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Bridge to Copernicus

Final project report

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Scientific report

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Summary

NILU has a mandate to monitor air quality and particularly its changes over time, both nationally through Miljødirektoratet (MD) and internationally through the European Monitoring and Evaluation Programme (EMEP). Satellite data related to atmospheric composition are increasingly used for monitoring as they provide long time series of spatially continuous observations. It is therefore essential for NILU to begin preparing for the upcoming Copernicus missions. Here, we evaluate methane products from AIRS, TES, TANSO-FTS and SCIAMACHY as added value for GHG monitoring in Norway and Svalbard. As expected, due to the low sensitivity of the sensors to ground-level methane concentrations, large deviations are seen when comparing satellite observations to *in situ* data from Birkenes and Ny-Ålesund. Higher level products (L4), combining satellite and ground-based information, seem more appropriate for future reporting purposes. Further, we investigated the usability of the current set of long-term operational ground-based MAX-DOAS stations worldwide for inter-comparing their NO_2 observations to those of satellite-based instruments, in particular OMI and GOME-2A. The two data sources agree very well for sites located in rural, non-polluted regions. For sites located in polluted areas, we found strong systematic biases, large random errors, or slightly shifting systematic biases. The systematic biases can be explained primarily by the strong spatial gradients in NO_2 levels in urban areas in conjunction with the large differences in the spatial representativity of the measurements. We evaluated the possibility to use the now relatively long time series of MAX-DOAS observations to fit a statistical trend model and to directly compare the resulting trends to those obtained for the satellite-based time series for the same area and time period. It was found that the sites with approximately 50 months of valid data for both data sources showed quite similar long-term trends and that sites with fewer than 30 months of valid data exhibited significant discrepancies in the resulting trends.

Sammendrag

NILU har flere prosjekter for overvåkning og evaluering av luftkvalitet, både nasjonalt gjennom Miljødirektoratet og internasjonalt gjennom «European Monitoring and Evaluation Programme» (EMEP). Satellitter som måler atmosfærens sammensetning, blir stadig oftere brukt i overvåkingen, og de kan gi lange tidsserier med kontinuerlige observasjoner over et stort område. Det er derfor viktig for NILU å være forberedt på kommende oppdrag fra Copernicus-programmet. Vi har studert metanprodukter fra AIRS, TES, TANSO-FTS og SCIAMACHY, som et supplement til overvåkningen av drivhusgasser i Norge og på Svalbard. Som forventet, på grunn av lav satellitt sensorfølsomhet ved bakkenivå, blir det relativt store avvik mellom satellittdata og *in situ* data fra Birkenes og Ny-Ålesund. Nye produkter, som kombinerer satellitt- og bakkebasert informasjon, synes mer hensiktsmessig for fremtidige rapporteringsformål. Videre har vi studert bakkebaserte MAX-DOAS stasjoner over hele verden og sammenlignet med NO_2 observasjoner fra satellitt, spesielt OMI og GOME-2A. Satellitt- og bakkebaserte målinger samsvarer bra i rurale områder med lite forurensning. For områder med mer forurensning fant vi imidlertid systematiske avvik. Dette er primært knyttet til romlig representativitet og store NO_2 gradienter i urbane områder. MAX-DOAS observasjonene har blitt tilpasset en statistisk trendmodell og sammenlignet med satellittbaserte trender for samme område og tidsperiode. Det ble funnet at stasjoner med mer enn 50 måneder med data ga nokså samsvarende trender for MAX-DOAS og satellitt. Stasjoner med mindre enn 30 måneder med data ga derimot sprikende trendresultater.

Bridge to Copernicus
Final project report

1 WP 1: Satellite observations of methane above Norway and the Norwegian Arctic region

Ann Mari Fjæraa and Tove Svendby

1.1 Introduction: satellite and *in situ* methane (CH_4) data

Satellite products can, in their current state, not replace the ground-based monitoring of greenhouse gases, but they give an important contribution to regional and global GHG mapping. Earlier studies (Vik et al., 2011, Stebel et al., 2013) have shown that the uncertainty in many satellite products, particularly in polar areas, are too large to be used alone in reporting to national authorities and international projects and programs. However, satellite products make it possible to investigate the geographical extent of e.g. enhanced methane concentrations, emission and long-range transport. In the project “Bridge to Copernicus”, jointly financed by the Norwegian Space Center and NILU, Norwegian Institute for Air Research, various methane (CH_4) satellite products have been investigated, particularly focusing on Norway and the Norwegian Arctic region. Results from the project are summarized here. Table 1 gives an overview of the methane measuring satellites and their time of operation, together with the duration of *in situ* measurements at Zeppelin mountain, Ny-Ålesund, and Birkenes. The ground-based (GB) observations at Zeppelin started in 2001, two years prior to the starting date in Table 1.

Table 1: Overview of methane measuring satellites and their time of operation. The duration of Norwegian ground-based measurements are also marked. GB data are available in near-real time (NRT) from 2011 onward.

GB station, Satellite instrument	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016
Zeppelin *in situ*									NRT	NRT	NRT	NRT	NRT	NRT
Birkenes *in situ*									NRT	NRT	NRT	NRT	NRT	NRT
SCIAMACHY / ENVISAT														
TANSO-FTS / GOSAT														
AIRS / AQUA														
TES / AURA														
ISAI / MetOp														
TROPOMI / Sentinel-5p														

A number of satellite sensors are able to detect CH_4 in the atmosphere, but for many of those the sensitivity to the surface level is very limited. Instruments operating in the thermal infrared (TIR), e.g. TES on Aura, AIRS on AQUA or IASI on MetOp have largest sensitivity to the mid-upper tropospheric CH_4. Near-IR (NIR) CH_4 instruments, e.g. SCIAMACHY on ENVISAT and TANSO on GOSAT have largest sensitivity to the total column amount of CH_4. Limb sensors like MIPAS observe CH_4 in the upper troposphere and stratosphere.

In the current project, we have focused on methane products from four satellites: AIRS, SCIAMACHY, TES, and TANSO. Table 2 lists the availability of the data, the resolution, and the product versions studied for each of these satellites. The satellite data have been compared to ground-based *in situ* data from the two Norwegian monitoring stations[1].

For the methane GB/satellite inter-comparison an area of 2x2 degrees around Zeppelin and Birkenes were defined, and the satellite pixels closest to the observational sites were selected.

Table 2: Overview of methane satellite products used.

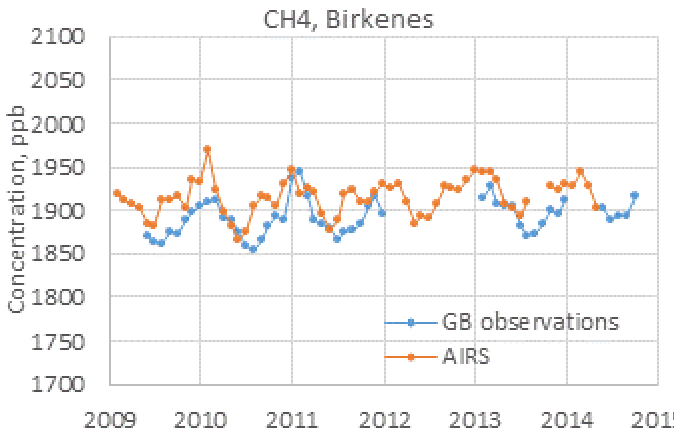
Satellite	Frequency	Year	Layers	grid	Product version
AIRS (AQUA)	Monthly	2007-2014	Profile(L24)	1 x 1°	L3, AIRX3STM, NASA
TES (AURA)	Daily	2004-2012	Profile(L15)	2 x 4°	L3, R11.01.00, NASA
	Monthly	2004-2011	Profile(L15)	2 x 4°	L3, R13.01.00, NASA
TANSO (GOSAT)	Daily	2009-05.2011	Profile(L17)	2.5 x 2.5°	L4, v01.01, NIES
SCIAMACHY (ENVISAT)	Daily	2003-04.2012	total column	Swath	IMAP v6.0, TEMIS

1.2 AIRS

The Atmospheric Infrared Sounder, AIRS, onboard the AQUA satellite (NASA) was launched in 2002 and is still in operation. Monthly mean data, product AIRX3STM, are available for the period 2007-2014. AIRS provides both total column and CH_4 profile data. Figure 2a (upper panel) shows monthly mean methane data from the CH4_VMR_A ascending product (orange dots) and ground based observations (blue dots) at Birkenes. The correlation and bias between satellite and GB measurements are 0.62 and 22 ppb, respectively, the satellite data being higher than the GB observations. For Zeppelin (Figure 2b, lower panel) the deviation between AIRS satellite values and GB observations are larger. Since the Zeppelin observatory is located at an altitude of 474 m, satellite data at the two lowest layers are used in the study; altitude level 0 (L0: 0-750 m.a.s.l.) and level 1 (L1: ~750-1450 m.a.s.l.), marked as orange and grey lines, respectively. The correlations between AIRS and GB measurements at Zeppelin are 0.65 for L0 and 0.73 for L1. The corresponding biases are quite large: 68 and 56 ppb for L0 and L1, respectively.

[1] Note that the Near Real Time ground based methane data for 2013 and 2014 are preliminary and minor adjustments might occur after data finalization.

a)



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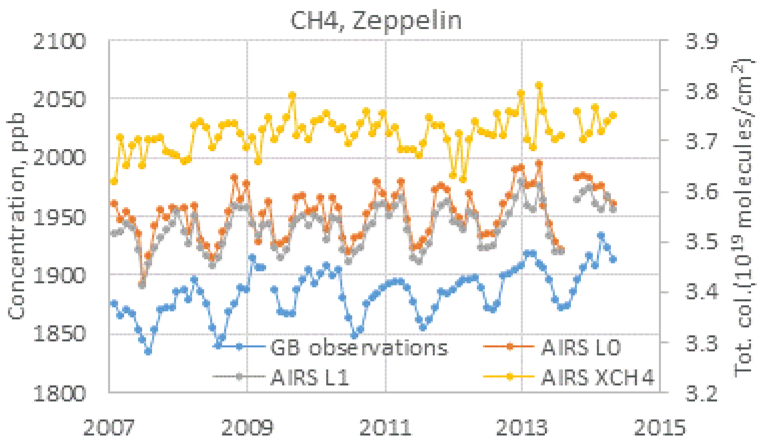


Figure 1: ***a)*** *(upper panel): Monthly mean ground-based CH_4 measurements at Birkenes (blue) and the corresponding satellite observations from AIRS surface layer (orange),* ***b)*** *(lower panel**)**: Monthly mean CH_4 data at Zeppelin. Blue line represents GB observations, orange line is AIRS data from surface layer L0, grey line is AIRS data from layer L1, whereas yellow line is CH_4 total column values from AIRS.*

AIRS sensitivity in the polar region is usually smaller than in the mid-latitude and tropics. Small sensitivities indicate that the retrieved CH_4 will be closer to the first-guess (Xiong, 2008). Figure 2 shows monthly mean methane from AIRS L0 in December 2013. The high AIRS methane concentrations in the polar region are likely caused by the reduced sensitivity and uncertain retrieval algorithms at high latitudes, also evident from Figure 1b.

Total column methane data from AIRS, product XCH4_A, is shown as yellow line in Figure 1b. As expected the correlation between total column data and ground-based surface concentrations are not as good as for L0 and L1. A summary of the AIRS statistics, compared to *in situ* measurements, are presented in Table 2. AIRS total column values are higher than L0 and L1 values.

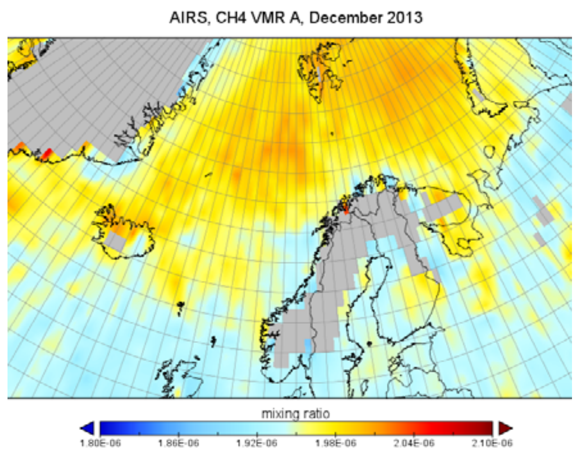


Figure 2: AIRS average methane concentrations (L0 surface layer) in December 2013.

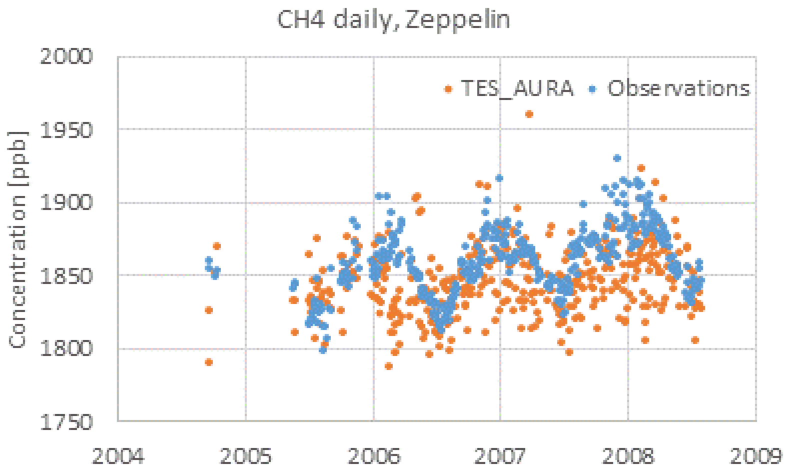
Table 3: Bias, scatter and correlation (R) between AIRS AQUA satellite and ground-based methane data.

Monthly	Bias	Scatter	R	N
Zeppelin (L0)	67.5	17.3	0.65	85
Zeppelin (L1)	56.4	14.5	0.73	85
Birkenes (L0)	22.1	18.7	0.62	42

1.3 TES

The Tropospheric Emission Spectrometer, TES, onboard the AURA satellite (NASA) was launched in 2004 and is still in operation. Data is currently available from September 2004 to October 2012. The data coverage has varied significantly over time, and since mid 2009 no observations have been available at high latitudes. Consequently, we have not enough Birkenes *in situ* measurements for a proper satellite/GB inter-comparison.

a)



b)

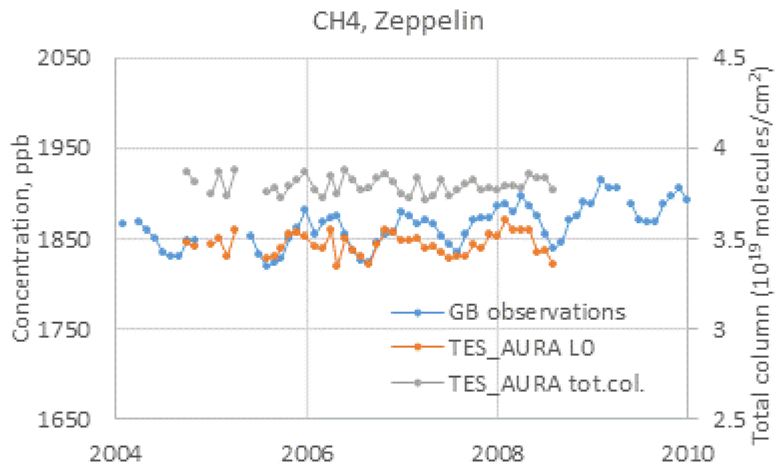


*Figure 3: **a** (upper panel)**)**: Daily ground-based CH_4 measurements at Zeppelin (blue dots) and the corresponding satellite observations from TES (AURA) surface layer (orange dots), **b)** (lower panel): Monthly mean CH_4 data at Zeppelin.*

Figure 3a shows daily TES AURA methane measurements (orange dots) and GB observations (blue dots) during days of simultaneous TES/GB measurements. The correlations and biases are listed in Table 4. Contrary to AIRS, the TES satellite data are in general lower than the GB observations. The bias between AIRS daily data (surface level, L0) and GB observations is -13 ppb and the correlation is 0.33. For the monthly mean data (Figure 3b) the correlation is significantly higher, 0.61 for the lowest TES level (L0) and 0.64 for the second lowest level (L1). The bias between satellite and GB observations are around -15 and –19 ppb for L0 and L1, respectively. Total column values are higher than the L0 and L1 values.

Table 4: Bias, scatter and correlation (R) between TES AURA satellite and ground-based methane data.

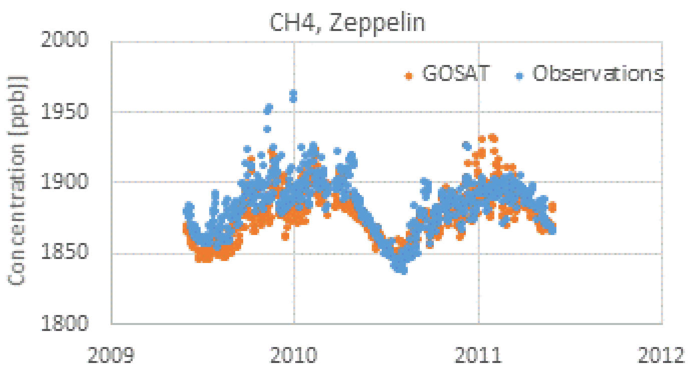
Daily	**Bias**	**Scatter**	**R**	**N**
Zeppelin (L0)	-12.9	26.5	0.33	423
Monthly	**Bias**	**Scatter**	**R**	**N**
Zeppelin (L0)	-15.4	15.9	0.61	39
Zeppelin (L1)	-18.8	16.3	0.64	39

1.4 TANSO

The Greenhouse Gases Observing Satellite, GOSAT, was launched in 2009 and is still in operation. The GOSAT project is a joint effort of JAXA, MOE and NIES (Japan). For comparison between GOSAT and the Norwegian ground-based measurements, we have used GOSAT Level 4 data product of CH_4. This product consists of the Level 4A (L4A) surface CH_4 flux data estimated with GOSAT and ground-based observations and the Level 4B three-dimensional global CH_4 distributions predicted with the estimated L4A fluxes. The data content is described in more detail in the GOSAT (L4) Data Product Format Description document (http://data.gosat.nies.go.jp/).

Table 5a and Table 5b show comparisons of GOSAT satellite data (orange dots) and *in situ* observations (blue dots) at Zeppelin and Birkenes. As expected, the correlations are very good: 0.78 and 0.72 for Zeppelin and Birkenes, respectively. The overall biases are relatively small, within ±7 ppb. A summary of the GOSAT statistics, compared to the Norwegian *in situ* measurements, are summarized in Table 5. The good agreement between the GOSAT L4 product and the GB measurements as expected as global ground-based observations are taken into account.

a)



b)

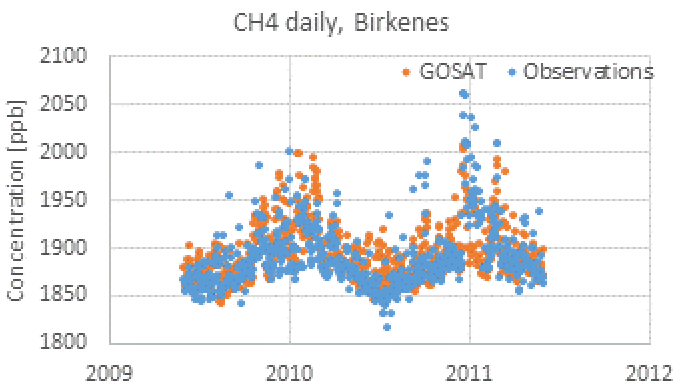


*Figure 4: **a)** (upper panel): Daily ground-based CH_4 measurements at Zeppelin (blue) and the corresponding satellite observations from GOSAT surface layer (orange),*
***b)** (lower panel): GOSAT and GB daily measurements at Birkenes.*

Table 5: Bias, scatter and correlation (R) between GOSAT and ground-based methane data

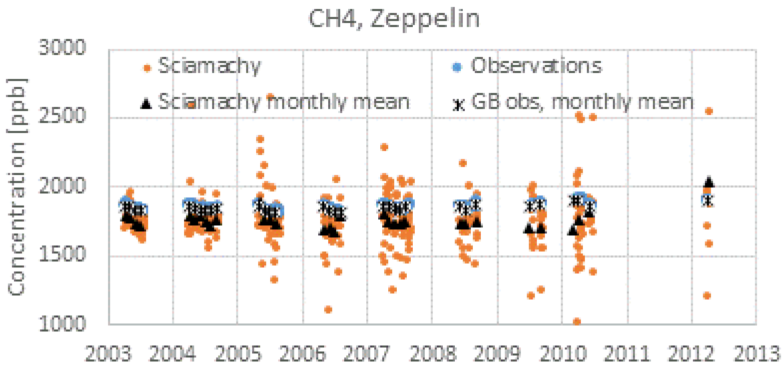
Daily	Bias	Scatter	R	N
Zeppelin (L0)	-6.5	12.5	0.77	682
Zeppelin (L1)	-6.7	12.1	0.78	682
Birkenes (L0)	4.8	24.3	0.72	724

1.5 SCIAMACHY

SCIAMACHY, SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY, onboard ENVISAT was launched in 2002 and was in operation until April 2012. Only total CH_4 column data (average ppb) are retrieved from SCIAMACHY, which makes the day-to-day comparison to GB surface measurements less valid as a perfect fit is not expected. Figure 5 shows daily and monthly mean SCIAMACHY methane measurements (orange dots) and GB observations (blue dots) during all days of simultaneous GB/satellite measurements.

As seen from the figure the day-to day variations in the SCIAMACHY data are much larger than the GB observations. For example, in 2012 the SCIAMACHY CH_4 total column at Zeppelin varied from 1206 ppb to 3528 ppb. Whereas the IR (AIRS and TES) total column values are larger than the *in situ* ground-based data, on average the SCIAMACHY total column monthly mean are smaller than the *in situ* vales.

a)



b)

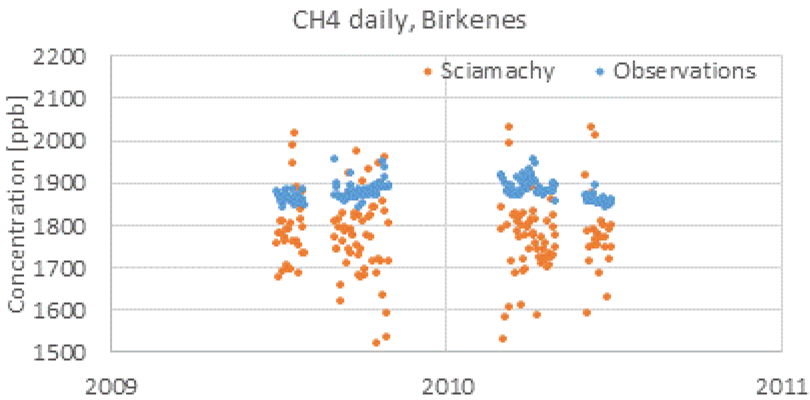


Figure 5: ***a)*** *Upper panel: Daily ground-based CH_4 measurements at Zeppelin (blue) and the corresponding total column satellite observations from SCIAMACHY (orange). Black triangles and stars represent monthly mean values from SCIAMACHY and GB observations, respectively,* ***b)*** *Lower panel: Daily CH_4 GB data at Birkenes (blue) and from SCIAMACHY (orange).*

An evaluation of methane products from SCIAMACHY is also described in the Product Validation and Intercomparison Report (PVIR) from ESA-CCI, 2013 (available on www.esa-ghg-cci.org). Two different SCIAMACHY methane retrieval products were studied, the WFMD (Weighting Function Modified DOAS, Buchwitz et al., 2006) and the IMAP (Iterative Maximum A Posteriori-DOAS, Frankenberg et al., 2005) products, and compared to total column FTIR observations at 12 different sites. One of the stations, Sodankyla (67.37ºN, 26.63ºE), can be classified as "Arctic" and is relevant for our study. For the IMAP and WFMD retrieval products, the correlation between FTIR and SCIAMACHY satellite data were 0.24 and -0.21, respectively. The corresponding scatters were 54 and 140. This shows that SCIAMACHY has relatively poor correlation and high scatter for high latitude stations, even compared to ground-based FTIR total columns measurements. This is especially the case for the WFMD products.

The PVIR report and a study from Petersen (2010) conclude that FTIR observations of methane in the tropics agree well with SCAIAMACHY observations retrieved with the IMAP algorithm. However, SCIAMACHY is not able to detect methane emissions of biomass burning due to the retrieval method used.

The methane observations at Zeppelin and Birkenes are surface concentrations. Saad et al. (2014) studied the Total Carbon Column Observing Network (TCCON), a global ground-based network of total CH_4 measurements, and calculated the contribution of stratospheric to the total column methane. These results give an indication of the expected agreement between the Norwegian ground-based measurements and total column CH_4 values from satellite. For the high latitude station at Sodankyla, Saad et al. (2014) showed that the tropospheric CH_4 concentration in average was 60-70 ppb higher than the total column mixing ratio. These finding can partly explain the high negative bias seen in Figure 5. Saad et al. (2014) also showed that the seasonal cycle of CH_4 tropospheric and total column differ.

1.6 Conclusion and outlook

Comparisons between Norwegian ground-based methane measurements and satellites data indicate a relatively good monthly mean correlation. However, on a day-to-day basis the agreement is less satisfactory. An exception is the GOSAT L4 methane product, which uses global ground-based observations in the data processing.

It is important with continued focus on the evaluation of Arctic satellite data. The current study reveals large methane differences between various satellite products, e.g. between AIRS and TES. The deviation seems to increase with latitude. Improved retrieval algorithms, especially in the polar region, will most likely improve the satellite products in upcoming years and make them more useful for methane assessments. Continued evaluation against high latitude ground-based observations is crucial.

The longest satellite based methane time series are around 9 years, which is too short for reliable trend studies. Thus, we have focused on GB/satellite methane comparisons rather than trend evaluations in our study. With a few more years of data, trend studies with regional and global coverage may be performed with relative high confidence.

1.7 References

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2 WP 2: Preparing for the use of air quality observations from Sentinel-5p for air quality monitoring over Europe

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2.1 Introduction

NILU has a mandate to monitor air quality and particularly its changes over time, both nationally through Miljødirektoratet (MD) and internationally through the European Monitoring and Evaluation Programme (EMEP). Satellite data related to atmospheric composition are increasingly used for monitoring as they provide long time series of spatially continuous observations (Hilboll et al., 2013; Richter et al., 2005; Schneider and van der A, 2012; Schulz et al., 2013). It is therefore essential for NILU to begin preparing for the upcoming Copernicus programme and, in particular, for evaluating the data expected from the Sentinel 5-P satellite. In addition to Sentinel-3, this will be the main Copernicus satellite platform used at NILU for the next few years.

As nitrogen dioxide (NO_2) is one of the most prominent atmospheric pollutants (Schneider and van der A, 2012; Schulz et al., 2013), the main focus of this WP was on NO_2 and its change over time; however, methodologies to be developed in WP2 apply to other atmospheric pollutants such as SO_2 and O_3. Tools developed at NILU to obtain global trend maps for NO_2 from the archive of SCIAMACHY, OMI, and GOME-2 data were used to estimate NO_2 trends for selected regions in Europe. To estimate the uncertainty of satellite-based trends and to establish a framework for evaluating upcoming TROPOMI data, it is essential to compare such data to ground-based remote sensing observations. This involved compiling a ground-based dataset, developing a comparison methodology, and implementing software tools to carry out these tasks. All tools used in WP2, as well as those developed within the framework of the SatMonAir-1 and -2 projects, will be made available for future monitoring activities at NILU within the Copernicus programme.

2.2 Data and Methodology

In the following section we briefly describe the datasets that were used within the framework of this project and the methodology that was used to process and analyse them.

2.2.1 Satellite data

Operational satellite remote sensing of NO_2 has been carried out since 1995 when the Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999; Richter and Burrows, 2002) was first launched. Beginning in 2002, the observations were continued by the SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY) sensor onboard of Envisat (Bovensmann et al., 1999;

Gottwald et al., 2011), and subsequently complemented in 2004 by the Ozone Monitoring Instrument (OMI) (Levelt et al., 2006) as well as the Global Ozone Monitoring Experiment-2 (GOME-2) instrument in 2006 (Munro et al., 2006) and now onboard of both MetOp-A and MetOp-B. Table 6 provides an overview of the primary characteristics of the various satellite instruments.

Table 6: Overview of the most important past and near-future satellite instruments relevant for observing tropospheric NO_2 and other trace gases.

Sensor	GOME	SCIAMACHY	OMI	GOME-2	TROPOMI
Platform	ERS-1	ENVISAT	Aura	MetOp	Sentinel-5P
Data availability	1996 to 2003	2002 to 2012	2004 to present	2006 to present	2015 to ?
Spatial resolution at nadir	320 km x 40 km	60 km x 30 km	13 km x 24 km	80 km x 40 km	7 km x 7 km
Daily coverage	Near-Global	Partial (due to alternating nadir/limb observation)	Near-Global (significantly reduced due to instrument failure since 2007)	Near-Global	Global
Overpass time	10:20 LST	10:00 LST	13:45 LST	09:30 LST	13:30 LST

Two of the listed spaceborne instruments were used here: OMI and GOME-2A. The Ozone Monitoring Instrument (OMI) is based on the experiences acquired from both GOME and SCIAMACHY. It combines their advantages, measuring the complete spectrum in the UV/VIS wavelength range at a comparatively high spatial resolution of 13 km × 24 km, while providing daily global coverage. The OMI instrument is flying on the National Aeronautics and Space Administration's Earth Observing System Aura platform as part of the A-train constellation of satellites. In contrast to the other instruments measuring NO_2, which have equator crossing times around 10:00 local time, OMI has an equator crossing time of approximately 1:45 LST in the afternoon, and therefore probes the Earth's atmosphere under different conditions. Aura/OMI was launched in 2004 and has been continuously providing data. Beginning in June 2007, OMI has suffered from several row anomalies affecting the quality of the Level 1B and Level 2 data products. Level-3 products are produced after filtering for the affected anomalies.

The Global Ozone Monitoring Experiment-2 (GOME-2) is a scanning spectrometer onboard of the MetOp series of satellites. As a modified and improved successor of ERS-2's GOME instrument, GOME-2 measures in a spectral range of 240 nm to 790 nm with a varying spectral resolution between 0.24 nm and 0.53 nm (Callies et al., 2000). The spatial resolution of the instrument is 80 km × 40 km.

The full global archives of NO_2 satellite data were acquired for two products. The daily NASA OMNO2d product was chosen for OMI, whereas the daily TEMIS product was chosen for GOME-2A. The data for the latter instrument was acquired from the TEMIS service operated by KNMI (www.temis.nl). For GOME-2A data, version 2.3 of the combined modelling/retrieval/assimilation approach (Boersma et al., 2004) was used. Monthly averages including uncertainties were computed from the daily data.

2.2.2 MAX-DOAS data

The selection process was mainly based on the length of the available time series, with the main selection criterion being that the time series should at least be on the order of 4 to 5 years long so they can be used to some extent for some trend analysis. The data owners were subsequently contacted and in most cases provided their permission for the use of the data.

Overall, datasets from seven sites were acquired. These included two stations operated by BIRA-IASB, namely Observatoire de Haute Provence, France, and Xianghe in China (although the latter ended up not being used). Data from five stations were acquired from the MADRAS network (Irie et al., 2012; Kanaya et al., 2014). These stations include three sites in Japan, namely Yokosuka, Cape Hedo, and Fukue, one site in Korea (Gwangju) and one site in Russia (Zvenigorod). Two other suitable sites operated by the University of Bremen were identified to be suitable for the purposes of this study, but while permission to use the data was granted, no data was delivered up to this point. Figure 6 gives an overview over the locations of the sites for which MAX-DOAS data could be acquired within the framework of this project.

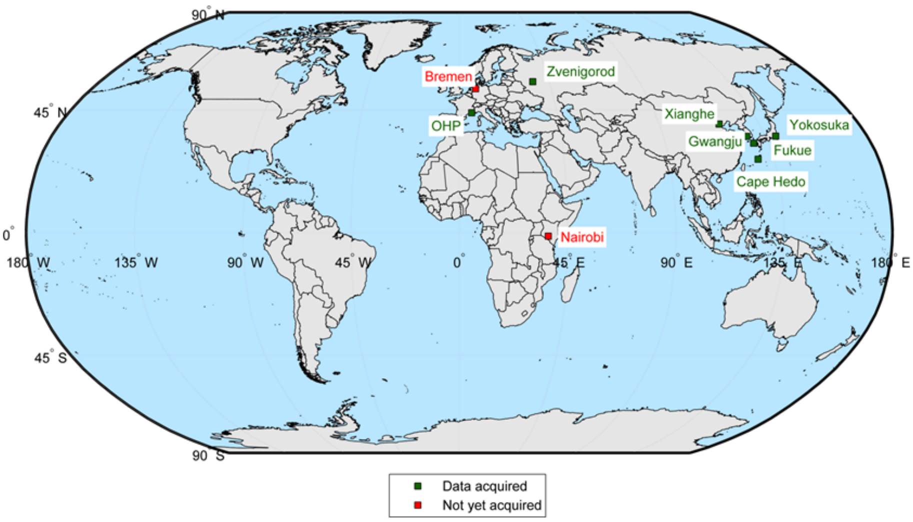

Figure 6: Global overview map of MAX-DOAS sites with time series of at least 4-5 years in length. Some of the stations such as Bremen and Nairobi have suitable time series but the data could not be obtained within the framework of this project.

The processing of the MAX-DOAS data involved finding the right matchup time between satellite MAX-DOAS observations and a subsequent filtering of the MAX-DOAS time series for all data points within plus/minus 30 minutes of the satellite overpass. Then, monthly averages were computed from these filtered datasets and these could then be directly compared to the monthly averaged satellite data which were calculated from daily observations..

2.2.3 Trend analysis

The trend analysis methodology largely follows the one used by Schneider and van der A (2012) and Schneider et al. (2014). In order to compute trends from the satellite data we follow the methodology suggested by Weatherhead et al. (1998) and later applied by van der A et al. (2006), Good et al. (2007), and van der A et al. (2008) for fitting a seasonal signal and a linear trend to monthly data. The monthly average NO_2 tropospheric column Ct at time t (in months) was thus modeled as

$$C_t = \mu + S_t + \frac{1}{12}\omega t + R_t \qquad (1)$$

where μ is a constant, S_t is a seasonal component, ω is a linear trend and R_t is the residual variability. The seasonal component S_t is modeled as

$$S_t = \sum_{j=1}^{4}\left[\beta_{1,j}\sin\left(\frac{2\pi jt}{12}\right) + \beta_{2,j}\cos\left(\frac{2\pi jt}{12}\right)\right] \qquad (2)$$

where $\beta_{1,1}$ through $\beta_{2,4}$ are coefficients of the fit. The residual variability R_t is assumed to be autoregressive of order 1 and was modeled as

$$R_t = \phi R_{t-1} + \epsilon_t \qquad (3)$$

where ϕ is the first order autocorrelation and ϵ is a random error component.

The significance of the trend (Santer et al., 2000) was computed based on the suggestion of Tiao et al. (1990) and Weatherhead et al. (1998) such that a trend ω is considered to be significant and to represent a real geophysical trend with a 95% confidence when $|\omega/\sigma_\omega| > t_\omega$, where σ_ω is the uncertainty of the trend and t_ω is the value of the Student's t-distribution for a significance level of $\alpha = 0.05$ and the degrees of freedom given for the time series (Santer et al., 2000). This approach slightly differs from previous studies, which assume a constant value of $t_\omega = 2$ (Tiao et al., 1990; Weatherhead et al., 1998; van der A et al., 2006). Finally, σ_ω is approximated according to Weatherhead et al. (1998) as

$$\sigma_\omega = \left[\frac{\sigma_r}{n^{3/2}}\sqrt{\frac{1+\phi}{1-\phi}}\right] \qquad (4)$$

where σ_r is the standard deviation of the de-trended residuals, n is the number of years with available data, and ϕ is the first-order autocorrelation. In order to eliminate spurious significant trends for time series with extremely low long-term averaged NO_2 column values C that are obviously below the uncertainty threshold of the satellite data (primarily over the oceans), the uncertainty for such time series was computed differently. If σ_ω as computed in Equation 4 was found to be less than a minimum uncertainty value of $\sigma_{min} = 0.65 + 0.3 \cdot C$ (Boersma et al., 2009) with C given in $\times 10^{15}$ molecules cm^{-2}, σ_ω was set equal to σ_{min}. In addition, trends were only computed for grid cells that exhibited a 9-year average of at least 1×10^{15} molecules cm^{-2}.

3 Results and Discussion

In the following section we present some of the general validation results obtained from comparing both OMI and GOME-2A observations against MAX-DOAS data at several ground-based sites and subsequently some initial attempts at using the multi-year time series of MAX-DOAS observations to perform a direct validation of trends derived from satellite data.

3.1.1 General validation

A general validation of the OMI and GOME-2A NO_2 products was undertaken in order to achieve a general idea about the quality level of the satellite data but also in order to investigate to what extent the different sampling methodologies for the ground-based and satellite datasets have an impact on the results. The validation was carried out at the monthly average level. Several time series and scatterplots were created and summary statistics were calculated.

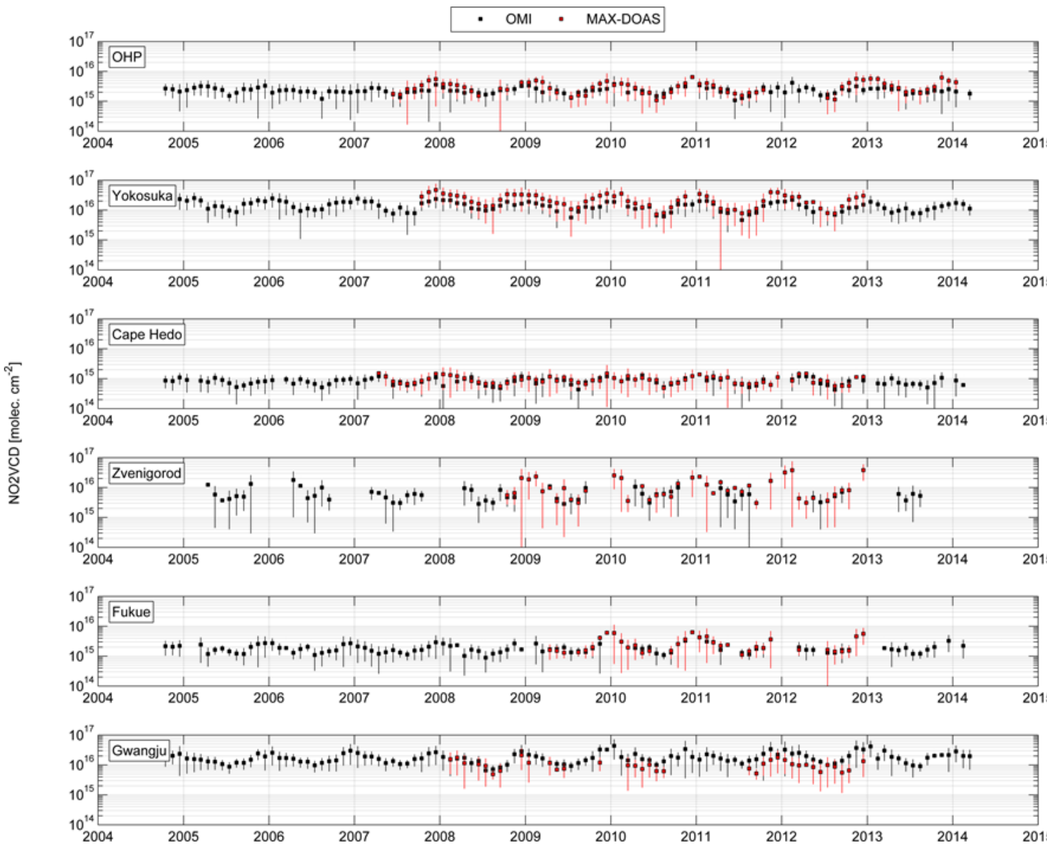

Figure 7: Time series of monthly averages of the full available datasets from MAX-DOAS and OMI at 6 MAX-DOAS sites.

Figure 7 shows the monthly time series of OMI tropospheric NO_2 and monthly averaage MAX-DOAS observations at the 6 sites with available data. At the OHP site, the time series of the two datasets agree reasonably well although a tendency towards underestimation by OMI is visible. At the Yokosuka site in urban Japan, the MAX-DOAS observations clearly exhibit significantly higher value of tropospheric NO_2 than OMI. While there is a systematic bias, most likely caused by the strong spatial gradients in urban areas which the satellite is not able to pick up, the random error is actually not very high and the two time series agree quite well in terms of temporal patterns such as the seasonal cycle. At the rural site Cape Hedo the two data source agree remarkably well. At the Zvenigorod site in Russia the comparison of the two datasets shows much more dissimilarity. In particular, large data gaps are visible in both time series, predominantly during the winter months. The Fukue station in rural Japan, on the other hand, once again shows a very good agreement between the two time series, although the MAX-DOAS time series is unfortunately relatively short at this station. Finally, the Gwangju station in South Korea shows an interesting behavior in the sense that it mostly exhibits a systematic bias between satellite retrieval of tropospheric NO_2 and MAX-DOAS observation, however this systematic difference increases over time, with smaller discrepancies in the years 2008 and 2009 and the bias increasing significantly over later years.

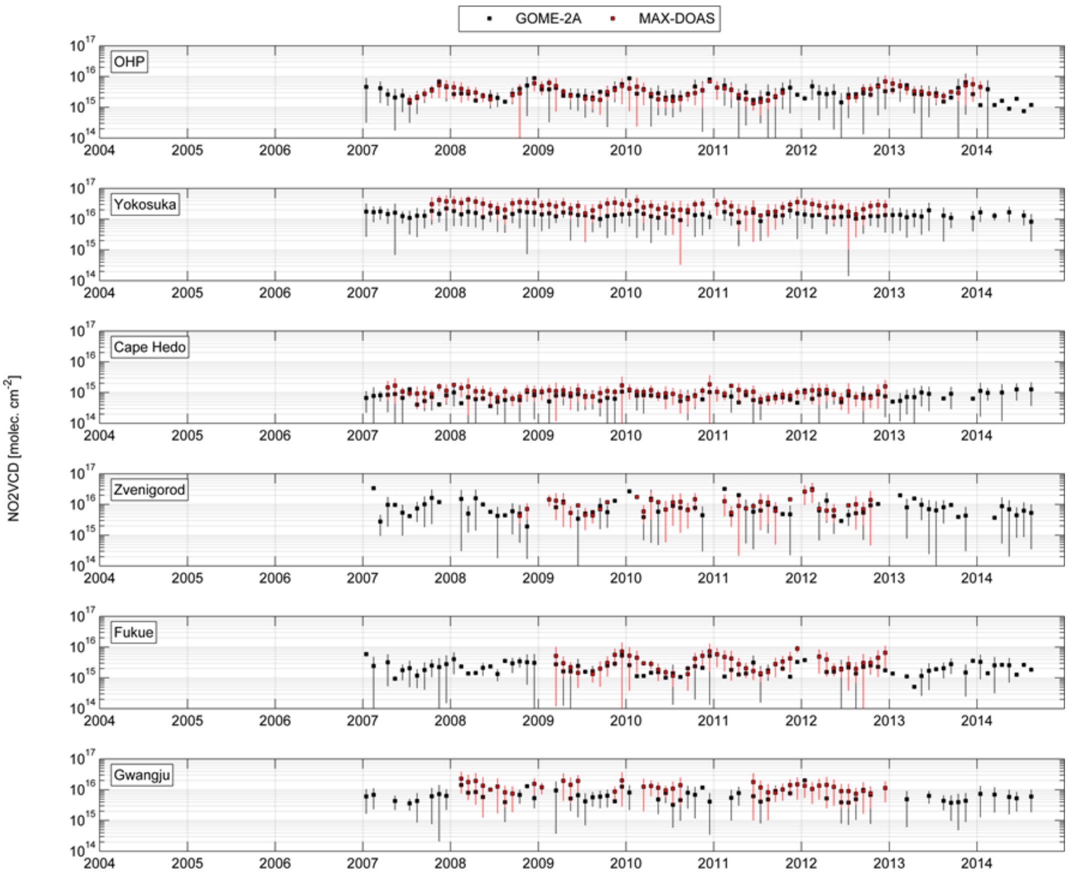

Figure 8: Time series of monthly averages of the full available datasets from MAX-DOAS and GOME-2A at 6 MAX-DOAS sites.

Figure 8 shows the corresponding results for the comparison between the GOME-2A tropospheric NO_2 product and the MAX-DOAS observations. In general the results tend to be similar as for OMI but there is slightly more random error. One significant difference is that for the Gwangju station the satellite retrievals are lower than the MAX-DOAS observations, whereas the opposite was true for the comparison with OMI.

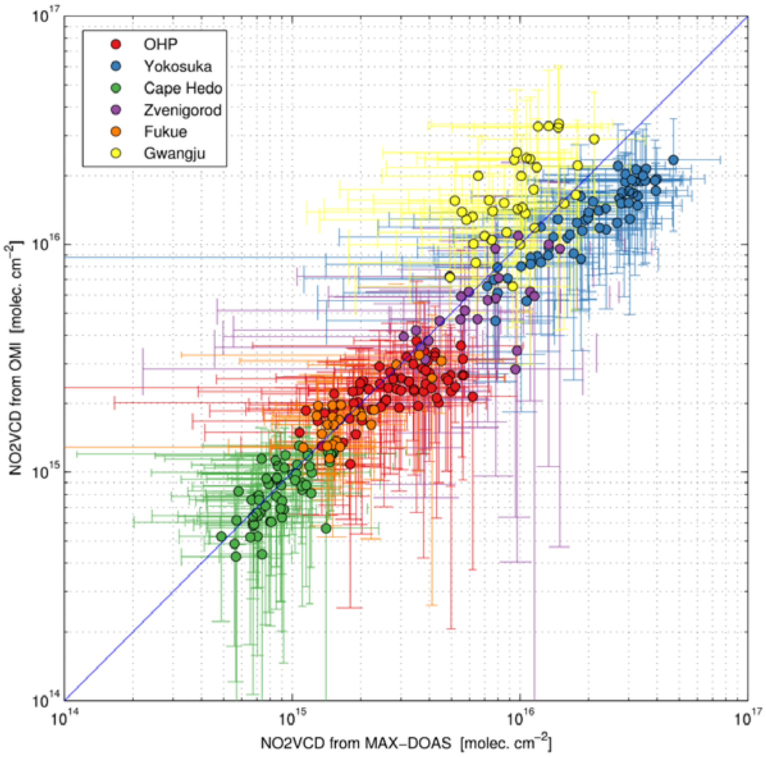

Figure 9: Scatter plots of monthly averages of the full available datasets from MAX-DOAS and OMI at 6 MAX-DOAS sites. Note that both axes use a logarithmic scale.

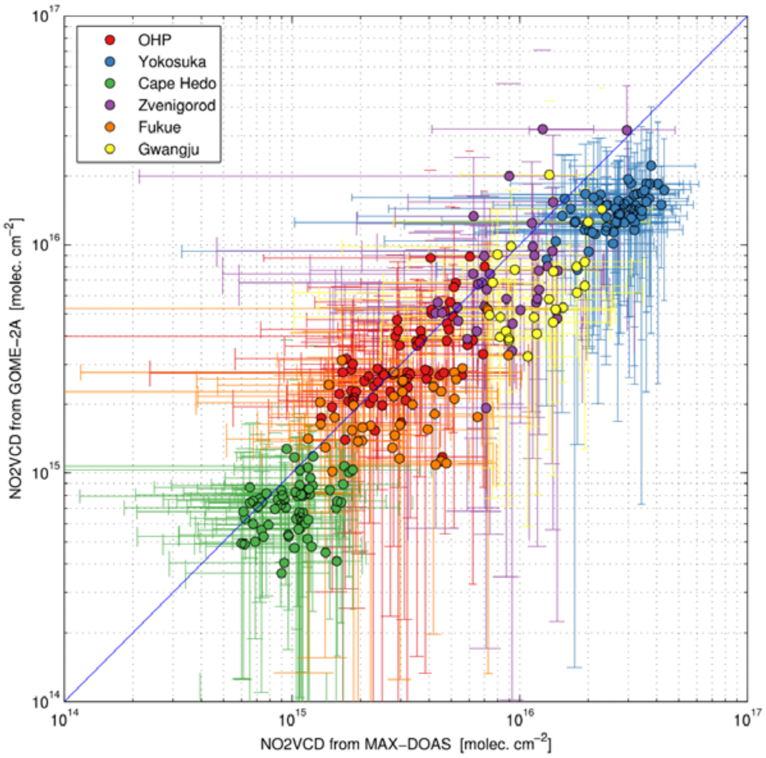

Figure 10:Scatter plots of monthly averages of the full available datasets from MAX-DOAS and GOME-2A at 6 MAX-DOAS sites. Note that both axes use a logarithmic scale.

The results were further visualized in form of scatterplots directly comparing the monthly mean NO_2 observations calculated from the MAX-DOAS instruments with the monthly averages computed from the daily tropospheric NO_2 columns from the satellite datasets. Figure 9 and Figure 10 show the behaviour mentioned earlier more efficiently in the form of scatterplots which clearly indicate low and high biases between the two datasets. Once again it is obvious for both OMI and GOME-2A that the random errors is lowest for the least polluted sites in rural regions, such as Cape Hedo and Fukue and, to some extent, OHP. The two highly polluted sites (Yokosuka and Gwangju) show quite large biases. Interestingly the bias for Gwangju is positive for OMI and negative for GOME-2A. The reason for this behaviour is not entirely clear but it could be related to the different overpass times of the satellite platforms.

In general, the OMI product shows slightly less random error, in particular for the less polluted sites. To some extent this can be attributed to the higher spatial resolution at which OMI provides tropospheric NO_2 columns.

Table 7: Summary statistics for the general validation of monthly average OMI tropospheric NO_2 data against MAX-DOAS station data observed at the same time of the satellite overpass. All units except for N, slope, and R^2 are in × 10^{15} molecules cm^{-2}.

Station	N	Bias	Std Dev	RMSE	Offset	Slope	R^2
OHP	66	0.75	1.07	1.30	1.53	0.26	0.36
Yokosuka	58	8.98	6.09	10.82	3.94	0.43	0.81
Cape Hedo	57	0.07	0.20	0.22	0.29	0.60	0.46
Zvenigorod	23	1.45	2.66	2.98	2.77	0.42	0.36
Fukue	24	0.13	0.52	0.52	0.78	0.54	0.72
Gwangju	35	-7.51	6.23	9.70	5.66	1.18	0.36
Average	**44**	**0.64**	**2.80**	**4.26**	**2.50**	**0.57**	**0.51**

Table 8: Summary statistics for the general validation of monthly average GOME2-A tropospheric NO_2 data against MAX-DOAS station data observed at the same time of the satellite overpass. All units except for N, slope, and R^2 are in × 10^{15} molecules cm^{-2}.

Station	N	Bias	Std Dev	RMSE	Offset	Slope	R2
OHP	64	3.34	1.41	1.30	0.92	0.88	0.52
Yokosuka	61	27.28	7.18	14.28	7.90	0.23	0.37
Cape Hedo	68	1.05	0.30	0.38	0.69	0.14	0.05
Zvenigorod	35	9.69	4.71	5.38	0.30	0.89	0.38
Fukue	37	3.43	1.94	1.57	1.38	0.37	0.54
Gwangju	25	12.81	4.30	7.13	2.67	0.35	0.16
Average	**48**	**9.60**	**3.31**	**5.01**	**2.31**	**0.48**	**0.34**

Table 7 shows the quantitative results of the general validation of tropospheric NO_2 from OMI against MAX-DOAS. The results confirm the previous findings that the polluted sites at Yokosuka and Gwangju have large biases whereas the biases at most other stations are quite small (less than 2 × 10^{15} molecules cm^{-2}). For the OHP site it was found that the OMI NO_2 product to some extent underestimates the observations acquired by MAX-DOAS. The authors found out recently that a bias of approximately 30% was found for the *in situ* data at OHP which was caused by a slightly wrong view angle (G. Pinary, personal communication). This could to some extent explain the discrepancy found for the OHP station.

The largest standard deviation and RMSE was found for Yokosuka, whereas the lowest was found for Cape Hedo, the least polluted site. The overall bias for all sites is a quite low 0.64 × 10^{15} molecules cm^{-2}. Interestingly, however, the overall agreement between the two datasets as expressed through the coefficient of determination (R2) was found for Yokosuka as well with a value of 0.81, whereas the sites with much lower overall errors only showed R2 values of less than 0.5.

Table 8 shows the same summary statistics for the comparison between the GOME-2A tropospheric NO_2 product and the MAX-DOAS observations at the same time as the satellite overpass. The results indicate a worse accuracy than was found for the OMI comparison, with biases up to 27.3 × 10^{15} molecules cm^{-2} and an average bias of 9.6 × 10^{15} molecules cm^{-2}. However, with exception of the

Yokosuka station, the standard deviations and RMSE results are approximately similar to what was found for OMI. The R2 values are, however, significantly lower than for OMI. To a large extent this discrepancy can be explained by the much larger spatial footprint of the GOME-2A instrument which cannot capture the relatively fine spatial detail that OMI is able to detect.

The main conclusion for the general validation is that the ground-based MAX-DOAS and satellite datasets agree quite well for sites that are located in very clean regions with station characteristics are mostly indicative of the overall background concentrations. At very polluted sites strong systematic biases between the two datasets can be detected (e.g. Yokosuka) and in some case larger random errors (e.g. Zvenigorod) and even shifting systematic biases (e.g. Gwangju). This is at first glance somewhat counter-intuitive as it would be expected that the satellite products are most accurate for very polluted sites and are limited by the instruments' detection limits in very clean regions.

However, at very polluted sites, which tend to be located in urban areas the small-scale spatial gradients in NO_2 concentrations can be quite significant. As such, since the MAX-DOAS instruments are sensitive to very local emission sources and are representative for at most a distance of about 10 km, they can display strong systematic differences as compared to the satellite data which have a spatial footprint that is significantly larger (13 km × 24 km at nadir for OMI and even 80 km × 40 km at nadir for GOME-2A).

3.1.2 Direct validation of trends

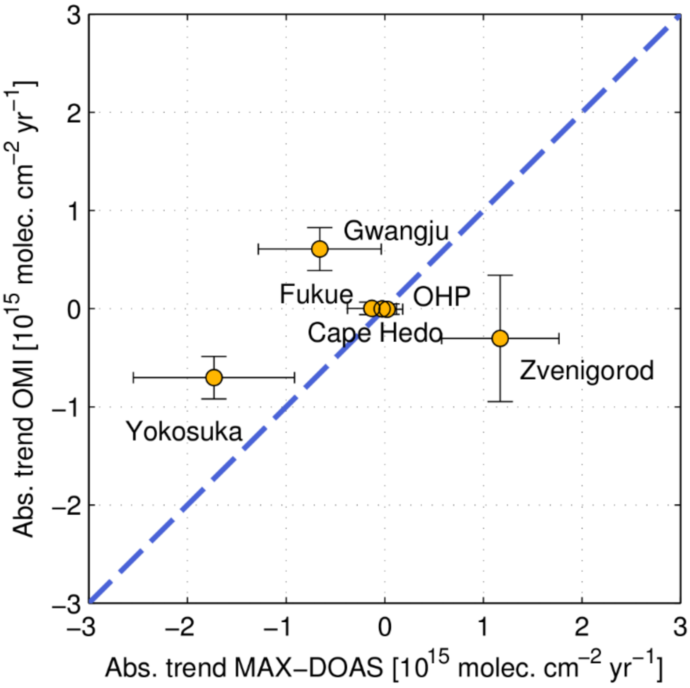

Figure 11:Scatterplot showing the direct comparison of absolute trends in tropospheric NO_2 derived from MAX-DOAS data versus the corresponding trends derived from OMI data.

In addition to the general validation reported on in the previous section, a direct validation of the trends in tropospheric NO_2 was attempted. This has not been done so far as most of the MAX-DOAS have relatively short time series length. However some sites now have reached time series lengths of 5 years or more and together with the in some cases quite rapidly changing tropospheric NO_2 levels (Schneider and van der A, 2012; Schneider et al., 2014) have the potential to be used as a direct reference against which the linear trends derived from satellite datasets can be compared against.

For this purpose, a statistical trend model including a seasonal cycle and a linear trend (Schneider and van der A, 2012; Schneider et al., 2014) was fitted to both the MAX-DOAS and satellite time series at each site. The resulting values of absolute and relative trends obtained for both datasets were subsequently compared.

Figure 11 shows the direct comparison of absolute trends derived using the statistical trends model for the MAX-DOAS time series against those derived for the OMI satellite data. It is obvious that there is a substantial amount of scatter. Three stations in clean background conditions, namely Fukue, Cape Hedo, and OHP fall close to the 1:1 line, however they are also stations where the absolute trend is very close to zero for both datasets, i.e. no strong change in NO_2 levels was observed at these sites.

The situation is different for the more polluted sites. Both Yokosuka and Gwangju follow the 1:1 line with a positive systematic offset indicating that OMI overestimates the absolute trends as compared to the trends derived from the ground-based MAX-DOAS sites. Yokosuka exhibits a negative trend, i.e. decreasing NO_2 levels, as measured by both MAX-DOAS as well as OMI, although the negative trend obtained from MAX-DOAS is significantly more rapid by a factor of two. The Gwangju station shows a slightly negative trend for the ground-based data but a slightly positive trend for OMI data, so the two datasets disagree here. Finally, the Zvenigorod station shows a positive trend of over 1×10^{15} molecules cm^{-2} for the ground-based MAX-DOAS data but a trend near zero was found for the OMI data.

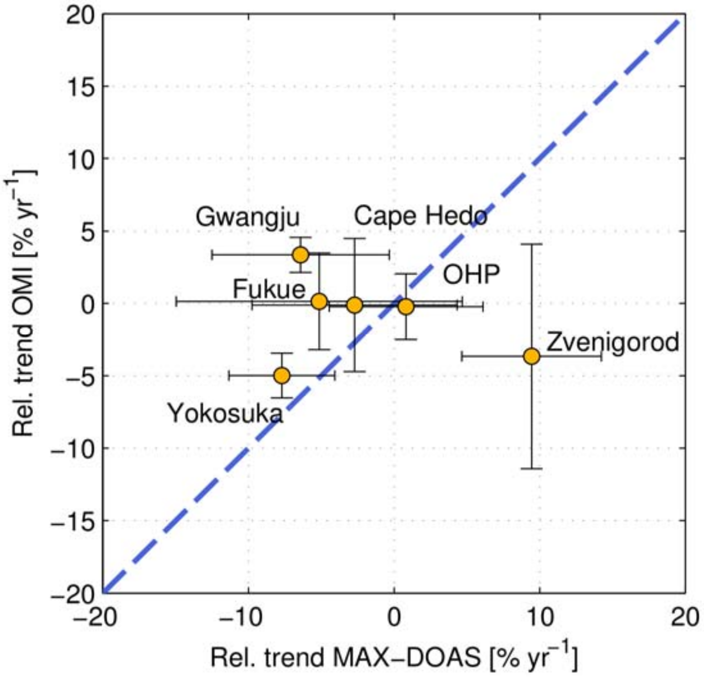

Figure 12:Scatterplot showing the direct comparison of relative trends in tropospheric NO_2 derived from MAX-DOAS data versus the corresponding trends derived from OMI data.

Figure 12 shows the corresponding relative trends in tropospheric NO_2 derived from the two data sources. The relative trends were computed not in relation to a specific reference year but with reference to the long-term average NO_2 concentration at each site in order to be less susceptible to outliers at the pixel level in the reference year.

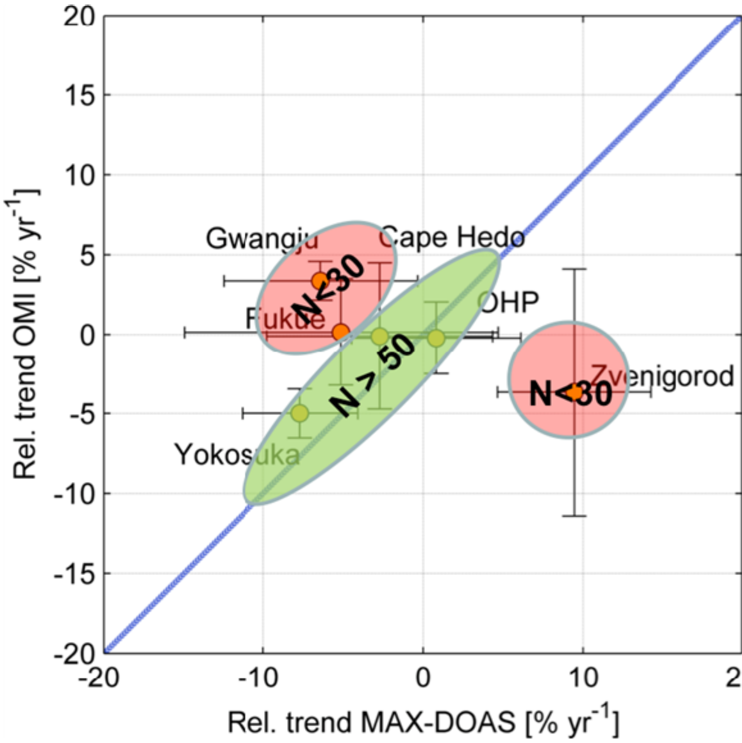

Figure 13:Same as Figure 12 but with annotations indicating the number of months for which valid data was available at the various stations. Note that the sites with more than 50 months of valid data fall close to the 1:1 line whereas sites with much fewer valid data points tend to exhibit large discrepancies between the trends computed from both datasets.

Figure 13 shows the same Figure but with annotations indicating the number of months for which the time series at the individual sites had valid data from both the ground-based MAX-DOAS instruments as well the satellite datasets. It is quite obvious that the sites with more than 50 months of valid observations from both instruments have trend that agree quite well with each other and therefore fall reasonably close to the 1:1 line. On the other hand, sites that were found to be quite far away from the 1:1 line and which thus show quite different trends values from the two datasets all have a significantly lower number of months with valid data (less than 30 months).

These results of course clearly indicate that overall the length of the time series that are currently available from the MAX-DOAS sites that were investigated here (and which to our knowledge are the sites worldwide with the longest record), are not sufficiently long yet to be used for direct validation of the trends obtained from satellite-based instruments. The uncertainty of the trends obtained from the MAX-DOAS sites is at this point still considerably too high to draw any meaningful conclusions regarding the quality of the satellite-based trends. Furthermore, while MAX-DOAS sites at this point provide measurements that are currently considered to be the closest possible to what a satellite-derived tropospheric column provides, large difference in sampling methodology remain and further complicate such comparisons.

3.2 Conclusions and recommendations

In this study we investigated the usability of the current set of long-term operational ground-based MAX-DOAS stations worldwide for inter-comparing their observations to those of satellite-based instruments, in particular OMI and GOME-2A. The full time series of MAX-DOAS data were acquired for a total of seven stations worldwide, which all have been operating on the order of approximately 5 years or more, although significant data gaps do occur for several of the sites. We first performed a general validation of the satellite datasets. The results indicate that the two data sources agree very well for sites located in rural, non-polluted regions. For sites located in polluted areas we found strong systematic biases, large random errors, or slightly shifting systematic biases. The systematic biases can be explained primarily by the strong spatial gradients in NO_2 levels in urban areas in conjunction with the large differences in the spatial representativity of the measurements. While the MAX-DOAS data are representative for distances on the order of several kilometers (and up to distances of approximately 10 km (M. van Roozendael, personal communication)), the satellite observations are averaged over significantly larger areas and therefore are not able to measure the fine spatial detail from local emissions sources that the MAX-DOAS sites are able to capture. Overall GOME-2A data showed mostly similar patterns as OMI in terms of biases however the random error was higher for GOME-2A, which can to some extent be attributed to the significantly coarser spatial footpring of the instrument.

After performing a general validation, we evaluated the possibility to use the now relatively long time series of MAX-DOAS observations to fit a statistical trend model and to directly compare the resulting trends to those obtained for the satellite-based time series for the same area and time period. The results indicate that, while the time series are long enough to fit a trend model and obtain some rough correspondence in trends, the uncertainties associated with the resulting trends are too high to draw any firm conclusions and certainly to use them for direct validation of satellite-based trends. The main issue that was found was that while the MAX-DOAS time series now reach lengths of 5 years and more, a significant amount of data gaps exist in the time series which increase the uncertainty in the obtained trends. It was found that the sites with approximately 50 months of valid data for both data sources showed quite similar long-term trends and that sites with fewer than 30 months of valid data exhibited significant discrepancies in the resulting trends.

It is recommended to revisit this topic in approximately 2 years since at this point the sites which in this study did not have enough months with valid observations will most likely exceed the threshold of approximately 50 months of valid data at which the derived trends become more comparable with satellite trends. At that point there are also likely to be more stations worldwide which provide operational long-term MAX-DOAS datasets. Once the MAX-DOAS time series are long enough for a significant number of stations worldwide it is anticipated that they will provide the first method for directly validating the tropospheric NO_2 trends obtained from satellite-based platforms without the need for indirect validation using models or similar techniques which introduce significant amounts of additional uncertainty.

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ABSTRACT NILU has a mandate to monitor air quality and particularly its changes over time, both nationally through Miljødirektoratet (MD) and internationally through the European Monitoring and Evaluation Programme (EMEP). Satellite data related to atmospheric composition are increasingly used for monitoring as they provide long time series of spatially continuous observations. It is therefore essential for NILU to begin preparing for the upcoming Copernicus missions. Here, we evaluate methane products from AIRS, TES, TANSO-FTS and SCIAMACHY as added value for GHG monitoring in Norway and Svalbard. As expected, due to the low sensitivity of the sensors to ground-level Artic large deviations are seen when comparing to *in situ* data from Birkenes and Ny-Ålesund. Higher level products (L4), combining satellite and ground-based information, seem more appropriate for future reporting purposes. Further, we investigated the usability of the current set of long-term operational ground-based MAX-DOAS stations worldwide for inter-comparing their NO_2 observations to those of satellite-based instruments, in particular OMI and GOME-2A. The two data sources agree very well for sites located in rural, non-polluted regions. For sites located in polluted areas we found strong systematic biases, large random errors, or slightly shifting systematic biases. The systematic biases can be explained primarily by the strong spatial gradients in NO_2 levels in urban areas in conjunction with the large differences in the spatial representativity of the measurements. We evaluated the possibility to use the now relatively long time series of MAX-DOAS observations to fit a statistical trend model and to directly compare the resulting trends to those obtained for the satellite-based time series for the same area and time period. It was found that the sites with approximately 50 months of valid data for both data sources showed quite similar long-term trends and that sites with fewer than 30 months of valid data exhibited significant discrepancies in the resulting trends.

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ABSTRACT NILU har flere prosjekter for overvåkning og evaluering av luftkvalitet, både nasjonalt gjennom Miljødirektoratet og internasjonalt gjennom «European Monitoring and Evaluation Programme» (EMEP). Satellitter som måler atmosfærens sammensetning, blir stadig oftere brukt i overvåkingen, og de kan gi lange tidsserier med kontinuerlige observasjoner over et stort område. Det er derfor viktig for NILU å være forberedt på kommende oppdrag fra Copernicus-programmet. Vi har studert metanprodukter fra AIRS, TES, TANSO-FTS og SCIAMACHY, som et supplement til overvåkningen av drivhusgasser i Norge og på Svalbard. Som forventet, på grunn av lav satellitt sensorfølsomhet ved bakkenivå, blir det relativt store avvik mellom satellittdata og *in situ* data fra Birkenes og Ny-Ålesund. Nye produkter, som kombinerer satellitt- og bakkebasert informasjon, synes mer hensiktsmessig for fremtidige rapporteringsformål. Videre har vi studert bakkebaserte MAX-DOAS stasjoner over hele verden og sammenlignet med NO2 observasjoner fra satellitt, spesielt OMI og GOME-2A. Satellitt- og bakkebaserte målinger samsvarer bra i rurale områder med lite forurensning. For områder med mer forurensning fant vi imidlertid systematiske avvik. Dette er primært knyttet til romlig representativitet og store NO2 gradienter i urbane områder. MAX-DOAS observasjonene har blitt tilpasset en statistisk trendmodell og sammenlignet med satellittbaserte trender for samme område og tidsperiode. Det ble funnet at stasjoner med mer enn 50 måneder med data ga nokså samsvarende trender for MAX-DOAS og satellitt. Stasjoner med mindre enn 30 måneder med data ga derimot sprikende trendresultater.

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