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Publication on air pollution in hotspot areas

During CityZen a large number of papers on air pollution in the selected hotspot areas of CityZen have been published. Additional papers are either submitted or in preparation and will be published after the end of CityZen.

The following sections present those papers, which are published or accepted (except four papers which were, as of 19 August 2011, still under review). The publications are sorted by hotspot area, along with their abstracts. Web links are provided where applicable. Not included in this deliverable are those CityZen publications that do not specifically address air pollution in hot spot areas but rather deal with techniques and methods within modelling, observations, and emission inventories.

1) General (not related to one specific CityZen hotspot)

Yoon, J., W. von Hoyningen-Huene, M. Vountas, and J. P. Burrows: Analysis of linear long-term trend of Aerosol Optical Thickness derived from SeaWiFS using BAER over Europe and South China, Atmos. Chem. Phys. Discuss., 11, 20757-20792, 2011.

<http://www.atmos-chem-phys-discuss.net/11/20757/2011/acpd-11-20757-2011.pdf>

Abstract. The main purpose of the present paper is to derive and discuss linear long-term trends of Aerosol Optical Thickness (AOT) at 443 and 555 nm over regions in Europe and South China. These areas are densely populated and highly polluted. The study uses the Bremen Aerosol Retrieval (BAER) and Sea-viewing Wide Field-of-view Sensor (SeaWiFS) data for AOT retrievals in the specified regions from October 1997 to May 2008. In order to validate the individually retrieved AOTs and the corresponding trends, Aerosol RObotic NETwork (AERONET) level 2.0 data have been used. The retrieved AOTs were in good agreement with those of AERONET ($0.79 \leq R \leq 0.88$, $0.08 \leq \text{RMSD} \leq 0.13$). The contamination of BAER aerosol retrievals and/or AERONET observations by thin clouds can significantly degrade the AOT and lead to statistically non-representative monthly-means, especially during cloudy seasons. Therefore an inter-correction method has been developed and applied. The “corrected” trends for both BAER SeaWiFS and AERONET AOT were similar having an average of relative error $\approx 25.19\%$. In general terms, negative trends (decrease of aerosol loading) were mainly observed over European regions, with magnitudes up to -0.00453 (-1.93%) and -0.00484 (-2.35%) per year at 443 and 555 nm, respectively. In contrast, the trend in Pearl River Delta was positive, most likely attributed to rapid urbanization and industrialization. The magnitudes of AOT increased by $+0.00761$ ($+1.24\%$) and $+0.00625$ ($+1.15\%$) per year respectively at 443 and 555 nm.

Noguchi, K., Richter, A., Bovensmann, H., Hilboll, A., Burrows, J.P., Irie, H., Hayashida, S., Morino, Y., A feasibility study for the detection of the diurnal variation of tropospheric NO₂ over Tokyo from a geostationary orbit, Advances in Space Research, In Press, Accepted Manuscript. Available online 5 July 2011:

<http://www.sciencedirect.com/science/article/pii/S0273117711004674>

Abstract. We have conducted a feasibility study for the geostationary monitoring of the diurnal variation of tropospheric NO₂ over Tokyo. Using NO₂ fields from a chemical transport model, syn-

thetic spectra were created by a radiative transfer model, SCIATRAN, for summer and winter cases. We then performed a Differential Optical Absorption Spectroscopy (DOAS) analysis to retrieve NO₂ slant column densities (SCDs), and after converting SCDs into vertical column densities (VCDs), we estimated the precision of the retrieved VCDs. The simulation showed that signal-to-noise ratio (SNR) ≥ 500 is needed to detect the diurnal variation and that SNR ≥ 1000 is needed to observe the local minimum occurring in the early afternoon (LT13–14) in summer. In winter, the detection of the diurnal variation during LT08–15 needs SNR ≥ 500 , and SNR ≥ 1000 is needed if early morning (LT07) and early evening (LT16) are included. The currently discussed sensor specification for the Japanese geostationary satellite project, GMAP-Asia, which has a horizontal resolution of 10km and a temporal resolution of 1hr, has demonstrated the performance of a precision of several percent, which is approximately corresponding to SNR=1000~2000 during daytime and SNR ≥ 500 in the morning and evening. We also discuss possible biases caused by the temperature dependence of the absorption cross section utilized in the DOAS retrieval, and the effect of uncertainties of surface albedo and clouds on the estimation of precisions.

Colette, A. C. Granier, Ø. Hodnebrog, H. Jakobs, A. Maurizi, A. Nyiri, B. Bessagnet, A. D'Angiola, M. D'Isidoro, M. Gauss, F. Meleux, M. Memmesheimer, A. Mieville, L. Rouil, F. Russo, S. Solberg, F. Stordal, and F. Tampieri, *Atmos. Chem. Phys. Discuss.*, 11, 19029-19087, 2011.

<http://www.atmos-chem-phys-discuss.net/11/19029/2011/acpd-11-19029-2011.pdf>

Abstract. We discuss the capability of current state-of-the-art chemistry and transport models to reproduce air quality trends and inter annual variability. Documenting these strengths and weaknesses on the basis of historical simulations is essential before the models are used to investigate future air quality projections. To achieve this, a coordinated modelling exercise was performed in the framework of the CityZEN European Project. It involved six regional and global chemistry-transport models (Bolchem, Chimere, Emep, Eurad, OsloCTM2 and Mozart) simulating air quality over the past decade in the Western European anthropogenic emissions hotspots.

Comparisons between models and observations allow assessing the skills of the models to capture the trends in basic atmospheric constituents (NO₂, O₃, and PM₁₀). We find that the trends of primary constituents are well reproduced (except in some countries – owing to their sensitivity to the emission inventory) although capturing the more moderate trends of secondary species such as O₃ is more challenging. Apart from the long term trend, the modelled monthly variability is consistent with the observations but the year-to-year variability is generally underestimated.

A comparison of simulations where anthropogenic emissions are kept constant is also investigated. We find that the magnitude of the emission-driven trend exceeds the natural variability for primary compounds. We can thus conclude that emission management strategies have had a significant impact over the past 10 yr, hence supporting further emission reductions strategies.

von Schneidemesser, E., P. S. Monks, C. Plass-Duelmer: Global comparison of VOC and CO observations in urban areas, *Atmos. Environ.*, 44, 5053-5064, 2010.

Abstract. Speciated volatile organic compound (VOC) and carbon monoxide (CO) measurements from the Marylebone Road site in central London from 1998 through 2008 are presented. Long-term trends show statistically significant decreases for all the VOCs considered, ranging from -3% to -26% per year. Carbon monoxide decreased by -12% per year over the measurement period. The VOC trends observed at the kerbside site in London showed greater rates of decline relative to trends from monitoring sites in rural England (Harwell) and a remote high-altitude site (Hohenpeis-

senberg), which showed decreases for individual VOCs from -2% to -13% per year. Over the same 1998 through 2008 period VOC to CO ratios for London remained steady, an indication that emissions reduction measures affected the measured compounds equally. Relative trends comparing VOC to CO ratios between Marylebone Road and Hohenpeissenberg showed greater similarities than absolute trends, indicating that emissions reductions measures in urban areas are reflected by regional background locations. A comparison of VOC mixing ratios and VOC to CO ratios was undertaken for London and other global cities. Carbon monoxide and VOCs (alkanes greater than C5, alkenes, and aromatics) were found to be strongly correlated (>0.8) in the Annex I countries, whereas only ethene and ethyne were strongly correlated with CO in the non-Annex I countries. The correlation results indicate significant emissions from traffic-related sources in Annex I countries, and a much larger influence of other sources, such as industry and LPG-related sources in non-Annex I countries. Yearly benzene to ethyne ratios for London from 2000 to 2008 ranged from 0.17 to 0.29 and compared well with previous results from US cities and three global megacities.

Konovalov, I. B., Beekmann, M., Richter, A., Burrows, J. P., and Hilboll, A.: Multi-annual changes of NO_x emissions in megacity regions: nonlinear trend analysis of satellite measurement based estimates, *Atmos. Chem. Phys.*, **10, 8481-8498, doi:10.5194/acp-10-8481-2010, 2010.**

<http://www.atmos-chem-phys.net/10/8481/2010/acp-10-8481-2010.pdf>

Abstract. Hazardous impact of air pollutant emissions from megacities on atmospheric composition on regional and global scales is currently an important issue in atmospheric research. However, the quantification of emissions and related effects is frequently a difficult task, especially in the case of developing countries, due to the lack of reliable data and information. This study examines possibilities to retrieve multi-annual NO_x emissions changes in megacity regions from satellite measurements of nitrogen dioxide and to quantify them in terms of linear and nonlinear trends. By combining the retrievals of the GOME and SCIAMACHY satellite instrument data with simulations performed by the CHIMERE chemistry transport model, we obtain the time series of NO_x emission estimates for the 12 largest urban agglomerations in Europe and the Middle East in the period from 1996 to 2008. We employ then a novel method allowing estimation of a nonlinear trend in a noisy time series of an observed variable. The method is based on the probabilistic approach and the use of artificial neural networks; it does not involve any quantitative a priori assumptions. As a result, statistically significant nonlinearities in the estimated NO_x emission trends are detected in 5 megacities (Bagdad, Madrid, Milan, Moscow and Paris). Statistically significant upward linear trends are detected in Istanbul and Tehran, while downward linear trends are revealed in Berlin, London and the Ruhr agglomeration. The presence of nonlinearities in NO_x emission changes in Milan, Paris and Madrid is confirmed by comparison of simulated NO_x concentrations with independent air quality monitoring data. A good quantitative agreement between the linear trends in the simulated and measured near surface NO_x concentrations is found in London.

Vrekoussis, M., Wittrock, F., Richter, A., and Burrows, J. P.: GOME-2 observations of oxygenated VOCs: what can we learn from the ratio glyoxal to formaldehyde on a global scale?, *Atmos. Chem. Phys.*, **10, 10145-10160, doi:10.5194/acp-10-10145-2010, 2010.**

<http://www.atmos-chem-phys.net/10/10145/2010/acp-10-10145-2010.pdf>

Abstract. Collocated data sets of glyoxal (CHO.CHO) and formaldehyde (HCHO) were retrieved for the first time from measurements of the Global Ozone Monitoring Experiment-2 (GOME-2) during the first two years of operation in 2007 and 2008. Both oxygenated Volatile Organic Com-

pounds, OVOC, are key intermediate species produced during the oxidation of precursor hydrocarbons. Their short lifetime of a few hours in the lower troposphere links them to emission sources and makes them useful tracers of photochemical activity. The global composite maps of GOME-2 HCHO and CHO.CHO have strong similarities confirming their common atmospheric and/or surface sources. The highest column amounts of these OVOCs are recorded over regions with enhanced biogenic emissions (e.g. tropical forests in South America, Africa and Indonesia). Enhanced OVOC values are also present over areas of anthropogenic activity and biomass burning (e.g. over China, N. America, Europe and Australia). The ratio of CHO.CHO to HCHO, RGF, has been used, for the first time on a global scale, to classify the sources according to biogenic and/or anthropogenic emissions of the precursors; RGF between 0.040 to 0.060 point to the existence of biogenic emissions with the highest values being observed at the highest Enhanced Vegetation Index, EVI. RGFs below 0.040 are indicative of anthropogenic emissions and associated with high levels of NO₂. This decreasing tendency of RGF with increasing NO₂ is also observed when analyzing data for individual large cities, indicating that it is a common feature. The results obtained for RGF from GOME-2 data are compared with the findings based on regional SCIAMACHY observations showing good agreement. This is explained by the excellent correlation of the global retrieved column amounts of CHO.CHO and HCHO from the GOME-2 and SCIAMACHY instruments for the period 2007–2008.

2) East Mediterranean

Gerasopoulos, E., Amiridis, V., Kazadzis, S., Kokkalis, P., Eleftheratos, K., Andreae, M. O., Andreae, T. W., El-Askary, H., and Zerefos, C. S.: Three-year ground based measurements of aerosol optical depth over the Eastern Mediterranean: the urban environment of Athens, *Atmos. Chem. Phys.*, 11, 2145-2159, doi:10.5194/acp-11-2145-2011, 2011.
<http://www.atmos-chem-phys.net/11/2145/2011/acp-11-2145-2011.pdf>

Abstract. Three years (2006–2008) of ground-based observations of the Aerosol Optical Depth (AOD) in the urban environment of Athens, in the Eastern Mediterranean, are analysed in this work. Measurements were acquired with a Multi-Filter Rotating Shadowband Radiometer at five wavelengths. The daily average AOD at 500 nm is 0.23, and the mean Ångström coefficient calculated between 415 and 867 nm is 1.41. 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). An additional important contribution (23%) is dust from Africa, 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%. This is 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.

Hatzianastassiou, N., A. Gkikas, N. Mihalopoulos, O. Torres, B. D. Katsoulis: Natural versus anthropogenic aerosols in the eastern Mediterranean basin derived from multi-year TOMS and MODIS satellite data, *J. Geophys. Res.*, 114, D24202, doi:10.1029/2009JD011982, 2009.

Abstract. In the present study we investigate the spatial and temporal variation of aerosol optical thickness (AOT) in the eastern Mediterranean basin, and more specifically in the area extending from 28.5°N to 42.5°N and from 18.5°E to 35.5°E, which includes large urban areas and megacities such as Cairo, Istanbul, Athens, Izmir, Ankara, and Thessaloniki. For this purpose we use long-term AOT data from the Total Ozone Mapping Spectrometer (TOMS) version2 converted to 500 nm (AOT₅₀₀) for the period 1980–2001 and Collection 005 AOT data at $\lambda = 550$ nm (AOT₅₅₀) from Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra and Aqua satellites for the periods 2000–2005 and 2002–2005, respectively. The spatial and temporal variation of AOT shows a good agreement between TOMS and MODIS, in terms of geographical patterns, which maximizes the usefulness of TOMS AOT product given its long temporal coverage (climatological). According to MODIS-Terra, the annual mean AOT₅₅₀ over the studied region equals 0.22 ± 0.05 (monthly values ranging from 0.14 to 0.32) and shows strong spatial inhomogeneities. The smallest values (down to 0.1) occur over the western, and especially in the northwestern, part of Greece, over the northern part of Anatolian peninsula and also over the sea of east Mediterranean. The largest AOT values (up to about 1.0) occur over northern Africa, Middle East, and the adjacent coasts, and over the Anatolian plateau. These high aerosol loadings are of natural origin, mainly desert dust. However, very large values (up to 0.8) are also found over large urban areas surrounding megacities, associated with anthropogenic, apart from natural desert, aerosols. By using TOMS AOT data as proxy for the dust source, and the difference of MODIS-TOMS AOT as an indicator of the anthropogenic aerosol component, the relative contribution of natural versus anthropogenic sources of aerosols has been derived for the eastern Mediterranean area. Limitations of this approach are discussed and the associated uncertainties are also evaluated and discussed.

Theodosi, C., U. Im, A. Bougiatioti, P. Zarmpas, O. Yenigun, N. Mihalopoulos, Aerosol chemical composition over Istanbul, *Science of the Total Environment*, 408, p. 2482–2491, 2010.

Abstract. This study examines the chemical composition of aerosols over the Greater Istanbul Area. To achieve this 325 (PM₁₀) aerosol samples were collected over Bosphorus from November 2007 to June 2009 and were analysed for the main ions, trace metals, water-soluble organic carbon (WSOC), organic (OC) and elemental carbon (EC).

PM₁₀ levels were found to be in good agreement with those measured by the Istanbul Municipality air quality network, indicating that the sampling site is representative of the Greater Istanbul Area. The main ions measured in the PM₁₀ samples were Na⁺, Ca²⁺ and non-sea-salt sulphates (nss-SO₄²⁻). On average, 31% of Ca²⁺ was found to be associated with carbonates. Trace elements related to human activities (as Pb, V, Cd and Ni) obtained peak values during winter due to domestic heating, whereas natural origin elements like Al, Fe and Mn peaked during the spring period due to dust transport from Northern Africa. Organic carbon was found to be mostly primary and elemental carbon was strongly linked to fuel oil combustion and traffic. Both OC and EC concentrations increased during winter due to domestic heating, while the higher WSOC to OC ratio during summer can be mostly attributed to the presence of secondary, oxidised and more soluble organics. Factor analysis identified six components/sources for aerosol species in PM₁₀, namely traffic/industrial, crustal, sea-salt, fuel–oil combustion, secondary and ammonium sulfate.

Im, U., K. Markakis, A. Unal, T. Kindap, A. Poupkou, S. Incecik, O. Yenigun, D. Melas, C. Theodosi, N. Mihalopoulos, Study of a winter PM episode in Istanbul using the high resolution WRF/CMAQ modeling system, Atmospheric Environment, 44, p. 3085-3094, 2010.

Abstract. High winter-time PM₁₀, sulfate, nitrate and ammonium levels in Istanbul were investigated using a high resolution WRF/CMAQ mesoscale model system. A model-ready anthropogenic emission inventory on 2 km spatial resolution was developed for the area and the present study is the first attempt to test these emissions. The results suggested that the system was capable of producing the magnitudes. PM₁₀ levels calculated by the model underestimated the observations with an average of 10 per cent at Bogazici University sampling station, whereas an overestimation of 12 per cent is calculated for all stations. High uncertainties, particularly in traffic and coal combustion, led to over estimations around emission hot spots. Base case results together with the sensitivity studies pointed significant contribution of local sources, pointing to the need of control strategies focusing on primary particulate emissions.

Im, U., Markakis, K., Poupkou, A., Melas, D., Unal, A., Gerasopoulos, E., Daskalakis, N., Kindap, T., and Kanakidou, M.: The impact of temperature changes on summer time ozone and its precursors in the Eastern Mediterranean, Atmos. Chem. Phys., 11, 3847-3864, doi:10.5194/acp-11-3847-2011, 2011a.

<http://www.atmos-chem-phys.net/11/3847/2011/acp-11-3847-2011.pdf>

Abstract. Changes in temperature due to variability in meteorology and climate change are expected to significantly impact atmospheric composition. The Mediterranean is a climate sensitive region and includes megacities like Istanbul and large urban agglomerations such as Athens. The effect of temperature changes on gaseous air pollutant levels and the atmospheric processes that are controlling them in the Eastern Mediterranean are here investigated. The WRF/CMAQ mesoscale modeling system is used, coupled with the MEGAN model for the processing of biogenic volatile organic compound emissions. A set of temperature perturbations (spanning from 1 to 5 K) is applied on a base case simulation corresponding to July 2004. The results indicate that the Eastern Mediterranean basin acts as a reservoir of pollutants and their precursor emissions from large urban agglomerations. During summer, chemistry is a major sink at these urban areas near the surface, and a minor contributor at downwind areas. On average, the atmospheric processes are more effective within the first 1000m above ground. Temperature increases lead to increases in biogenic emissions by $9\pm 3\%K^{-1}$. Ozone mixing ratios increase almost linearly with the increases in ambient temperatures by 1 ± 0.1 ppb O₃ K⁻¹ for all studied urban and receptor stations except for Istanbul, where a 0.4 ± 0.1 ppb O₃ K⁻¹ increase is calculated, which is about half of the domain-averaged increase of 0.9 ± 0.1 ppb O₃ K⁻¹. The computed changes in atmospheric processes are also linearly related with temperature changes.

Koçak, M., Theodosi, C., Zarpas, P., Im, U., Bougiatioti, A., Yenigün, O., Mihalopoulos, N.: Particulate matter (PM₁₀) in Istanbul: Origin, source areas and potential impact on surrounding regions, Atmos. Environ., doi:10.1016/j.atmosenv.2010.10.007, 2010.

Abstract. Water-soluble ions (Cl⁻, NO₃⁻, SO₄²⁻, C₂O₄⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺, Ca²⁺), water soluble organic carbon (WSOC), organic and elemental carbon (OC, EC) and trace metals (Al, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb) were measured in aerosol PM₁₀ samples above the megacity of Istanbul between November 2007 and June 2009. Source apportionment analysis using Positive Matrix Factorization (PMF) indicates that approximately 80% of the PM₁₀ is anthropogenic in

origin (secondary, refuse incineration, fuel oil and solid fuel combustion and traffic). Crustal and sea salt account for 10.2 and 7.5% of the observed mass, respectively. In general, anthropogenic (except secondary) aerosol shows higher concentrations and contributions in winter. Mean concentration and contribution of crustal source is found to be more important during the transitional period due to mineral dust transport from North Africa. During the sampling period, 42 events exceeding the limit value of 50 mgm^{-3} are identified. A significant percentage (91%; $n=38$) of these exceedances is attributed to anthropogenic sources. Potential Source Contribution Function analysis highlights that Istanbul is affected from distant sources from Balkans and Western Europe during winter and from Eastern Europe during summer. On the other hand, Istanbul sources influence western Black Sea and Eastern Europe during winter and Aegean and Levantine Sea during summer.

Koçak, M., Kubilay, N., Tuğrul, S., and Mihalopoulos, N.: Atmospheric nutrient inputs to the northern levantine basin from a long-term observation: sources and comparison with riverine inputs, Biogeosciences, 7, 4037-4050, doi:10.5194/bg-7-4037-2010, 2010.

Abstract. Aerosol and rainwater samples have been collected at a rural site located on the coastline of the Eastern Mediterranean, Erdemli, Turkey between January 1999 and December 2007. Riverine sampling was carried out at five Rivers (Ceyhan, Seyhan, Göksu, Berdan and Lamas) draining into the Northeastern Levantine Basin (NLB) between March 2002 and July 2007. Samples have been analyzed for macronutrients of phosphate, silicate, nitrate and ammonium (PO_4^{3-} , Si_{diss} , NO_3^- and NH_4^+). Phosphate and silicate in aerosol and rainwater showed higher and larger variations during the transitional period when air flows predominantly originate from North Africa and Middle East/Arabian Peninsula. Deficiency of alkaline material have been found to be the main reason of the acidic rain events whilst high pH values (>7) have been associated with high Sidiss concentrations due to sporadic dust events. In general, lowest nitrate and ammonium concentrations in aerosol and rainwater have been associated with air flow from the Mediterranean Sea. Comparison of atmospheric with riverine fluxes demonstrated that DIN and PO_4^{3-} fluxes to NLB have been dominated by atmosphere ($\approx 90\%$ and $\approx 60\%$ respectively) whereas the input of Si was mainly derived from riverine runoff ($\approx 90\%$). N/P ratios in the atmospheric deposition (233); riverine discharge (28) revealed that NLB receives excessive amounts of DIN and this unbalanced P and N inputs may provoke even more phosphorus deficiency. Observed molar Si/N ratio suggested Si limitation relative to nitrogen might cause a switch from diatom dominated communities to non-siliceous populations particularly at coastal NLB.

Kanakidou, M., N. Mihalopoulos, T. Kindap, U. Im, M. Vrekoussis, E. Gerasopoulos, E. Dermizaki, A. Unal, M. Koçak, K. Markakis, D. Melas, G. Kouvarakis, A. F. Youssef, A. Richter, N. Hatzianastassiou, A. Hilboll, F. Ebojie, F. Wittrock, C. von Savigny, J. P. Burrows, A. Ladstaetter-Weissenmayer, H. Moubasher, Megacities as hot spots of air pollution in the East Mediterranean, Atmospheric Environment, doi:10.1016/j.atmosenv.2010.11.048, 2010.

Abstract. This paper provides a comprehensive overview of the actual knowledge on the atmospheric pollution sources, transport, transformation and levels in the East Mediterranean. It focuses both on the background atmosphere and on the similarities and differences between the urban areas that exhibited important urbanization the past years: the two megacities Istanbul, Cairo and the Athens extended area. Ground-based observations are combined with satellite data and atmospheric modeling. The overall evaluation pointed out that long and regional range transport of natural and

anthropogenic pollution sources have about similar importance with local sources for the background air pollution levels in the area.

3) Pearl River Delta

Wang, Xuesong, Yuanhang Zhang, Yongtao Hu, Wei Zhou, Limin Zeng, Min Hu, Daniel S. Cohan and Armistead G. Russell, Decoupled direct sensitivity analysis of regional ozone pollution over the Pearl River Delta during the PRIDE-PRD2004 campaign, Atmos. Environ., doi:10.1016/j.atmosenv.2011.06.006, 2011.

Abstract. High-order decoupled direct method is applied to investigate O₃-precursor sensitivity during the Program of Regional Integrated Experiments of Air Quality over the Pearl River Delta (PRD) Region, China in October 2004 (PRIDE-PRD2004). First-order sensitivity coefficients of O₃ show a NO_x-inhibited chemistry along the polluted plumes in central and southern PRD areas, and a NO_x-limited condition over larger areas of the western, eastern and northern PRD. A nonlinearity ratio derived from O₃ sensitivity coefficients clearly describes the nonlinear feature of O₃ chemistry over PRD. The distributions of NO_x- and VOC-sensitive regimes are determined by near-ground flow conditions, and O₃ sensitivity is found to shift from VOC-sensitive conditions to NO_x-sensitive conditions from morning to afternoon in central and western areas of the PRD. Elevated surface O₃ around mid-afternoon in October mainly occurred in VOC-sensitive regions or those NO_x-limited areas in close vicinity of the VOC-sensitive regions. Ratios of H₂O₂/HNO₃ and peroxides/HNO₃ for indicating O₃-precursor sensitivity are evaluated and the transition values of 0.45 for H₂O₂/HNO₃ and 0.80 for peroxides/HNO₃ were found, with an overall accuracy of about 92% for sensitivity identification at elevated O₃ hours in afternoons during the campaign.

Wang, X., Zhang Y, Hu Y, Zhou W, Lu K, Zhong L, Zeng L, Shao M, Hu M, Russell AG. Process analysis and sensitivity study of regional ozone formation over the Pearl River Delta, China, during the PRIDE-PRD2004 campaign using the Community Multiscale Air Quality modeling system. Atmospheric Chemistry and Physics, 10: 4423-4437, 2010.
<http://www.atmos-chem-phys.net/10/4423/2010/acp-10-4423-2010.pdf>

Abstract. In this study, the Community Multiscale Air Quality (CMAQ) modeling system is used to simulate the ozone (O₃) episodes during the Program of Regional Integrated Experiments of Air Quality over the Pearl River Delta, China, in October 2004 (PRIDE-PRD2004). The simulation suggests that O₃ pollution is a regional phenomenon in the Pearl River Delta (PRD). Elevated O₃ levels often occurred in the southwestern inland PRD, Pearl River estuary (PRE), and southern coastal areas during the 1-month field campaign. Three evolution patterns of simulated surface O₃ are summarized based on different near-ground flow conditions. More than 75% of days featured interactions between weak synoptic forcing and local sea-land circulation. Integrated process rate (IPR) analysis shows that photochemical production is a dominant contributor to O₃ enhancement from 09:00 to 15:00 local standard time in the atmospheric boundary layer over most areas with elevated O₃ occurrence in the mid-afternoon. The simulated ozone production efficiency is 2–8 O₃ molecules per NO_x molecule oxidized in areas with high O₃ chemical production. Precursors of O₃ originating from different source regions in the central PRD are mixed during the course of transport to downwind rural areas during nighttime and early morning, where they then contribute to the daytime O₃ photochemical production. The sea-land circulation plays an important role on the regional O₃ formation and distribution over PRD. Sensitivity studies suggest that O₃ formation

is volatile-organic-compound-limited in the central inland PRD, PRE, and surrounding coastal areas with less chemical aging ($\text{NO}_x/\text{NO}_y > 0.6$), but is NO_x -limited in the rural southwestern PRD with aged air ($\text{NO}_x/\text{NO}_y < 0.3$).

Hu X., Li Y., Li J., Wang X., Zhang Y., Interaction of Ambient PM₁₀ among the Cities over the Pearl River Delta, 2011, Acta Scientiarum Naturalium Universitatis Pekinensis, 47(3), 519-524, (in Chinese).

Abstract. The Models-3/CMAQ modeling system was applied to investigate PM₁₀ pollution over the Pearl River Delta (PRD) region during October 2006. Sensitivity analysis was conducted to examine the relationship between the reduction of different source emissions and the consequent change of PM₁₀ concentrations, and to quantify the interaction of air pollution among adjacent cities. The results show that PM₁₀ pollution is a regional-scale issue in PRD. Guangzhou, Foshan Jiangmen and Dongguan, contribute to the PM₁₀ in PRD remarkably. The relative sensitivity coefficient was proposed to determine the impact of regional pollution sources. The PM₁₀ concentrations in Zhuhai, Jiangmen, Zhongshan, Foshan cities are significantly influenced by regional source emissions. The intercity transport has been the important factor of PM₁₀ pollution in PRD. To improve the air quality in PRD area, effective control of emission sources should be highly reinforced, together with consentaneous programming, intensive collaboration, joint prevention and control.

Xiao, R., N. Takegawa, M. Zheng, Y. Kondo, Y. Miyazaki, T. Miyakawa, M. Hu, M. Shao, L. Zeng, Y. Gong, K. Lu, Z. Deng, Y. Zhao, and Y. H. Zhang, Characterization and source apportionment of submicron aerosol with aerosol mass spectrometer during the PRIDE-PRD 2006 campaign, Atmos. Chem. Phys., 11, 6911-6929, 2011.

<http://www.atmos-chem-phys.net/11/6911/2011/acp-11-6911-2011.pdf>

Abstract. Size-resolved chemical compositions of nonrefractory submicron aerosol were measured using an Aerodyne quadrupole aerosol mass spectrometer (Q-AMS) at the rural site Back Garden (BG), located ≈ 50 km northwest of Guangzhou in July 2006. This paper characterized the submicron aerosol particles of regional air pollution in Pearl River Delta (PRD) in the southern China. Organics and sulfate dominated the submicron aerosol compositions, with average mass concentrations of $11.8 \pm 8.4 \mu\text{g m}^{-3}$ and $13.5 \pm 8.7 \mu\text{g m}^{-3}$, respectively. Unlike other air masses, the air masses originated from Southeast-South and passing through the PRD urban areas exhibited distinct bimodal size distribution characteristics for both organics and sulfate: the first mode peaked at vacuum aerodynamic diameters (D_{va}) ≈ 200 nm and the second mode occurred at D_{va} from 300–700 nm. With the information from AMS, it was found from this study that the first mode of organics in PRD regional air masses was contributed by both secondary organic aerosol formation and combustion-related emissions, which is different from most findings in other urban areas (first mode of organics primarily from combustion-related emissions). The analysis of AMS mass spectra data by positive matrix factorization (PMF) model identified three sources of submicron organic aerosol including hydrocarbon-like organic aerosol (HOA), low volatility oxygenated organic aerosol (LV-OOA) and semi-volatile oxygenated organic aerosol (SV-OOA). The strong correlation between HOA and EC indicated primary combustion emissions as the major source of HOA while a close correlation between SV-OOA and semi-volatile secondary species nitrate as well as between LV-OOA and nonvolatile secondary species sulfate suggested secondary aerosol formation as the major source of SV-OOA and LV-OOA at the BG site. However, LV-OOA was more aged than SV-OOA as its spectra was highly correlated with the reference spectra of fulvic acid, an indicator of aged and oxygenated aerosol. The origin of HOA and OOA (the sum of LV-OOA and SV-OOA) has

been further confirmed by the statistics that primary organic carbon (POC) and secondary organic carbon (SOC), estimated by the EC tracer method, were closely correlated with HOA and OOA, respectively. The results of the EC tracer method and of the PMF model revealed that primary organic aerosol (POA) constituted $\approx 34\text{--}47\%$ of OA mass and secondary organic aerosol (SOA) constituted $\approx 53\text{--}66\%$ of regional organic aerosol in PRD during summer season. The presence of abundant SOA was consistent with water soluble organic carbon (WSOC) results (accounting for $\approx 60\%$ of OC on average) by Miyazaki et al. (2009) for the same campaign. OOA correlated well with WSOC at the BG site, indicating that most OOA were water soluble. More specifically, approximately 86% of LV-OOA and 61% of SV-OOA were estimated as water soluble species on the basis of carbon content comparison.

4) BeNeLux

Hodnebrog, Ø., Stordal, F., and Berntsen, T. K.: Does the resolution of megacity emissions impact large scale ozone?, Atmospheric Environment, In Press, 10.1016/j.atmosenv.2011.01.012, 2011.

Abstract. The importance of using high resolution when modelling the impact of megacity emission on large scale tropospheric ozone has been investigated using the regional WRF-Chem model. Two 3-day summer periods in July and August 2003 have been simulated for three megacities; London, Ruhr, and Cairo.

Simulations have been performed with constant model resolution ($9 \times 9 \text{ km}^2$) and with megacity emission resolutions at $9 \times 9 \text{ km}^2$, $27 \times 27 \text{ km}^2$, and $81 \times 81 \text{ km}^2$, by using detailed emission inventories averaged to the appropriate resolution. In order to quantify the megacities' contributions to regional ozone, simulations without megacity emissions were also performed.

In general, results from the six case studies show that the inaccuracies that arise on a large scale when using megacity emission resolution at a typical scale of a global Chemistry-Transport Model ($81 \times 81 \text{ km}^2$) were relatively small (12% or less). However, the impact of resolution on ozone changes caused by megacity emissions depends strongly on meteorological conditions. The case with the largest impact (London, July 2003) showed that net ozone formed from the megacity were 12% higher when using $81 \times 81 \text{ km}^2$ rather than $9 \times 9 \text{ km}^2$ emission resolution, due to stronger ozone production since the NO_x emissions were more rapidly diluted to regions where the ozone production was NO_x limited in the coarse resolution case. Our study suggests that high resolution is more important for local air pollution studies than for large scale ozone changes relevant for climate studies, since a change in megacity emission resolution induces small-scale spatial changes in ozone fields, but relatively small changes when integrating over a large volume. These results indicate that parameterization of megacity emissions in large scale models may be unnecessary. However, the impact of a finer resolution of the meteorology is not studied here and could possibly give larger effects seen from a climate perspective.

5) Po Valley

The following publication was submitted in Italian to the SAPERE journal (<http://sapere.galileonet.it/>). The abstract is provided below in English.

Tampieri, F. A. Maurizi, F. Russo. Import/export di inquinanti dalle grandi aree urbanizzate, Submitted to SAPERE, 2011.

Abstract. The import/export of pollutants to and from hot-spots is an important issue for local communities, to define mitigation actions in order to limit air pollution. Moreover, the hot-spots role in the budget of atmospheric composition is a question of general interest.

A tracer study has been performed to shed some light on the problem. Emissions modulated according to CO sources are marked differently in reference to points within (tracer “i”) or outside (tracer “o”) the selected areas (here, the Po Valley has been mainly used, and some comparisons with BeNeLux are made).

The mass of the tracers emitted inside and outside the selected area present on the air column above the same area is computed using BOLCHEM. A one year run (using meteorology and emissions of 2007) has been performed over a European domain, with a resolution of 50 x 50 km.

The ratio of the mass of “i” over the mass of “o” in the entire column has a modal value of about 1, meaning that the contribution due to transport from remote sources becomes higher than the local one in half of the year. At the ground level (the lowest model level) the contribution to the total columnar mass from the local emission is larger than the remote (the average value of the ratio is about 3) but for about 15% of time the contribution from outside is larger than the local.

Since the Po Valley is mostly surrounded by high mountains, the advection at low levels is reduced and thus the local effects may be more important than in other areas. To get some comparison, a similar investigation has been performed for the BeNeLux hot spot. The qualitative results are very similar, despite the very different topographical features.

A careful investigation of the concentration patterns reveals local inhomogeneities. Therefore what has been noted to stand for the area averaged results is not necessarily true for each point. In this case it is evident that the air quality in areas inside the Po Valley with relatively low emissions can be strongly affected by the long range transport of pollution.