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The sensitivity of Western European NO_2 columns to interannual variability of meteorology and emissions: a model – GOME study

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Abstract

Interannual variability (IAV) in tropospheric species concentrations can be driven by variability in emissions, chemistry, transport and UV radiation. In a 3D CTM study we have found good agreement between the IAV of NO_2 columns observed by the GOME satellite instrument and model simulations over Western Europe from 1996 to 2000. We find that meteorological variability is an important factor during this period. Averaged 10 m wind speeds from the European Centre for Medium-Range Weather Forecasts (ECMWF) operational analysis are a good proxy for the overall meteorology driving the IAV during the studied period of 1996–2000. Copyright © 2008 Royal Meteorological Society

Keywords: interannual variability; GOME; model; NO_2

1. Introduction

Interannual variability (IAV) of tropospheric composition has been the subject of many recent studies using both observations and modeling. Creilson *et al*. (2003) showed a relationship between transport of ozone across the North Atlantic into Europe and the positive phase of the North Atlantic Oscillation (NAO). Duncan and Bey (2005) studied IAV of export from Europe and found that export of pollution was more frequent for the positive phase of the NAO.

Tropospheric ozone production in many areas is NO_x limited (Sillman, 1999 and references therein), so a key component of the IAV of ozone is the interannual variation in NO_2. Richter *et al*. (2005) recently showed a large positive trend in NO_2 over China and a smaller downward trend over Europe. Uno *et al*. (2007) studied the sensitivity of NO_2 columns in eastern Asia to meteorology and emissions and found that errors in estimating emission trends were larger for autumn and winter because of the variability of meteorology. Our objective in this study is to understand the sensitivity of tropospheric NO_2 columns to variability in both emissions and meteorology over Western Europe. Using GOME data we validate the modeled IAV of tropospheric NO_2 and, by performing a sensitivity study, we investigate if emissions or dynamical processes are dominant drivers of the IAV of NO_2 in Western Europe.

2. Model details

This study uses the global offline chemistry transport model p-TOMCAT which is an updated version (O'Connor *et al*., 2004) of the TOMCAT model (Law *et al*., 2000; Savage *et al*., 2004). In this study p-TOMCAT was run with a $2.8° \times 2.8°$ horizontal resolution and 31 vertical levels from the surface to 10 hPa with offline meteorology from the operational analyses of the European Centre for Medium-Range Weather Forecasts (ECMWF) model for 1996–2000.

The model used annually and monthly varying emissions for industry, transport, shipping and biomass burning from the RETRO emissions database (Schultz *et al*., 2007a). Further details of the RETRO biomass burning emissions can be had from Schultz *et al*. (2007b). The terrestrial anthropogenic emissions in the RETRO database were produced using the TNO Emissions Assessment Model (Pulles and van het

Bolscher, 2007). There was a minor error in the seasonality of the anthropogenic emissions but this had a negligible effect on this study. Lightning emissions of NO_x are based on the parameterization of Price and Rind (1992) as implemented by Stockwell *et al.* (1999) with an average emission for 1996–2000 of 3.9 Tg (N) per year. The modeled heterogeneous removal of N_2O_5 on sulfate aerosol is based on that in the MOZART model as described in Tie *et al.* (2003). Offline sulfate aerosol data were taken from the GOCART model (Chin *et al.*, 2002) and an uptake coefficient of 0.04 was assumed. The cloud fields used to calculate the offline photolysis rates are based on climatology and so do not contain IAV. 3D fields of NO_2 were output from the model at 10 : 30 local time. More details of model data processing can be obtained from Savage *et al.* (2004).

3. Scope of the study

The model setup we are using is suitable to describe the contributions to IAV of NO_2 due to variations in emissions and large scale dynamic meteorology. The use of precalculated photolysis rates based on monthly mean cloudiness will naturally suppress any variability due to variations in cloudiness. This model limitation clearly prevents us from answering the question of how variability in cloudiness contributes to NO_2 differences, and is an important simplification of the system. Nevertheless, it also reduces the complexity of the analysis by relating modeled NO_2 differences simply to either large scale differences in the flow field or to emissions. Our intention is to isolate the impact on NO_2 variability of these two factors; we accept that other factors (e.g. cloud-induced variations in photolysis) will also play a role. Nevertheless, any good agreement between observations and our model results here will suggest that an important component of observed NO_2 variability must be related to changes in emissions and/or large scale meteorology.

4. GOME data

The GOME instrument as described by Burrows *et al.* (1999) measures each location at the same local time, e.g. the northern midlatitudes at around 10 : 45. Monthly mean GOME data for the period 1996–2000 were used for this study. Daily data from the stratospheric Chemistry Transport Model (CTM) SLIMCAT (Chipperfield, 1999) were used to account for the variability in stratospheric NO_2. Other data used in the retrieval were the surface albedo climatology of Koelemeijer *et al.* (2003) and monthly air mass factors based on tropospheric aerosol classified according to maritime, rural and urban locations, and vertical NO_2 profiles for 1997 from the MOZART model (Horowitz *et al.*, 2003). A correction for topography effects was made and a 0.2 cloud cover threshold based on the FRESCO algorithm (Koelemeijer *et al.*, 2001) used to select cloud free pixels. More details can be found in Richter *et al.* (2005).

5. Experimental set up

To evaluate the reasons for IAV of ozone and its precursors, two particularly important contributions have been investigated – meteorology (except cloudiness) and emissions. A base run has been performed using the monthly and annually varying RETRO emissions and ECMWF operational analyses for the 1996–2000 period. Two further runs were then completed: ‘Met. Fix.’ where the emissions varied according to the RETRO database for 1997–2000, but the meteorology for 1996 was repeated annually and ‘Emi. Fix.’ where the emissions for 1996 were repeated for each model year, but the meteorology varied for 1997–2000 as in the base run.

For all years, 3-month averages (JFM, AMJ, JAS, OND) from both GOME (regridded to T42 resolution) and the model runs have been calculated. This averaging helps reduce the effects of random error in GOME retrievals. In addition, for differences from the 5-year mean, any consistent systematic errors will cancel. Because of instrument problems there was no GOME data available for January 1998. In the comparisons shown below when calculating the seasonal average for JFM 1998 January’s data was replaced by the average of January 1996, 1997, 1999 and 2000. The same procedure was applied to the model data to ensure consistency.

6. Results

Figure 1 shows the IAV in NO_2 columns over Western Europe in JFM from GOME, the model base run and the run with fixed emissions for the 1996–2000 period. These anomalies should be compared to average GOME NO_2 columns in this region at this time of year of 6–8 × 10^{15} molecules cm^{-2}. The model captures well the main features of the IAV. For 1996 and 1997 a positive anomaly can be seen in northwestern Europe. The size and position of the deviation from the mean is similar in both model and GOME for both years and in both datasets the anomaly is smaller in 1997 than in 1996. In 1998 GOME has a large negative anomaly and although the differences in p-TOMCAT are smaller than those in GOME there is a negative anomaly in the same location as the largest negative deviations in GOME. Both 1999 and 2000 also show generally negative anomalies in NO_2 although in 2000 there is a positive anomaly over Southern England not seen in the model. The results from the run with fixed emissions give a very similar pattern of anomalies to the base run indicating that much of the variability in this region come from IAV of meteorology.

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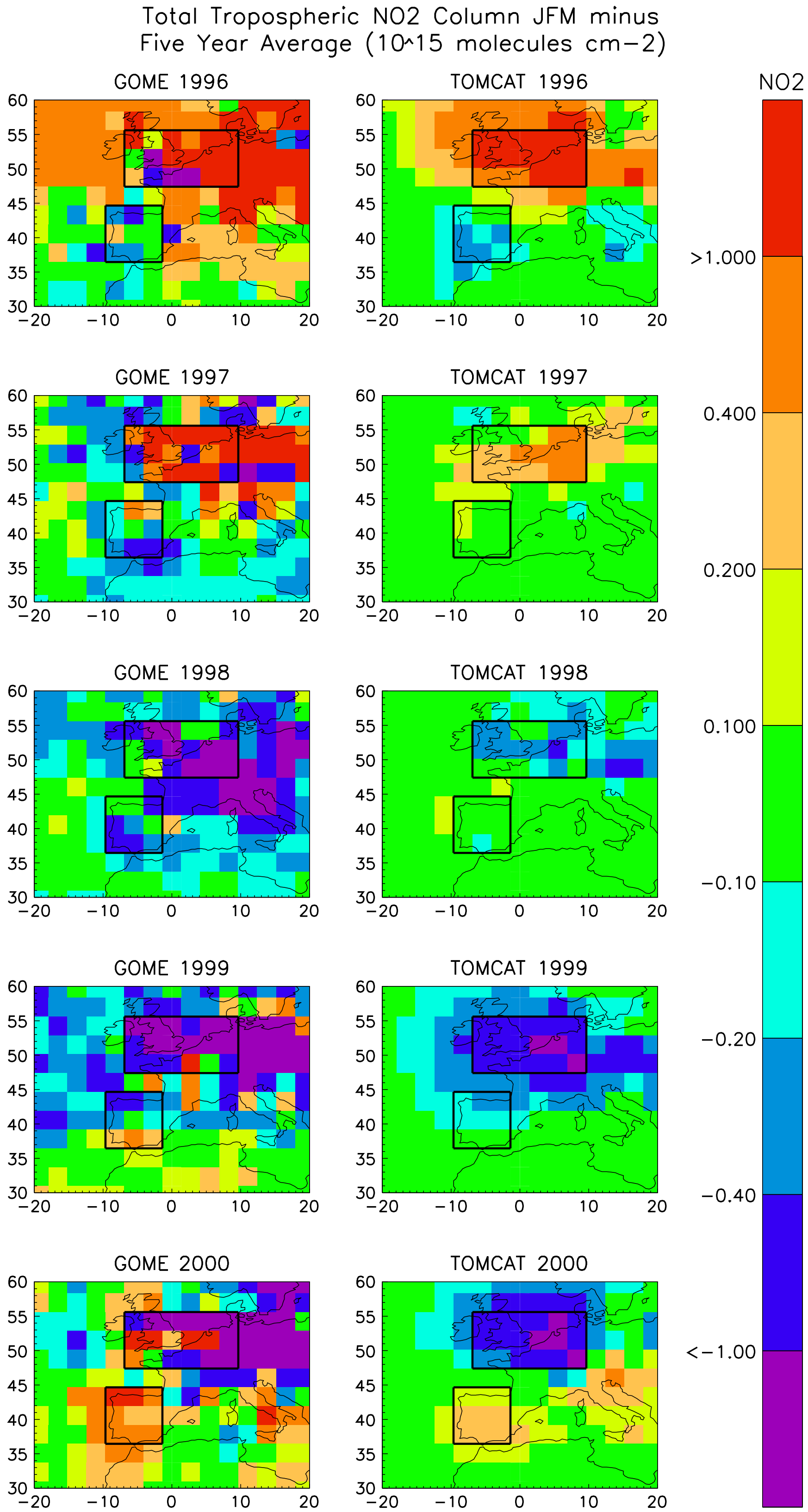

Figure 1. Tropospheric NO_2 column anomalies over Western Europe for JFM 1996–2000 from GOME observations (left) and p-TOMCAT data (middle: base case; right: 'Emi. Fix.' run). Rectangles indicate the areas for which area averaged temporal correlations were calculated.

Area averages of the anomalies have been calculated for two regions: northwestern Europe (18 grid model boxes) and Iberia (9 grid boxes), as indicated in Figure 1. The average NO_x emissions were 1.5 Tg(N) per year for northwestern Europe and 0.34 Tg(N) per year for Iberia. Emission trends in the two regions are quite different – weakly negative for northwestern Europe (−1.2% per year) and positive for Iberia

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(+4.7% per year). In Iberia, there is an increase in emissions every year. In northwestern Europe, the emissions decrease every year except in 2000 when there is a small increase to a value between those of 1998 and 1997. The total year to year variability in emissions is therefore small.

Temporal correlations between GOME and p-TOMCAT were then determined for these regions (Table I). In addition, the ordinary least squares bisector best-fits between model values and GOME have been calculated. As well as correlations the table shows the intercept (c) and gradient (m) of this fit. As this is a relatively short period the correlations are not expected to be statistically significant but still illustrate the good overall agreement between GOME and the base run of p-TOMCAT for certain seasons. For JFM the correlation between the NO_2 column anomalies for the base run are good for both northwestern Europe and Iberia (0.84 and 0.79 respectively) while it is actually even better (0.90) in northwestern Europe for the AMJ season. For other seasons the correlations are lower, falling to only 0.18 for Iberia in JAS. For the base run the gradient of the best-fit line is between 0.43 and 0.94 in northwestern Europe while that in Iberia lies between 0.36 and 1.0. This indicates that the model does not capture all of the IAV but in general has the right signal in these regions. The best correlations are also seen for the periods and regions where there is the greatest IAV. In northwestern Europe the worst correlations are in JAS, which is also the season with the smallest IAV.

Although for JFM the correlations for northwestern Europe are similar for all runs (0.84 for the base run, 0.86 for the 'Met. Fix.' and 0.84 for the 'Emi. Fix.' runs) the amplitude of the variation is much smaller in the 'Met. Fix.' run. The magnitude of the modeled IAV is indicated by the gradient of the best-fit line. If the model could explain all the observed variation the slope of the best-fit line would be equal to 1 and have a correlation of 1. A model which captures the sign of the IAV but underpredicts its magnitude by a constant factor would have a correlation coefficient of 1 but a gradient less than 1. For the 'Met. Fix.' experiment the gradient is only 0.1 compared to 0.6 for the base run and 0.54 in the run with fixed emissions. These results are illustrated in Figure 2, which shows the seasonal mean anomalies for the three runs plus the best-fit line for all three experiments. The best-fit line for the emissions and the base case are very close to each other as are the anomalies in all years. From these amplitudes, and from the fact that the pattern of anomalies in the 'Emi. Fix.' run closely resembles those in the base run and in GOME, we conclude that in this region the variability in JFM is dominated by meteorological factors. In cases with high correlation coefficients, the result of the base run is, to a good approximation, the sum of the 'Met. Fix.' and 'Emi. Fix.' runs. This linearity supports our interpretation of the importance of meteorology for the IAV of NO_2.

One of the chemical processes driving the NO_2 variability could be the increased formation of peroxyacetylnitrate (PAN) and/or HNO_3. However, the

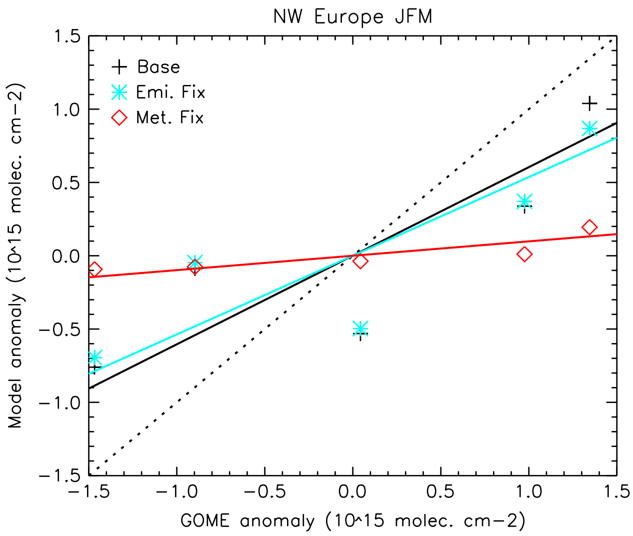

Figure 2. Scatter plot of JFM area averaged anomalies for northwestern Europe GOME observations *versus* p-TOMCAT model results. The light dotted line indicates the 1 : 1 line.

Table I. Intercept (c, 10^{15} molecules cm^{-2}), gradient (m) and temporal correlations of area averaged deviations from the 5-year mean of tropospheric NO_2 columns between GOME data and p-TOMCAT base case. Areas considered are marked in Figure 1

	Base			Emi. Fix.			Met. Fix.		
NW Europe	***c***	***m***	***r***	***c***	***m***	***r***	***c***	***m***	***r***
JFM	1.25E-07	0.60	0.84	2.66E-08	0.54	0.84	1.60E-07	0.10	0.86
AMJ	−1.79E-07	0.87	0.90	−3.00E-08	0.67	0.88	2.90E-09	0.26	0.91
JAS	−9.65E-10	0.94	0.63	−9.45E-08	0.81	0.63	2.15E-08	0.36	0.47
OND	−3.84E-08	0.43	0.51	−7.12E-09	0.37	0.56	5.30E-09	0.21	0.20
Iberia	***c***	***m***	***r***	***c***	***m***	***r***	***c***	***m***	***r***
JFM	−2.72E-08	0.43	0.79	2.62E-09	0.37	0.51	−1.29E-08	0.17	0.77
AMJ	−1.38E-10	0.36	0.36	−7.05E-09	−0.28	−0.47	−2.14E-08	0.25	0.78
JAS	1.89E-08	1.00	0.18	−5.61E-10	−0.88	−0.81	2.69E-08	1.37	0.64
OND	2.08E-08	0.42	0.44	1.08E-08	0.29	0.77	−2.70E-09	−0.36	−0.34

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anomalies in the columns of PAN and HNO_3 are positively correlated with the NO_2 column anomalies so that rapid conversion of NO_2 to PAN or nitric acid cannot explain the periods of lower NO_2.

These model runs included a radon tracer (lifetime 5.5 days, emissions mostly over land) based on the experiment described in Jacob *et al.* (1997). Figure 3 shows the anomalies in the surface concentrations of NO_2 and radon in the base run for JFM. The patterns of the radon and NO_2 anomalies are very similar and the anomalies in surface NO_2 closely resemble the anomalies seen in the total NO_2 column. For northwestern Europe the temporal correlation coefficient in JFM between NO_2 and radon is 0.993 and for Iberia it is 0.722. Figure 3 also shows the anomalies in averaged 10 m wind speed. In northwestern Europe the high NO_2 and radon columns in 1996 and 1997 correspond to low wind speeds in most of this region, while the lower columns in this region seen in the other years also correspond to generally higher than average wind speeds in this region. For northwestern Europe the temporal correlation coefficient in JFM between NO_2 and wind speed is −0.89 and for Iberia it is −0.21. We conclude that the IAV of NO_2 columns

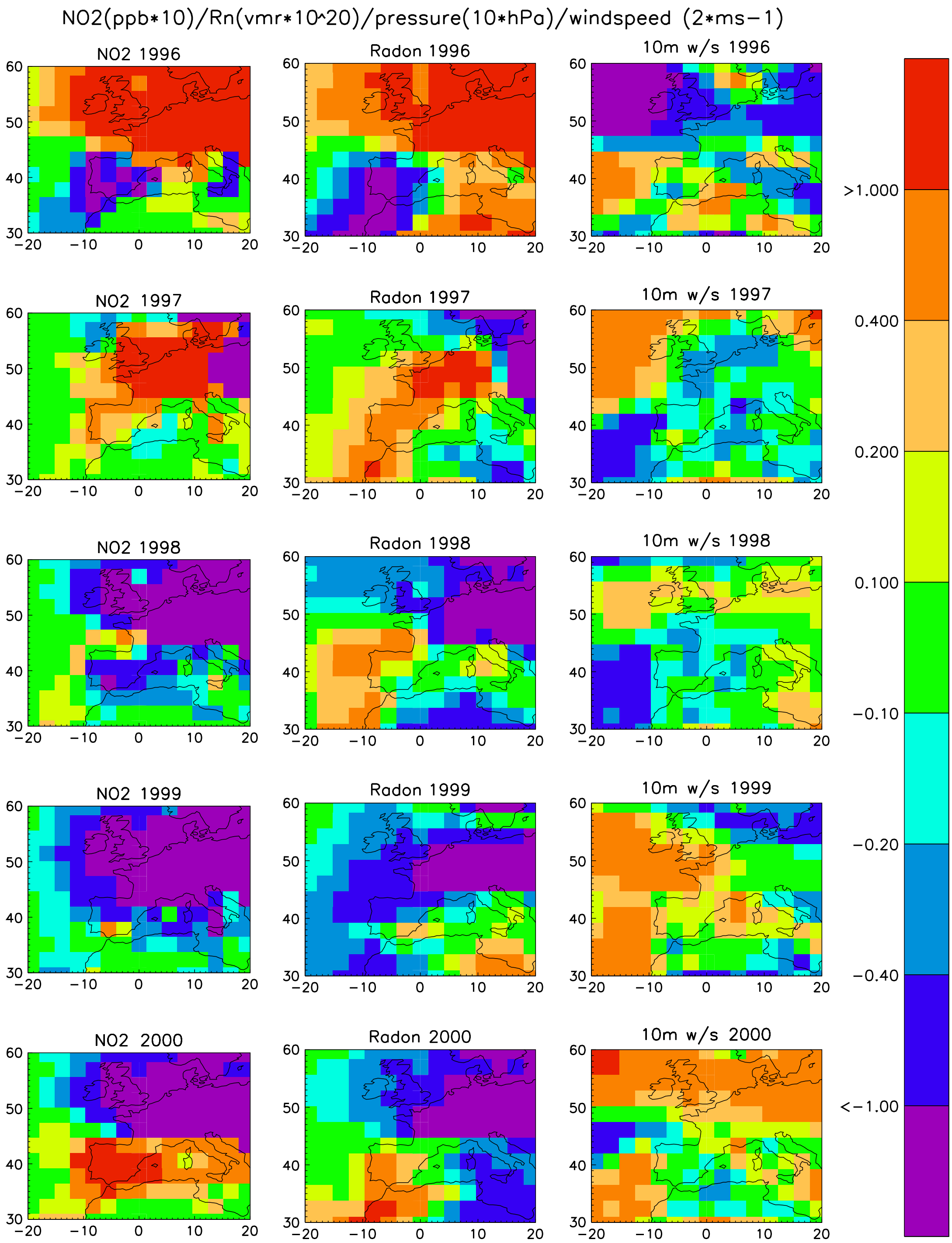

Figure 3. JFM anomalies from 5-year mean for surface NO_2, radon and averaged 10 m wind speed.

in northwestern Europe is mostly driven by the transport of NO_x emissions away from their sources. This may be related directly to the wind speeds themselves, e.g. faster horizontal transport in the lower troposphere being associated with higher wind speeds or in addition, the averaged 10 m wind speed may be a proxy for other aspects of meteorology such as turbulence (as far as the models, ECMWF and p-TOMCAT can represent those processes) with, for example, higher wind speeds being associated with greater vertical mixing and more effective release of tracers into the free troposphere and subsequent (stronger) advection of the emissions from source regions. The source of variability is less clear for Iberia but given the strong correlation with radon transport processes are clearly involved and wind direction may be more critical for this region.

The link between meteorology and IAV seems particularly strong in Western Europe. This may be a combination of the facts that trends in NO_x emissions over this period were small in this region and that midlatitude weather systems have a strong influence on wind direction in northwestern Europe thus potentially having a large influence on air mass origins.

7. Conclusions

The p-TOMCAT model has been used to study IAV of NO_2. Model variability driven by meteorology is a more important factor than emission changes with deviations from the 5-year mean NO_2 column of the order of 1×10^{15} molecules cm^{-2} from meteorology alone. We have not considered variability in cloud, which could be another important driver of interannual variations in composition. Nevertheless, the model reproduces well many aspects of the IAV of NO_2 columns seen in GOME data over Western Europe. The agreement is particularly good in JFM and AMJ. There appears to be a relationship between the averaged 10 m wind speed and NO_2 columns with higher than average wind speeds leading to lower than average NO_2 columns. The steady increase/decrease in GOME NO_2 columns over China/Western Europe of $\sim 5 \times 10^{15}$ molecules cm^{-2} from 1996 to 2002 (Figure 3 of Richter *et al.*, 2005) is significantly larger than the interannual variations due to meteorology alone shown here. However the variations from meteorology alone are large enough that any use of GOME NO_2 columns for trend analysis must consider the potential influence of meteorology or rely on summer measurements where the sensitivity to meteorology is smaller as a result of the shorter NO_2 lifetime.

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