Assessment of NO_2 satellite observations for en-route aircraft emissions detection

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Abstract

In this work the possible use of satellite remote sensors for the detection of the air traffic emissions produced during the en-route segment of flight in the Upper Troposphere/Lower Stratosphere region (8000-12000 m) has been examined. NO_2 has been considered as the tracer of aircraft's plumes with highest possibility of being successfully detected from space. An analysis of the technical potential of the current orbital sensors capable of measuring NO_2 in the proximity of the tropopause has been developed. In order to estimate an upper bound for NO_2 column related to aircraft emissions, the Canary Islands Corridor has been selected to conduct a simple emission calculation exercise based on real air traffic and operational data and assuming an ideal atmospheric scenario. The result obtained in this approximation has been compared with the actual information retrieved from space sensors. An in-depth inspection of the NO_2 column data for two particular areas (Canary Islands Corridor and North Atlantic Flight Corridor) produced in the last years by SCIAMACHY and OMI has also been carried out.

The general conclusions of this viability study are not optimistic. The estimated maximum NO_2 column value attributable to aircraft emissions at cruise altitudes were lower than the detection limits associated to SCIAMACHY and OMI for NO_2 column measurements. As a consequence,

detecting and quantifying the actual NO_2 levels in aircraft corridors by space remote sensing is a very challenging task

1. Introduction

Aviation is a worldwide activity which contribution to the total global anthropogenic CO_2 emissions is about 2% (IPCC, 1999), that is, it has a not a very relevant weight in global terms, but with the specificity that a great fraction of its emissions are injected almost directly into the upper free troposphere and lower stratosphere (UT/LS) (Hoinka et al. 1993). The IPCC, as well as other bodies, has been warning of the environmental effects that air traffic emissions in this region could generate, especially for their influence, in the mid to long-term, on climate change (Brasseur et al., 1998; IPCC, 1999; Schumann, 2002).

The concern is fundamentally based on results obtained through various research projects which started at the beginning of the 90's (Brenninkmeijer, 2006; Brunner et al., 2001; Cammas & Volz-Thomas, 2007; Marenco et al., 1998; Schumann et al., 2000). These scientific researches have widely documented the behaviour of the atmosphere around the tropopause, which corresponds to the cruise segment of flight (8-12 km). As a result of this great research effort, knowledge on the emissions produced by aircraft, together with their influence on the chemical equilibrium of the atmospheric region and their potential implications, in relation to a possible climate change induced by anthropogenic emissions, has greatly improved (Brasseur et al., 1998; IPCC, 2007; Lee et al., 2009; Schumann, 2000). All this knowledge has been achieved through specific research programs, so the documentation of these issues have had no continuity in time.

Important efforts have been dedicated to achieve a theoretical quantification of the fuel burn from commercial air traffic as well as the quantification of atmospheric emissions generated by air traffic (Carlier & Jelinek, 2006; Eyers et al., 2004; Jelinek et al., 2004; Kim et al., 2005).

However, the quality of the results greatly depends on the data supplied by the aircraft manufacturers, with the inherent difficulty in obtaining an authentic guaranteed validation of the emission indices used.

Unlike other productive sectors, in aviation not many means and tools exist to monitor and control real emissions. At present no methodology is available to document in a systematic way the real emissions coming from commercial aviation and the role that space remote sensing could play to this purpose should be evaluated. In this work we present an assessment of the use of satellite remote sensing tools for the detection of NO_2 aircraft emissions produced in the UT/LS region during the en-route segment of flight.

2. Aircraft emissions: background information

In general, the results of the international projects carried out in the last years demonstrate that the measured concentrations of gaseous tracers in the studied air traffic corridors or their immediate surroundings display a strong variability in space and time and, therefore, a great heterogeneity (Brasseur et al., 1998; Brunner et al., 2001; Emmons et al., 2000; Grewe et al., 2001). Among the multiple causes that explain this, the following are emphasized: the variability of the dispersive conditions; the dependency of the height of the tropopause with season and latitude (9-12 km at midlatitudes, and about 17 km in the tropics); chemical regimes and lifetime of gaseous species; and the interference generated by the transport of pollutants from the surface to the higher troposphere.

Up to present, one of the better studied air traffic corridors has been the one crossing the North Atlantic Ocean (North Atlantic Flight Corridor, NAFC). The corresponding results obtained from the in-situ measurements support that aircraft emissions accumulate at corridor altitudes leading to an enhancement of NO_x and particles concentrations in atmospheric regions affected by heavy air traffic. However, the impact of CO_2, CO, H_2O and SO_2 emissions on the background concentrations is hardly measurable (Ferry et al., 1999; Paladino et al., 2000; Schlager et al., 1999; Schumann et al., 2000; Ziereis et al., 1999, Ziereis et al., 2000;). Table 1 summarizes some data that allow explaining these experimental results (IPCC, 1999; Schumann et al., 1998).

It can be observed that the emission of NO_x (NO+ NO_2) per kg of fuel consumed by an airplane is 100 to 200 times lower than the corresponding CO_2 quantity. As the natural background for this last one is 10^6 times higher than the first, the CO_2 emission only just influences the closest vicinity of each specific flight trajectory whereas the emissions of NO_x generate a remarkable contrast on the existing background at route scale. Moreover, NO_x lifetime in the upper troposphere (few days up to a week) favours the contrast between NO_x source regions and background regions. The situation for H_2O is similar to the one for CO_2.

Due to the global spatial distribution of the air traffic, the concentration of NO_x in the background for the 8-12 km layer displays a clear latitudinal gradient in the Northern Hemisphere, with minimum values (20-40 pptv) near the Equator and maximum (200-300 pptv) in latitudes 50º-60º N. Within the strong general variability, the detected concentrations are usually higher in the areas of influence of the air traffic corridors, as demonstrated by the data corresponding to the vertical profiles obtained with in-situ measurements. These results show an inhomogeneous vertical distribution of emission tracers with a significant increase of NO_2 concentrations in the 8-12 km layer (Ziereis et al., 1999; Ziereis et al., 2000).

3. Methodology

The assessment of the use of satellite remote sensing tools for the detection of NO_2 generated in the UT/LS region from en-route aircraft emissions has been tackled by applying the following two different and complementary approaches:

1) Firstly the most powerful orbital sensors capable of measuring atmospheric NO_2 have been selected. The associated products with more possibilities of containing information about the NO_2 produced in the vicinity of the UT/LS have been identified. Subsequently, a simple exercise of air traffic emission calculation has been performed in order to establish an upper bound for NO_2 that typically could be associated to any congested air route. From this result an estimation of the corresponding selected NO_2 satellite product has been calculated as retrieved from the selected space sensors. Finally, these results have been compared with the actual space sensor features and measurements. To conduct the calculation exercise real air traffic and operational data from Canary Islands Corridor (CIC) have been used due to its high and stable traffic demand and its data accessibility. Moreover, an ideal atmospheric scenario characterised by unrealistic stable conditions has been considered in order to favour the maximum possible accumulation of NO_2 at cruise altitudes.

2) Secondly, a research of any evidence of air traffic emissions in the selected space sensors data sets has been done. OMI and SCIAMACHY instruments have been used for this purpose and a thorough analysis of the respective NO_2 column data retrieved from their measurements over some oceanic corridors has been carried out. CIC and NAFC have been selected as representative of the most congested ones and where spatial gradients of NO_2 concentration are expected. For this analysis available yearly and monthly mean NO_2 column data have been inspected and compared with the spatial global pattern of aircraft NO_2 emissions obtained from emission inventories. This strategy is similar to that one successfully used for the identification of NO_2 ship emissions from space remote sensors for congested maritime routes (Beirle et al., 2004; Franke et

al., 2009; Richter et al., 2004). Finally, daily mean NO_2 column maps have been also analysed if no NO_2 signature is detected in the monthly mean NO_2 column data.

3.1 Calculation exercise for analysis of space sensor capability to detect aircraft emissions in the UT/LS

In this section the results of the first mentioned approach, based on a comparison of actual NO_2 measuring potential of current space sensors vs. an upper bound calculated for NO_2 concentration in the UT/LS due to aircraft emissions are presented.

3.1.1 Current satellite sensors for NO_2 detection.

The characteristics of space sensors devoted to NO_2 measurement are presented in Table 2. These systems were specifically designed for obtaining the best yield in terms of space/time resolution and detection limits, according to the state-of-the-art technology in their respective dates. Among these sensors, only MIPAS onboard ENVISAT has been specifically designed for detection of aircraft emissions. MIPAS is a limb sounder which performs measurements in different configurations modes. The named “Aircraft Emission mode” (AE) has been optimized to explore lines of sight parallel to some aircraft corridors. This measuring strategy enables long optical paths in the 7-13 km altitude range with the purpose of detecting the contribution of the air traffic emissions to the existing background concentrations of NO_2 (Fischer et al., 2008). Although supposedly this kind of measurements would allow the detection of aircraft emissions in congested corridors, up to now the analysis of the available data of the most promising case (NAFC) has been unsatisfactory (Holmes, 2007).

Apart from MIPAS, there are several sensors capable of performing NO_2 measurements in atmospheric regions that cover the UT/LS, providing two kinds of products: column (total, tropospheric and stratospheric) density values and concentration vertical profiles (see Table 2). The lower altitude involved in the calculations of NO_2 concentration vertical profile is nominally 10 km, however, only reliable measurements are obtained above 15 km. Consequently, the information related to gaseous compounds present at altitudes near the tropopause can only be found in column data being SCIAMACHY and OMI the sensors which offer the best technical performance in NO_2 measurements. Table 3 shows the main instrumental characteristics for both systems (Bovensmann et al., 1999; Levelt et al., 2006; OMI Team, 2009; Sierk et al., 2006). In our work, OMI and SCIAMACHY NO_2 tropospheric column density data available from TEMIS and IUP Bremen have been used (TEMIS, 2010; SCIAMACHY DOAS nadir data browser, 2010).

The algorithms for tropospheric NO_2 vertical column retrievals developed for SCIAMACHY and OMI data have common steps. Briefly, the first step is the determination of the total amount of NO_2 along the effective line of sight (Slant Column, SC) from a spectral fit to the Earth reflectance spectrum by using the Differential Optical Absorption Spectroscopy (DOAS) approach (Platt & Stutz, 2008). The second step is the estimation of the stratospheric contribution to the SC. Finally, the remaining tropospheric SC obtained by subtraction the stratospheric contribution to the total SC is converted to tropospheric vertical column by using a tropospheric air mass factor (AMF), parameter that strongly depends on many factors such as surface albedo, a priori NO_2 profile shapes, cloud retrieval and aerosols (Boersma et al., 2004; Martin et al., 2003; Richter & Burrows, 2002).

The primary uncertainties in satellite NO_2 column measurements arise from the slant column fitting, which defines the instrumental detection limit. The most promising areas for space-borne detection of aircraft emissions are the air traffic corridors located over oceanic areas. These areas can be considered as “clean zones” with associated low tropospheric NO_2 column values. In this situation and for cloud free situations, the overall retrieval uncertainty is dominated by the errors in the total slant column and the stratospheric component of the total slant column (Boersma et al., 2004). For instance, in the case of the DOMINO TEMIS (Dutch OMI NO_2) product the uncertainty in the stratospheric slant column is around 1-2 10^{14} molec cm^{-2}, much smaller than the one associated to the total slant column (7 10^{14} molec cm^{-2}) (Boersma et al., 2007). Hence the detection limit of the DOMINO method is mainly determined by the random total slant column error that can be averaged out by taking large numbers of observations. Similar results have been obtained for SCIAMACHY observations with values ranging 0.5-1 10^{15} molec cm^{-2} (Richter et al., 2005b; Boersma et al., 2008).

Regarding the calculation of the stratospheric component there are different procedures to do it. The Reference Sector Method (RSM) is the simplest one and is based on two principles: 1) assuming the longitudinal homogeneity of the stratospheric NO_2 layers; 2) assuming the total NO_2 column measured on an unpolluted zone (Pacific or Atlantic Ocean) has a negligible tropospheric contribution. The difference between the measured total column and the value determined in the reference sector on the same day at the same latitude is interpreted as a value of tropospheric column. This assumption is considered reasonable at low and middle latitudes but at high latitudes longitudinal variations are not negligible close to the Polar Vortex or during major changes in stratospheric dynamics (Boersma et al., 2004; Richter & Burrows, 2002; Richter et al., 2005a). Consequently, the tropospheric NO_2 column data obtained following this calculation procedure could have a considerable level of uncertainty. As an example, Fig.1 shows the differences between the stratospheric NO_2 vertical column obtained from 2003 to 2006 in two World Meteorological Organisation’s (WMO) ground based stations of the Network for the

Detection of Atmospheric Composition Change (NDACC): Izaña (Canary Islands, 28.30°N 16.48°W) and Mauna Loa (Hawaii, 19.54°N, 155.58°W) with similar latitudes (only 8º of difference) (NDACC, 2009). These results clearly show that assuming that stratospheric NO_2 is zonally homogeneous and extrapolating stratospheric column data obtained over Pacific Ocean sectors to other zones of similar latitude is not realistic, even for middle-low latitudes.

Another procedure to obtain the stratospheric component from the total column of NO_2 consists of assimilating NO_2 slant columns into a global chemistry and transport model (CTM) that include stratospheric chemistry and temperature and wind fields. In this approach the stratospheric zonal variability is taken into account by the calculation reducing the uncertainties due to stratospheric dynamics (Boersma et al., 2004; Boersma et al., 2007; Dirksen et al., 2008).

Another source of error in the retrieved tropospheric NO_2 columns is the use of the tropospheric AMF for the conversion of the tropospheric slant column into vertical one. These errors define the relative error for columns well above the detection limit (Boersma et al., 2004; Martin et al., 2003; Richter & Burrows, 2002). Although this error source is the most important for polluted conditions, it also contributes in cases with low tropospheric NO_2 columns. In this situation the main source of error for the calculation of AMF is the uncertainty in profile shapes. The influence of this inaccuracy has been estimated for different conditions. On the one hand, when NO_2 is located in boundary layer the AMF uncertainty is typically 10 %, although for high latitude regions it can be larger than 50% (Boersma et al 2004). On the other hand, when NO_2 is located at higher altitudes, as is the case of lightning, the uncertainty could be even higher (Beirle et al., 2006; Beirle et al., 2009; Boersma et al., 2005; Bucsela et al., 2010; Martin et al., 2006).

The use of a priori tropospheric NO_2 profile shapes obtained by CTM are currently the best option, as measurements of tropospheric NO_2 profile shapes are very scarce. SCIAMACHY and OMI TEMIS products use predicted profile shapes from CTM TM4 model (Dirksen et al., 2008;

van der A. et al., 2010) that are based on anthropogenic and natural NO_x emissions including free tropospheric air traffic emissions and lightning activity.

The important uncertainties related to these profile shapes, such as the undersampling of the model relative to the SCIAMACHY and OMI pixel size, lead to representativeness errors (Boersma et al., 2007; van der A et al., 2010).

For cloudy scenes, the AMF uncertainties due to the presence of clouds range from approximately 20% for low cloud fraction to 50-80% for high cloud fraction, depending on profile shape and cloud top height (Wenig et al., 2008). In general, tropospheric NO_2 vertical column information available from OMI or SCIAMACHY measurements corresponds to cloud free pixels. This filtering criterion is generally adopted because most of tropospheric NO_2 is located near the surface and the presence of clouds above NO_2 avoids its detection. However, when tropospheric NO_2 is located above clouds the higher albedo of the scene increases the sensitivity for NO_2 detection and therefore the AMF values. There are several works which demonstrate the ability of nadir viewing satellite observations for detecting NO_2 at the top and above clouds which is generally associated to biomass burning outflow or lightning activity (Beirle et al., 2006; Beirle et al., 2009; Boersma et al., 2005; Martin et al., 2002; Ritcher & Burrows, 2002; van der A et al., 2008). Considering that UT/LS level is generally above cloud formation height, the presence of clouds in the field of view of satellite sensors could favour, in principle, the detection of aircraft emissions produced in the vicinity of the tropopause.

Therefore and despite the uncertainty of this product, the retrieved NO_2 tropospheric vertical column, for all scenes (cloud free and cloudy), could content good information about the presence of aircraft emissions in the UT/LS.

3.1.2 Emission calculation exercise

Following the proposed methodology, an air traffic emission calculation exercise has been performed in order to estimate a theoretical maximum NO_2 column value related to aircraft emissions of a representative congested corridor. The CIC, with an on-route length of 800 km approximately and a mean traffic frequency of 400 flights/day, has been selected for this purpose. This corridor connects the Iberian Peninsula and Europe to the Canary Islands (Fig.2). It is composed of a system of four nearly parallel routes (UN866, UA/UN873, UN858 and UA/UN857) that keep constant lateral position within ± 9.25 km and vertical position within 300 m (for altitude level above 884 m) (BOE 95, 2002; BOE 64, 2002). These routes are not uniformly spaced, having a minimum separation of 80 km (UN873-UN858), 98 km (UN866-UN873) and 96 km (UN858-UN857). Although the dispersion of an aircraft emission plume is governed by atmospheric stratification and wind shears, and these conditions have a high spatial variability, the lateral dispersion of these kinds of plumes in the UT/LS reaches a typical extension of 15 km after 10 hrs and 200 m along the vertical dimension (Schumann & Konopka, 1994; Schumann et al., 2000). As a consequence, these routes of the CIC can be considered independent of each other in terms of spatial overlapping of their emissions.

Considering the most recent available data set about annual air traffic on the CIC (AENA 2007), the route with the average highest annual demand is UN858 through VASTO and the highest specific occupation day in this route was December 23rd with 225 aircrafts (see Fig. 3). These specific data have been taken into account to calculate a scenario of NO_x maximum emissions. The calculation has been done with the Advanced Emission Model III (AEM III) developed and supplied by EUROCONTROL (Carlier & Jelinek, 2006; Jelinek et al., 2004). This model is a state-of-the-art tool useful to assess the environmental impact of future airspace and route network planning scenarios. AEM III model allows the estimation of aviation emissions (CO_2,

H_2O, SO_x, NO_x, HC, CO, Benzene, Volatile Organic Compounds, and Total Organic Gases) and fuel burn. AEM III uses several databases (aircraft, aircraft engines, fuel burn rates and emission factors) provided by external data agencies in order to assure the quality of the information produced. Emission indices and fuel flow from ICAO Engine Exhaust Emissions Data Bank are adapted to the atmospheric conditions at altitude levels above 3000 feet (around 915 m). This information is combined with air traffic flight profiles. AEM III has been validated for fuel burn and the results are close to actual trip fuel data.

The total NO_x emissions generated on 23rd December 2006 along the route through VASTO have been calculated using as inputs in the model the real air traffic and flight data, the technical characteristics of the operating aircrafts (Table 4) and the on-route distance (743.48 km). The total emission estimated by the AEM III model for UN858 through VASTO route on this day was 5718.83 kg NO_x. This NO_x emission result has been used for estimating the maximum NO_2 mass that could be generated, assuming an ideal maximum yield of NO_2 production. For this purpose, the maximum ratio NO_2/NO_x reported at cruise altitudes, 40 %, (Ziereis et al., 1999) was used. The result was that the maximum NO_2 mass that could be generated in VASTO on 23rd December 2006 would be 2287.53 kg.

3.1.3 Cross-checking against satellite sensor detection limits and actual measurements over CIC.

Maximum NO_2 column values associated to the total NO_2 mass generated in VASTO on 23rd December 2006 have been calculated to be cross-checked against SCIAMACHY and OMI detection limits. For this calculation an unrealistic scenario characterised by the following assumptions has been considered: a) constant cruise speed of each aircraft along the route, b) all the emissions coming from all the aircrafts of the simulation day have been added up, c) in order to favour conditions for maximum accumulation of pollutants, atmospheric diffusion has been the only dispersion process taken into account and advection has not been considered. The input data have been: the total NO_2 mass virtually produced (2287.53 kg), the total on-route distance (743.48 km), the maximum horizontal dispersion of the global plume (15 km after ten hours) and the specific ground pixel sizes at nadir position (minimum observed area) of both sensors (see Table 3). The NO_2 column values obtained in our ideal scenario for SCIAMACHY and OMI minimum observed area were respectively:

- NO_2 column (for SCIAMACHY ground pixel at nadir): $0.67\ 10^{14}$ molec cm^{-2}

- NO_2 column (for OMI ground pixel at nadir): $1.68\ 10^{14}$ molec cm^{-2}

Taking into account the assumed considerations, these NO_2 column values can be taken respectively as upper bounds that could be also extrapolated to any congested corridor. Obviously, the actual column values should always be much lower than the ones obtained in this exercise. For any very high demand corridor, the actual conditions like the time distribution of flights, the existence of chemical conversion processes producing lower NO_2/NO_x ratios than 40%, and of course, the actual atmospheric dispersion phenomena will reduce the actual NO_2 columns produced. (Beirle et al., 2009; Meijer et al., 1997; Schumann et al., 2000)

Nevertheless, the maximum values obtained for the NO_2 column are lower than SCIAMACHY and OMI detection limits typically reported for individual NO_2 tropospheric column measurements ($0.5\text{-}1\ 10^{15}$ molec cm^{-2}) (Boersma et al., 2007; Richter et al., 2005b) and around the same order of magnitude ($0.4\text{-}2\ 10^{14}$ molec cm^{-2}) for temporally or spatially averaged NO_2 column measurements in some study cases (Boersma el at., 2004; Franke et al., 2009; Richter et al., 2004).

It can be concluded that even under these unrealistic favourable conditions, the detection of the

NO_2 columns due to air traffic emissions on congested corridors like the VASTO in the CIC would be difficult to achieve.

Regardless of this conclusion, an in-depth inspection of available daily data of NO_2 tropospheric vertical column from OMI and SCIAMACHY (TEMIS 2010) for year 2006 has been done in order to ascertain the presence of any NO_2 signature related to aircraft emissions over the area of the CIC. The results of this analysis have been always negative. For the particular case of 23rd December of 2006, unfortunately ENVISAT did not pass through the vertical of the CIC so there were not available SCIAMACHY data. The available OMI daily average values of NO_2 column density are shown in Fig 4.

The image available from the TEMIS web, corresponding to the tropospheric vertical column map for cloud free pixels (cloud radiance fraction < 50%) is shown in Fig. 4 left. It can be observed a lack of data for the area of the CIC due to the presence of clouds. As this cloudy situation could be favourable in terms of sensitivity of the sensor cloud free & cloudy pixels were analysed. Tropospheric vertical column data associated to cloudy scenes are available from the DOMINO product in a daily basis from the TEMIS web page in HE5 format (TEMIS, 2010; Boersma et al., 2009). DOMINO data are a pure Level 2 product, i.e. it provides geophysical information for each and every ground pixel observed by the instrument without any additional binning, averaging or gridding. These data were used in conjunction with ArcGis (v. 9.3) to generate NO_2 tropospheric vertical column maps for all scenes (cloud free and cloudy). Mean values are calculated for tropospheric vertical column data associated to areas where OMI orbits overlap.

The results for cloud free & cloudy pixels are shown in Fig.4 (right). The scale has been selected in order to differentiate among low tropospheric NO_2 column density values. Tropospheric vertical column values associated to cloudy pixels in the CIC are lower than $1\ 10^{15}$ molec cm^{-2}.

These values are very similar to the ones associated to other zones of the Atlantic Ocean far from the CIC and therefore not affected by aircraft emissions. There is only a spatial zone of the CIC in the south eastern coast of the Iberian Peninsula (35°-36°N, 7°-10°W) which presents values higher than 2 10^{15} molec cm^{-2} (3-10 10^{15} molec cm^{-2}). All these values are clearly higher than the NO_2 column results obtained for the ideal scenario of this work. Despite this fact an assessment of the possible sources of the NO_2 detected by OMI in the CIC has been done. For this purpose, three days backward trajectories calculations have been carried out by using HYSPLIT model from NOAA Air Resources Laboratory (Draxler & Rolph, 2003; Rolph, 2003). The trajectories (not shown here) for altitudes ranging from surface to flight level have been calculated for the time that OMI overpass the CIC on 23rd December for two geographical locations (32° N 12° W ;36° N 8° W). The tropospheric vertical column values retrieved over these points were representative of two different ranges <1 10^{15} molec cm^{-2} and >2 10^{15} molec cm^{-2} respectively. The results obtained for both cases are very similar and indicate that there is a narrow layer from surface to 500-1500 m altitude with air masses coming from centre and southwest of the Iberian Peninsula. NO_2 present in that area the previous days was transported to the CIC for the day of study. At cruise altitudes (8-12 km) the air masses arriving at those points maintain their height and come from the Atlantic Ocean and the Northern USA and consequently do not contribute to the NO_2 detected by the sensor.

It can be concluded that the upper bound obtained for NO_2 column value attributable to aircraft emissions is much lower than the actual retrieved vertical columns from OMI measurements for the zone of the CIC. NO_2 column values around 1 10^{15} molec cm^{-2} are the most frequently reported not only for the CIC but also for oceanic areas far from the influence of aircraft emissions. Backward trajectory calculations have indicated that tropospheric NO_2 detected by OMI for 23rd December 2006 in the area of the CIC is not associated to aircraft emissions but to transport processes from European continental areas.

3.2 Analysis of SCIAMACHY and OMI data in other specific air traffic corridors

Data from global aviation emissions inventories have been used for the identification of the most suitable corridors to be studied. Data of the fuel burn for all altitudes aggregated for year 2000 from SAGE (System for assessing Aviation's Global Emissions v1.5) are shown in Fig. 5 (Kim et al., 2005) and have been used for the selection of target corridors.

Finally, the NAFC has been considered the most promising corridor due to its high traffic demand and emissions and its geographical situation (45°-60°N, 10°-60°W). The position of air routes in this corridor is established every day taking into account the meteorological situation and are very frequently grouped in the 50°N-60°N latitude band, although some routes are occasionally located near 45°N (https://pilotweb.nas.faa.gov/common/nat.html). Although this corridor does not present the highest values of traffic demand, and emissions, its situation above the Atlantic Ocean connecting North America and Europe favours its possible identification from space remote sensors. Moreover, its geographical distribution almost perpendicular to OMI and SCIAMACHY orbits allows to be observed by these sensors several times per day. Consequently there are more possibilities of detecting aircraft emissions in this corridor than in other one with similar demand but situated with a different orientation, like the case of CIC.

Unlike continental corridors crossing Europe and North America, it can be assumed that NAFC is located over a zone nearly free of significant NO_2 surface emissions except the possible contribution of shipping and advection from continent. Although ship emissions can be considered as potential interference for measuring NO_2 from air traffic, their maxima values are produced further South than 50°N. The results from traffic emissions inventories show that NO_x

emissions from NAFC and the main shipping areas in the central north Atlantic ocean overlap only between 45º-50ºN, (Dalsoren et al., 2009; Eyring et al., 2010; Hoor et al., 2009; Kasibhatla et al., 2000). As a consequence, it is expected that a possible detection of NO_2 column in this area could be related essentially to the stratospheric background, aircraft emissions and lightning, this last one being a minor source (Brunner et al., 1998; Choi et al., 2005; Guerova et al., 2006; Kim et al., 2008; van der A et al., 2008; Richter et al., 2011).

A detailed inspection of yearly and monthly mean tropospheric NO_2 column density data from TEMIS and IUP Bremen (TEMIS, 2010; SCIAMACHY DOAS nadir data browser, 2010) has been done in order to find a pattern which corresponds to the aircraft emission spatial pattern estimated from SAGE (see Fig.5). A similar attempt with GOME data has been previously reported but the results were unsatisfactory (Beirle, 2004). In our case the results were also negative as no signature of NO_2 was observed for areas corresponding to NAFC.

An example that illustrates this conclusion is shown in Fig. 6 where mean OMI tropospheric NO_2 vertical column density values for February and July from 2006 to 2009 are depicted. These months have been selected as representative of low and high air traffic demand conditions in the NAFC respectively (Wilkerson et al., 2010). DOMINO data have been used in conjunction with ArcGis (v. 9.3) to obtain mean values for all scenes (cloud free and cloudy) following the same methodology used for Fig.4 (right). The scale has been selected in order to differentiate low tropospheric NO_2 column density values. The results obtained for both months show that the retrieved NO_2 column values over the corridor are quite homogeneous (below $7\ 10^{14}$ molec cm^{-2}) and without a marked spatial pattern of enhanced NO_2 which corresponds to the aircraft emission pattern in NAFC.

However, it can be observed in Figure 6, the existence of NO_2 column values around $0.5 – 1\ 10^{15}$ molec cm^{-2} located below 50°N near the southern limits of the NAFC which can be related to shipping traffic because of its overlap with one of the busiest shipping areas in the northern Atlantic (AMVER 2011; Eyring et al., 2010; Hoor et al., 2009), as it is shown in Fig. 7.

These NO_2 columns are observable not only in cloud free scenes but also in cloudy ones (not shown in the paper) for both averages (February and July 2006-2009). This fact has been also reported for tropospheric NO_2 column data associated to one of the most congested Indian maritime routes (Richter et al., 2010) showing that the reduction in NO_2 column values when using all data is much less than expected from cloud shielding. Nevertheless, long range transport of continental pollution and lightning can not be discarded as additional sources of the observed NO_2 in that area as both events are associated to the presence of clouds (Richter et al., 2011).

Despite these unsatisfactory results related to the study of the monthly mean NO_2 columns for NAFC area, an inspection of available OMI and SCIAMACHY NO_2 tropospheric column daily mean data from 2004 to 2009 has been done. The objective was to find specific days for which significant NO_2 tropospheric column densities were retrieved over the corridor. More than three hundred cases were identified and twenty were selected for an in-depth analysis. In any case the NO_2 could be associated to aircraft emissions in the NAFC. In very few cases the NO_2 source could be assigned definitely and always the associated process was pollution transport from continents. In order to illustrate this conclusion, three different and representative cases are presented here.

The first case shown in Fig. 8 corresponds to NO_2 tropospheric vertical column data from OMI for 11th February 2007. It can be observed that there is an extensive zone (50°-60°N, 15°-40°W) in which the NO_2 column values are $>2\ 10^{15}$ molec cm^{-2}. To determine the possible source of the NO_2 presented in that area, three days backward trajectories calculations have been carried out by using HYSPLIT model. The trajectories have been calculated for the time that OMI over passed the zone of interest and for several altitudes (from surface to flight altitudes). The results obtained at a representative point (58°N 30°W) of the NO_2 enriched area for 11th February 2007 are shown in Fig.9. There is a wide layer from 500 m to 3500 m altitude with air masses coming from northern European countries. It can be observed that NO_2 was generated in the continent during the previous days and transported to this area for the selected day. For higher altitudes the origin of the air masses is completely disconnected from the continent. At flight altitudes the air masses arriving at that point come from Canada and Greenland and consequently do not contribute to the NO_2 observed in the column measurement.

Similar results have been obtained for areas of the NAFC near North America. In the example shown in Fig. 10 corresponding to 23rd July 2006, there is a zone overlapping the corridor with NO_2 column values higher than the mean level found in the surrounding oceanic areas. The continuity of the column values observed from the continent would indicate again that there is a transport process involved. In order to confirm this hypothesis backward trajectories calculations have been done for different points of this area. The results obtained for the geographical location 45°N 42°W are shown in Fig. 11 Backward trajectories for the lower altitudes (500-3500 m) point out a well defined transport of air masses coming from the north-eastern area of USA. The trajectories indicate that NO_2 produced in 21st July at surface level over the continent reached the point of interest at 23rd July. The results for altitudes between 4000 and 8000 m indicate that probably there is no contribution of NO_2 coming from surface levels of northern and central USA because the altitudes of the air mass trajectories are higher than the associated mixed layer heights (not shown). Considering the uplift process undergone by trajectories ending in the layer between 8500-10000 m, a probable contribution to the NO_2 column values due to air masses coming from surface levels of southern USA areas could also exist. For altitudes up to 12000m the air masses come from zones free of NO_2 at the correspondent altitudes.

These two cases are examples of a phenomenon that occurs frequently in the zone associated to the NAFC and suggest that the observed NO_2 in this corridor is mainly transported from the continent (Europe or North America).

Other different patterns of NO_2 column values for the corridor area have been identified in OMI and SCIAMACHY data. A significant example of these other situations is shown in Fig.12. In this case NO_2 tropospheric column values >2 10^{15} molec cm^{-2} cover a wide area in nearly the whole corridor and this spatial pattern could be related somehow to the presence of aircraft emissions. In this particular case two geographical points have been selected to calculate backward trajectories. Fig. 13 and 14 show the results for 60°N 40°W and 55°N 20°W geographical points respectively. For 60°N 40°W, backward trajectories from 500 m to 5000 m indicate that air masses came from northern Canada and near polar zones giving no apportion of NO_2. However, air masses from surface levels located in eastern USA on 18th-19th January were uplifted to a layer between 6000 m and 7000 m during 19-20th and transported to the point of interest. The study of global maps (not shown in the text) of mean sea level pressure for 18th, 19th and 20th January 2009 obtained from the Ready Web server of the Air Resources Laboratory from NOAA (National Oceanic and Atmospheric Administration, USA) (http://ready.arl.noaa.gov/ready2-bin/metmap1a.pl) allows to explain this transport process. These maps show the presence of a persistent low pressure system located southern Greenland and a high pressure system over the eastern coast of USA during 18-19th producing a mixed layer height around 1000 m keeping most of pollutants located near surface. The high pressures over this area were lowering during 19th and 20th January developing a new low pressure system that became connected with the one near Greenland. This fact produced the uplift and transport of pollutants from continental surface up to higher altitudes in oceanic areas. In these conditions, the NO_2 content of these masses could contribute to the observed NO_2 column values. On the contrary, the trajectories of 7500-12000 m keep their altitude levels over passing Canada and polar zones and do not contribute to the enhancement of the NO_2 column values in NAFC. Concerning the second test point presented in Fig.14, the exploration of backward trajectories calculations from surface to flight levels gave similar conclusions. The results for 500 up to 5000 m altitude indicate that Canada and polar areas are the origin of the air masses. For the altitude range 7000-12000 m, only a thin layer between 6000 to 6500 m could transport part of the NO_2 generated at surface level near the east coast of USA to the point of interest. The rest of calculated trajectories keep their altitude in a roughly constant value and cover an extensive area from north-south to north of the middle east of North America.

These last examples support again the conclusion that transport from continental source areas is one of the most probable phenomena related to the increment of NO_2 column levels observed in the NAFC. Nevertheless, it has to be mentioned that besides the other sources previously mentioned (shipping and lightning) the observed NO_2 column values in this corridor could be due to other origin. Considering the vicinity of the Arctic polar vortex, the inflow of polar air masses could produce false NO_2 enhancements due to possible artifacts of the stratospheric assimilation, especially in wintertime.

4. Conclusions

Results from previous research projects show that aircraft emissions can produce significant increases in the concentration of NO_2 at corridor altitudes in atmospheric regions affected by heavy air traffic. This fact opens the possibility of using this gaseous compound as tracer for detecting air traffic emissions in tropospheric column data obtained by space remote sensing.

In this work Canary Island Corridor (CIC) has been taken as representative of high demand corridors. Actual emission data corresponding to maximum demand circumstances in this corridor have been used to estimate upper limits for the associated NO_2 column values. This exercise has been done assuming unrealistic atmospheric dispersive conditions to allow the development of maximum NO_2 concentration in the corridor. Under these constraints, upper bounds for NO_2 columns have been obtained (0.7 10^{14} molec cm^{-2} for SCIAMACHY and 1.7 10^{14} molec cm^{-2} for OMI). These values are lower than the typical reported SCIAMACHY/OMI detection limits for individual NO_2 tropospheric column measurements (0.5-1 10^{15} molec cm^{-2}) and similar to the limits obtained in some studies for temporally or spatially averaged NO_2 column measurements (0.4-2 10^{14} molec cm^{-2}). Obviously, the real physico-chemical atmospheric dynamics is more likely to be considerable for reducing the actual NO_2 concentrations in the UT/LS with respect to the calculated maximum quantities. This conclusion can be extrapolated to air traffic emissions in any oceanic corridor.

This fact has been confirmed from the analysis of available yearly, monthly or daily mean tropospheric NO_2 column data obtained for CIC and NAFC corridors. No spatial or temporal pattern related to aircraft emissions have been observed in these oceanic corridors. The background tropospheric NO_2 column values (~ 2.5-7.5 10^{14} molec cm^{-2} for monthly mean) retrieved for these oceanic areas are clearly higher than those NO_2 column upper bounds obtained in the emission calculation exercise with CIC data and ideal atmospheric scenario.

Other difficulties for remote sensing of NO_2 related to aircraft activity are intrinsically linked to its presence in the stratosphere or to the existence of multiple potential anthropogenic and natural tropospheric sources. For instance, the influence on the NO_2 column values retrieved for different areas of the NAFC due to long range transport of pollutants from continents and to shipping emissions has been documented in this work. Moreover, the calculation procedures used for retrieving both the NO_2 stratospheric and tropospheric columns also introduce significant errors in these products. These errors are even higher for cloudy scenes which are in principle the most suitable for the detection of NO_2 associated to aircraft emissions at cruise levels.

As a consequence of all these circumstances, detecting and quantifying the low NO_2 column density values associated to air traffic in the UT/LS is a very challenging task for space remote sensing.

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References

AENA: Aeropuertos Españoles y Navegación Aérea, Air Traffic Data Base. (2007),

AMVER: Atlantic Merchant Vessel Emergency Reporting system (2011). Available in http://www.amver.com/density.asp

Beer, R. (2006). TES on the Aura mission: Scientific objectives, measurements, and analysis overview. *Ieee Transactions on Geoscience and Remote Sensing, 44,* 1102-1105.

Beirle, S. (2004). Estimating source strengths and lifetime of Nitrogen Oxides from satellite data. Dissertation, Ruperto-Carola University of Heidelberg, Germany.

Beirle, S., Platt, U., von Glasow, R., Wenig, M., & Wagner, T. (2004). Estimate of nitrogen oxide emissions from shipping by satellite remote sensing. *Geophysical Research Letters, 31,*L18102.

Beirle, S., Spichtinger, N., Stohl, A., Cummins, K.L., Turner, T., Boccippio, D., Cooper, O.R., Wenig, M., Grzegorski, M., Platt, U., & Wagner, T. (2006). Estimating the NO_x produced by lightning from GOME and NLDN data: a case study in the Gulf of Mexico. *Atmospheric Chemistry and Physics, 6,* 1075-1089

Beirle, S., Salzmann, M., Lawrence, M.G., & Wagner, T. (2009). Sensitivity of satellite observations for freshly produced lightning NO_x. *Atmospheric Chemistry and Physics, 9,* 1077-1094.

Bernath, P.F., McElroy, C.T., Abrams, M.C., Boone, C.D., Butler, M., Camy-Peyret, C., Carleer, M., Clerbaux, C., Coheur, P.F., Colin, R., DeCola, P., Bernath, P.F., McElroy, C.T., Abrams, M.C., Boone, C.D., Butler, M., Camy-Peyret, C., Carleer, M., Clerbaux, C., Coheur, P.F., Colin, R., DeCola, P., DeMaziere, M., Drummond, J.R., Dufour, D., Evans, W.F.J., Fast, H., Fussen, D., Gilbert, K., Jennings, D.E., Llewellyn, E.J., Lowe, R.P., Mahieu, E., McConnell, J.C., McHugh, M., McLeod, S.D., Michaud, R., Midwinter, C., Nassar, R., Nichitiu, F., Nowlan, C., Rinsland, C.P., Rochon, Y.J., Rowlands, N., Semeniuk, K., Simon, P., Skelton, R., Sloan, J.J., Soucy, M.A., Strong, K., Tremblay, P., Turnbull, D., Walker, K.A., Walkty, I., Wardle, D.A., Wehrle, V., Zander, R., & Zou, J. (2005). Atmospheric Chemistry Experiment (ACE): Mission overview. *Geophysical Research Letters, 32,* L15S01.

BOE 64. (2002). RESOLUCIÓN de 25 de febrero de 2002, de la Dirección General de Aviación Civil, relativa a la aprobación operacional y criterios de utilización de sistemas para la Navegación de Área Básica (RNAV Básica) en el espacio aéreo europeo (Circular operativa 1/98 Rev.1).

BOE 95. (2002). RESOLUCIÓN de 12 de marzo de 2002, de la Dirección General de Aviación Civil, relativa a la emisión de aprobaciones operacionales RVSM genéricas (Circular Operativa 03/02).

Boersma, K.F., Eskes, H.J., & Brinksma, E.J. (2004). Error analysis for tropospheric NO_2 retrieval from space. *Journal of Geophysical Research-Atmospheres, 109,* D04311.

Boersma, K.F., Eskes, H.J., Meijer, E.W., & Kelder, H.M. (2005). Estimates of lightning NO_x production from GOME satellite observations. *Atmospheric Chemistry and Physics, 5,* 2311-2331.

Boersma, K.F., Eskes, H.J., Veefkind, J.P., Brinksma, E.J., van der A, R.J., Sneep, M., van den Oord, G.H.J., Levelt, P.F., Stammes, P., Gleason, J.F., & Bucsela, E.J. (2007). Near-real time retrieval of tropospheric NO_2 from OMI. *Atmospheric Chemistry and Physics, 7,* 2103-2118.

Boersma, K.F., Jacob, D.J., Eskes, H.J., Pinder, R.W., Wang, J., & van der A, R.J. (2008). Intercomparison of SCIAMACHY and OMI tropospheric NO_2 columns: Observing the diurnal evolution of chemistry and emissions from space. *Journal of Geophysical Research-Atmospheres, 113,* D16S26.

Boersma, K.F., Dirksen, R.J., Veefkind, J.P., Eskes, H.J., van der A, R.J. (2009). Dutch OMI NO_2 (DOMINO) data product. HE5 data file user manual.

Bovensmann, H., Burrows, J.P., Buchwitz, M., Frerick, J., Noel, S., Rozanov, V.V., Chance, K.V., & Goede, A.P.H. (1999). SCIAMACHY: Mission objectives and measurement modes. *Journal of the Atmospheric Sciences, 56*, 127-150.

Brasseur, G.P., Cox, R.A., Hauglustaine, D., Isaksen, I., Lelieveld, J., Lister, D.H., Sausen, R., Schumann, U., Wahner, A., & Wiesen, P. (1998). European scientific assessment of the atmospheric effects of aircraft emissions. *Atmospheric Environment, 32*, 2329-2418.

Brenninkmeijer, C.A.M. (2006). Civil aircraft in global atmospheric chemistry research and monitoring. *Journal De Physique IV, 139*, 321-336.

Brunner, D., Staehelin, J., & Jeker, D. (1998). Large-scale nitrogen oxide plumes in the tropopause region and implications for ozone. *Science, 282*, 1305-1309.

Brunner, D., Staehelin, J., Jeker, D., Wernli, H., & Schumann, U. (2001). Nitrogen oxides and ozone in the tropopause region of the Northern Hemisphere: Measurements from commercial aircraft in 1995/1996 and 1997. *Journal of Geophysical Research-Atmospheres, 106*, 27673-27699.

Bucsela, E.J., Pickering, K.E., Huntemann, T.L., Cohen, R.C., Perring, A., Gleason, J.F., Blakeslee, R.J., Albrecht, R.I., Holzworth, R., Cipriani, J.P., Vargas-Navarro, D., Mora-Segura, I., Pacheco-Hernandez, A., & Laporte-Molina, S. (2010). Lightning-generated NOx seen by the Ozone Monitoring Instrument during NASA's Tropical Composition, Cloud and Climate Coupling Experiment (TC4). *Journal of Geophysical Research-Atmospheres, 115,* D00J10.

Burrows, J.P., Weber, M., Buchwitz, M., Rozanov, V., Ladstatter-Weissenmayer, A., Richter, A., DeBeek, R., Hoogen, R., Bramstedt, K., Eichmann, K.U., & Eisinger, M. (1999). The global ozone monitoring experiment (GOME): Mission concept and first scientific results. *Journal of the Atmospheric Sciences, 56,* 151-175.

Callies, J., Corpaccioli, E., Eisinger, M., Hahne, A., & Lefebvre, A. (2000). GOME-2 – Metop's Second-Generation Sensor for Operational Ozone Monitoring. *ESA bulletin, Vol. 102,* (pp. 28-36).

Cammas, J. P., & Volz-Thomas, A. (2007). The MOZAIC Program (1994-1997). *International Global Atmospheric Chemistry (IAGC) Newsletter, Issue 37*.

Carlier, S., & Jelinek, F. (2006). GAES - Advanced Emissions Model (AEM3) v1.5 - Validation Exercise #4. *EUROCONTROL Experimental Centre GAES report,* EEC/SEE/2006/007.

Choi, Y., Wang, Y.H., Zeng, T., Martin, R.V., Kurosu, T.P., & Chance, K. (2005). Evidence of lightning NO_x and convective transport of pollutants in satellite observations over North America. *Geophysical Research Letters, 32,* L02805.

Dalsoren, S.B., Eide, M.S., Endresen, O., Mjelde, A., Gravir, G., & Isaksen, I.S.A. (2009). Update on emissions and environmental impacts from the international fleet of ships: the contribution from major ship types and ports. *Atmospheric Chemistry and Physics, 9,* 2171-2194

Dirksen, R., Eskes, H., Boersma, F., Levelt P., Veefkind P., & van der A R. (2008). Derivation of ozone monitoring instrument tropospheric NO_2 in near-real time (DOMINO). *Final report of the User Support project DOMINO and DMINO-2, NIVR.*

Draxler, R. R., & Rolph, G. D. (2003). HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) *Model access via NOAA ARL READY Website* *(http://www.arl.noaa.gov/ready/hysplit4.html). NOAA Air Resources Laboratory, Silver Spring, MD.*

Emmons, L.K., Hauglustaine, D.A., Muller, J.F., Carroll, M.A., Brasseur, G.P., Brunner, D., Staehelin, J., Thouret, V., & Marenco, A. (2000). Data composites of airborne observations of tropospheric ozone and its precursors. *Journal of Geophysical Research-Atmospheres, 105,* 20497-20538.

Eyring, V., Isaksen, I.S.A., Berntsen, T., Collins, W.J., Corbett, J.J., Endresen, O., Grainger, R.G., Moldanova, J., Schlager, H., & Stevenson, D.S. (2010). Transport impacts on atmosphere and climate: Shipping. *Atmospheric Environment, 44*, 4735-4771

Eyers, C. J., Norman, P., Middel, J., Plohr, M., Michot, S., Atkinson, K., &. Christou, R. A. (2004). AERO2K Global Aviation, Emissions Inventories for 2002 and 2025, *Qinetic/04/01113*.

Ferry, G.V., Pueschel, R.F., Strawa, A.W., Kondo, Y., Howard, S.D., Verma, S., Mahoney, M.J., Bui, T.P., Hannan, J.R., & Fuelberg, H.E. (1999). Effects of aircraft on aerosol abundance in the upper troposphere. *Geophysical Research Letters, 26*, 2399-2402.

Fischer, H., Birk, M., Blom, C., Carli, B., Carlotti, M., von Clarmann, T., Delbouille, L., Dudhia, A., Ehhalt, D., Endemann, M., Flaud, J.M., Gessner, R., Kleinert, A., Koopman, R., Langen, J., Lopez-Puertas, M., Mosner, P., Nett, H., Oelhaf, H., Perron, G., Remedios, J., Ridolfi, M., Stiller, G., & Zander, R. (2008). MIPAS: an instrument for atmospheric and climate research. *Atmospheric Chemistry and Physics, 8,* 2151-2188.

Franke, K., Richter, A., Bovensmann, H., Eyring, V., Jockel, P., Hoor, P., & Burrows, J.P. (2009). Ship emitted NO_2 in the Indian Ocean: comparison of model results with satellite data. *Atmospheric Chemistry and Physics, 9,* 7289-7301

Grewe, V., Brunner, D., Dameris, M., Grenfell, J.L., Hein, R., Shindell, D., & Staehelin, J. (2001). Origin and variability of upper tropospheric nitrogen oxides and ozone at northern mid-latitudes. *Atmospheric Environment*, *35*, 3421-3433.

Guerova, G., Bey, I., Attie, J.L., Martin, R.V., Cui, J., & Sprenger, M. (2006). Impact of transatlantic transport episodes on summertime ozone in Europe. *Atmospheric Chemistry and Physics, 6*, 2057-2072.

Hoinka, K.P., Reinhardt, M.E., & Metz, W. (1993). North Atlantic Air Traffic Within the Lower Stratosphere: Cruising Times and Corresponding Emissions. *Journal of Geophysical Research-Atmospheres, 98*, 23113-23131.

Holmes, M. (2007). Detection of Aircraft Emission Signatures in Atmospheric Spectra, *EODG MPhys Reports*, University of Oxford.

Hoor, P., Borken-Kleefeld, J., Caro, D., Dessens, O., Endresen, O., Gauss, M., Grewe, V., Hauglustaine, D., Isaksen, I.S.A., Jockel, P., Lelieveld, J., Myhre, G., Meijer, E., Olivie, D., Prather, M., Poberaj, C.S., Shine, K.P., Staehelin, J., Tang, Q., van Aardenne, J., van Velthoven, P., & Sausen, R. (2009). The impact of traffic emissions on atmospheric ozone and OH: results from QUANTIFY. *Atmospheric Chemistry and Physics, 9*, 3113-3136

IPCC (1999). *Aviation and the Global Atmosphere*. In J. E. Penner, D. H. Lister, D. J. Griggs, D. J. Dokken, & M. Mc-Farland M. (Eds.). Intergovernmental Panel on Climate Change. Cambridge University Press.

IPCC (2007). *Climate change2007. The physical science basis.* In S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor & H.L. Miller. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC). Cambridge University Press.

Jelinek, F., Carlier, S., & Smith, J. (2004). The Advanced Emission Model (AEM3) Version 1.5 – Validation Report. , *EUROCONTROL Experimental Centre report,* EEC/SEE/2004/004.

Kasibhatla, P., Levy, H., Moxim, W.J., Pandis, S.N., Corbett, J.J., Peterson, M.C., Honrath, R.E., Frost, G.J., Knapp, K., Parrish, D.D., & Ryerson, T.B. (2000). Do emissions from ships have a significant impact on concentrations of nitrogen oxides in the marine boundary layer? *Geophysical Research Letters, 27*, 2229-2232.

Kim, B. Y., Fleming, G., Balasubramanian, S., Malwitz, A., Lee, J., Waitz, I., Klima, K., Locke, M., Holsclaw, C., Morales, A., McQueen, E., & Gillette, W. (2005). System for assessing Aviation's Global Emissions (SAGE) Version 1.5, Global Aviation Emissions Inventories for 2000 through 2004, *FAA-EE-2005-02*.

Kim, S.Y., Talbot, R., Mao, H., Blake, D., Vay, S., & Fuelberg, H. (2008). Continental outflow from the US to the upper troposphere over the north Atlantic during the NASA INTEX-NA airborne campaign. *Atmospheric Chemistry and Physics, 8*, 1989-2005.

Lee, D.S., Fahey, D.W., Forster, P.M., Newton, P.J., Wit, R.C.N., Lim, L.L., Owen, B., & Sausen, R. (2009). Aviation and global climate change in the 21st century. *Atmospheric Environment, 43*, 3520-3537.

Levelt, P.F., Van den Oord, G.H.J., Dobber, M.R., Malkki, A., Visser, H., de Vries, J., Stammes, P., Lundell, J.O.V., & Saari, H. (2006). The Ozone Monitoring Instrument. *Ieee Transactions on Geoscience and Remote Sensing, 44*, 1093-1101.

Llewellyn, E., Lloyd, N.D., Degenstein, D.A., Gattinger, R.L., Petelina, S.V., Bourassa, A.E., Wiensz, J.T., Ivanov, E.V., McDade, I.C., Solheim, B.H., McConnell, J.C., Haley, C.S., von Savigny, C., Sioris, C.E., McLinden, C.A., Griffioen, E., Kaminski, J., Evans, W.F.J., Puckrin, E., Strong, K., Wehrle, V., Hum, R.H., Kendall, D.J.W., Matsushita, J., Murtagh, D.P., Brohede, S., Stegman, J., Witt, G., Barnes, G., Payne, W.F., Piche, L., Smith, K., Warshaw, G., Deslauniers, D.L., Marchand, P., Richardson, E.H., King, R.A., Wevers, I., McCreath, W., Kyrola, E., Oikarinen, L., Leppelmeier, G.W., Auvinen, H., Megie, G., Hauchecorne, A., Lefevre, F., de La Noe, J., Ricaud, P., Frisk, U., Sjoberg, F., von Scheele, F., & Nordh, L. (2004). The OSIRIS instrument on the Odin spacecraft. *Canadian Journal of Physics, 82*, 411-422.

Marenco, A., Thouret, V., Nedelec, P., Smit, H., Helten, M., Kley, D., Karcher, F., Simon, P., Law, K., Pyle, J., Poschmann, G., Von Wrede, R., Hume, C., & Cook, T. (1998). Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, An overview. *Journal of Geophysical Research-Atmospheres, 103*, 25631-25642.

Martin, R.V., Chance, K., Jacob, D.J., Kurosu, T.P., Spurr, R.J.D., Bucsela, E., Gleason, J.F., Palmer, P.I., Bey, I., Fiore, A.M., Li, Q.B., Yantosca, R.M., & Koelemeijer, R.B.A. (2002). An improved retrieval of tropospheric nitrogen dioxide from GOME. *Journal of Geophysical Research-Atmospheres, 107 D20,* 4437.

Martin, R.V., Jacob, D.J., Chance, K., Kurosu, T.P., Palmer, P.I., & Evans, M.J. (2003). Global inventory of nitrogen oxide emissions constrained by space-based observations of NO_2 columns. *Journal of Geophysical Research-Atmospheres, 108,* 4537-4548.

Martin, R.V., Sioris, C.E., Chance, K., Ryerson, T.B., Bertram, T.H., Wooldridge, P.J., Cohen, R.C., Neuman, J.A., Swanson, A., & Flocke, F.M. (2006). Evaluation of space-based constraints on global nitrogen oxide emissions with regional aircraft measurements over and downwind of eastern North America. *Journal of Geophysical Research-Atmospheres, 111,* D15308.

Meijer, E.W., vanVelthoven, P.F.J., Wauben, W.M.F., Beck, J.P., & Velders, G.J.M. (1997). The effects of the conversion of nitrogen oxides in aircraft exhaust plumes in global models. *Geophysical Research Letters*, *24*, 3013-3016.

NDACC: Network for the Detection of Atmospheric Composition Change. (2009) Available on http://www.ndsc.ncep.noaa.gov/

OMI Team. (2009). Ozone Monitoring Instrument (OMI) Data User's Guide, *OMI-DUG-2.0*.

Paladino, J.D., Hagen, D.E., Whitefield, P.D., Hopkins, A.R., Schmid, O., Wilson, M.R., Schlager, H., & Schulte, P. (2000). Observations of particulates within the North Atlantic Flight Corridor: POLINAT 2, September-October 1997. *Journal of Geophysical Research-Atmospheres*, *105*, 3719-3726.

Platt, U., & Stutz, J. (2008). *Differential Optical Absorption Spectroscopy. Principles and Aplications*. Berlin Heidelberg: Springer-Verlag.

Ratier, G., Levrini, G., Popescu, A., Paulsen, T., Readings, C., & Langen, J. (1999). GOMOS: Envisat's Contribution to Measuring Long-Term Trends in Ozone and Other Trace Gases. *ESA Bulletin 97*.

Richter, A., & Burrows, J.P. (2002). Tropospheric NO_2 from GOME measurements. *Advances in Space Research*, *29*, 1673-1683.

Richter, A., Eyring, V., Burrows, J.P., Bovensmann, H., Lauer, A., Sierk, B., & Crutzen, P.J. (2004). Satellite measurements of NO_2 from international shipping emissions. *Geophysical Research Letters*, *31*, L23110.

Richter, A., Burrows, J.P., Nuss, H., Granier, C., & Niemeier, U. (2005 a). Increase in tropospheric nitrogen dioxide over China observed from space. *Nature*, *437*, 129-132.

Richter, A., Burrows, J.P., Nuss, H., Granier, C., & Niemeier, U. (2005 b). Supplementary Discussion for Increase in tropospheric nitrogen dioxide over China observed from space. *Nature, 437*, 129-132.

Richter, A., Hilboll, A., Zien, A., & Burrows, J.P. (2010). GOME-2 satellite observations of NOx emissions from ships. *Geophysical Research Abstracts, 12,* EGU2010-12919.

Richter, A., Hilboll, A., Zien, A., & Burrows, J.P. (2011). Cloud effects in satellite observed tropospheric NO_2. *Geophysical Research Abstracts, 13,* EGU2011-8161.

Rolph, G. D. (2003). Real-time Environmental Applications and Display sYstem (READY). *Website (http://www.arl.noaa.gov/ready/hysplit4.html). NOAA Air Resources Laboratory, Silver Spring, MD.*

Russell, J.M., Gordley, L.L., Park, J.H., Drayson, S.R., Hesketh, W.D., Cicerone, R.J., Tuck, A.F., Frederick, J.E., Harries, J.E., & Crutzen, P.J. (1993). The Halogen Occultation Experiment. *Journal of Geophysical Research-Atmospheres*, *98*, 10777-10797.

Schlager, H., Schulte, P., Flatoy, F., Slemr, F., van Velthoven, P., Ziereis, H., & Schumann, U. (1999). Regional nitric oxide enhancements in the North Atlantic flight corridor observed and modeled during POLINAT 2 - a case study. *Geophysical Research Letters, 26*, 3061-3064.

Schoeberl, M.R., Douglass, A.R., Hilsenrath, E., Bhartia, P.K., Beer, R., Waters, J.W., Gunson, M.R., Froidevaux, L., Gille, J.C., Barnett, J.J., Levelt, P.E., & DeCola, P. (2006). Overview of the EOS Aura Mission. *Ieee Transactions on Geoscience and Remote Sensing, 44*, 1066-1074.

Schumann, U., & Konopka, P. (1994). A simple estimate of the concentration field in a flight corridor. In Impact of Emissions From Aircraft and Spacecraft Upon the Atmosphere. In U. Schumann & D.Wurzel (Ed.), *DLR-Mitt 94-06, DLR, Köln, Germany*, (pp. 354-359).

Schumann, U., Schlager, H., Arnold, F., Baumann, R., Haschberger, P., & Klemm, O. (1998). Dilution of aircraft exhaust plumes at cruise altitudes. *Atmospheric Environment, 32*, 3097-3103.

Schumann, U. (2000). Effects of aircraft emissions on ozone, cirrus clouds, and global climate. *Air & Space Europe, 2*, 29-33.

Schumann, U., Schlager, H., Arnold, F., Ovarlez, J., Kelder, H., Hov, O., Hayman, G., Isaksen, I.S.A., Staehelin, J., & Whitefield, P.D. (2000). Pollution from aircraft emissions in the North Atlantic flight corridor: Overview on the POLINAT projects. *Journal of Geophysical Research-Atmospheres, 105*, 3605-3631.

Schumann, U. (2002). Aircraft Emissions. Vol3. Causes and consequences of global environmental change. In I. Douglas (Ed.), *Encyclopedia of Global Environmental Change* (pp. 178-186). Chichester: John Wiley & Sons, Ltd.

SCIAMACHY DOAS nadir data browser. (2010). http://www.iup.physik.uni-bremen.de/doas/scia_data_browser.htm.

Sierk, B., Richter, A., Rozanov, A., Von Savigny, C., Schmoltner, A.M., Buchwitz, M., Bovensmann, H., & Burrows, J.P. (2006). Retrieval and monitoring of atmospheric trace gas concentrations in nadir and limb geometry using the space-borne SCIAMACHY instrument. *Environmental Monitoring and Assessment, 120*, 65-77.

Thomason, L.W., & Taha, G. (2003). SAGE III aerosol extinction measurements: Initial results. *Geophysical Research Letters, 30*, 1631.

TEMIS: Tropospheric Emission Monitoring Internet Service. (2010). http://www.temis.nl/airpollution/no2.html

van der A, R.J., Eskes, H.J., Boersma, K.F., van Noije, T.P.C., Van Roozendael, M., De Smedt, I., Peters, D., & Meijer, E.W. (2008). Trends, seasonal variability and dominant NO_x

source derived from a ten year record of NO_2 measured from space. *Journal of Geophysical Research-Atmospheres, 113*, D04302.

van der A, R. J., Eskes, H. J., Van Roozendael, M., De Smedt, I., Blond, N., Boersma, F., Weiss, A., &. van Peet, J. C. A. (2010). Algorithm Document Tropospheric NO_2, *TEM/AD1/001, issue 1.0*.

Wenig, M.O., Cede, A.M., Bucsela, E.J., Celarier, E.A., Boersma, K.F., Veefkind, J.P., Brinksma, E.J., Gleason, J.F., & Herman, J.R. (2008). Validation of OMI tropospheric NO_2 column densities using direct-Sun mode Brewer measurements at NASA Goddard Space Flight Center. *Journal of Geophysical Research-Atmospheres, 113,* D16S45.

Wilkerson, J.T., Jacobson, M.Z., Malwitz, A., Balasubramanian, S., Wayson, R., Fleming, G., Naiman, A.D., & Lele, S.K. Analysis of emission data from global commercial aviation: 2004 and 2006. *Atmospheric Chemistry and Physics, 10,* 6391-6408.

Ziereis, H., Schlager, H., Schulte, P., Kohler, I., Marquardt, R., & Feigl, C. (1999). In situ measurements of the NO_x distribution and variability over the eastern North Atlantic. *Journal of Geophysical Research-Atmospheres, 104*, 16021-16032.

Ziereis, H., Schlager, H., Schulte, P., van Velthoven, P.F.J., & Slemr, F. (2000). Distributions of NO, NO_x, and NOy in the upper troposphere and lower stratosphere between 28 degrees and 61 degrees N during POLINAT 2. *Journal of Geophysical Research-Atmospheres, 105,* 3653-3664.