retrieved from the TIR channels; the expected CH₄ products are still unavailable due to instrument calibration problems (Drummond and Mand, 1996). SCIAMACHY measurements in the NIR have the important advantage over TIR measurements that they are sensitive down to the earth's surface, where most emission sources are located, whereas thermal infrared measurements have a reduced sensitivity in the boundary layer. It is very important therefore to thoroughly investigate the potential capabilities of SCIAMACHY in its NIR channels.

The purpose of the current validation is to identify quantitatively to what extent the SCIAMACHY NIR products generated by various scientific institutes in Europe can be exploited for global geophysical studies. It therefore addresses the consistency of the data to represent the variations of the CO, CO₂, CH₄ and N₂O fields with season, latitude, surface altitude, etc. This is done by comparing the available SCIAMACHY data with correlative, i.e., close in space and time, independent data – in casu from a remote-sensing network of ground-based FTIR spectrometers. Other complementary validation efforts have been made, such as comparisons with data from other satellites, e.g., with CO data from MOPITT, or with analyses from global chemistry models such as TM3 (Heimann and Körner, 2003) or TM5 (Krol et al., 2005), and have been reported by Buchwitz et al. (2005a); Gloudemans et al. (2005); Straume et al. (2005).

The SCIAMACHY data for CO, CH₄, CO₂ and N₂O total columns investigated in this paper have been produced by the algorithms WFM-DOAS v0.4 and v0.41 for CH₄ (Weighting Function Modified DOAS, Institute for Environmental Physics, University of Bremen; Buchwitz et al., 2005a, b, 2000, 2004), IMLM v5.5 (Iterative Maximum Likelihood Method, SRON; Schrijver, 1999; Gloudemans et al., 2005, 2004) and IMAP-DOAS v0.9 (Iterative Maximum A Posteriori-DOAS, University of Heidelberg; Franken-

berg et al., 2004, 2005). So far, CO₂ and N₂O total column data have been provided by WFM-DOAS only. The data provided for this validation exercise cover the January to December 2003 time period, and thus offer a much better basis for validation than the limited data set that was available for previous exercises (De Mazière et al., 2004).

5 Since then, some algorithm updates have also been implemented. For more in depth information about the SCIAMACHY retrieval algorithms and data products, the reader is referred to the above cited references.

The characteristics of the correlative ground-based FTIR data are described in the next section. Section 3 presents the conditions that have been verified for carrying out the comparisons. The comparison methodology and the results of the comparisons are discussed in Sect. 4, successively for CO, CH₄ and N₂O and CO₂. Conclusions are drawn in Sect. 5.

2. The ground-based correlative data

The ground-based (g-b) correlative data are collected from 11 FTIR spectrometers that are operated at various stations of the Network for the Detection of Stratospheric Change (NDSC, <http://www.ndsc.ws>). They have been submitted to the Envisat Cal/val database at NILU or directly to BIRA-IASB and have been compiled by us as part of the commitment in the Envisat AO ID 126 'Validation of ENVISAT-1 level-2 products related to lower atmosphere O₃ and NO. Fig. 1 and Table 1 identify the locations of the contributing stations.

The g-b FTIR data are obtained from daytime solar absorption measurements under clear-sky conditions. G-b FTIR data can also be obtained from lunar absorption measurements at near full noon, e.g., in polar night conditions at high northern and southern latitude stations: such lunar absorption data are not included in the present dataset however.

Figures 2a to d show the database of the CO, CH₄, N₂O and CO₂ g-b data products, respectively, available at BIRA-IASB for the present validation exercise. For compar-

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ison purposes, all data have been normalized to zero station altitude, as explained hereinafter.

Regarding CO (Fig. 2a) seasonal variations are quite pronounced (amplitude of about 50%), with a maximum by the end of local spring. Large excursions in the CO column amounts are observed at Wollongong: they can probably be attributed to biomass burning events. Also, the g-b FTIR data (Fig. 2) clearly illustrate the inter-hemispheric gradient of CH₄ that amounts to 15% going from the South Pole (Arrival Heights) to the maximum values at northern mid-latitudes (Egbert). This gradient is high compared to model predictions and in-situ surface observations that observe an interhemispheric gradient of order 3 to 8%, respectively (Dlugokencky et al., 1994): the apparent discrepancy can probably be explained by some inhomogeneity over the network, as discussed below. One also observes a seasonal variation of CH₄ that is more distinct in the northern hemisphere than in the southern one. The CH₄ minimum in the northern hemisphere occurs at the beginning of the year, i.e., around mid-winter. N₂O has a very small seasonal variation; also the variability over the entire data set is less than 15%. The CO₂ dataset is limited to 3 ground stations, with only very few data at Ny.Alesund.

In view of the use of the g-b FTIR data for the validation of SCIAMACHY data products, three issues must be clarified, that are, (1), the data availability, (2) the precision and accuracy of the data, and (3), how to deal with different ground station altitudes.

(1) The first issue (data availability) concerns the amount of available g-b data. One must remember that the g-b FTIR observations require clear-sky conditions. Consequently the g-b FTIR database does not represent a daily coverage, even if most stations are operated on a quasi-continuous basis. This limits of course the number of possible coincidences with SCIAMACHY overpasses. Moreover for several ground-based stations the available datasets do not cover the entire January till December 2003 time period. To maximize data overlap between SCIAMACHY observations and FTIR g-b measurements, and ensure a statistically significant correlative data set, the SCIAMACHY data that meet the spatial collocation criteria (see Sect. 3) are not com-

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pared on the basis of temporal overlap with the g-b data. Instead, we developed an alternative method in which the SCIAMACHY measurements are compared with the corresponding (in time) interpolated value of a third order polynomial fit through the FTIR g-b data, rather than with the FTIR data themselves. To ensure consistency between all stations, all FTIR data points, if not already daily averages, have been converted to daily averages prior to any further manipulations such as the 3rd order polynomial fitting procedure. This third order polynomial fit gives a good representation of the seasonal variability (see example in Fig. 3), but loss of information as to daily variability and as to possible short term events cannot be avoided. The data comparisons have been limited to the time periods during which g-b data is available to avoid gross extrapolation errors. This explains why there are no g-b data available for intercomparisons during the dark period in local winter at high-latitude stations. This method, which significantly increases the number of coincident data, allows us to study the latitudinal dependence over a wider range of stations whereas the usual validation method considering only daily coincidences failed to provide sufficient, if at all, data for stations near the poles. The standard deviations of the ground-based data with respect to their 3rd order fit, or

$$std \left(\frac{GB - PF}{PF} \right) \quad (1)$$

(with GB, the individual daily averaged FTIR ground based measurements and PF, their corresponding values derived from the third order polynomial fit), are, on average, 9.6% for CO (the average standard deviation drops to 7.6% when excluding the Wollongong measurements), 3.3% for CH₄, 1.6% for N₂O and 1.3% for CO₂.

(2) The second issue of data precision and accuracy has been discussed partially hereabove. Individual g-b FTIR data for N₂O, CO, CO₂ and CH₄ have a precision in the order of some percent (<5%). Because of the adopted approach to use interpolated (fit) values instead of original measurement data, the effective precision of the g-b correlative data is set by the values listed above.

The accuracies considering the entire FTIR network are estimated to be of order 3%

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for N₂O and CO₂, and 7% for CO and CH₄. Network accuracies can be improved in future by imposing some homogenization as to the spectral data analysis among the contributing stations, especially for CH₄. The current rather limited accuracy for CH₄ probably explains why the interhemispheric gradient in the g-b data appears too large.

(3) The third issue concerns a normalisation with altitude that has been applied to all g-b data. Because the target molecules have most of their total concentration in the lower troposphere, the total column amount is strongly dependent on the observatory's altitude. To eliminate any apparent differences or variations in the data set that are due to this altitude dependence, we have normalised all data to sea level altitude, using the following simplified formula, which is a variation of the hypsometric equation (Wallace and Hobbs, 1977):

$$C_0 = C_Z \exp\left(\frac{Z}{7.4}\right) \quad (2)$$

Herein Z is the station's altitude (in km), C_Z is the observed total column amount, and C₀ is the corresponding normalised (to sea level) total column amount. The same normalisation has been applied to the overpass SCIAMACHY data, with Z being the mean altitude of the observed ground pixel. This normalisation procedure is, while the best at hand in the absence of auxiliary information, relatively coarse, inducing possible errors up to about 15% for high altitude stations such as Zugspitze, Jungfraujoeh and Izaña. The approximation is best for CO₂, having a nearly constant volume mixing ratio (VMR) throughout the whole atmosphere, relatively good for CH₄ and N₂O with an almost constant tropospheric VMR, but worse for CO that has a more variable VMR in the troposphere.

Moreover the column measured by SCIAMACHY is an average column above the area covered by a SCIAMACHY pixel which extends beyond the location of the g-b station. For channel 8 products (see further in Sect. 3), the pixel size is 30×120 km², for channel 6 products 30×60 km² (see Table 2 for the used SCIAMACHY channels for each algorithm). Consequently for a mountainous g-b station, the SCIAMACHY column also samples to some extent the valleys around the station that often harbour

significantly higher concentrations of pollutants compared to the mountain site. This might create an apparent bias between the FTIR and SCIAMACHY measurements. In addition to that, to obtain a statistically significant dataset, the spatial collocation criteria include all SCIAMACHY pixels centred within $\pm 2.5^\circ$ latitude and $\pm 5^\circ$ or $\pm 10^\circ$ longitude of the FTIR ground-station coordinates (for the small grid and large grid collocation, respectively – see Sect. 4), thus covering an even wider area, which in turn may lead to an inherent apparent increase of the data scatter as compared to that of the FTIR g-b measurements.

3. The SCIAMACHY data and selection criteria for comparison

The retrieval methods (WFM-DOAS, IMLM and IMAP) discussed in this paper not only use different mathematical retrieval algorithms, but also obtain their data for CO, CH₄, N₂O and CO₂ from different spectral channels and wavelength regions: an overview hereof is given in Table 2. Each of the channels/windows has its own distinct features and associated problems. For instance the SCIAMACHY NIR channels 7 and 8 are affected by ice layer build-up on the detectors, which is countered by regular decontamination of the instrument (Bovensmann et al., 2004). Also, not all the SCIAMACHY data sets considered for the present comparisons cover the complete year 2003: the IMLM data set contains no data for the April till August 2003 period, while the CH₄ IMAP data set is limited to the August till November 2003 time period. The CO IMAP data lacks measurements for August and December, and WFM-DOAS only contains data from January till October. Due to the fact that the January to December 2003 time frame includes periods of lower transmission and ice decontamination of the SCIAMACHY instrument, differences in the considered time periods may lead to apparent differences in the final comparison results when evaluating the algorithms. Some differences may also occur because of the seasonal variation of the inter-hemispheric latitudinal gradient of some species, notably CO.

The data products for SCIAMACHY give reliable total column values only for cloud-

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free pixels because the clouds are not transparent in the NIR (Buchwitz et al., 2000, 2004; Gloudemans et al., 2005; Straume et al., 2005) and thus effectively take over the role of the earth's surface. Since the highest concentrations of the target species are found close to the earth's surface, where the air pressure is the highest and where the sources and sinks are located, interpreting cloud-contaminated columns as total columns can lead to large errors in the analysis. The different algorithms investigated here use different cloud detection schemes (Buchwitz et al., 2004, 2005a; Schrijver et al., 1999; Gloudemans et al., 2005) resulting in different cloud masks. In some cases they do not mask all cloudy pixels and in other cases they may be too restrictive, because they cannot distinguish between ice- or snow-covered surfaces and clouds, resulting in loss of data. This implies that some comparisons with g-b FTIR data may still suffer from the presence of clouds in the SCIAMACHY observation. The current IMAP method does not contain a cloud detection algorithm for CO. For CH₄, the filtering is done based on a lower threshold for the height-corrected CO₂ column: the column must be at least 89% of the expected total column assuming constant CO₂. This method effectively filters high-altitude clouds. A more definite cloud detection algorithm will be implemented in the near future. As for the algorithms themselves, all research groups are continuously improving their cloud filtering methods.

In addition to that, for low albedo values, the precision of the cloud-free SCIAMACHY data is strongly influenced by the albedo of the observed ground-pixel, because it determines to a large extent the signal-to-noise ratio of the corresponding observed spectra. This explains why data over ocean (water) are less reliable than data over land. Also measurements with a high solar zenith angle (typically at the Earth's poles), lead to low signal to noise ratios, and thus larger errors in the retrieved total columns. A restriction on the accepted solar zenith angles therefore contributes to the observed limited dataset for northern and southern high-latitude stations.

It must also be noted that for WFM-DOAS and IMAP, in most cases (CH₄, CO₂ and N₂O) the products discussed here, namely the total column values, are not the final data products of these algorithms. Their final data products, denoted as XCH₄, XCO₂

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and XN_2O , are the total column values of said species divided by the total column values of either CO_2 or O_2 (scaled to be a proxy for dry air). This normalisation should improve the data quality, given the fact that systematic retrieval errors, such as column variations due to changes in surface pressure or topography, or residual cloud contamination, are eliminated to a large extent from the ratio product. Unfortunately at present, we do not dispose of the appropriate correlative measurements to evaluate these final products.

The criteria adopted for temporal and spatial ‘collocation’ stem from choosing the best compromise between achieving better or worse statistics and keeping more or less natural variability in the data. Spatial collocation has been defined as data being within $\pm 2.5^\circ$ latitude and $\pm 10^\circ$ longitude of the FTIR ground station. Data that have been taken closer to each other (within $\pm 2.5^\circ$ latitude and $\pm 5^\circ$ longitude of each other) have been looked at in particular. The spatial collocation criteria adopted here are loose; however making them more stringent would have made the number of coincidences too small, especially at the high-latitude stations.

Additional selection criteria have been applied to the SCIAMACHY data, based on confidence limits given by the data providers. These confidence limits are different for the different algorithms, because they estimate the errors differently. For example, WFM-DOAS includes spectral fit errors in the final error estimate, whereas the error reported by IMLM only accounts for instrument-noise related errors, and therefore appears to be smaller. The additional selection criteria that have been applied to the SCIAMACHY data from each algorithm are listed in Table 3.

In summary, the comparisons are limited to (1) cloud-free SCIAMACHY data, according to the individual cloud detection schemes from individual algorithms, (2) having the centre of the SCIAMACHY pixel within the spatial collocation area around the location of the g-b site, as outlined above, and (3) satisfying the additional selection criteria listed in Table 3. Temporal coincidence has been defined as data being taken at the same time, in which the real g-b FTIR data set has been approximated by a continuous set of interpolated values, as explained in Sect. 2.

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Before making the comparisons, we have verified that the total column averaging kernels of both data products (g-b FTIR and SCIAMACHY) are very similar, showing a rather uniform sensitivity close to 1 from the ground to the stratosphere (Rodgers and Connor, 2003; Buchwitz et al., 2004, private communication). Therefore we have compared the data products as such, without taking the averaging kernels explicitly into account.

4. The comparisons between timeseries of g-b FTIR network and SCIAMACHY data of CO, CH₄, CO₂ and N₂O total column amounts

4.1. Comparison methodology

Time series of the relative differences between SCIAMACHY individual data points (SCIA) and the corresponding values from the 3rd order polynomial interpolation through the g-b FTIR daily network data (PF), i.e., [(SCIA-PF)/PF] have been made for all the different SCIAMACHY algorithms and target products. An example for CH₄ from the WFM-DOAS algorithm at the Toronto station is shown in Fig. 4. An overall bias, being

$$\text{Bias} = \text{mean} \left(\frac{\text{SCIA} - \text{PF}}{\text{PF}} \right) \quad (3)$$

over the considered time period was calculated for each target product, algorithm and station; these biases are listed in Tables 5 to 7. A globally averaged bias (i.e., a mean over all stations) was calculated as well and is also listed in the same corresponding Tables. The errors reported on the biases are the statistical 1σ standard deviations on the [(SCIA-PF)/PF] values, not on the original SCIAMACHY measurements themselves.

It should be mentioned that the number of correlative data points can vary greatly from station to station (from 1 to several thousands). These numbers of correlative data points are indicated also in Tables 5 to 7. It must be kept in mind that the obtained absolute value of the overall bias can often be explained by slightly wrong slit

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functions and/or spectral parameters (GlouDEMANS et al., 2005); in some cases even the SCIAMACHY data have been scaled according to a chosen reference (Buchwitz et al., 2004). Similarly the indicated error is strongly influenced by the exact choice of error criteria.

We have also evaluated the scatter of the SCIAMACHY measurements themselves, σ_{scat} , for each station, algorithm and target species, for comparison with the corresponding ones of the FTIR data. To this end, the individual SCIAMACHY measurements have been averaged per day, and a daily bias value, called Biasday hereinafter, has been calculated according to Eq. (3), now using the daily averaged SCIAMACHY values. σ_{scat} is then obtained as the statistical 1σ standard deviation, of the daily averaged SCIAMACHY data (SCIAday) with respect to the polynomial interpolation of the daily FTIR data, corrected for the daily bias (Biasday), according to:

$$\sigma_{scat} = std \left(\frac{SCIAday - \{(1 + Biasday) \times PF\}}{\{(1 + Biasday) \times PF\}} \right) \quad (4)$$

The resulting values of σ_{scat} for the large collocation grid are summarized in Table 4, together with the scatter on the g-b FTIR data and the desired target precision for each species. These targets have been set in order to accurately detect the global sources and sinks of these species. The complete set of σ_{scat} values, including those from small grid collocated measurements, are listed in Tables 5 to 7.

It is also very important to verify whether SCIAMACHY is able to reproduce the seasonal and latitudinal variations of the target species. A useful marker for this ability is the correlation coefficient (R) between the SCIAMACHY measurements and the 3rd order polynomial interpolated FTIR g-b measurements. It turns out to be impossible to produce meaningful R values for the individual stations, given the limited temporal variation of the g-b data as compared to the scatter present in the SCIAMACHY measurements, and the limited number of data points per station. However, the overall correlation coefficient per retrieval method over all stations and time does provide useful information. The value of the correlation coefficient depends not only on the effective

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correlation but also on the number of data points and their scatter. Therefore, the R value may appear small even though the datasets do correlate and R is still significant. Therefore, next to the correlation coefficient R, we also tested for the hypothesis of no correlation. The latter is expressed by the P-value, also given in Tables 5 to 7, which is the probability of getting a correlation R as large as the observed value by random chance, supposing the true correlation is zero. If P is small, say less than 0.05, then the correlation R is significant. The P-value is computed by transforming the correlation to a t-statistic having $n-2$ degrees of freedom, with n the number of data points. The calculated R and P values give us a clear indication of how successful SCIAMACHY is in reproducing the overall variations in the g-b FTIR data. These variations include the temporal variations as well as the latitudinal variations.

A separate look at the latitudinal variation in the SCIAMACHY data can be easily derived from Tables 5 to 7 (in combination with Table 1) and is illustrated in Figs. 5 and 8, showing the bias as a function of latitude, per algorithm, for CO and CH₄, respectively.

To have a clearer view on the ability of SCIAMACHY to reproduce temporal variations we have calculated monthly averages of both the original ground-based data (without a polynomial fitting procedure) and the SCIAMACHY data, on the large collocation grid and satisfying all selection criteria. Time series of the relative differences ($[\text{scia-gb}]/\text{gb}$) have been plotted in Figs. 6, 9 and 10, again for all target products, algorithms and stations. The errors depicted on these Figures represent the statistical 1σ standard deviations derived through error propagation from the SCIAMACHY and ground based standard deviations of the respective monthly ensembles of individual data points.

Hereinafter the results summarized in the Tables and Figures are discussed in detail, per molecule.

4.2. Results for CO

Before discussing the results in Table 5, it must be noted that only the IMAP algorithm allows us to make some statistics at the polar stations Ny-Alesund and Arrival Heights.

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This is due to the abovementioned weakness of the cloud detection algorithms used in WFM-DOAS and IMLM in that they cannot distinguish clouds from ice and snow. Therefore, the conclusions drawn hereinafter as to latitudinal variations exclude these polar latitudes.

5 From the calculated global bias listed in Table 5, one notices that WFM-DOAS apparently overestimates the CO total column values. One must remember however that the absolute column values for the WFM-DOAS data have been scaled according to a chosen reference (Buchwitz et al., 2004). IMAP data exhibit a significant negative bias. The results for IMLM on the other hand seem to have the lowest bias, but with
10 a rather substantial difference between the results for the small (12.0%) and the large grid (1.1%). But as the data spread is very large – the largest of the three algorithms –, this bias difference between the two collocation grids is not significant. Stricter error criteria led to a significant reduction of the data spread but at the cost of a large reduction in the number of data points. In all cases, when considering the data spread
15 and bias, one has to keep in mind (1) that SCIAMACHY is aiming at a 10% precision on the CO column data, and (2) that there is already a significant standard deviation of 9.6% on the CO ground-based data themselves. The standard deviation of the IMAP measurements, σ_{scat} , is only about 2.5 times larger than that of the ground-based FTIR data, closely followed by WFM-DOAS.

20 Table 5 and Fig. 5 also show that for both WFM-DOAS and IMAP, the biases observed in the southern hemisphere are systematically more positive than those observed in the northern hemisphere, which indicates that WFM-DOAS and IMAP somewhat underestimate the latitudinal variability of CO. However, given the actual large errors associated with the obtained bias values and the 7% accuracy of the g-b network data, this preliminary conclusion deserves confirmation. There is no such apparent
25 latitudinal dependence of the bias in the IMLM dataset. This is reflected also in the highest R and lowest P values among the three retrieval methods, even if it exhibits the highest scatter. IMAP on the other hand has a particularly low correlation coefficient R: this is probably caused to some extent by the complete lack of cloud correction on

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the IMAP CO measurements, which automatically reduces the correlation and probably also partly explains the observed negative bias. However, although the P value is relatively high compared to the other algorithms it is still well below the 0.05 mark which indicates that the IMAP SCIAMACHY and FTIR measurements do correlate. As already mentioned before, the R and P values give an indication of the response of SCIAMACHY measurements towards latitudinal and temporal variations of the FTIR data.

When looking at the WFM-DOAS time series of monthly biases (Fig. 6) we observe a consistent rise of the relative bias during the June to September time period over almost all Northern hemisphere stations, pointing towards some deviation from a correct seasonal behaviour of the SCIAMACHY WFM-DOAS data.

Having an in depth look into the datasets, we can explain the apparent latitudinal (and seasonal) discrepancies in WFM-DOAS and IMAP by the fact that both algorithms overestimate the CO columns as these become lower – a feature that is not observed in the (more limited) IMLM dataset. To verify the existence of such a gradual overestimation (or lesser underestimation) with decreasing CO total column values, the calculated monthly mean biases have been plotted against the monthly mean CO FTIR total column values, for all algorithms, as shown in Figs. 7a, b and c. The Figures confirm that both WFM-DOAS and IMAP exhibit such gradual column overestimation, and that the effect is more pronounced in WFM-DOAS: slopes fitted to the ensemble of data points amount to -27×10^{-18} and $-47 \times 10^{-18} \% \text{ molec}^{-1} \text{ cm}^2$ for IMAP and WFM-DOAS, respectively. Such a dependence of the bias on the total column amount is not found for any other species-algorithm combination.

Another factor which could contribute to the apparent temporal bias increase in WFM-DOAS is the problem of ice-build-up associated with the SCIAMACHY spectral channel 8, and subsequent decontamination around mid-August. As we will see in the next section, version v0.41 implemented for CH_4 to deal with this problem is a great improvement over v0.40 (as still used for CO). Although not demonstrated yet, it is possible that the implementation of the corrected version v0.41 is worthwhile also for

the retrieval of CO (see e.g. Gloudemans et al., 2005).

4.3. Results for CH₄

As for CO (Table 5) it must be noted that only the IMAP algorithm allows us to make some statistics at the Ny-Alesund and Arrival Heights stations. Also, the lack of Zugspitze and Wollongong IMAP measurements is due to the limited IMAP CH₄ dataset (only covering August to November 2003).

It is clear from Table 6 and Fig. 8 that all three algorithms exhibit very small biases, especially when taking into account the 3.3% standard deviation on the CH₄ ground based measurements. Notice that the target precision for SCIAMACHY, based on requirements for inverse modelling applications, is 1%. The overall scatter on the SCIAMACHY measurements, σ_{scat} , is the lowest for IMAP (almost identical to the ground based measurements scatter) closely followed by IMLM. Scatter on the WFM-DOAS measurements is only slightly larger (factor ~ 2.3). In addition one doesn't observe any latitudinal dependence of the bias for any of the algorithms, thus indicating that the latitudinal variations of CH₄ are well reproduced by SCIAMACHY. Given the limited datasets for IMLM and IMAP it is difficult to draw any binding conclusions from the obtained R and P values. All P values indicate a strong correlation of the measurements. The lowest value occurs for the IMAP measurements, but given the fact that it covers a time period of only 4 months and that all algorithms are still undergoing improvements, it would be very premature to draw stringent conclusions based on this value. The differences in data quality between the three retrieval algorithms appear to be minimal, which indicates that the ice issue problem on spectral channel 8 of SCIAMACHY, impacting both WFM-DOAS and IMLM but not IMAP CH₄ measurements, has been well handled by both affected algorithms. The actually used version v0.41 of WFM-DOAS proves to be a significant improvement over the WFM-DOAS v0.4 algorithm: it manages to almost completely erase the -10% bias found with v0.4, by improving the way it deals with the problem of icing and consequent loss of transmission.

From the time series of monthly mean biases (Fig. 9), even though sometimes lim-

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ited, we cannot observe any apparent deviations from the g-b seasonal evolution of CH₄.

Plots of the SCIAMACHY monthly biases versus the monthly FTIR g-b columns (not shown), analogous to Fig. 7, confirm that the bias for CH₄ does not depend on the absolute value of the total column; anyway, the latter changes are of the order of only 20% over the whole FTIR dataset.

4.4. Results for N₂O and CO₂

For N₂O and CO₂, only WFM-DOAS v0.4 measurements have been available for the present study. Furthermore, the ground-based dataset for CO₂ is limited to three stations only, which makes it impossible to draw any conclusions regarding the latitudinal dependence of the CO₂ measurements. However, Table 7 shows that the results for these stations are fairly consistent, indicating a significant negative bias of the order of 11% of the SCIAMACHY measurements relative to the 3rd order polynomial fit through the ground based-FTIR measurements. Also the obtained correlation R and probability P indicate that there is a good correlation.

The biases for N₂O are also summarized in Table 7. They are not significant. From Table 7 we observe no obvious latitudinal dependence of the bias.

The standard deviation of the N₂O SCIAMACHY measurements, σ_{scat} , is a significant 15 times larger than that of the ground-based FTIR measurements and also the R and P values indicate a relatively weak correlation, although the P values are still well below the 0.05 mark. The CO₂ standard deviations on the other hand are less than 5 times larger than that of the ground-based FTIR measurements.

From the time series of monthly mean biases in Fig. 10, again we cannot see any apparent deviations from the temporal evolution, indicating that the monthly variability of N₂O is well reproduced by SCIAMACHY. It is impossible to draw any definite conclusions for CO₂, given the extremely limited dataset, although a very small negative curvature over time could be noticed. Additional measurements are needed to confirm this trend. As for CH₄, neither the N₂O nor the CO₂ measurements exhibit a

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dependence of the bias on the absolute value of the total column.

5. Conclusions

The present comparisons covering the period January to December 2003 between SCIAMACHY data for CO, CH₄, CO₂, and N₂O total column amounts from three different algorithms (WFM-DOAS v0.4 (v0.41 for CH₄), IMLM v5.5, and IMAP v0.9) and correlative FTIR g-b data have demonstrated the capability of the SCIAMACHY instrument to detect the variability of these important atmospheric species. They show that scientific teams have developed retrieval algorithms that succeed in deriving the total columns of these species from the instrument's NIR channels, despite the calibration problems inherent with these NIR spectra (Buchwitz et al., 2000, 2004, 2005a, b; Schrijver et al., 1999; Frankenberg et al., 2004, 2005; Gloudemans et al., 2005, 2004).

The actual status of the SCIAMACHY products discussed here is such that related geophysical studies on a global scale can be initiated, at least from a qualitative perspective.

Overall all algorithms give good descriptions of the seasonal and latitudinal variability of the gas species involved. However for CO, WFM-DOAS as well as IMAP seem to underestimate the latitudinal variability of CO resulting in increased biases for the southern hemisphere. Also worth mentioning is the apparent bias increase in the June to September time period for WFM-DOAS CO measurements. These latitudinal (and seasonal) deviations from the g-b measurements, as observed in the WFM-DOAS and IMAP CO measurements, seem to relate to a gradually increased overestimation of the CO total columns by these algorithms, with decreasing CO total column values.

The precision of the data, in the case of CH₄, is approaching that of the FTIR data, but is still a factor of order 3 above the target precision of 1%. For CO, the scatter on the daily mean data is still at least a factor of 2 worse than that of the g-b FTIR measurements that is about the target precision (10%). The precisions of the N₂O and CO₂ data from WFM-DOAS appear to be worse by a factor 15 to 4, respectively. When

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assessing the scatter on the daily means of SCIAMACHY measurements, one has to keep in mind that the observed scatter contains the temporal scatter (also included in the FTIR measurements), as well as scatter coming from natural spatial variability due to the non-restrictive spatial collocation parameters. The latter spatial variability includes variations related to topography as well as real variability of the concentrations. Therefore, species with a high spatial variability such as CO, will automatically exhibit a higher standard deviation on their daily mean SCIAMACHY data than on their daily mean FTIR measurements, even though the inherent data quality approaches that of FTIR. Thus, while the daily averages of FTIR are averages in time, the daily averages for SCIAMACHY are mostly averages in space, due to the limited time coverage of the overpassing ENVISAT satellite. However, the scatter on the daily mean measurements did not change dramatically, even slightly improved, between the small and large spatial collocation grids. This could indicate that the potential worsening of the data quality by increasing the spatial variability is to a large degree (or even completely) countered by the ever increasing number of collocated data in the large grid dataset. However the small grid already covers a substantial geographical area and thus the increase in spatial variability from small to large grid could be minimal, while an increase from single point (as the FTIR measurements) to the small grid could be more significant. The remaining quantitative uncertainties will probably be reduced in future algorithm improvements, having acquired a better comprehension of the instrument/spectral problems.

It must be stressed once more that the actual conclusions are based on a limited number of data coincidences, that the collocation criteria were not very stringent, and that an approximate correction for the surface altitude has been applied that may add additional uncertainties. Some comparisons may still suffer from the presence of clouds because of imperfect cloud algorithms associated to the satellite data retrieval. The dry-air normalized data products of WFM-DOAS and IMAP have not been evaluated. Additional features that have not been taken into account in the comparisons are possible small sensitivity differences due to slightly different total column averag-

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ing kernels, spectroscopic uncertainties, etc. One must remember also that it is not possible yet to make an honest evaluation of the performances of the three algorithms among them, because the delivered data products do not cover exactly the same time periods, the time periods covered are still very limited (especially for the evaluation of the seasonal variability) and all the algorithms are still being constantly improved.

Based on the conclusions drawn here and in other papers in this same volume, one can state that SCIAMACHY provides an added value to the actually deployed fleet of satellite instruments, especially for tropospheric chemistry research on a global scale.

Acknowledgements. This work has been performed in the frame of the Envisat AOID126 'Validation of ENVISAT-1 level-2 products related to lower atmosphere O₃ and NO_y', and the EC project EVERGREEN. It has been possible thanks to financial support from ESA, the European Commission, the Belgian Federal Science Policy Offices, the Bundesministerium für Bildung und Forschung and PRODEX. The work could not have been carried out without the scientific and technical support from J.-C. Lambert, T. Jacobs, J. Granville and P. Gérard at the Belgian Institute for Space Aeronomy.

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Table 1. Spatial Coordinates of the g-b FTIR stations depicted in Fig. 1.

Station	Lat N	Lon E	Altitude(m)
NY.ALESUND	78.91	11.88	20
KIRUNA	67.84	20.41	419
HARESTUA	60.22	10.75	580
ZUGSPITZE	47.42	10.98	2964
JUNGFRAUJOCH	46.55	7.98	3580
EGBERT	44.23	-79.78	251
TORONTO	43.66	-79.40	174
IZAÑA	28.30	-16.48	2367
WOLLONGONG	-34.45	150.88	30
LAUDER	-45.05	169.68	370
ARRIVAL.HEIGHTS	-77.85	166.78	190

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Table 2. Selection of spectral channels and microwindows for the retrieval of CO, CH₄, N₂O and CO₂ in the different retrieval methods considered.

	WFM-DOAS v0.4 (v0.41 for CH ₄)	IMLM v5.5	IMAP v0.9
CO	Channel 8: 2359.0–2370.0 nm	Channel 8: 2327.00–2339.3 nm	Channel 8: 2324.2–2334.9 nm
CH ₄	Channel 8: 2265.0–2280.0 nm	Channel 8: 2327.00–2339.3 nm	Channel 6: 1630–1670 nm
N ₂ O	Channel 8: 2265.0–2280.0 nm		
CO ₂	Channel 6: 1558.0–1594.0 nm		

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Table 3. Selection criteria associated with accepted error levels for the SCIAMACHY data included in the comparisons with g-b data.

Algorithm	Selection criteria (in addition to spatial and temporal collocation criteria)
WFM-DOAS	Cloud-free, Over land (altitude>0), Solar Zenith Angle<85 deg, Error (fitting)<10% for CH ₄ and CO ₂ , <60% for CO and N ₂ O
IMLM	Cloud-free, Albedo≥0.01, Instrument-noise related Error<2E18 molec cm ⁻² for CH ₄ (~7%) and <1.5E18 molec cm ⁻² for CO (~70%), Solar Zenith Angle<80°
IMAP	For CH ₄ data: [Vertical Column Density of CO ₂ /exp(-surface elevation(m)/8000)]>7E21 molec cm ⁻² and variance of fit residual<0.5% For CO data: variance of the fit residual (without weighting)<0.017, weighted variance of the fit residual between 10 and 0.1, error <7E17 molec cm ⁻² and <30%

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Table 4. Percentage scatter on the daily mean FTIR and SCIAMACHY data, collocated on the large spatial grid. Also indicated are the target precisions set for the SCIAMACHY data.

	FTIR	WFM-DOAS	IMLM	IMAP	Desired precision
CO	9.6	25.4	39.7	24.5	5 (10)
CH ₄	3.3	6.56	5.03	3.60	1
N ₂ O	1.6	25.1			10
CO ₂	1.3	5.75			1

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Table 5. A. Calculated bias (in %) of the SCIAMACHY data relative to the 3rd order polynomial fit through the ground based FTIR data for CO, using the small grid (SG= $\pm 2.5^\circ$ LAT, $\pm 5^\circ$ LON) and large grid (LG= $\pm 2.5^\circ$ LAT, $\pm 10^\circ$ LON) spatial collocation criteria. The indicated errors represent the 1σ standard deviations of the ensemble of individual biases. B. The number of correlative SCIAMACHY data. C. σ_{scat} , the percentage 1σ standard deviation of the daily averaged SCIAMACHY measurements towards the bias corrected polynomial FTIR fit (see Eq. 4).

Algorithm→ Station ↓		WFM-DOAS, SG	WFM-DOAS, LG	IMLM, SG	IMLM, LG	IMAP, SG	IMAP, LG
Ny. Alesund	A	/	/	/	/	-32.3±9.64	-36.8±10.5
	B	0	0	0	0	6	22
	C	/	/	/	/	15.0	12.3
Kiruna	A	37.0±55.2	35.6±50.7	22.4±74.3	-1.94±69.7	-35.9±18.8	-35.9±17.4
	B	51	95	48	87	632	1252
	C	43.3	39.6	61.2	58.9	24.7	18.3
Harestua	A	21.4±35.0	20.8±37.4	-14.1±62.7	-14.9±63.0	-36.0±20.7	-36.8±19.5
	B	72	110	55	61	700	1466
	C	33.4	33.6	78.8	65.9	23.8	21.9
Zugs Spitze	A	21.3±27.9	18.7±25.8	22.3±58.1	20.5±68.1	-32.5±21.9	-30.9±22.8
	B	162	361	84	224	610	1329
	C	20.1	17.2	24.4	43.4	24.0	25.6
Jungfraujoch	A	32.5±32.4	32.5±32.7	24.2±70.7	23.7±72.7	-19.1±29.9	-18.7±31.2
	B	468	921	792	1546	725	1675
	C	19.0	18.1	53.4	43.9	29.7	27.7
Egbert	A	12.3±33.9	9.06±30.7	24.1±71.6	16.8±69.6	-38.5±17.9	-38.9±19.2
	B	193	480	874	1682	976	1873
	C	34.4	28.8	27.2	33.7	26.0	25.2
Toronto	A	6.74±33.7	0.92±29.1	14.7±65.3	8.32±63.5	-43.5±17.1	-43.7±18.3
	B	199	511	990	1872	967	1923
	C	35.5	29.9	26.5	36.8	27.3	26.9
Izana	A	33.3±41.7	27.0±27.4	-17.3±28.4	-17.7±30.4	-18.1±21.7	-22.4±20.2
	B	461	1769	420	2237	1097	2910
	C	19.2	16.9	16.2	23.4	23.4	24.3
Wollongong	A	41.4±32.1	33.5±23.8	-33.6±51.6	-41.9±38.2	4.04±44.9	-7.71±33.7
	B	86	484	284	904	78	349
	C	15.8	8.00	42.9	36.1	37.7	20.4
Lauder	A	98.46±81.4	98.46±81.4	56.1±128.3	57.6±127.5	-2.82±31.4	-1.26±33.0
	B	21	21	52	53	136	364
	C	45.8	45.8	61.1	59.3	28.0	26.2
Arrival Heights	A	/	/	/	/	0.13±32.5	-3.00±29.4
	B	0	0	0	0	12	23
	C	/	/	/	/	28.0	25.9
Global	A	27.3±39.0	23.8±32.0	12.0±68.1	1.1±62.5	-30.5±24.7	-30.1±24.8
	B	1713	4752	3599	8666	5939	13 186
	C	28.3	25.4	42.1	39.7	25.8	24.5
R	0.23	0.30	0.31	0.35	0.07	0.08	
P	1.24E-22	4.70E-100	2.27E-79	4.28E-246	1.29E-8	4.74E-20	

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Table 6. A. Calculated bias (in %) of the SCIAMACHY data relative to the 3rd order polynomial fit through the ground based FTIR measurements for CH₄, using the small grid (SG=±2.5° LAT, ±5° LON) and large grid (LG=±2.5° LAT, ±10° LON) spatial collocation criteria. The indicated errors represent the 1σ standard deviations of the ensemble of individual biases. B. The number of correlative SCIAMACHY data. C. σ_{scat} , the percentage 1σ standard deviation of the daily averaged SCIAMACHY measurements towards the bias corrected polynomial FTIR fit (see Eq. 4).

Algorithm→ Station ↓		WFM-DOAS (v0.41) , SG	WFM-DOAS (v0.41), LG	IMLM, SG	IMLM, LG	IMAP, SG	IMAP, LG
Ny. Alesund	A	/	/	/	/	/	/
	B	0	0	0	0	0	0
	C	/	/	/	/	/	/
Kiruna	A	-2.41±10.8	-3.05±9.91	3.57±8.53	3.55±9.94	-5.25±2.61	-4.49±3.12
	B	405	760	32	52	39	106
	C	13.9	9.95	6.75	14.1	2.40	2.72
Harestua	A	-2.55±7.46	-3.46±8.60	-3.86±8.86	-3.86±8.87	-7.60±2.05	-5.35±3.87
	B	379	562	25	25	95	193
	C	6.06	6.68	9.82	9.82	1.93	2.70
Zugspitze	A	5.48±8.57	3.37±7.54	4.70±7.28	4.42±5.97	/	/
	B	332	667	84	228	0	0
	C	6.93	6.76	9.39	8.55	/	/
Jungfraujoch	A	0.24±8.52	-0.33±7.29	-1.73±6.31	-1.85±6.39	-3.12±3.97	-3.37±3.91
	B	1048	2248	834	1569	386	835
	C	6.70	5.76	6.60	5.20	3.70	3.76
Egbert	A	-6.04±6.16	-6.08±6.24	-4.69±6.11	-5.85±6.90	-9.19±3.30	-9.23±3.49
	B	929	1877	608	1142	369	790
	C	5.38	4.48	5.30	5.22	3.49	3.74
Toronto	A	2.26±7.04	2.36±7.03	3.01±6.57	1.67±7.04	-1.34±4.92	-1.52±5.08
	B	899	1852	658	1280	403	848
	C	6.46	5.46	4.96	4.45	4.20	4.78
Izana	A	-8.54±7.54	-3.57±7.14	-3.26±2.27	-3.44±2.22	-1.57±4.63	-1.06±3.70
	B	696	2029	418	2245	760	2191
	C	5.33	5.49	1.36	1.37	3.95	3.40
Wollongong	A	-3.80±7.07	-2.02±5.89	-1.11±3.13	-1.53±2.69	/	/
	B	122	591	357	1038	0	0
	C	8.52	6.67	3.77	3.45	/	/
Lauder	A	-11.6±11.7	-2.16±11.6	-0.77±6.02	-0.77±6.02	-3.31±3.74	-3.90±3.22
	B	215	219	64	64	71	196
	C	10.1	9.68	5.45	5.45	2.82	3.04
Arrival Heights	A	/	/	/	/	-4.89±0.00	-5.33±1.76
	B	0	0	0	0	1	9
	C	/	/	/	/	0.00	1.48
Global	A	-2.46±9.22	-1.92±8.11	-1.21±6.39	-2.05±5.92	-3.53±5.14	-3.10±4.87
	B	5025	10 805	3080	7643	2124	5168
	C	7.76	6.56	5.25	5.03	3.43	3.60
R		0.13	0.14	0.18	0.19	0.29	0.37
P		5.72E-20	3.11E-49	1.22E-20	4.63E-56	5.71E-42	2.66E-164

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Table 7. A. Calculated bias (in %) of the SCIAMACHY measurements relative to the 3rd order polynomial fit through the ground based FTIR measurements for N₂O and CO₂ WFM-DOAS, using the small grid (SG=±2.5° LAT, ±5° LON) and large grid (LG=±2.5° LAT, ±10° LON) spatial collocation criteria. The indicated errors represent the 1σ standard deviations of the ensemble of individual biases. B. the number of correlative SCIAMACHY data. C. σ_{scat} , the percentage 1σ standard deviation of the daily averaged SCIAMACHY measurements towards the bias corrected polynomial FTIR fit (see Eq. 4).

Species→ Station ↓		N ₂ O, SG	N ₂ O, LG	CO ₂ , SG	CO ₂ , LG
Ny. Alesund*	A	/	/	-14.3±15.2	-14.7±12.1
	B	0	0	7	11
	C	/	/	16.4	10.7
Kiruna	A	-9.51±25.9	-8.92±26.3		
	B	473	887		
	C	24.7	24.0		
Harestua	A	-5.53±27.1	0.52±41.8		
	B	468	730		
	C	18.6	38.0		
Zugspitze	A	3.15±23.8	1.07±20.4		
	B	351	687		
	C	16.2	14.5		
Jungfraujoch*	A	-1.18±20.0	-1.60±22.6	-11.3±8.97	-12.0±7.42
	B	1117	2362	2846	6150
	C	12.7	14.9	7.36	5.80
Egbert*	A	1.57±28.4	1.53±27.1	-10.5±5.47	-10.3±5.60
	B	935	1881	2232	4520
	C	19.6	20.2	6.66	5.46
Toronto	A	3.51±28.8	4.16±27.5		
	B	897	1852		
	C	20.2	18.7		
Izana	A	4.12±45.2	1.50±29.1		
	B	802	2142		
	C	24.9	20.4		
Wollongong	A	-5.14±14.4	-1.53±13.7		
	B	132	588		
	C	9.26	9.27		
Lauder	A	-4.32±55.4	-3.08±56.8		
	B	267	275		
	C	50.2	50.4		
Arrival Heights	A	/	/		
	B	0	0		
	C	/	/		
Global	A	-0.22±31.4	0.13±28.2	-10.9±7.66	-11.3±6.77
	B	5442	11 404	5085	10 681
	C	24.1	25.1	7.30	5.76
R		0.10	0.09	0.26	0.24
P		1.58E-14	2.34E-23	5.01E-79	7.59E-141

* CO₂ g-b measurements available for these stations only

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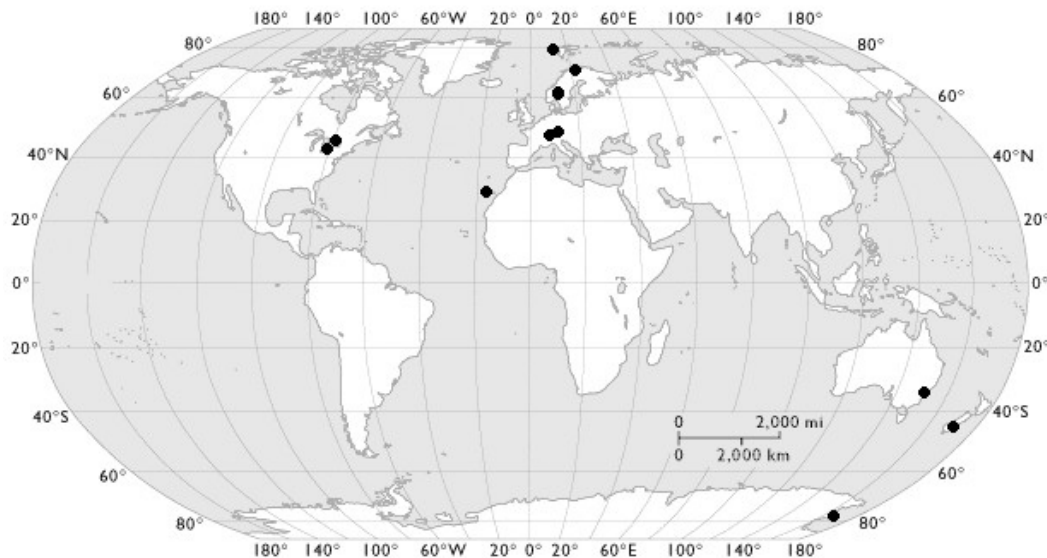


Fig. 1. Distribution of stations contributing to the delivery of correlative g-b FTIR data for comparisons with SCIAMACHY products – see also Table 1.

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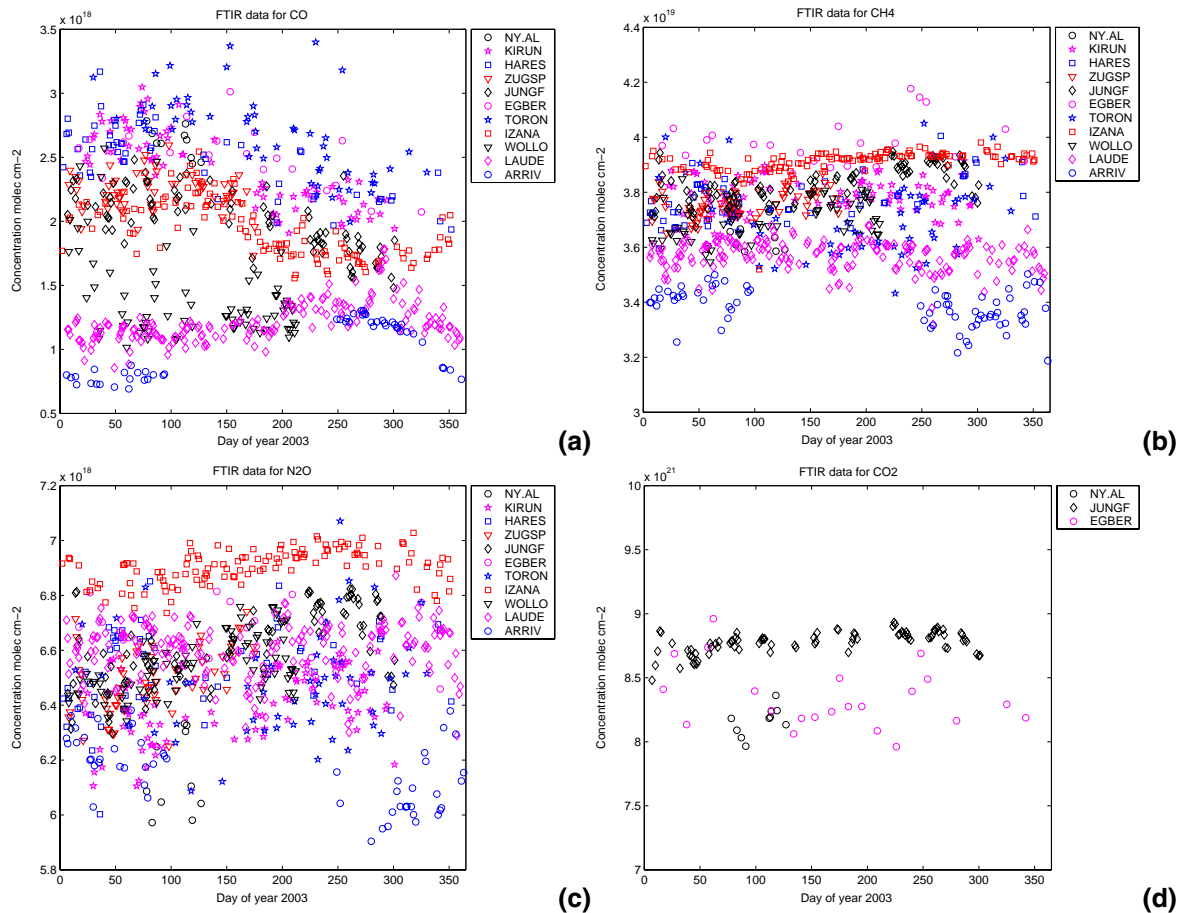


Fig. 2. Ground-based NDSC FTIR data of total column amounts of (a) CO, (b) CH₄, (c) N₂O, and (d) CO₂ for the year 2003 compiled at BIRA-IASB for the present validation exercise. In the plots, the column data have been normalized to zero station altitude according to a simple exponential law using a scaling height of 7.4 km (see text).

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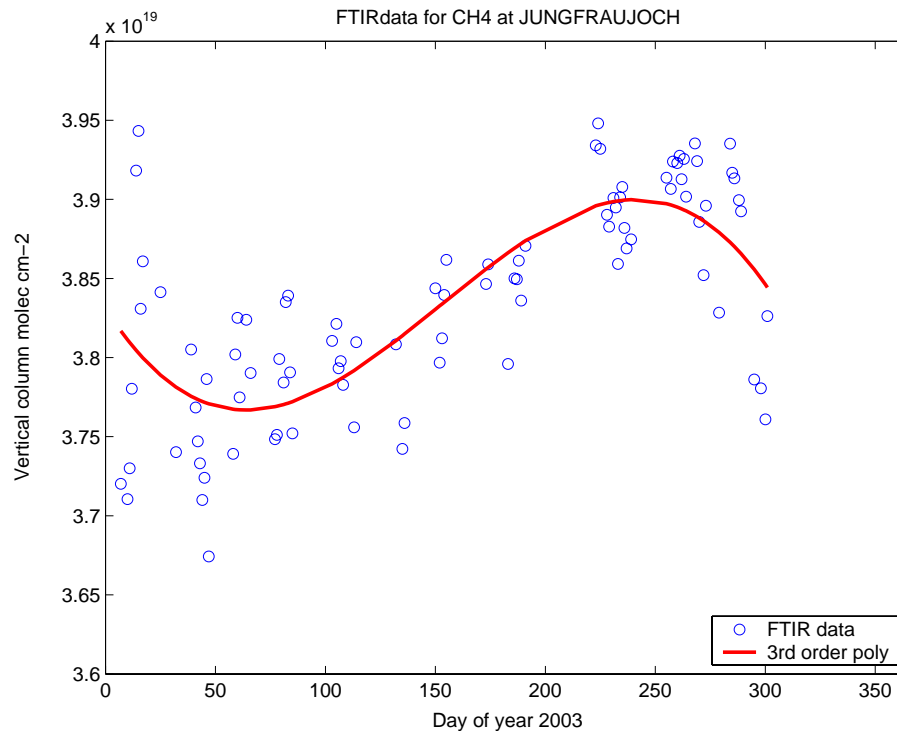


Fig. 3. The daily mean ground-based FTIR measurements for CH₄ at the Jungfraujoch (open circles), and the corresponding third order polynomial interpolation (solid line).

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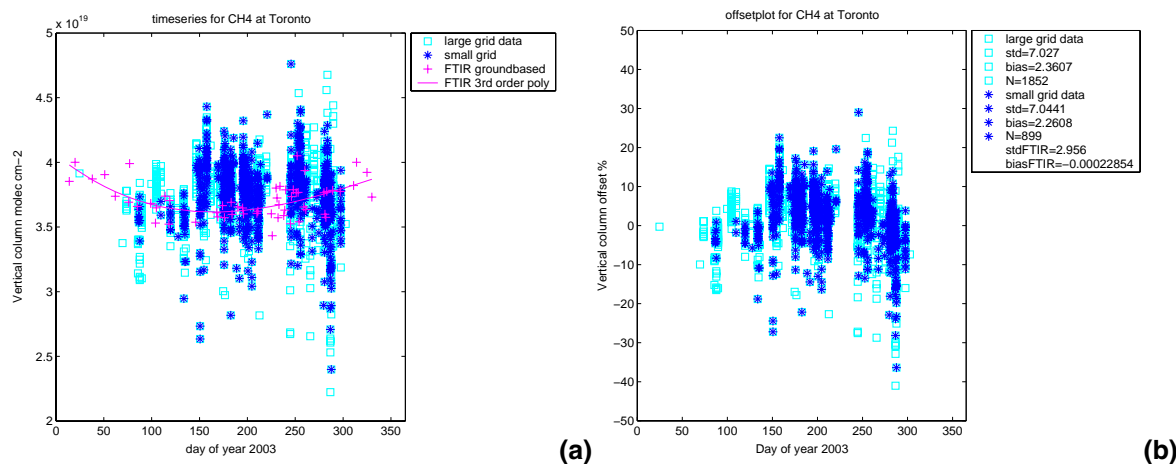


Fig. 4. Time series of CH₄ measurements at Toronto from g-b FTIR (+) and SCIAMACHY WFM-DOAS (open squares for large collocation grid; * for small collocation grid). Left-hand side: original data points (symbols) and 3rd order polynomial fit through the FTIR ground-based data (solid line). Right-hand side: Corresponding time series of relative biases of WFM-DOAS versus g-b interpolated data. Listed in the legend are the average bias, the standard deviation and the number of data points for the WFM-DOAS data sets as well as the average bias and standard deviation of the FTIR data relative to their polynomial fit.

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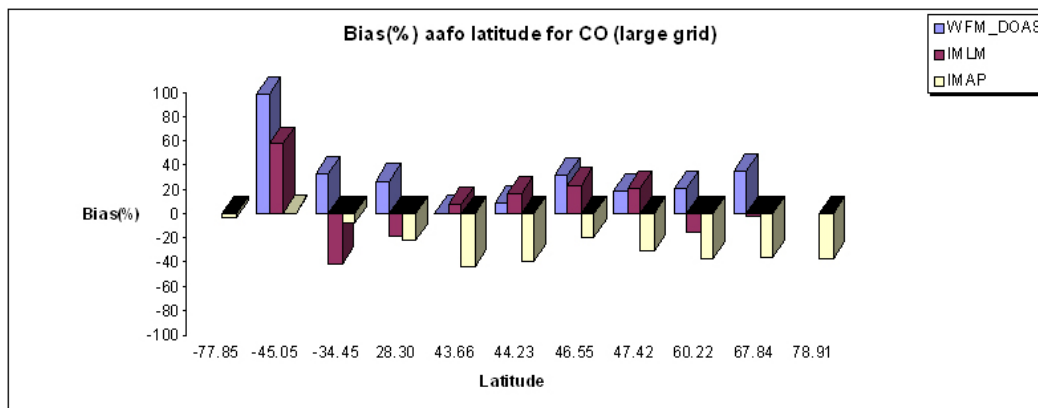


Fig. 5. The calculated percentage bias for CO as a function of latitude for a large grid collocation, for all three algorithms.

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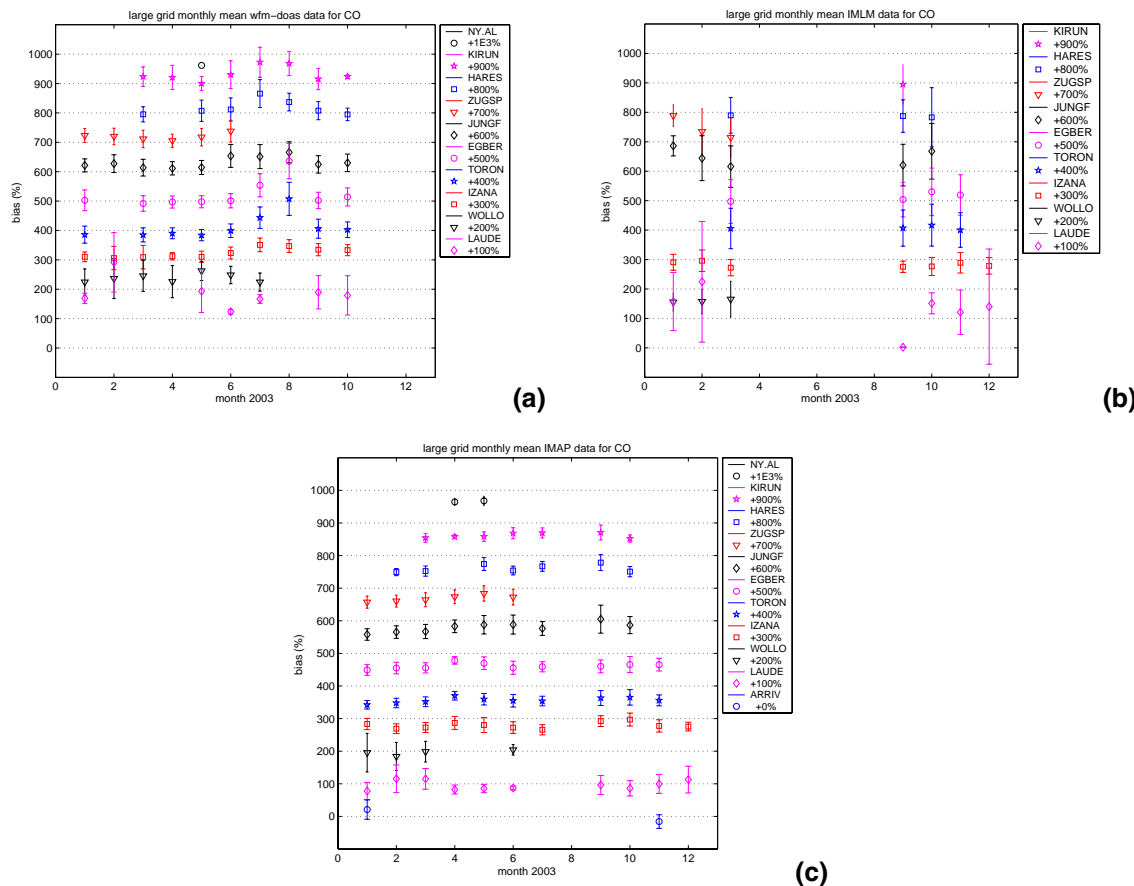


Fig. 6. Monthly mean biases for CO at all stations as a function of time for the year 2003, for the 3 algorithms (a) WFM-DOAS, (b) IMLM and (c) IMAP. For plotting purposes, the bias values for each station (whether they contain data or not) have been incremented by 100% starting from Arrival Heights (+0%), Lauder (+100%) till Ny.Alesund (+1000%) following the ordering of the stations of Table 1. The large grid was chosen for the spatial collocation criterium.

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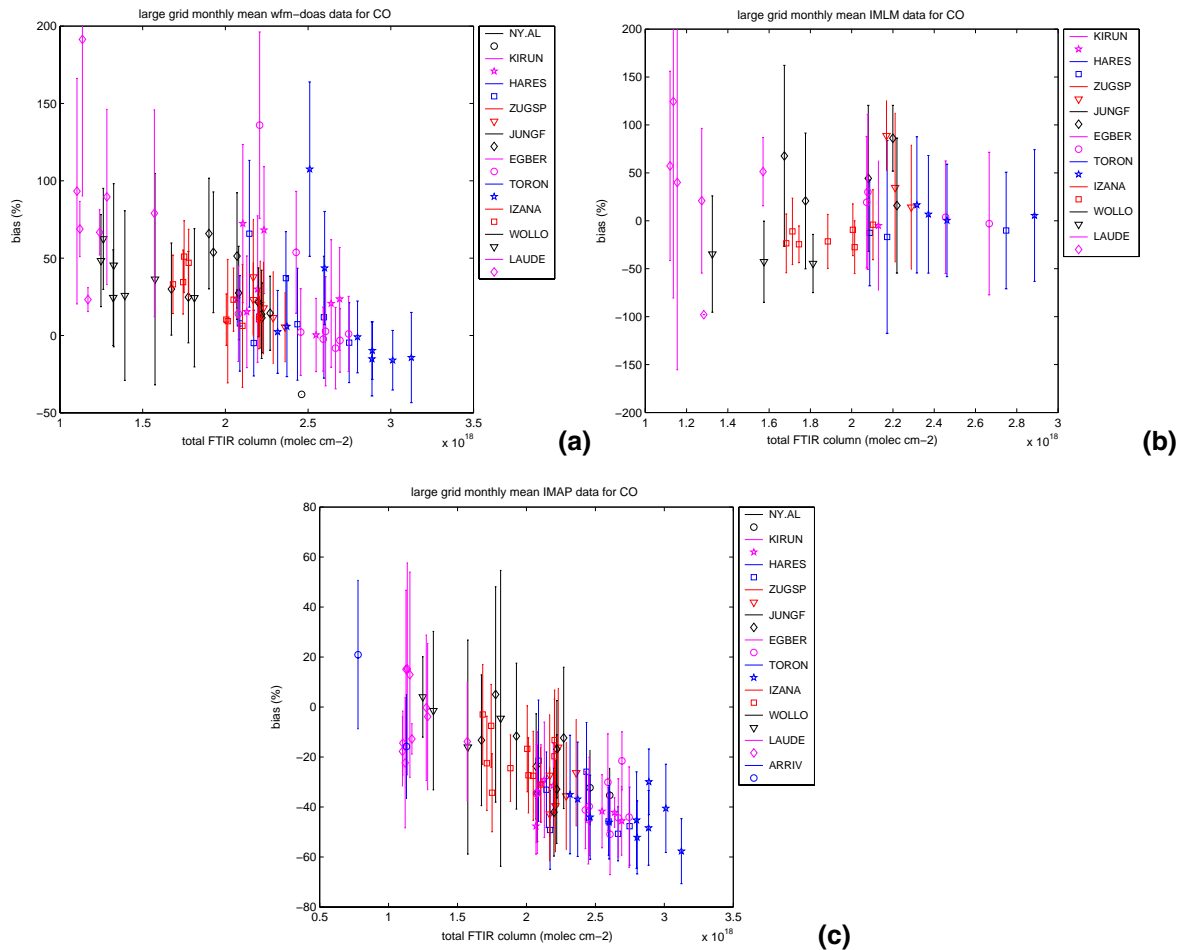


Fig. 7. Monthly mean biases for CO at all stations as a function of monthly mean FTIR total column values, for the 3 algorithms (a) WFM-DOAS, (b) IMLM and (c) IMAP. Note that the Y-axes do not cover the same range.

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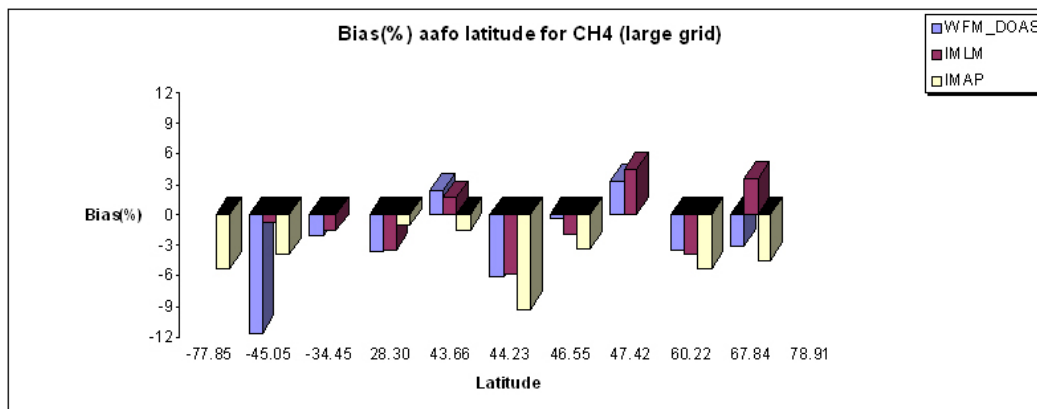


Fig. 8. The calculated percentage bias for CH₄ as a function of latitude for a large grid collocation, for all three algorithms.

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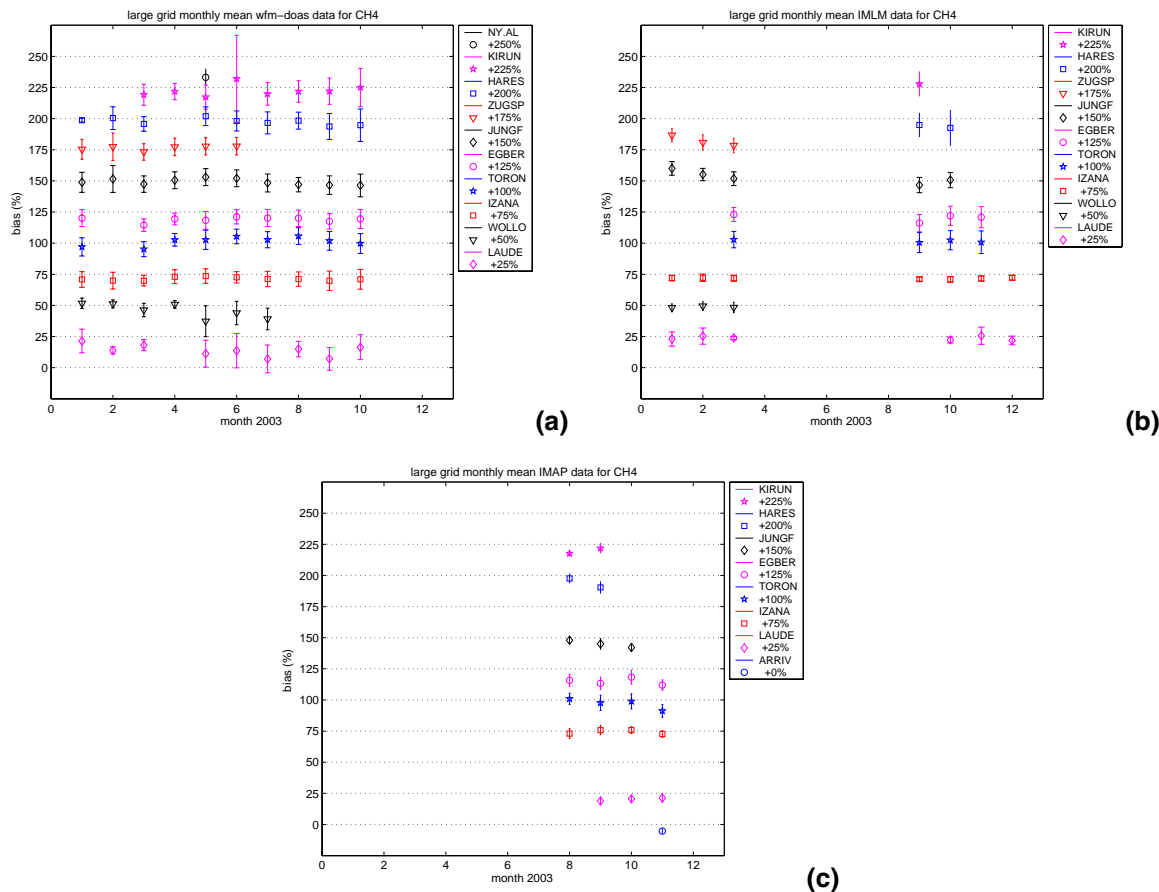


Fig. 9. Monthly mean biases for CH₄ at all stations as a function of time for the year 2003, for the 3 algorithms (a) WFM-DOAS, (b) IMLM and (c) IMAP. For plotting purposes, the bias values for each station (whether they contain data or not) have been incremented by 50% starting from Arrival Heights (+0%), Lauder (+50%) till NyAlesund (+500%) following the ordering of the stations of Table 1. The large grid was chosen for the spatial collocation criterium.

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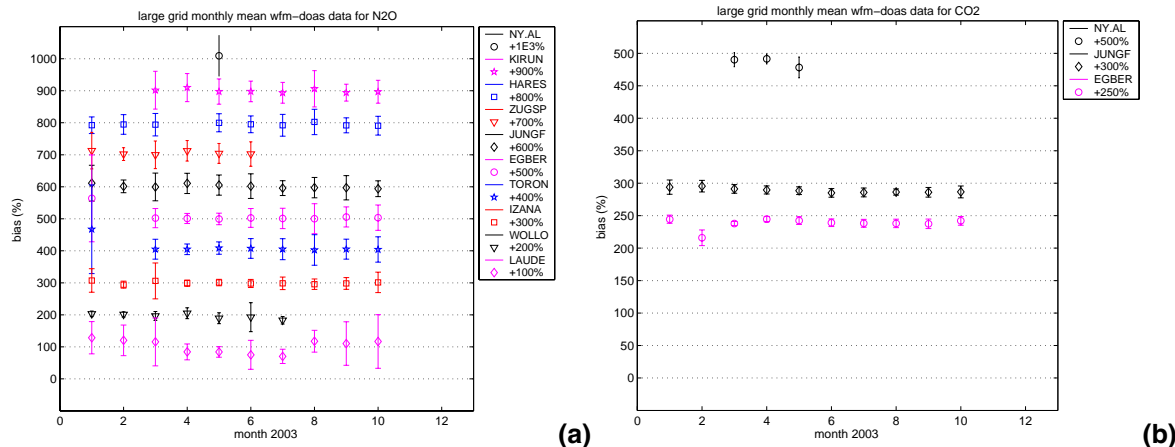


Fig. 10. Monthly mean WFM-DOAS biases for (a) N₂O and (b) CO₂ at all stations as a function of time for the year 2003. For plotting purposes, the bias values for each station (whether they contain data or not) have been incremented by 100% in case of N₂O or 50% in case of CO₂, starting from Arrival Heights (+0%), Lauder (+100% or 50%) till Ny.Alesund (+1000% or 500%) following the ordering of the stations of Table 1. The large grid was chosen for the spatial collocation criterium.

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