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megaCITY - Zoom for the Environment

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Name of lead beneficiary for this deliverable: IUP-UB

Scientist(s) responsible for this deliverable: Michael Gauss, Andreas Richter

Contributing authors: Erika von Schneidemesser (ULeic), Michael Gauss (met.no), Andreas Richter (IUP-UB), Augustin Colette (INERIS), Paul Monks (ULeic), Alberto Maurizi (CNR-ISAC), Andreas Hilboll (IUP-UB), Øivind Hodnebrog (UiO), Frode Stordal (UiO), Evangelos Gerasopoulos (NOA), Mihalis Vrekoussis (NOA, IUP-UB), Maria Kanakidou (ECPL)

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PU	Public	X
PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

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Final project assessment of changes in air pollution in hotspot areas for the last decade derived from nested modelling, remote sensing and in-situ measurements

The due date of this deliverable was set at month 34, i.e. June 2011 or two months before the end of the CityZen project. This report summarizes the findings that have been obtained by the end of the contractual end of the CityZen project. The final report of CityZen, to be issued by the end of October 2011, will build on these findings and provide more consolidated policy messages from the project.

CityZen modelling efforts have had their focus on European emission hotspots, related to the fact that most models in CityZen are regional in scale and cover the European domain. The satellite work has been done with global coverage, including trends also for the Chinese hotspots selected in CityZen. The in-situ measurements collected and/or analysed within CityZen deal mainly with the agglomerations of Istanbul, Athens and London, although it has to be noted that the project has collected ground-based measurement data also for other metropolitan areas.

The present report is, in line with its title, structured into three different sections, addressing the findings from modelling (chapter 1), remote sensing (chapter 2) and in-situ measurements (chapter 3) performed and/or analysed within CityZen. Chapter 2 on remote sensing also includes a special section on satellite-model inter-comparisons performed within CityZen and involving both regional and global models.

1 Modelling

1.1 Coordinated trend calculations

When looking at past trends or the present state of the atmosphere, the main purpose of modelling is to understand the mechanisms behind what is observed. For instance, if models manage to reproduce trends in air pollution, they can be used to investigate the processes that have contributed to the trends. In a modelling world, emission strategies and other forcings to air pollution, such as climate change, can be assessed separately. The rationale behind the CityZen modelling studies was to use ensembles of different models covering different scales, but with the same set of emissions.

Regarding the assessment of air pollution changes in hotspot areas during the last decade by means of modelling, the coordinated multi-model study of Colette et al. (2011) has played a central role in the project. The study involved all CityZen modelling partners, covering the regional to the global spatial scales and using a set of time-varying emissions, which is commonly accepted as a good reference, namely the EMEP data base of European emissions. The paper was included as annex to deliverable report D1.2.3 (“Publication on scale errors in global model simulations based on comparison with regional/Lagrangian model results and also observations”), but the main findings are summarized here.

Colette et al. (2011) found that the trends of primary constituents were well reproduced by the models (except in some countries – owing to their sensitivity to the emission inventory). Apart from the long term trend, the modelled monthly variability was consistent with the observations but the year-to-year variability was generally underestimated.

The modelled trends are shown in Figures 1 and 2 for NO_2 and ozone, respectively. The CTMs proved to be quite successful in capturing the decreasing trend of primary pollutants, especially in the emission hotspot areas around the Benelux region. Downwards trends of NO_2 were successfully captured at 73% of the stations on average for all models. PM10 trends were also quite well captured, although the validation could not be as quantitative because of the relative lack of long term measurements. Ozone trends turned out to be much more challenging to reproduce, partly because the trends are small in magnitude during the period under consideration. Nevertheless, the models captured the trend in the majority of stations and we could discuss ozone evolution in terms of photochemical regimes. It was found that the NOx-reduction policy yields moderate increases in ozone over the Benelux hotspot of emissions. Given the photochemical regimes dominating there, more ambitious VOC reduction measures should be considered in future policies.

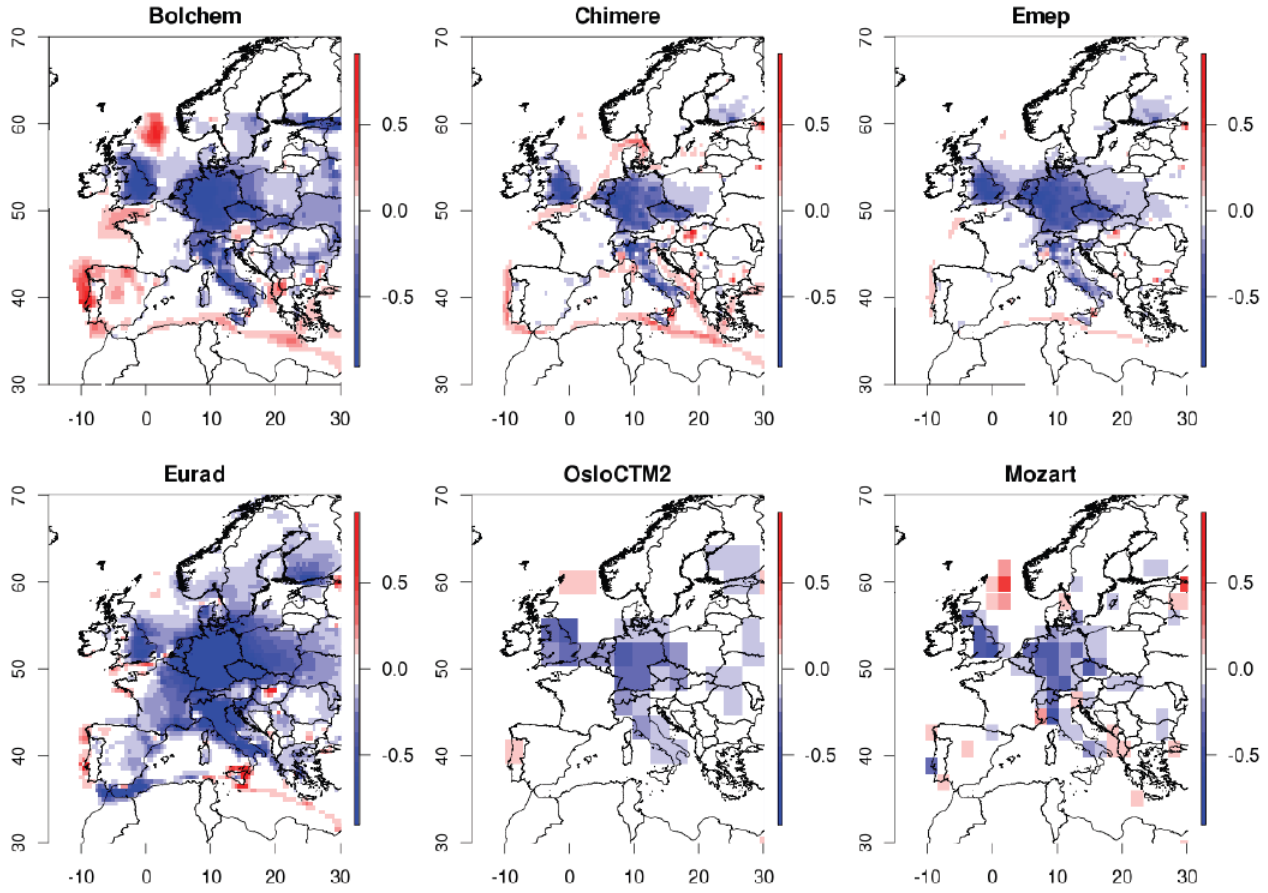

Figure 1: Modelled NO2 trend (μgm^{-3} yr^{-1}) for each CTM and at each grid point computed on the basis of monthly means of daily means over the 1998–2007 period with a linear least square fit of deseasonalised values (from Colette et al., 2011).

A comparison of simulations where anthropogenic emissions were kept constant was also made. The authors found that the magnitude of the anthropogenic NO_2 decrease exceeds the natural variability over most of Europe. This demonstrates that emission reduction strategies enforced over the past decade led to the reduction of NO_2 background levels. Consequently, this result suggests that ambitious environmental policies have a beneficial impact on NO_2 ambient concentrations, even if this effect was not as large as expected when the emission control strategies were decided (partly because of an increased proportion of diesel engines). Indeed, increasing trends in NO_2 have been registered recently in the vicinity of roads in megacity areas, like London due to changes in car technology that favour NO_2 emissions instead of NO. Such changes enhance ozone production in the source area instead of downwind. Amplification of this effect might lead to O_3 becoming an air pollution issue for the urban core, and not only the downwind location.

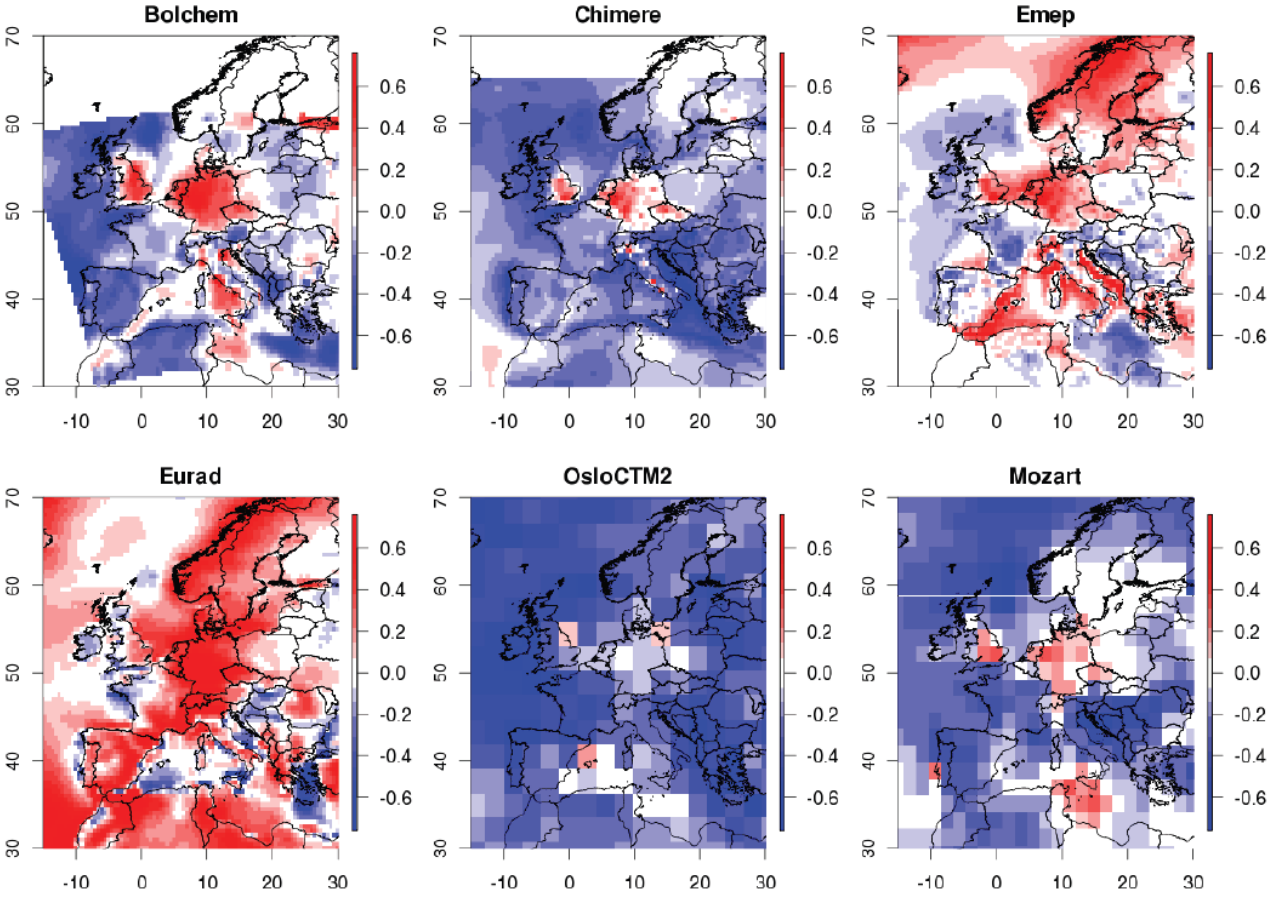

Figure 2: As Figure 1, but for ozone (from Colette et al., 2011).

To summarize, the trend assessment conducted by Colette et al. (2011) showed that reductions of anthropogenic emissions of nitrogen oxides and particulate matter effectively lead to reductions of atmospheric loading of primary constituents. However, the insufficient efforts on volatile organic compounds lead to localised increases of ozone, especially over the most urbanised areas. The model assessment proved that the models were efficient at capturing the trend of primary species but the more limited magnitude of ozone changes was more challenging to reproduce.

1.2 A special focus on the Eastern Mediterranean region

Deliverable report 1.4.3 ("Model evaluated trends of ozone, PM and deposition patterns over the recent 10 to 20 years, discrimination between anthropogenic and natural contributions") focused especially on the Eastern Mediterranean region. Several publications have been (and are being) produced by CityZen for this hotspot (as listed in deliverable report D1.1.7).

As described by Kanakidou et al. (2011) enhanced levels of air pollution and increasing trends over the last decade are observed over the East Mediterranean and over the Middle East and Cairo. Background tropospheric ozone levels in the area are high, particularly in spring and summer, depending on the meteorological conditions since they are controlled by large-scale, long-range transport and photochemical formation. Background particulate matter (PM) levels are also high due to a significant contribution of Sahara dust aerosol but also transported pollution. In the urban atmosphere due to the high levels of primary pollutants, like PM and nitrogen oxides (NOx), maintained by the anthropogenic emissions, ozone titration by reaction with nitrogen oxide (NO) is lead-

ing to very low ozone levels over city centers, whereas NOx and PM remain high. This effect is stronger in the urban core area of the Istanbul megacity than that of Athens because Athens experiences a higher regional background of O_3 due to its more central location in the East Mediterranean. Primary pollutants decrease in downwind hot spot areas where ozone and secondary aerosols build up photochemically. In the urban regions, the temporal variability of primary gaseous pollutants reflects the high emissions during winter time and the faster photochemical destruction during summer time.

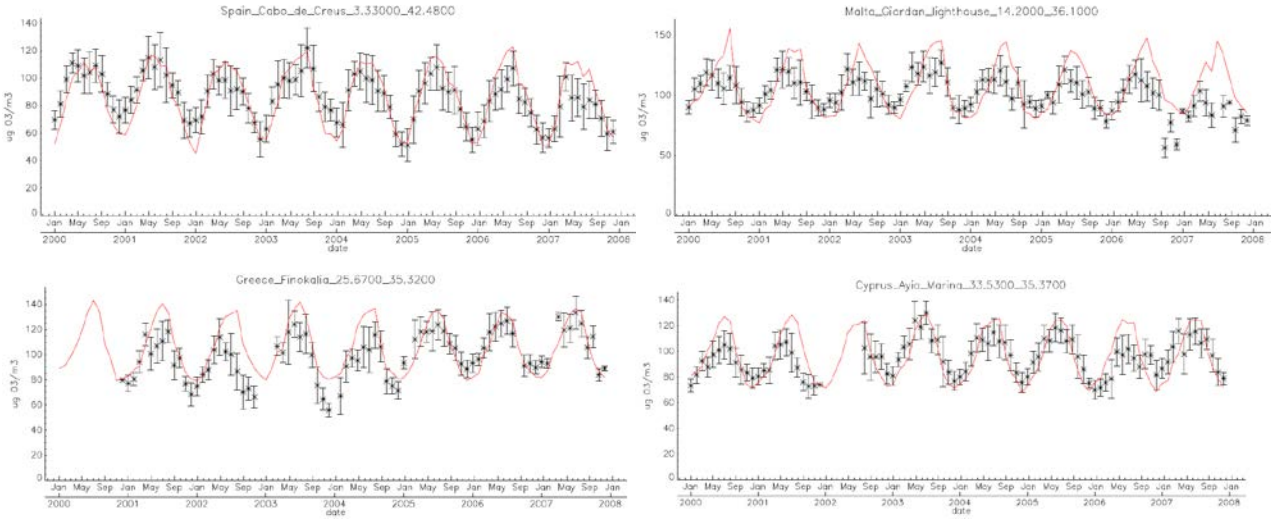

Figure 3: Simulated and observed inter-annual variability in background surface O_3 in the Mediterranean –coastal locations. Top left: Cabo de Creus, Spain; top right: Malta; bottom left: Finokalia, Greece; bottom right: Agia Marina, Cyprus (in $\mu g/m^3$) (Daskalakis et al., in preparation 2011).

TM4-ECPL global model results are compared in Figure 3 with ozone observations at EMEP stations with focus on coastal sites in the Mediterranean and its Eastern basin. The model is satisfactorily capturing the overall ozone variability in recent years and levels in the studied background stations. It also satisfactorily simulates the PM10 levels at Finokalia, Greece (CityZen deliverable report D1.4.3). Similar conclusions can be drawn for background stations in the suburbs of Athens where, however, PM10 levels are somehow underestimated partially due to dust re-suspension that is not satisfactorily represented in the global model. Indeed, based on aerosol chemical composition size-segregated simultaneous observations at urban and suburban stations in Athens and at the background site of Finokalia (Theodosi et al., 2011) the contribution of dust from local source (mainly resuspension) to local PM_{10} mass has been evaluated to be up to 33%. Further targeted simulations with different emission scenarios/meteorology and comparison to observations are being performed to analyze these observed and simulated patterns and link them to past anthropogenic emission changes due to measures taken for air quality improvement.

2 Remote sensing

2.1 Methodology

Trace Gases

Remote sensing of tropospheric constituents can be performed using the specific absorption signatures of molecules such as O_3, NO_2, or SO_2 in nadir observations of backscattered solar light in the UV and visible spectral ranges. Depending on wavelength, a significant fraction of the photons observed at the satellite have penetrated the boundary layer and therefore provide information on pollution in the lower troposphere. The first satellite observations of tropospheric NO_2 became possible with the GOME instrument in 1996, and data at better spatial resolution has been delivered by the SCIAMACHY instrument since August 2002. Even better spatial resolution is available from the OMI instrument launched in 2004, but as the Aura satellite is in an early afternoon orbit while the other two instruments measure in the early morning, the data are not fully comparable.

The basic measurement quantity of satellite observations are the spectrally resolved earth-shine radiance and the solar irradiance. From these to spectra, integrated absorber columns along the light path can be determined using absorption spectroscopy. These so called Slant Columns need first to be corrected for the stratospheric contribution, which, in the case of O_3 and NO_2, is large. The resulting Tropospheric Slant Columns are then converted to Tropospheric Vertical Columns by application of an air mass factor which accounts for the length of the light path through the troposphere. It is important to realise that in this rather indirect retrieval, a priori assumptions have to be made which have a significant impact on the results. Also, the quantity retrieved is integrated over the entire troposphere and cannot easily be separated into a boundary layer part (associated to emissions and advection) and a free tropospheric part (linked of convection, long range transport, and transformation processes).

Linking the tropospheric columns derived from satellite observations to air pollution is based on the assumption, that most of the absorber is located in the boundary layer where it is a pollutant and eventually contributes to air quality. The validity of this assumption depends on the atmospheric life time of the quantity of interest, and, in the case of NO_2, is often fulfilled, in particular in summer. However, during transport episodes, care must be taken to differentiate between signals from the surface which are relevant for air quality and those from higher altitudes, which are pollution but do not affect air quality directly. Examples are the export of SO_2 from the industrialised areas of China linked to passing frontal systems or the build-up of ozone over the Atlantic as result of biomass burning and lighting in Africa.

While satellite data provide unprecedented near global spatial coverage on a daily or weekly basis, they still have limited spatial resolution (30 x 60 km^2 in the case of SCIAMACHY) and cannot resolve localised pollution hot-spots or the details of pollution distributions within cities, and even less the effects of street canyons. Therefore, they are best used to investigate the large-scale patterns of pollution and its changes over time. Integration over larger areas is also needed to reduce the noise introduced by measurement uncertainties and the sparse spatial and temporal sampling of the observations.

The uncertainty of the observations can further be reduced by using data from only one instrument, as this excludes the effects expected for changes of spatial resolution, satellite overpass time, and instrument design associated to the switch between instruments. By analysing only relative changes, some of the assumptions needed in the retrieval cancel, avoiding some systematic error sources.

Aerosols

Aerosols in the troposphere can be retrieved from UV/vis/NIR satellite observations using their extinction properties, similar to the trace gas retrievals discussed above. However, the spectral signature of aerosols is mostly smooth and therefore differences exist between the two methods. First of all, a broad spectral coverage is needed to identify aerosol extinction and to separate it from the effects of clouds and surface spectral reflectance. In order to minimise the effect of clouds, strict cloud screening is needed which necessitates small ground-pixels in order to increase the probability for cloud free scenes. Also, good knowledge of the surface spectral reflectance is essential for quantitative aerosol retrieval.

As in the case of trace gases, only integrated aerosol properties can be retrieved, most importantly the Aerosol Optical Thickness, AOT. If the surface spectral reflectance is well known, the wavelength dependence of the AOT also provides information on the size distribution of the aerosol. These quantities can be validated by comparison with ground-based sun-photometer observations from the AERONET network. Direct comparison to size specific particle concentrations at ground-level (the quantity relevant for air quality assessments) is not possible because of the integrated nature of the measurements. However, approximations can be made on the vertical distribution of the aerosols and the depth of the mixing layer, and with these assumptions, an estimate of surface particle concentrations can be made.

For the accuracy of the AOT measurements, the most important quantity is cloud contamination. A compromise has to be found between strict cloud screening and good coverage. Also, the distinction between thick aerosol and a thin cloud is not always clear, in particular in the surrounding of clouds or in heavily polluted scenes. Too strict cloud screening can lead to a low bias in the retrieved aerosol averages. As in the case of trace gas observations, individual measurements have large uncertainties and averages over longer time periods and larger areas are needed to reduce the errors in the products.

2.2 Results

Trace Gases, NO_2

For nitrogen dioxide, the overall changes in pollution between 2003 and 2010 can be summarized from Figures 4 and 5 as follows:

1. A very large upward trend is evident over the industrialising parts of China, continuing the development already reported from the GOME-observations covering data from 1996 – 2003. This is linked to increases in NOx emissions from increased use of fossil fuels for energy production and transportation which are so large that they override the effects of significant reductions in specific emissions achieved by the introduction of cleaner technologies. Changes in biomass burning practice could also contribute to the observed changes. Interestingly, the area around Hong-Kong as well as the island of Taiwan show negative trends in NO_2 levels.
2. Large upward trends are also apparent over many large cities in India and the Middle East, but in absolute terms, the NO_2 values and their changes are small compared to China. These upward trends are expected from increasing use of fossil fuels.
3. For many large cities in developing and emerging countries, clear upward trends can be observed albeit at even lower absolute levels and limited to comparatively small areas. In the GOME time-series, this was not detected, presumably because of the much lower spatial resolution of 320 x 40 km^2. While upward trends in pollution levels are expected for these

cities, the absolute values are relatively small compared to the severity of the air quality problems in many of these areas. This apparent discrepancy is probably explained by the differences in emission sources between developing cities and China or Western Europe, where NOx from cars and power plants are the dominant emitters.

4. Over the US, a strong downward trend is observed in all polluted areas. This is in contrast to the GOME time-series where the NO_2 trend in the US was less pronounced. This trend is probably explained by the success in denoxification of power plants and a general decrease of NOx emissions from cars. No comparable downward trend is found over Canada or Mexico.
5. Over Japan, there is a continuing downward trend.
6. Over Europe, there is a downward trend but it is not as pronounced as in the GOME time-series. While NOx emissions are thought to have decreased significantly over this time period, there has been a change in NO/NOx ratio that leads to increased NO_2 at the same level of NOx emissions.

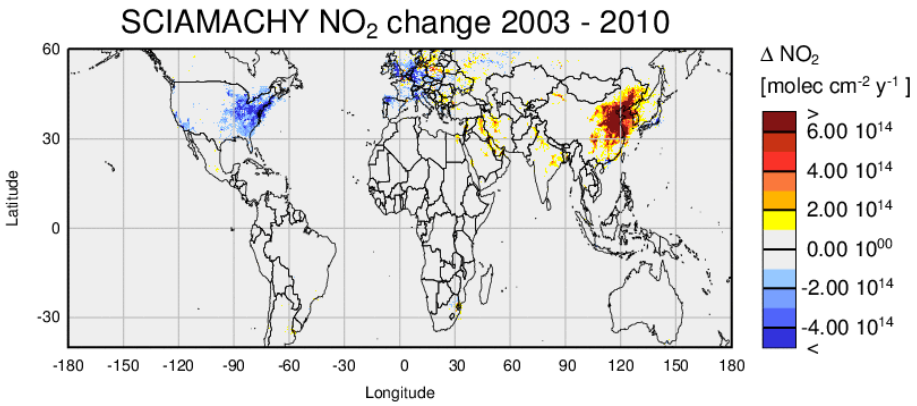

Figure 4: Annual changes in tropospheric NO_2 column as derived from the SCIAMACHY measurements from 2003 to 2010. Data have been gridded on 0.125° x 0.125° and a linear regression performed on annual averages. The resulting slope is displayed with red colours for increasing, blue colours for decreasing trends.

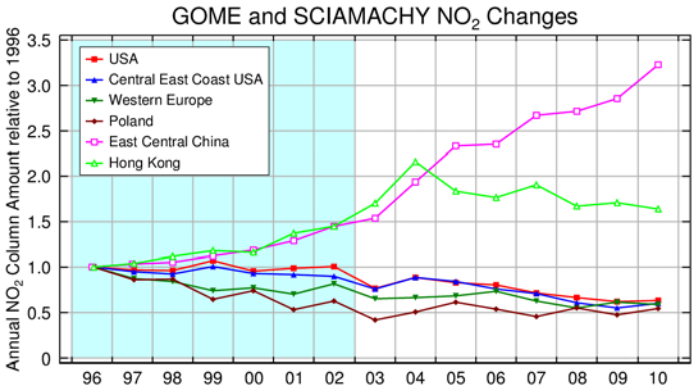

Figure 5: Changes of GOME and SCIAMACHY NO2 columns relative to 1996. This figure is an updated version of the results shown for GOME data in Richter et al., 2005 (blue shaded area). It should be noted that decreasing values are less apparent in this display as the ratio is shown.

Trace Gases, SO_2

The evolution of SO_2 over China has been particularly dramatic, and in spite of the measures taken for emission reductions before and after the Olympic Games in Beijing, only a temporary slowdown of the continuing increase was achieved in 2008 / 2009. This is linked to the dramatic increase in fossil fuel use as the Chinese economy is developing and the standard of living is increasing. As a large part of the emissions is from coal fired power plants, SO_2 is also emitted in large quantities. However, in contrast to NO_2, SO_2 is now removed from the emissions of most of the large power plants in China, leading to a significant reduction of observed SO_2 levels since 2007. As can be seen from Figure 6, SO_2 levels are still larger than in 1996 and show indication of a small increase in the last year. This is in agreement with reports that other SO_2 sources are still increasing and now have the largest share in total SO_2 emissions.

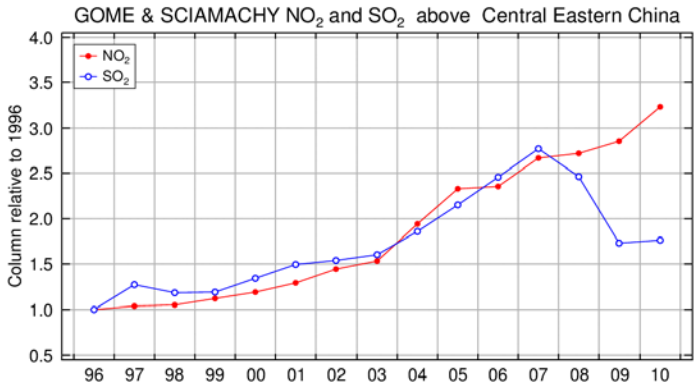

Figure 6: Evolution of annual NO_2 and SO_2 columns over Central Eastern China (30 – 40°N, 110 – 123°E) relative to the measurements from 1996. Data from GOME and SCIAMACHY have been combined.

It should be pointed out that SO_2 is not only a pollutant but also an important precursor for aerosols. Sulphuric acid aerosols have a large single scattering albedo, leading to overall stronger cooling effects. As SO_2 emissions in China have been increasing over the last decade, some of the expected warming resulting from increasing greenhouse gas emissions has been offset by the aerosol cooling. With the significant SO_2 emission reductions over the last 4 years, less scattering aerosol will be present over China and an accelerated warming can be expected.

Aerosols

Aerosol Optical Thickness trends have been derived from more than a decade of SeaWiFS data using the BAER algorithm over five CITYZEN regions. These data have been validated against ground-based AERONET data, and very good agreement was found. In some cases, discrepancies were identified and explained by cloud interference and misrepresentations of surface reflectance, and these data were excluded. The resulting satellite time agreed with the ground-based data not only in their average values, but also in the temporal evolution.

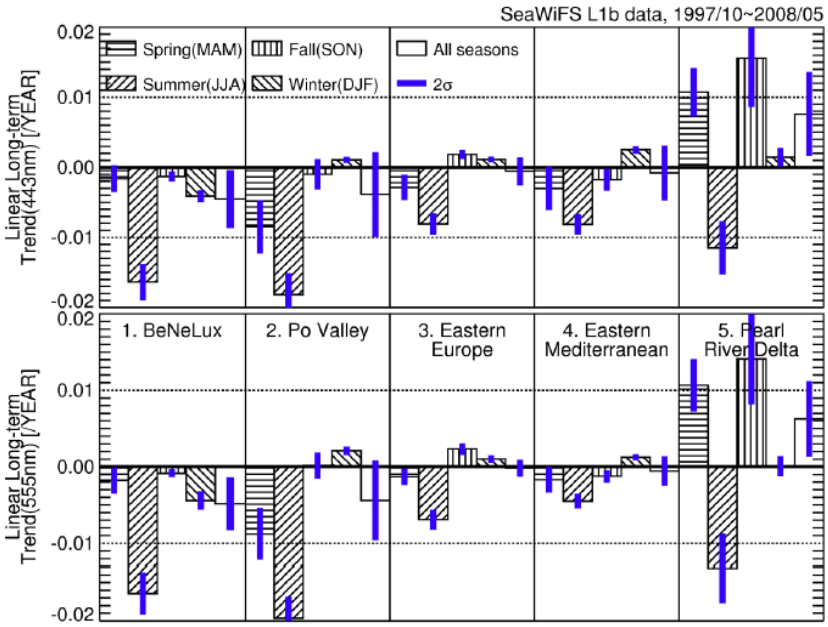

Figure 7: Linear trends in AOT at two wavelengths as derived from SeaWiFS data from October 1997 to May 2008 using the BAER algorithm. Trends are separated by season and shown for 5 regions. The upper panel is for AOT at 443 nm, the lower at 555nm. (Figure from Yoon et al., 2011.)

As shown in Figure 7, downward trends in AOT are found over the BeNeLux area, the Po Valley, and to lesser degree also over Eastern Europe. In contrast, an upward trend in AOT is apparent over the Pearl River Delta. These tendencies in AOT are in agreement with expectations and are explained by decreasing anthropogenic emissions in Europe, in particular in the highly industrialised BeNeLux and Po Valley regions and the rapid economic development in China which leads to large increases in aerosol levels.

Closer inspection of the satellite data revealed problems with cloud contamination in some observations. In order to investigate this, seasonal trends were also computed (see Figure 7) and compared. A surprisingly large variability in aerosol trends is found over Western Europe, probably related to a mixture of seasonal differences in anthropogenic emissions of aerosols and aerosol precursors (e.g. VOCs which undergo rapid photochemistry in summer, leading to the formation of SOA) and retrieval problems in the cloudy seasons. In the Pearl River Delta, the sign of the trend found for AOT in summer (the cloudy season) is negative while it is positive in all other seasons. This result is not yet fully understood but is probably linked to cloud interference and changes in cloud frequencies and distribution.

As pointed out in the section on SO_2 trends, there is indication for a significant reduction in SO_2, and in consequence, a reduction in aerosol AOT is expected. This effect could not yet be identified in the aerosol data as the time period analysed ends in 2008, just when the changes started to show effect. Also, the trend analysis performed is linear and not well suited to detect such non-linear changes. As soon as a longer data set is available, this question will be investigated in more detail.

2.3 A special focus on satellite-model intercomparisons

The aim of the satellite-model inter-comparison study for CityZen was to evaluate the consistency of CityZen models and satellite data for tropospheric NO_2 distribution, seasonality and trends.

The following models have participated:

- OsloCTM2 (global)
- CHIMERE (regional, Europe)
- BOLCHEM (regional, Europe)

The model data were interpolated to satellite time of overpass and integrated to the tropopause or top of model atmosphere. In order to be comparable to the satellite data, the reference region over the Pacific (180 – 210° longitude) was also subtracted from the modelled fields. Re-gridding was made to a uniform lat-long grid for BOLCHEM**.** GOME and SCIAMACHY IUP Bremen NO_2 tropospheric columns, re-gridded to model-resolution, were used as satellite data. Figure 8 shows SCIAMACHY and OsloCTM2 data for tropospheric NO_2 columns in 2007.

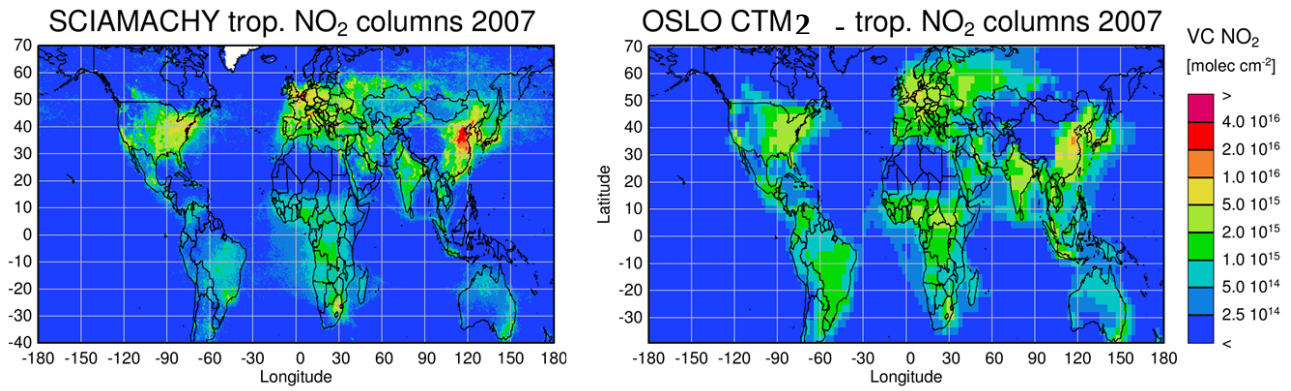

Figure 8: Tropospheric columns of NO_2 observed by SCIAMACHY (left) and modeled by OsloCTM2 (right). Units: molecules(NO_2)/cm^2.

We see a good overall agreement of patterns, although the peak values are not fully reproduced by the model, in particular over China. However, we do find a reasonable correlation when the data are gridded to the same resolution. The trend estimates over Central East-China shown in Figure 9 show reasonable agreement although the model underestimates also the trend, which is consistent with Figure 8.

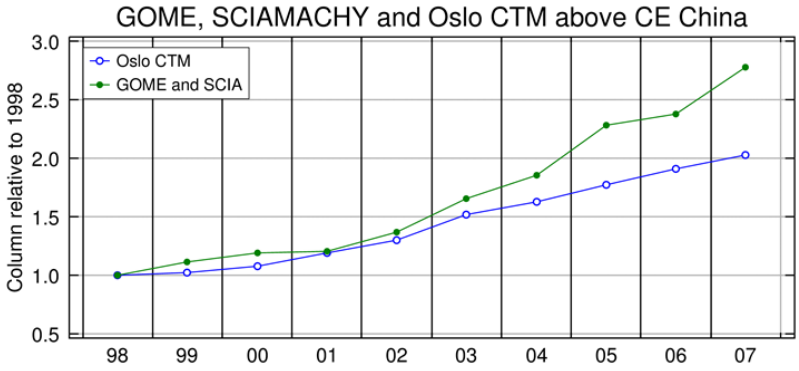

Figure 9: Trends of tropospheric NO_2 columns over Central East-China relative to 1998, observed by SCIAMACHY (green line) and modelled by OsloCTM2 (blue line).

Figure 10 shows European maps of tropospheric NO_2 columns for summer 2006, also including the regional models CHIMERE and BOLCHEM. Summer values over Europe are in good agreement with a good representation of the spatial distribution in the models, but possibly too large NOx values along ship tracks. Also, BOLCHEM underestimates NO_2 over Eastern Europe. The Oslo CTM2 underestimation over hot spot regions is partly due to the relatively coarse horizontal resolution (T42). This could also lead to an underestimation of plume processes as the NOx emissions are smeared out over a larger area in the model.

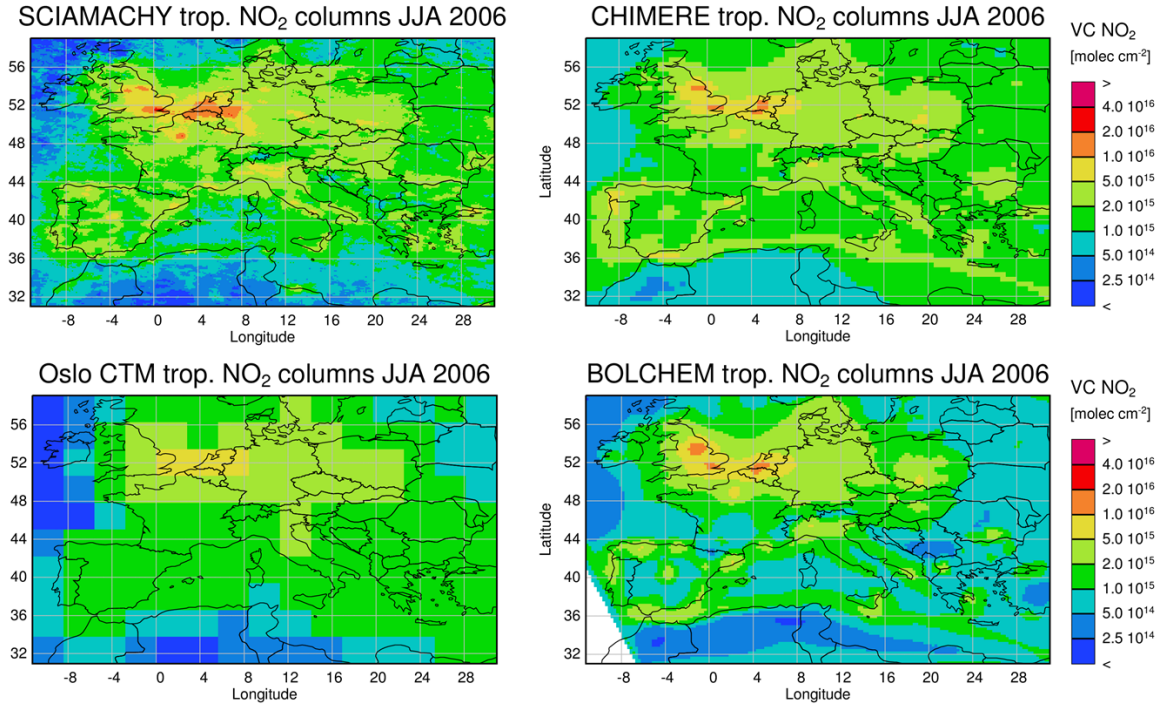

Figure 10: Tropospheric NO2 columns in summer 2006 as observed by SCIAMACHY (top left) and the three models involved in this inter-comparison. Units: molecules(NO_2)/cm^2.

Figure 11 shows trends from 1998 to 2007 of NO_2 vertical columns over Central Europe, for satellite data and for model data covering the GOME and SCIAMACHY periods. Satellite values show large variability in winter, probably due to sampling problems in the presence of persistent clouds and possibly also snow contamination. Oslo CTM gives a good representation of NO_2 over Central

Europe but underestimates winter values; the same is true for the CHIMERE model. BOLCHEM gives a very good representation of NO_2 over Central Europe in summer and winter. It also reflects some of the large variability in winter values.

Trends for summer Central Europe are shown in Figure 12. Summer NO_2 values over Central Europe from satellites and models agree well. The downward trend is clearly more pronounced in models than in satellite data. This finding is similar to what Colette et al. (2011) found, but here for columns, not surface concentrations. This could be related to uncertainties in NOx emission inventories regarding Diesel engines which might have significantly larger emissions in everyday use than expected from standard tests. As many countries in Europe have introduced incentives for cars with Diesel engines, this can potentially lead to an increasing mismatch between emission inventories and real NOx emissions.

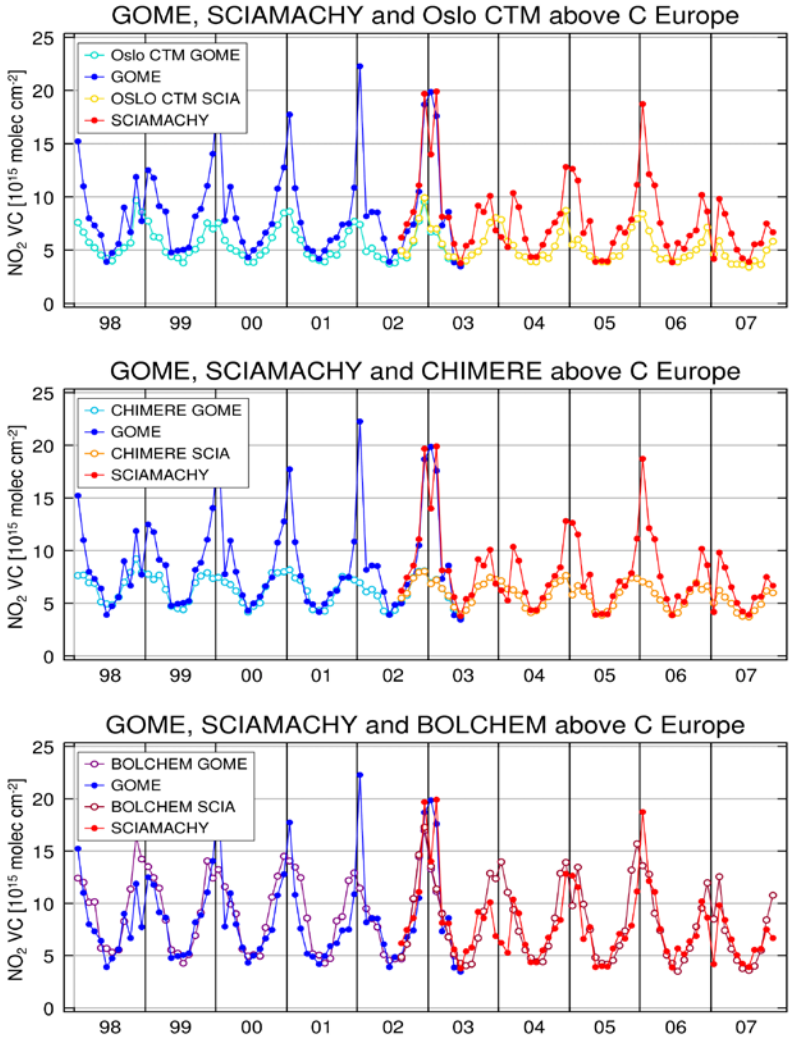

Figure 11: Observed and modelled NO_2 vertical columns above Central Europe (defined here as the region 45°N–55°N, 2°E–15°W). Top: OsloCTM2, middle: CHEMERE, bottom: BOLCHEM. Units: 10^{15} molecules(NO_2)/cm^2.

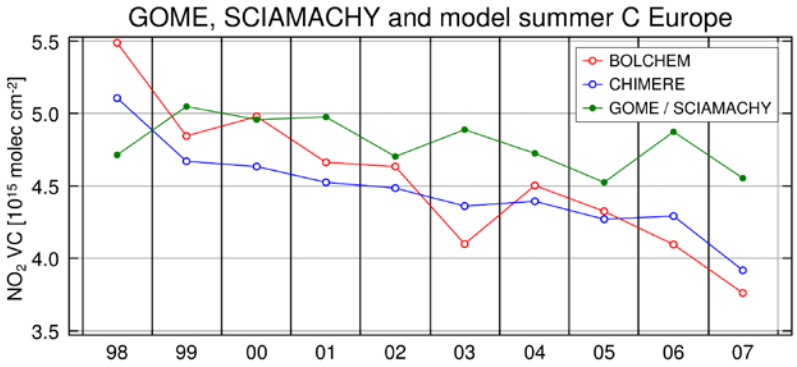

Figure 12: Trends for NO_2 vertical columns in summer over Central Europe (defined here as the region 45°N–55°N, 2°E–15°W), observed by GOME/SCIAMACHY (green line) and modeled by BOLCHEM (red line) and CHIMERE (blue line). Units: 10^{15} molecules(NO_2)/cm^2.

In summary, the comparison of models and satellite NO_2 columns has shown, that the models represent the NO_2 distribution in recent years reasonably well, that there are some issues with the seasonality of NOx and that the modelled trends in NO_2 (which are to a large degree determined by NOx emissions) are in reasonably good agreement with observed trends.

3 In-situ measurements

CityZen has collected in-situ measurements from many sources and for many areas. Detailed descriptions have been given for the Po Valley in deliverable report D1.6.1 (”Data compilation for the Po Valley”), for the Eastern Mediterranean hotspot in deliverable reports D1.4.1 (“First observational data compilation for the East-Mediterranean area”) and D1.4.2 (“New targeted observations in the East-Mediterranean area”), for the Rhine-Ruhr area in deliverable report D1.5.1 (“Data compilation for the Benelux/Ruhr area”), and for the Pearl River Delta (China) in deliverable report D1.7.1 (“Data compilation for the Pearl River Delta (PRD)”).

One of the milestones of CityZen was a database, which was originally intended to focus on the summer of 2003, but now deals with a larger data set hosted at NILU. A report was written as milestone document M2.2.1 available to the CityZen consortium and Commission services on the CityZen wiki space.

In the analysis of in-situ measurements, CityZen has concentrated mainly on Athens, Istanbul and London. In particular, for the Greater Athens Area (GAA), the observations since 1985 from fourteen monitoring stations of the Greek Ministry of Environment air pollution monitoring network have been collected and categorized into four different groups namely urban traffic (Urb-Trf), urban-Background (Urb-Bg), suburban industrial (Sub-ind) and suburban background (Sub-BG). For the Greater Istanbul Area (GIA) the observations of ten stationary monitoring stations operating since 1998 have been categorized to up to three subgroups (urban traffic data, suburban industrial and suburban background) depending on the availability of the observations. The findings can be summarized as follows (see Figure 13):

NO_2: Nitrogen dioxide over Athens shows a decreasing trend for the urban traffic regions. The highest decline in NO_2 levels is found for the period 1985-1994. For the present period (2003-2008) this decline is almost negligible. The other three sub-categories encountered an overall increase in NO_2 levels from 1985-1994 and a decrease of about 1 μg m^{-3} NO_2 per year for the years 1995-2008.

Contrary, for GIA an important increase of 8 μg m^{-3} NO_2 per year is recorded for the suburban background areas during 2003 to 2008.

NO: The mean annual values of nitrogen oxide over Athens indicate a significant decrease on their levels from 1985 to 2008. Especially for the urban traffic regions a reduction by 42% has been observed during this period; the highest reduction in μg m^{-3} occurred from 1994-2002. In Istanbul the NO measured at the suburban traffic areas decreased significantly (≈60%).

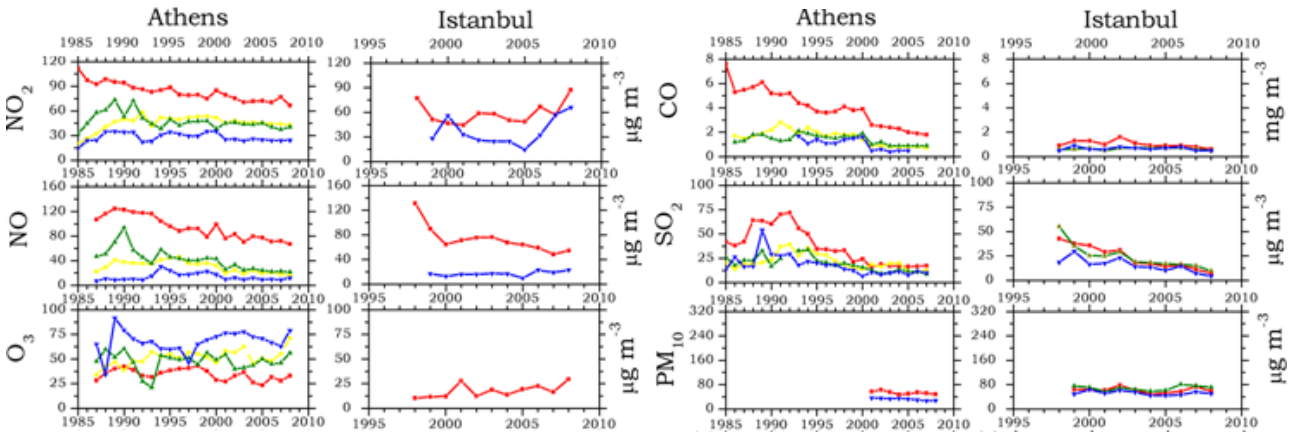

Figure 13: Surface concentrations changes of air pollutants as recorded in Athens and Istanbul municipalities' stations.

O_3: Ozone trend is less profound than the nitrogen oxides' one. The highest values are observed for the suburban background regions and the lowest for the urban traffic ones due to the titration with the NOx species. On average ozone remained constant throughout the last 25 years except for the urban background areas where an important increase of about 35 μg m^{-3} has been recorded. A more straightforward behaviour of the ozone monitored at the urban traffic areas of Istanbul showing an overall increase of 2 μg $m^{-3}y^{-1}$.

Since 1998, background O_3 levels have been recorded in the East Mediterranean at Finokalia station on the island of Crete (Figure 14; see also Figure 3 for comparisons with model results for 2001-2008). This 13-year record shows an O_3 decline by 5.6%/yr during the first 5-years from 1998 to 2002 (Gerasopoulos et al., 2005), succeeded by an abrupt increase in 2003 and no clear trend afterward since it is marked by the heat waves of 2003 and 2007 and the intensive fires in Greece in 2007 and 2009 that followed the large fires in 2000. Transport from the European continent was identified as the main mechanism that controls ozone levels in the eastern Mediterranean in agreement with modeling analysis (see also deliverables 1.4.3 and 1.4.4). Global model simulations for the year 2008 indicate that on annual mean basis, about 25% of the ozone levels at Finokalia can be attributed to the East Mediterranean anthropogenic, biogenic and biomass burning emissions, the remaining is maintained by mid and long range transport to the area from upwind locations. East Mediterranean anthropogenic emissions alone contribute by about 15% with their impact maximizing (~20%) in summer (paper in preparation). The results show also significant involvement of biogenic emissions in ozone formation during the warm months of the year. These results are in agreement with Im et al. (2011) mesoscale study for summer 2004 regional anthropogenic emissions suppress ozone in Athens and Istanbul whereas they increase the regional summertime ozone by about 19%.

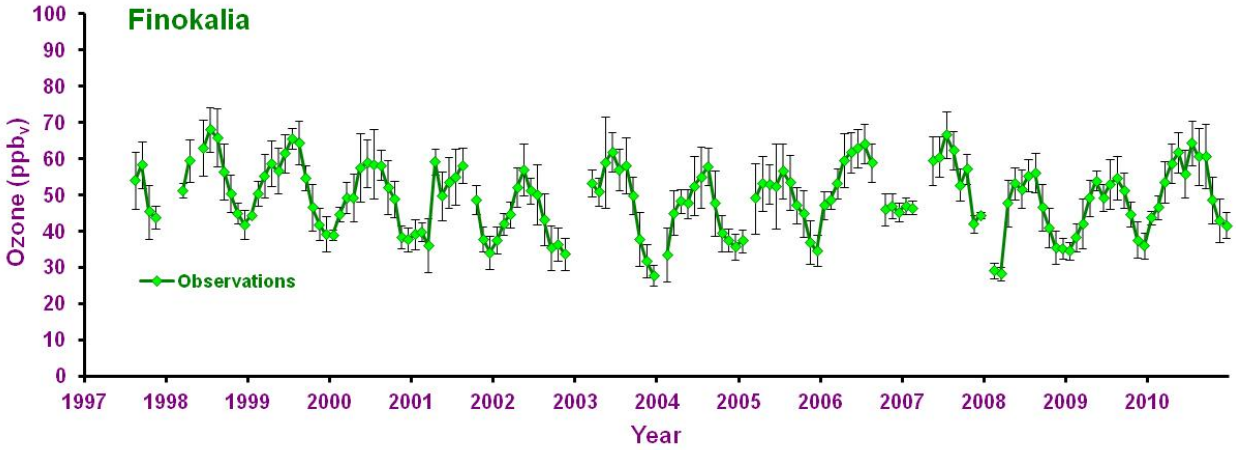

Figure 14. Surface ozone record at Finokalia station, Crete, Greece in the middle of the East Mediterranean since 1997 (Mihalopoulos et al., unpublished data).

CO: Carbon monoxide levels are constantly dropping for the urban traffic areas of Athens presenting a decrease of 65% during the last 25 years. A smaller scale decrease is observed for the other three categories. For GIA, CO presented an increasing trend until 2003 and a decreasing one until present time. For both areas the CO levels of the urban traffic regions are greater than the ones of the other categories.

SO_2: For the GAA the increase of the sulfur dioxide levels during 1998-1992 in almost all groups of interest has been followed by a drastic decrease mainly during the decade 1993-2002. These significant improvements reflect emission changes by the 1990-1994 replacement of old cars and reallocation of large industries out of the Athens and use of filters. After 2002, the SO_2 levels have been stabilized at about 15 μg m^{-3}. Although the observational record is not going back enough in time to fully register air quality changes due to the wide use of liquefied petroleum gas for traffic starting 1998, a continuous decrease in SO_2 levels has been recorded also for the GIA from 1998 to 2008. Especially for the industrialized areas this decrease is drastic and equals to about 90%.

Gerasopoulos et al. (2011), studied air pollution changes in the greater Athens region. Three years (2006–2008) of ground-based observations of the Aerosol Optical Depth (AOD) in the urban environment of Athens, in the Eastern Mediterranean, were analysed in their work. Average values were derived and a source attribution was made. It was found that the annual variability of AOD has a spring maximum dominated by coarse dust particles from the Sahara (AOD 0.34–0.42), while the diurnal pattern is typical for urban sites, with AOD steadily increasing throughout the day. The greatest contribution to the annually averaged AOD, accounting for almost 40%, comes from regional and local sources (namely the Istanbul metropolitan area, the extended areas of biomass burning around the north coast of the Black Sea, power plants spread throughout the Balkans and the industrial area in the Po valley, with average daily AOD in the range of 0.25–0.35). As an additional important contribution (23%) dust from Africa has been identified, whereas the rest of Europe contributes another 22%. The geographical distribution of the above sources in conjunction with the prevailing synoptic situation and contribution of local sources, lead to mixed types of aerosols over Athens, with highly variable contribution of fine and coarse particles to AOD in the range 10%–90%. The data presented by Gerasopoulos et al. (2011) represent the first long-term, ground based data set available for Athens, and it has also been used for the validation of satellite derived AOD by MODIS, showing good agreement on an annual basis, but with an overestimation of satellite AODs in the warm period.

The remainder of this section is devoted to a case study for London, which, in CityZen was considered as part of the 'greater BeNeLux' focus area, as the regions are closely connected through transboundary transport of air pollution.

In situ measurements for London

Erika von Schneidemesser, Paul S. Monks (ULeic)

Methodology

Data (NO_2, NO, CO, O_3, and speciated VOCs) were retrieved from the UK Air Quality Archive website (http://www.airquality.co.uk/). Data processing for all species was done using R and openair (Carslaw and Ropkins, 2009; Team, 2008). The time period considered was from January 1, 1998 to December 31, 2009. Speciated VOCs were only available for Marylebone Road (urban roadside site) and Eltham (suburban background site).

Data distributions were assessed and determined to be log-normal using q-q plots. Therefore, trends were calculated by fitting a simple linear fit to the natural log of the data. The slope was then able to be interpreted as a percent decrease per year. Log-normalized data trends were assessed for all data using the Mann Kendall function in openair. Ninety-fifth percentile confidence intervals and statistical significance were determined for all trends. All trend analysis required 50% of possible hourly data each month to be present in order for a trend to be calculated; all trends, except those of VOCs, were deseasonalized.

The VOCs measured included only non-oxygenated C_2-C_8 hydrocarbons. In order to scale up the measured VOCs to calculate an estimated sum of total VOCs, emission inventory data was used from the UK National Atmospheric Emission Inventory of non-methane VOCs. The compounds for which data was available from the monitoring sites that overlapped with speciated emission inventory data were summed and a ratio determined relative to the total VOC emissions for each year. This was then applied to the measured data to estimate total VOCs.

Detailed information on data coverage is listed in Table 1 for all sites. Only statistically significant trend values are discussed, unless specifically mentioned otherwise. Many of the data sets end as of October, 2007, which is approximately 10 years. Years of coverage will not be mentioned unless data sets are shorter than this ~10 year period. VOC data for London Eltham were available only from 2003 onwards.

Detailed information on the UK Automatic Urban and Rural Network monitoring sites can be found at http://www.bv-aurnsiteinfo.co.uk/default.asp. For this study NO_2, NO, CO, and O_3 data were included from 17 urban roadside, urban center, suburban, and urban background sites in Greater London. Data were not available for all species at all sites. In addition, ozone data from 20 sites in southeast England were also included. The majority of these sites were rural and urban background sites. All basic site information can be found in Table 1.

Table 1. UK Site Information (end dates are not inclusive).

Site (London Site Code)	Classification	Species and Data Coverage					Latitude	Longitude
		NO2	NO	O3	CO	VOCs		
A3 Roadside, London (A3)	roadside	1998 – 10/2007	1998 – 10/2007	n.m.	1998 – 10/2007	n.m.	51.37348	-0.29186
Bexley, London (BX)	suburban	1998 – 2010	1998 – 2010	1998 – 10/2007	1998 – 2010	n.m.	51.46603	0.1848
Bloomsbury, London (BB)	urban center	1998 – 2010	1998 – 2010	1998 – 2010	1998 – 2010	n.m.	51.52229	-0.12589
Bournemouth	urban background	-	-	2/2003 – 2010	-	-	50.739576	-1.826749
Brent, London (BR)	urban background	1998 – 10/2007	1998 – 10/2007	1998 – 10/2007	1998 – 10/2007	n.m.	51.58963	-0.27551
Bristol St. Paul's	urban background	-	-	6/2006 – 2010	-	-	51.462828	-2.584542
Bromley, London	roadside	1998 –	1998 –	n.m.	1998 –	n.m.	51.40554	-0.01887

(BL)		10/2007	10/2007		10/2007			
Coventry Memorial Park	urban background	-	-	2/2001 – 2010	-	-	52.394327	-1.519591
Cromwell Road, London (CR)	roadside	1998 – 2008	1998 – 2010	n.m.	1998 – 2010	n.m.	51.49546	-0.17868
Eltham, London (LE)	suburban	1998 – 2010	1998 – 2010	1998 – 2010	n.m.	10/2003 – 2010	51.45257	-0.07078
Hillingdon, London (HL)	suburban	1998 – 2008	1998 – 2010	1998 – 2010	1998 – 10/2007	n.m.	51.49633	-0.46087
Hackney, London (HK)	urban center	1998 – 10/2007	1998 – 10/2007	1998 – 10/2007	1998 – 10/2007	n.m.	51.55877	-0.0566
Harwell	rural	1998 – 2010	1998 – 2010	1998 – 2010	n.m.	1998 – 2010[1]	51.570967	-1.325121
Leamington Spa	urban background	-	-	1998 – 2010	-	-	52.288807	-1.533134
Leominster	suburban	-	-	7/2005 – 2010	-	-	52.221704	-2.736834
Lewisham, London (LH)	urban center	1998 – 10/2007	1998 – 10/2007	n.m.	n.m.	n.m.	51.44541	-0.02015
Lullington Heath	rural	-	-	1998 – 2010	-	-	50.793698	0.1812412
Market Harborough	rural	-	-	12/2003 – 2010	-	-	52.554553	-0.772284
Marylebone Road, London (MR)	kerbside	1998 – 2010	1998 – 2010	1998 – 2010	1998 – 2010	1998 – 2010	51.52253	-0.15461
North Kensington, London (NK)	urban background	1998 – 2010	1998 – 2010	1998 – 2010	1998 – 2010	n.m.	51.52106	-0.21344
Northampton	urban background			3/2003 – 2010			52.27349	-0.885938
Portsmouth	urban background	-	-	4/2003 – 2010	-	-	50.828802	-1.068584
Reading New Town	urban background	-	-	10/2003 – 2010	-	-	51.45309	-0.944080
Rochester Stoke	rural	-	-	1998 – 2010	-	-	51.456167	0.6348761
Sibton	rural/remote	-	-	1998 – 2010	-	-	52.294385	1.463499
Southend-on-Sea	urban background	-	-	7/2000 – 2010	-	-	51.544149	0.67831
Southampton Centre	urban centre	-	-	1998 – 2010	-	-	50.908138	-1.395785
Southwark, London (SW)	urban center	1998 – 10/2007	1998 – 10/2007	1998 – 10/2007	1998 – 10/2007	n.m.	51.49055	-0.09668
St. Osyth	rural	-	-	5/2002 – 2010	-	-	51.777846	1.0489468
Sutton, London (ST)	suburban	1998 – 5/2002	1998 – 5/2002	n.m.	n.m.	n.m.	51.36789	-0.1655
Teddington, London (TD)	urban background	1998 – 2010	1998 – 2010	n.m.	n.m.	n.m.	51.42099	-0.33966
Thurrock	urban background	-	-	1998 – 2010	-	-	51.47707	0.317956
Wandsworth, London (WW)	urban center	1998 – 10/2007	1998 – 10/2007	n.m.	n.m.	n.m.	51.45696	-0.19117
Westminster, London (WM)	urban background	07/2001 – 2010	07/2001 – 2010	07/2001 – 2010	07/2001 – 2010	n.m.	51.49466	-0.13194
Weybourne	rural	-	-	5/2001 – 2010	-	-	52.950481	1.1220111
Wicken Fen	rural	-	-	1998 – 2010	-	-	52.298494	0.2909191
Yarner Wood	rural	-	-	1998 – 2010	-	-	50.597594	-3.716521

[1]from 01/2002 to 05/2007 only a limited number of aromatic VOCs were measured.

Results

The geographical distribution of sites across Greater London is shown in Figure 15. The change in concentration of total scaled-up VOCs for the two sites with VOC data are shown in Figure 16. A comparison of all NO_2, NO, CO, and O_3 data for the 17 sites in greater London is shown in Figure 17.

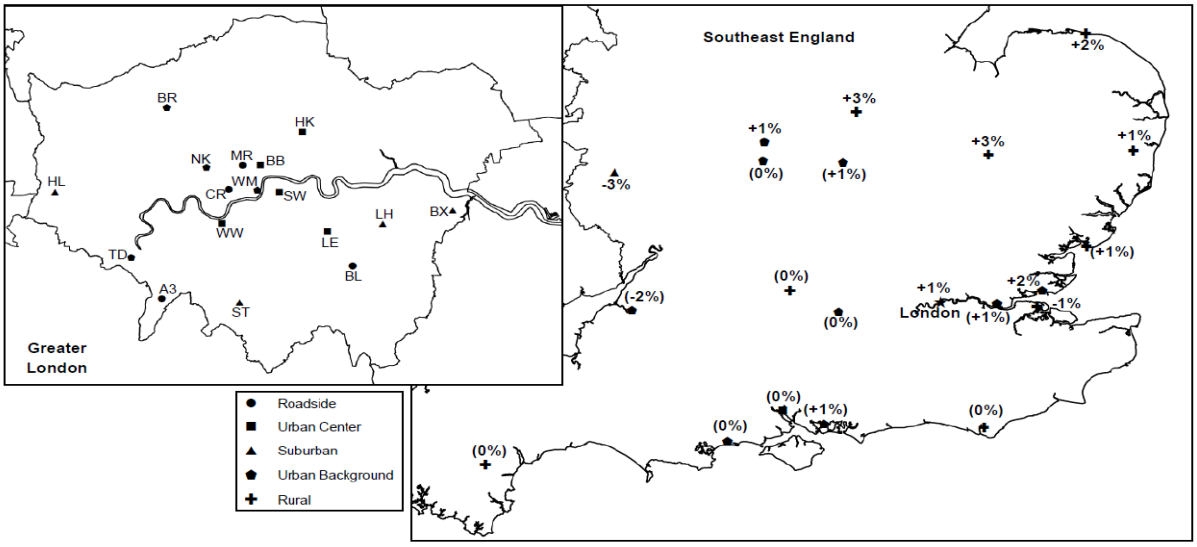

Figure 15: The box on the left shows the greater London area and the monitoring sites therein. The two letter codes correspond to the sites listed in Table 1. The shape of the site point designates the classification of the site. The boundary shown designates greater London which encompasses the City of London and the 32 surrounding London boroughs. The larger box on the right shows all of southeast England and the mainly urban background and rural sites surrounding the London area. The values given represent ozone trends observed at those sites, with statistically insignificant trends shown in parentheses. The London trend shown is an average of all ozone trends within London.

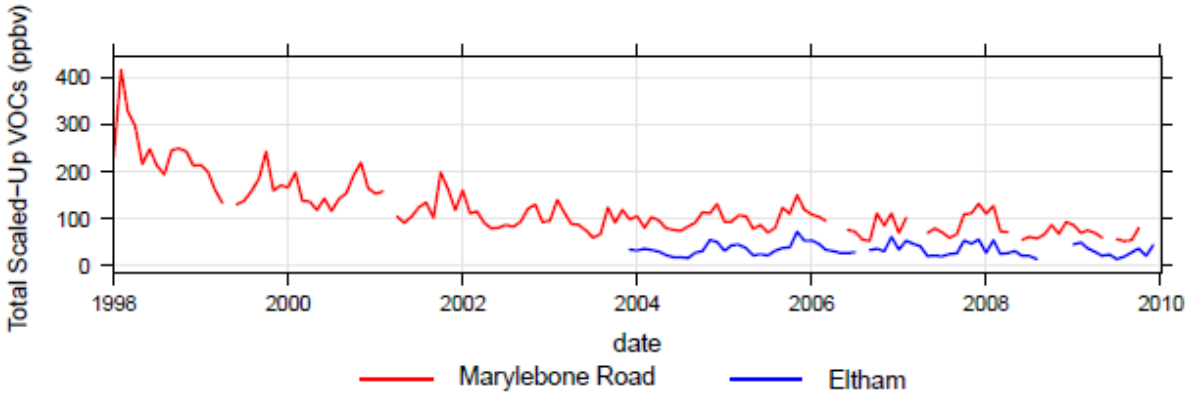

Figure 16: Monthly mean total VOC concentrations for Marylebone Road (urban roadside) and Eltham (suburban background). The concentrations are 'scaled up' so that the total amount of VOCs is represented, and not limited to those 26 non-methane hydrocarbons monitored. Scaling up was done using NAEI data (see Methodology for explanation).

Among all of the London sites, the observed trends in NO_2 were decreasing at all sites, with the exception of the urban roadside site at Marylebone Road (see Table 2). Nitrogen dioxide annual average (geometric mean) concentrations ranged from 24 ug m^{-3} to 85 ug m^{-3} in 1998 and from 15 ug m^{-3} to 94 ug m^{-3} in 2009 for Greater London. For both years cited, the lowest concentration was from Teddington (an urban background site), and the highest concentration was from Marylebone Road. The NO_2 decreases ranged from -5.1% per year to -1.0% per year. Overall, if considered as one continuous 12 year period, the NO_2 concentration increased by +2.0% per year at Marylebone Road. However, a noticeable change in the trend took place in early 2003, which corresponds to the

implementation of the congestion charging scheme and other traffic congestion and emissions reduction measures. The additional measures implemented, which were not limited to the congestion charging zone but incorporated all of Greater London, included additional bus lanes, the use of larger buses, particle traps on diesel buses, increased bus frequency, and changes to traffic light phases (Atkinson et al., 2009).

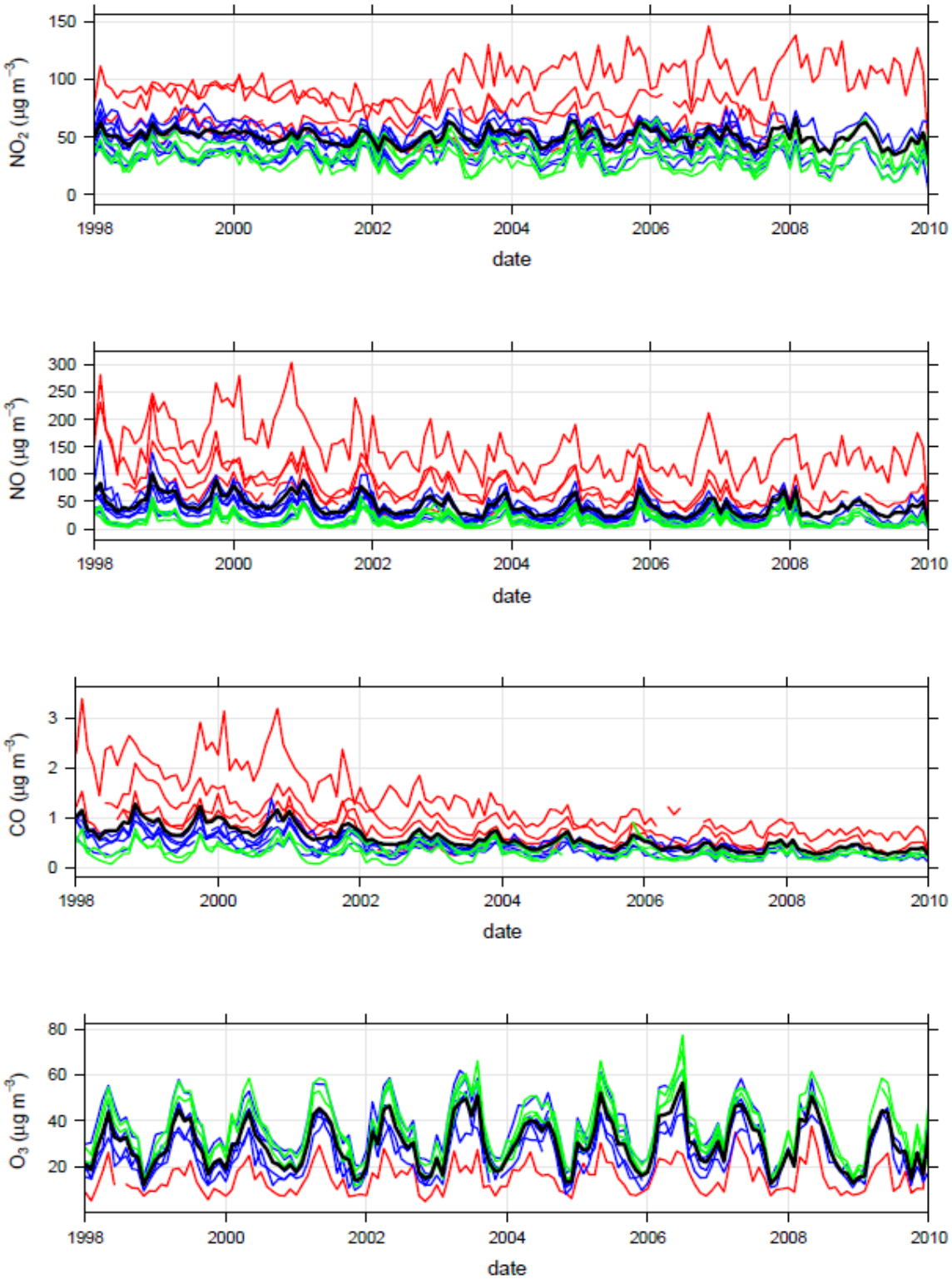

Figure 17: Monthly average concentrations of nitrogen dioxide, nitrogen oxide, carbon monoxide, and ozone for the 17 sites in Greater London. Not all 17 sites included all four species. Red lines are urban roadside sites, blue lines are urban center and suburban sites, green lines are urban background sites, and the thick black line shows the average of all London data.

If analyzed as two separate time periods (before 2003 and after 2003), the trend is very different. For the period from 1998 to 2003 NO_2 at Marylebone Road was decreasing at a rate of -4.1% per year (99.9% significance level), whereas from 2003 to 2010 a 0% per year trend in NO_2 was observed, but the trend was not significant. This would indicate that while the implemented changes may have positively influenced (reduced emissions) other species such as particulate matter, the impact on NO_2 actually reversed a decreasing trend at Marylebone Road. These results are in agreement with a study quantifying the changes due to the congestion charging scheme for the two years before and after its implementation, which observed increases in NO_2 and O_3 after implementation from monitors within the Congestion Charging Zone (CCZ) (Atkinson et al., 2009). The Marylebone Road monitoring station is on the boundary of the CCZ. No other sites have evidence of such a trend reversal.

Nitrogen oxide data showed decreasing trends at all sites in Greater London, which ranged from -20% per year (Bromley, roadside) to -3.0% per year (Bexley, suburban and Westminster, urban background). The largest per year decreases were observed among the roadside sites (see Table 1). 1998 concentrations ranged from 52 to 140 ug m^{-3} at roadside sites and decreased to 13 to 73 ug m^{-3} in 2007. The lowest concentrations were measured at the urban background site Teddington, where concentrations remained relatively steady at 3.7 ug m^{-3} in 1998 and 4.1 ug m^{-3} in 2007; the -1.0% per year trend was not significant.

Carbon monoxide data were available for 12 sites. Similar to NO, CO was decreasing at all sites, at rates ranging from -20% per year (Bromley, roadside) to -1.0% per year (North Kensington, urban background), with the largest decreases also among the roadside sites. Concentrations in 1998 and 2007 ranged from 0.24 ug m^{-3} at Brent (urban background) to 1.8 ug m^{-3} at Marylebone Road, and 0.14 ug m^{-3} at Hackney (urban center) to 0.67 ug m^{-3} at Marylebone Road, respectively.

Volatile organic compound measurements were available only for Marylebone Road and Eltham. As the measurements were limited to 26 non-oxygenated non-methane hydrocarbon species, yearly UK emission inventory data were used to scale-up to total VOCs, so as not to underestimate their role in ozone formation. Data from Marylebone Road showed decreases of -11% per year at 99.9% significance. The trend at Eltham (for the shorter time period) was not statistically significant. The VOC trends for Marylebone Road showed the most similarity to the CO trend.

Table 2. London site trends for NO_2, NO, CO, and O_3 from 1998 through 2009. 'nd' indicates no data was available; values in parentheses were not significant, all other values were significant to at least the 90th percentile; *indicates less than 10 years of data.

	Site	NO_2	NO	CO	O_3
Roadside	A3	(0%)	-12%	-11%	nd
	BL	-5.1%	-20%	-20%	nd
	CR	-2.0%	-7.3%	-14%	nd
	MR	+2.0%	-6.2%	-12%	+2.0%
Urban Center	BB	(-1.0%)	-7.3%	-9.4%	+3.0%
	HK	-3.0%	-8.3%	-13%	(+1.0%)
	LH	(-1.0%)	-8.3%	nd	nd
	SW	-3.0%	-9.4%	-5.1%	(+1.0%)
	WW	(0%)	-6.2%	nd	nd
Suburban	BX	-1.0%	-3.0%	-7.3%	+2.0%
	HL	-2.0%	-6.2%	-4.1%	+2.0%
	LE	-3.0%	-6.2%	nd	+1.0%
	ST	(0%)*	-8.3%*	nd	nd
Urban Background	BR	-3.0%	(-1.0%)	-4.1%	+3.9%
	NK	-3.0%	-5.1%	-1.0%	+3.0%
	TD	-3.0%	(-1.0%)	nd	nd
	WM	-2.0%*	-3.0%*	-13%*	+3.9%*

Ozone trends within London (for which data was available from 9 sites) showed increasing trends ranging from +2.0% per year to +3.9% per year (see Table 2). The highest increases in London were among the urban background sites. The overall average trend for all London ozone data was +1% per year. These results agree with recent findings from Bigi and Harrison (2010) who found that ozone showed a steady increase from 1996-2008 at North Kensington in London, unfortunately the trend was not quantified, so the rate of increase could not be compared. Ozone trends for sites surrounding London in Southeast England were also quantified. The majority of these sites were rural and urban background sites, with a few exceptions. These trends ranged from -3% per year to +3% per year (not all data sets were 10 years or longer, see Table 1 for data coverage dates). All of the surrounding sites that showed statistically significant increasing trends in ozone were in the predominant downwind region (to the northeast) from London. See Figure 18 for wind rose data for four London sites from 1998 to 2010.

Trends in atmospheric concentration are generally decreasing for ozone precursor species (NO_2, NO, VOCs, CO), with roadside trends typically showing larger reductions with respect to other types of monitoring sites throughout London (except for NO_2 at Marylebone Road). While concentrations of ozone precursors are decreasing throughout London, the subsequent trends in ozone in London and in the downwind regions of London are not showing corresponding decreases. However, there are many other factors in ozone formation, such as long-range transport and the contribution of hemispheric background ozone that must be considered, and this must be investigated further.

This data is in preparation for publication that will be submitted by the end of the year.

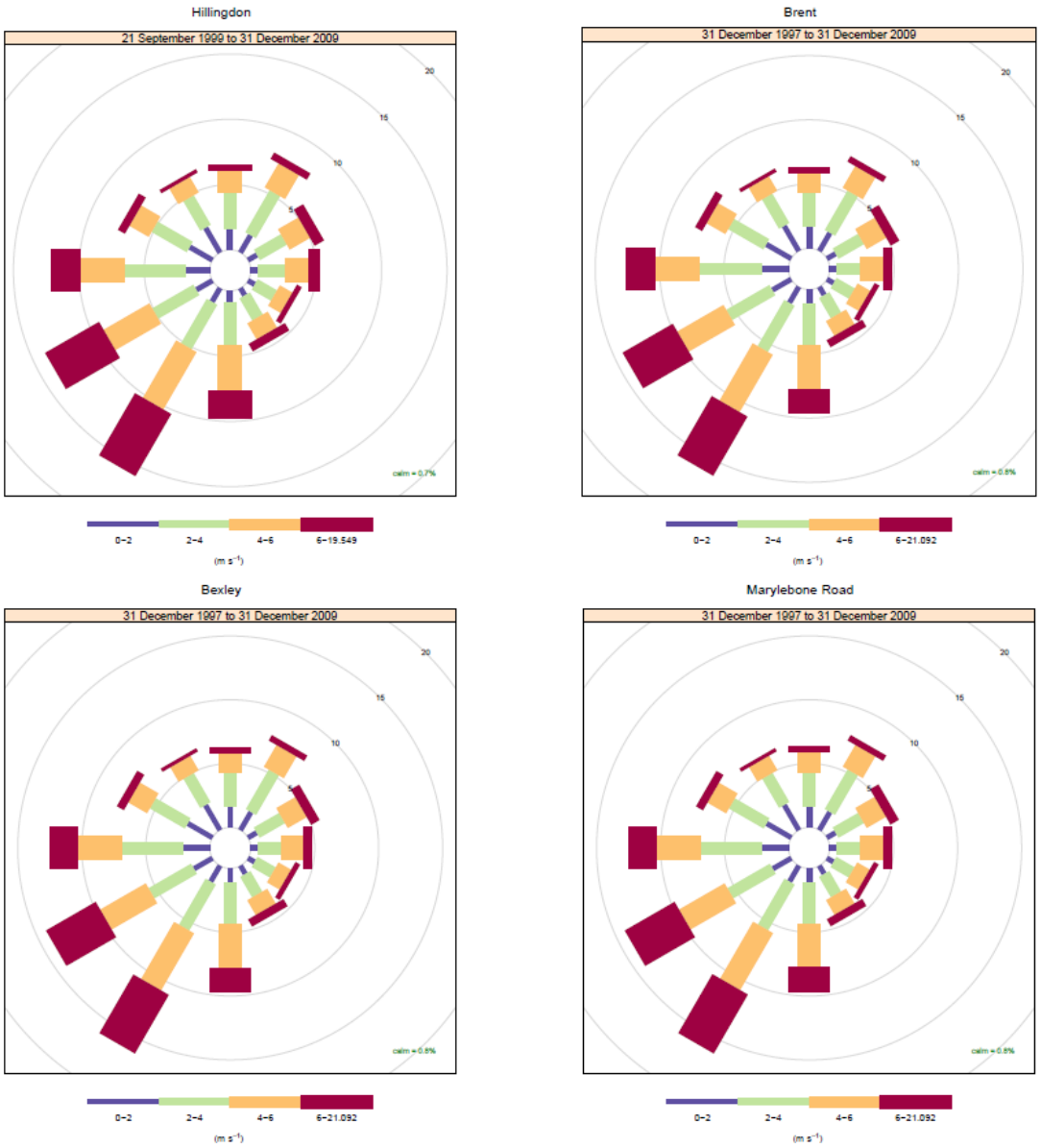

Figure 18: Wind data for four sites in Greater London. These sites are a representative of the wind data from London, showing that the winds are predominantly from the southwest to the northeast.

4 Conclusions and future work

CityZen has employed a large number of measurement techniques and modelling (using detailed time-varying emission data) to look at air pollution changes during the last decade. The analyses of the large amount of data gathered is still ongoing and will continue for several months after the Cityzen project has officially ended.

The focus in CityZen has been on ozone and particulate matter, which are particularly relevant for health, and primary pollutants such as nitrogen dioxide and carbon monoxide. The ozone trend in European emission hot spots during the last 10 to 20 years has attracted considerable interest within CityZen.

The trend analyses (both modelled and observed) performed within CityZen are meant to be helpful for the policy makers community, as they will help to explain the response of air concentrations to emission changes and to devise future air quality legislation. The overall conclusion from the analyses within CityZen seems to be that the trends for ozone and NO_2 are rather small. In particular they are smaller than was expected 10 to 15 years ago based on the emission reduction measures taken in Europe. Ozone and NO_2 concentrations remain high; the trends in ozone are very small or non-existent and negative trends in NOx are weak.

Several processes have been proposed to explain these results:

- climate change, interannual variability, forest fires
- increase in hemispheric background concentrations
- the NO/NO_2 ratio has decreased significantly
- Diesel engines are probably emitting significantly more NO_2 than expected
- Non-linear ozone production (relevant in and around densely populated areas)
- BVOC emissions, which depend on temperature
- Stratospheric ozone recovery

The data analyses are still ongoing, and the use of data generated during CityZen will continue beyond the end of the project. Further model studies are already underway to address some of the processes, which may explain the rather weak trend in ozone over Europe.

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