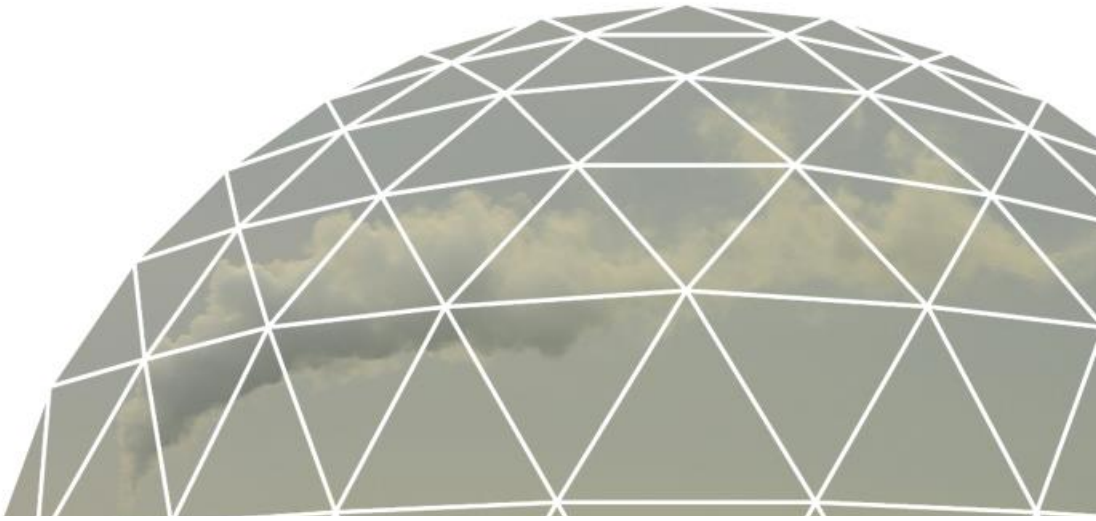
ECORYS

Services to assess the reasons for non-compliance of ozone target value set by Directive 2008/50/EC and potential for air quality improvements in relation to ozone pollution

Final report

Client: DG Environment

29 January, 2014



Risø DTU DTU MWH BUILDING A BETTER WORLD NATIONAL ENVIRONMENTAL RESEARCH INSTITUTE AARHUS UNIVERSITY milieu ENVIRONMENTAL LAW & POLICY UWE BRISTOL University of the West of England

Services to assess the reasons for non-compliance of ozone target value set by Directive 2008/50/EC and potential for air quality improvements in relation to ozone pollution

Final report

Client: DG Environment

21 January, 2014

Authors:

Hanne Bach, Jørgen Brandt, Jesper H. Christensen, Thomas Ellermann, Camilla Geels, Ole Hertel, Andreas Massling, Helle Ørsted Nielsen, Ole-Kenneth Nielsen, Claus Nordstrøm, Jacob K. Nøjgaard, and Henrik Skov,
DCE - Danish Centre for Environment and Energy, Aarhus University (AU), Denmark

Tim Chatterton, Enda Hayes, Jo Barnes, Duncan Laxen, Jimi Irwin and Jim Longhurst,
University of the West of England (UWE), Bristol

Florent Pelsy and Tony Zamparutti
Milieu, Belgium

AARHUS UNIVERSITY
DCE - DANISH CENTRE FOR ENVIRONMENT AND ENERGY

UWE BRISTOL University of the West of England

ECORYS

About Ecorys

At Ecorys we aim to deliver real benefit to society through the work we do. We offer research, consultancy and project management, specialising in economic, social and spatial development. Focusing on complex market, policy and management issues we provide our clients in the public, private and not-for-profit sectors worldwide with a unique perspective and high-value solutions. Ecorys' remarkable history spans more than 80 years. Our expertise covers economy and competitiveness; regions, cities and real estate; energy and water; transport and mobility; social policy, education, health and governance. We value our independence, integrity and partnerships. Our staff is dedicated experts from academia and consultancy, who share best practices both within our company and with our partners internationally.

Ecorys Netherlands has an active CSR policy and is ISO14001 certified (the internationally recognized quality standard for environmental management systems). Our sustainability goals translate into our company policy and practical measures, such as printing our documents on FSC certified paper and compensating our carbon footprint.

ECORYS Nederland BV
Watermanweg 44
3067 GG Rotterdam

P.O. Box 4175
3006 AD Rotterdam
The Netherlands

T +31 (0)10 453 88 00
F +31 (0)10 453 07 68
E netherlands@ecorys.com
Registration no. 24316726

W www.ecorys.nl

Table of Contents

1 Summary

This report presents the results of the project “Services to assess the reasons for non-compliance of ozone target value set by the EU Air Quality Directive 2008/50/EC (the Directive) and potential for air quality improvements in relation to ozone pollution” commissioned by the General Directorate (DG) Environment of the European Commission in December 2011 with the overall objective to support the review of the implementation of the Directive 2008/50/EC.

The Directive largely consolidates the provisions of EU Directive on ozone in ambient air, 2002/3/EC which established two target values for ozone, one based on the protection of human health (120 µg/m^3 as maximum daily 8-hour mean, not to be exceeded more than 25 days per year averaged over three years) and another for protection of vegetation (AOT40 (Accumulated Ozone over a Threshold of 40 ppb) 18,000 µg/m^3-h averaged over five years). The target values are levels to be attained where possible. They are in effect from January 2010, which means that data from the year 2010 is the first dataset to be used to evaluate compliance over the following three or five years relatively for the two target values. The Directive also established long-term objectives for the protection of health (120 µg/m^3 as maximum daily 8-hour mean and no exceedences) and vegetation (AOT40 of 6,000 µg/m^3-h) while not indicating the date for achieving these objectives. In addition, the Directive also set an information threshold of 180 µg/m^3 and an alert threshold of 240µg/m^3 (both hourly averages). The alert threshold requires exceedences to be measured or predicted for three consecutive hours to invoke the requirement short-term action plans under Article 24 of the Directive. Neither the target values nor the alert and information thresholds have been amended since they were first established in the Directive on ozone in ambient air (2002/3/EC).

Ground-level ozone

Ozone is a secondary pollutant formed through complex atmospheric chemical reactions of precursor gases nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOCs), carbon monoxide (CO), and methane (CH_4). The relationship between precursor emissions and formation of ground-level ozone is highly non-linear and is influenced by timescales for ozone formation and the transboundary nature of the pollutant and its precursors. All these complex factors make the management of ground-level ozone pollution challenging.

Due to local and regional ozone chemistry, the highest concentrations of ozone are found in rural areas and the lowest in urban areas where NOx emissions are highest. The exceptions to this are episodic events of high ozone concentration in cities with certain topography (valleys) and very low wind speeds in summer time. The highest ozone concentrations are generally found in southern Europe and the lowest in northern Europe due to a complex combination of emissions, ozone chemistry, transport and deposition.

The current situation for precursor emissions, ozone concentrations and human exposure

The general increasing trend in background ozone concentrations registered at least since the year 2000 is a global phenomenon, the main causes being increased global precursor emissions (especially NOx) and increased input from the stratosphere (estimated contribution in the order of 10%). The evidence points towards human activities as a major reason for the increasing trend in background ground-level ozone, but the source attribution of these increases remains uncertain.

Emission control legislation in Europe has achieved reductions between 30 and 60% in ozone precursor emissions over the last two decades (1990-2010), but the monitoring data indicates continuing difficulties in achieving target values among Member States.

The main reductions in ozone precursor emissions happened in the first decade of this period, while the rate of decrease slowed considerably in the later years. The distribution between source sectors has changed for some of the ozone precursors (NMVOC and CO) and for NOx the well-known, efficient measures such as low-NOx burners, DeNOx and three-way catalysts have now largely been implemented. In order to achieve further emission reductions, additional measures will have to be implemented in sectors other than those traditionally attracting the attention emission regulation e.g. small scale combustion ('NOx, NMVOC and CO), solvent use (NMVOC) and for CH_4, the agricultural sector will probably be the key sector for future reductions.

There is no clear single trend that can be detected in all metrics for ozone concentrations from 1996-2010 across Europe although concentrations in north-west Europe have tended to increase, and in central-eastern Europe and Scandinavia to decrease. Decadal mean concentrations (2001-2010) tend to be highest in southern and eastern Europe, whereas the highest 1-hour and 8-hour mean concentrations tend to show a different pattern with the highest concentrations mainly lying in a north-south band across central Europe. There is a tendency for ozone concentrations to have increased in areas of lowest decadal mean concentration (north-west Europe) and decreased in areas of highest decadal mean concentration (southeast Europe). When also considering exceedence statistics (information and alert thresholds, target value, long-term objective and AOT40) patterns of trends become even more complex. There is a discernible downward trend in some exceedence statistics (particularly target value and AOT40), but year-to-year variations in meteorological conditions disturb this picture. The years 2003 and 2006 appear as unusually high ozone years primarily due to meteorology those years with long periods of high pressure, low advection and high temperatures. Similarly there are also unusually low ozone years due to opposite meteorological factors, e.g. 2007, 2008 and 2009.

Analyses covering the period 2004-2010 estimate that around 12 % of the European population is exposed to exceedences of the target value, two thirds to exceedences of the information threshold, and over 99% of the population exposed to exceedences of the long-term objective. In terms of number of people exposed to exceedences, urban exposure is higher (in terms of number of exposed people) for all considered metrics than for the rural population. If urban and suburban areas are grouped, it is clear that in terms of human health, ozone pollution poses a disproportionate burden on 'non-rural' populations in terms of number of people affected. Therefore, although rural concentrations are in general the highest, fewer people are exposed to these levels

Table 2: Maximum exposure to exceedences of Thresholds/Objective/Target Value (2004-10) by 'Degree of Urbanisation' (DegUrba) Classification

	Urban Exposure		Suburban Exposure		Rural Exposure		Total Exposure	
	Population Exposed (millions)	% of Urban Population	Population Exposed (millions)	% of Suburban Population	Population Exposed (millions)	% of Rural Population	Population Exposed (millions)	% of Total Population
Information Threshold	**134.9**	67.4	**112.5**	70.7	**87.7**	61.3	**335.1**	66.7
Alert Threshold	**12.9**	6.4	**7.6**	4.8	**2.4**	1.7	**22.9**	4.5
Long-Term Objective	**199.2**	99.4	**158.3**	99.5	**142.6**	99.7	**500.0**	99.5
Target Value	**22.2**	11.1	**23.9**	15.1	**13.1**	9.1	**59.2**	11.8

Monitoring and modelling precursors and ground-level ozone

The analysis found that the data quality objectives of 90% data capture for ozone and NO_2 were fulfilled for 81 and 77% of the measurement sites respectively, whereas data capture for benzene was only 39%. A weakness in the Directive appears to be that, except for benzene, monitoring of non-methane volatile organic compounds is mandatory, but the substances listed are only recommended, and not mandatory.

This study identifies the poorly covered organic precursors of ozone, for which the current analysis has shown measurements to be characterised by a very low data capture, as a major weakness in fulfilling the provisions of the Directive with regard to ozone. Monitoring of organic precursors of ozone is central to the understanding of ozone formation in urban and suburban environments as well as in the background atmosphere. It is recommended that assessment of some of the 30 recommended substances is made mandatory, i.e. the medium-chain hydrocarbons (C5-C9 substances), which can be analysed using the automated instrument (GC "BTEX monitors") or laboratory instrument (GC-MS analysis of adsorbed substances). Other species with high ozone formation potential, e.g. short-chain hydrocarbons and oxygenated substances such as formaldehyde, are suggested to be made mandatory at a later stage.

However, issues related to data gaps do not appear to be a significant problem. A data gap analysis made in this study demonstrated that unless the data gaps were dominated by longer periods with no data, e.g. whole weeks, it is unlikely that the reported annual average concentrations, 8-hour running averages and 1-hour averages, or NOx concentrations would be significantly affected.

Moreover, the study concluded through an analysis of the European context that use of model calculations are useful and should be further encouraged, and within this context it should be considered to what extent fixed site measurements could be substituted by modelling results. The aim would be to reduce the number of monitoring sites at locations where these do not provide any value-added information (e.g. when concentration gradients are small). A better spatial and temporal coverage of the information on organic precursor emissions and concentrations would however be an important precondition for this goal, to ensure as far as possible accurate air quality modelling of ozone and its precursors. Both the opinions of Member States, and the findings of the literature review support the need to investigate and define an appropriate cost-efficient combination of modelling and monitoring of ozone and precursors.

Management of ozone pollution

Rising ground-level ozone in Europe reflects rising background tropospheric concentrations throughout the Northern Hemisphere, caused by increased emissions of the precursors of ozone formation (NOx and VOCs) in all regions in the hemisphere. Once formed, ozone can be transported far between regions and across the whole northern hemisphere. Local and regional management of precursor emissions is reported to have resulted in a reduction in the magnitude and frequency of peak ozone episodes across Europe (Royal Society, 2008; UNEP, 2011; Wilson et al., 2012; HTAP, 2010; EEA, 2012). However, the influence of hemispheric background ozone and the transboundary nature of ozone and its precursors have resulted in mean and 'background' (5th percentile) ozone concentrations remaining more or less constant or even, in some cases, increasing. This explains the continued exceedences of target values.

There is limited consistent or comparable information on Member States' policy responses to ozone exceedences. This may be due to the absence of coherent reporting requirements for ozone in the Directive. The policy focus on reducing precursors to manage ground-level ozone without considering the full picture of ozone chemistry appears to be inadequate, taking into consideration specific local and regional conditions and the complex nature of ozone chemistry in urban areas.

Measures to achieve the NEC Directive (2001/81/EC) NOx ceilings may even result in increased ozone concentrations in some urban areas, where public exposure is greatest. It has therefore been suggested, in accordance with Monks et al. (2012), that a more holistic approach to NOx/NO_2 and ozone management is pursued. Also, the views and experience of Member States suggest that national measures are not sufficient to tackle the issues related to the ozone precursors given their potential transboundary effects. Although international co-operation efforts were reported by some Member States there was little evidence of successful implementation of such co-operative measures, and it was pointed out that efforts to reduce background ozone in Europe are also hampered by the increasing hemispheric concentrations.

The reasons for Member States failing to meet the target values are hence complex and diverse. The increasing transboundary background ozone concentrations can often reach or exceed the target value, yet mitigation is beyond the control of Member States. In conclusion, there is a need for improved global/hemispheric co-operation between countries and within regions for widespread implementation of precursor emission management measures to effectively limit ozone concentrations. Existing legislation is focussed on addressing peak ozone episodes, while the NECD is designed to limit NO_x and VOC emissions and consequently baseline ozone concentrations there is less emphasis on policy measures to address hemispheric background concentrations. Furthermore, the regulation of ozone includes a "non-statutory" target value and therefore the level of priority it is given by Member States may not be sufficient to ensure significant action. While such a binding limit value would give more focus to Member States activities, a binding limit value is not seen as either achievable or desirable by many Member States. Evidence from literature and discovered during this study also indicates that ozone target values and long-term objectives are not technically attainable without disproportionate cost at the Member State level due both to the transboundary nature of the ozone pollution and the increasing global background concentrations.

Given the nature of ozone formation, efforts by Member States to manage NO_2 concentrations (via limiting NOx emissions) to meet the EU NO_2 limit values may have a detrimental effect on mean and background (5th percentile) ozone concentrations in urban areas where large populations are exposed. In order not to increase ozone concentrations, NOx emission controls need to be counterbalanced by sufficient VOC emission controls. This does not appear to be currently happening.

Article 24 of the Directive 2008/50/EC requires Member States to prepare a Short Term Action Plan where alert thresholds for ozone are exceeded, but only where there is significant potential to reduce the risk, duration or severity of the exceedence. Given the complex nature of ozone as a secondary pollutant and the influence of transboundary background ozone concentrations, it can be questioned if there are any actual benefits to the development of Short Term Action Plans. There is no evidence of Short Term Action Plans being developed so far of being successful in the mitigation of ozone pollution and these therefore could be considered a legislative weakness and distraction from more effective long-term actions.

International experiences on ozone management

Learning from other countries is important. Experiences from US, Japan, India and China provide some lessons for consideration.

The US sets strict requirements for the contents of State Implementation Plans (SIPs), and this approach may be considered as a model for more stringent EU requirements for planning to address zones that do not meet air quality standards. The enforcement mechanism between the federal government and states used in the US (withholding transport infrastructure grants) also

deserves attention. Though it may not fit in the EU budget practices, this form of conditionality between air quality measures and funding could be a valuable lever for action also in the EU. In addition, the US also has a voluntary approach to encourage states to go beyond attainment of air quality standards and support the preparation of more stringent future standards. A similar initiative might be considered for the EU. The US provisions to address cross-state ozone transport, while under revision, also merit close attention as cross-border transport is an important issue in parts of the EU.

An interesting Chinese approach to addressing air quality in general is that standards are implemented in a step-wise fashion – first addressing large urban and industrial areas, where air pollution problems are the most serious. Such a priority setting approach could be considered in the EU for non-legislative actions: for example, focusing attention (and possibly, pilot programmes) on local air pollution plans in large urban areas with high ozone levels and directing EU structural fund resources to these areas. Due to the transport of ozone and its precursors, these measures may need to be addressed at wide areas, possibly across several regions and Member States (e.g. the northern Italian plain, Benelux and perhaps surrounding regions), but likewise the benefits may also be felt more widely.

In addressing the sources of emission, Japan uses a voluntary approach to encourage the reduction of VOC emissions from industry, supplementing legal requirements. This programme is articulated at both regional and local levels. While this approach also reflects a specific Japanese policy context, it could be considered as a supplement to EU legal mechanisms. Japan and the US both have stringent standards on precursor emissions from motor vehicles – in the US, there are now standards also for low emissions vehicles – and these bear attention in setting EU standards of similar nature.

In general, the results for all four countries also highlight the importance of monitoring and scientific assessments of ozone trends and transport. Japan's plans to further assess non-anthropogenic sources of VOCs and non-identified VOCs may have lessons for the air quality management in other parts of the world, including the EU.

Japan and the US are also strongly focusing on research, including the development of new modelling systems to understand the formation of ozone and its transboundary movements, in order to take the adequate counter measures on ozone precursors. The EU might consider similar investments in ozone modelling and in cooperating with research in Japan and the US in this field.

Options for further reductions in ozone concentrations over Europe

The conclusion of this study is that the optimal strategy in Europe for reducing ozone concentrations in Europe is to continue reducing NOx and VOC emissions. This has already had a significant impact during the last ten years. According to modelling studies, if these ozone precursor emissions had not decreased in Europe, then ozone concentrations in Europe would have increased significantly instead of being approximately steady state or slightly increased. As mentioned above, the reason for the limited reduction in ozone is that the expected effect of precursor emission reductions in Europe has been counterbalanced by the increased precursor emissions elsewhere and increased hemispheric ozone concentrations.

The other two ozone precursors: CH_4 and CO have relatively long life times: 9-10 years and 2-3 months, respectively. Their effect on ozone is therefore primarily on the hemispheric scale and not on the European scale. Therefore, reducing emissions of CH_4 and CO in Europe will have a limited immediate effect on ozone concentrations within Europe.

The increase in the hemispheric background levels of ozone is primarily due to increased NOx and VOC emissions on the hemispheric scale and this increase in emissions is especially seen in parts of Asia. Also increased CH_4 and CO emissions contributes to the higher hemispheric background ozone concentration, however, the increase observed in these species can only partly explain the overall increase in the hemispheric background ozone concentrations. Since ozone has a lifetime in the atmosphere of about 21 days, ozone produced in the Northern Hemisphere can itself be transported by the wind around the globe. In conclusion, in order to decrease overall hemispheric background concentrations of ozone, the greatest potential thus lies within emission reductions of NOx and VOC in Asia.

Assuming that the implementation of the revised National Emissions Ceilings directive (NEC-II) will continue the reduction of NOx and VOC in Europe towards 2020 and beyond, there is likely to be a positive effect of this in Europe with respect to the future ozone concentrations. Model studies have previously been conducted using the DEHM model (Brandt et al., 2013a; b), comparing the ozone concentrations in the years 2000 and 2007 (using emissions reported to EMEP for the respective years) with 2020 (using agreed emission reduction commitments for 2020 in the amended Gothenburg Protocol). These studies showed a significant and considerable decrease in ozone concentrations, with related positive health impacts in 2020 over Europe.

In this context, it is important to improve the knowledge on which regions in Europe are NOx- or VOC-sensitive (or sensitive to both) for optimizing an emission reduction strategy. In some specific regions in Europe, the optimal strategy is to reduce VOCs, while small or no effects may be seen from reducing NOx. If the opposite is done (e.g. reducing NOx in VOC limited areas), there will be no or little effect on ozone (or even the risk of opposite effect of increasing ozone concentrations). In some regions both NOx and VOC emission reductions are required to reduce ozone. If information is missing on the exact locations of the NOx and VOC sensitive areas in Europe, both precursors have to be reduced across the whole of Europe, which is not always cost effective. The ideal solution for finding an optimal and cost effective emission reductions strategy in Europe with respect to ozone is hence to identify the NOx and VOC sensitive areas and reduce emission of either NO_x or VOC or both species simultaneously in these areas in accordance with this strategy. Since the NO_x/VOC sensitive regimes vary over Europe and evolve over time, to reduce both VOC and NO_x in all regions would most likely not be the optimal strategy. Reducing in a non-optimal way can even result in increases in ozone concentrations. For the purpose of establishing a solid and robust ozone strategy models will have to be used that allows both identification of the main options as well as the study of sensitivities in NOx/VOC reductions. Model sensitivity studies should preferably use a number of different models conducting the same emission reduction scenarios to produce a kind of ensemble and thereby reducing, or at least clarifying, model differences and uncertainties. Emission reductions at the European level are very costly and therefore using state-of-the-art modelling to identify the NO_x and VOC sensitive areas and to calculate reliable impacts from proposed emission reductions is recommended.

The same applies for CO and CH_4. Model studies are needed in order to quantify the impacts from emission reductions of CO, CH_4, VOC and NO_x, both individually and simultaneously, in order to be able to estimate the impacts of emission reductions both on ozone mean concentrations as well as daily maximum concentrations.

Studies with global/hemispheric models are also necessary to improve understanding of the sources and processes contributing to the positive trend in the ozone background concentrations on the hemispheric scale. The studies should focus on analysing the contribution from CO, CH_4, VOC and NOx emissions in different regions of the Northern Hemisphere and on identifying the important chemical and physical atmospheric processes to further understand the source allocation

from both anthropogenic and natural sources and their contribution to the general increase in the hemispheric background.

The current European modelling strategy used to identify optimal ozone precursor emission reduction strategies has some shortcomings. It is based on using the GAINS model to find the optimal reduction strategy utilising EMEP simulations to extract linearised source-receptor relationships. Since the VOC/NOx/CH_4/CO/ozone chemistry is highly non-linear, the extracted source-receptor relationships used in the GAINS model are applicable to the specific scenarios they have been estimated for (e.g. a specific year though accounting for weather variations using five meteorological years, a specific chemical regime over Europe, a specific emission reduction from a single country) and not necessarily applicable for multi-year, multi-species and multi-country emission reduction assessments. It is therefore recommended that the emission reduction strategy is compared to other state-of-the-art regional air pollution models, in order to assess the robustness of the strategy and whether the expected and desirable effect from the emission reduction strategy is obtained.

2 Introduction

2.1 Background

The EU is presently reviewing its air pollution policy. This will include reviews of the 2005 Thematic Strategy on Air Pollution, the EU Air Quality Directive 2008/50/EC (the Directive), the 4th Daughter Directive 2004/107/EC and the National Emission Ceilings Directive 2001/81/EC (NEC D), which according to plans are to be completed by 2013[1].

The Directive established two target values for ozone, one based on the protection of human health (120 µg/m^3 as maximum daily 8-hour mean, not to be exceeded more than 25 days per year averaged over three years) and another for protection of vegetation (AOT40 (Accumulated Ozone over a Threshold of 40 ppb) 18,000 µg/m^3-h averaged over five years). The target values are levels to be attained where possible. They are in effect from January 2010, which means that data from the year 2010 is the first dataset to be used to evaluate compliance over the following three or five years relatively for the two target values. The Directive also established long-term objectives for health (120 µg/m^3 as maximum daily 8-hour mean (no exceedences)) and vegetation (AOT40 of 6,000 µg/m^3-h) but no achievement date were defined. The Directive also set an information threshold of 180 µg/m^3 and an alert threshold of 240µg/m^3 (both hourly averages). The alert threshold requires exceedences to be measured or predicted for three consecutive hours to invoke the requirement short-term action plans under Article 24 of the Directive. Neither the target values nor the alert and information thresholds have been amended since they were first established in 2002.

2.2 Tropospheric ozone

Tropospheric, or ground-level, ozone (O_3) is formed as a so-called secondary pollutant in complex photochemical reactions in the atmosphere. Its formation involves nitrogen oxides (NO_x = NO + NO_2), carbon monoxide (CO), methane (CH_4), as well as both anthropogenic and biogenic volatile organic compounds (VOC).

Ozone is strongly coupled to nitrogen oxides. In the troposphere (the lowest layer of the atmosphere) photo-dissociation of nitrogen dioxide by sunlight is the only source of ozone (in a two-step reaction where oxygen radicals are formed that reacts further with molecular oxygen forming ozone). Close to sources of nitrogen oxides, ozone is depleted in the reaction with nitrogen monoxide leading to formation of nitrogen dioxide. These chemical reactions are sufficiently fast to take place even during the very short residence time inside urban streets (typically seconds to minutes).

In rural areas, chemical reactions that are degrading hydrocarbons lead to the formation of peroxy radicals that in turn convert nitrogen monoxide to nitrogen dioxide without depletion of ozone. These reactions are the causes of elevated ozone concentrations in the troposphere. However, the chemical processes leading to the formation of these peroxy radicals are generally too slow to take

[1] The new air pollution strategy was communicated the 18th December 2013.

place inside the urban domain (Hertel and Goodsite, 2009). The exceptions are episodic events of very low wind speeds in summer time with strong solar radiation in urban areas of e.g. southern Europe; episodes that may be favored by topography e.g. locations inside valleys and including sea breezes – as is known from Athens or the Los Angeles photochemical smog episodes.

Under certain conditions, influx of stratospheric ozone may also take place, and this contribution has previously been estimated to contribute 27% of the concentration in remote high northern latitudes during summer (Mauzerall et al., 1996). In general, long-range transport of ozone and formation of new ozone related to reactions within biomass burning plumes, both play major roles for the levels of tropospheric ozone.

Since ozone is a secondary pollutant formed through complex atmospheric chemical reactions the relationship between precursor emissions and ozone concentrations is highly non-linear and influenced by timescales for ozone formation and the transboundary nature of the pollutant and its precursors. As a consequence of its complex formation, ozone levels often behave in an opposite manner compared with levels of those pollutants that are directly emitted into the atmosphere. As an example the primary anthropogenic pollutants show the highest abundance in or close to urban areas where sources such as traffic and industry prevail, and the lowest levels in low emission rural and remote areas. For the ozone levels, the opposite distribution is often observed.

On a larger European scale, tropospheric ozone levels are generally highest in the south and decrease towards the north; a pattern that is a result of differences in incoming sunlight in combination with differences in the emission densities of ozone precursors. On a regional scale the highest concentrations of ozone are generally measured at rural stations. Somewhat lower levels are usually found at urban background stations, and the lowest concentrations are generally measured close to industrial and heavily trafficked sites, where the fast reaction with NO effectively removes ozone. Because of this chemical depletion, measurements of ozone levels at heavily trafficked urban street stations, primarily serves validation and research purposes, and not the purpose of being part of monitoring in relation to evaluation of the compliance with ozone target values in the protection of human health.

In urban and regional areas in southern Europe, reactive VOCs contribute largely to ozone formation, e.g. aromatic compounds from anthropogenic sources and isoprene and alkenes mainly originating from vegetation. The NO_x/O_3cycle is catalytic and fuelled by VOCs. However, the formation of tropospheric ozone is not linearly dependent on the concentrations of NO_x and VOCs. Rather, the initial concentrations of NO_2 and VOCs as well as the NO_x/VOC ratio, the identity of the abundant VOCs, photochemical aging, including cloud cover, humidity, sunlight and meteorological dispersion, determine the yield of tropospheric ozone (Sillman, 1999). Depending on the actual conditions, the chemistry is VOC sensitive or NO_x sensitive. In the VOC sensitive regime, a reduction (or increase) in NO_x can have little or no effect on tropospheric ozone formation. In fact, a reduction in NO_x in a VOC sensitive regime can in some cases increase tropospheric ozone formation. Still, ambient ozone can be reduced only by reducing emissions of its precursors. A reduction in VOC (and CO) is sometimes the only available means of abatement in the short term, which would typically be the case in central locations of larger cities. On the other hand, rural areas appear to be predominantly NO_x sensitive, though there are exceptions related to season and large urban plumes. A decrease in NO_x may therefore likely cause a reduction in the background tropospheric ozone on the northern hemisphere. An increase in NOx is expected to impact tropospheric ozone formation.

2.3 The current ozone situation

The European data show that even though substantial reductions in emissions of most anthropogenic ozone precursors have been seen this is not reflected in the observed annual average ozone concentrations, which do not show a clear downward trend in Europe between 1999 and 2010 (EEA, 2012). The differences in measured concentrations reported at different station types (rural, urban, high and low altitudes) illustrate the effects of ozone depletion close to local NO sources, with traffic stations reporting the lowest concentrations and rural stations the highest. Ozone levels are typically lower at rural stations at altitudes below 500 m (rural — low) than at mountainous stations at higher altitudes (rural — high). There is no clear trend (1999-2010) in the annual mean at any of the station types, although at rural stations there is a slight decreasing tendency since 2006 (EEA, 2012).

A 2012 report by the European Environment Agency (EEA, 2012) found that the long-term objective to protect human health (maximum daily eight-hour mean concentration of 120 µg/m^3) was exceeded in all EU Member States and in most of the other reporting European countries at least once during summer 2010. For the first time in four years, the information threshold (a one-hour average ozone concentration of 180 µg/m^3) was exceeded in northern Europe. In total 17 Member States are facing difficulties in meeting the target value for protecting human health. In all these countries, the maximum daily eight-hour mean ozone concentration of 120 µg/m^3 was exceeded on more than 25 days during summer 2010. A very obvious demarcation in concentrations was found between northern (low concentrations) and southern (high concentrations) Europe.

An analysis of historical ozone records (Wilson et al., 2012) indicates that tropospheric ozone levels in both hemispheres have increased by a factor of 3-4 over recent centuries. During the past 100 years, tropospheric ozone concentrations in the northern hemisphere have increased two to three fold because of the increase in anthropogenic emissions of its precursors (UNEP, 2011).

EEA (2012) highlighted that in the last decade (2001-2010), due to the implementation of emission control legislation, precursors emissions have fallen significantly in the EU27:

- Carbon Monoxide (33% reduction),
- Non-methane VOCs (27% reduction),
- NO_x (26% reduction) – however, 2010 NO_x emissions remained 12% above the National Emission Ceiling Directive ceiling. (2001/81/EC)
- Methane emissions slowed after 1990 but since 2007atmospheric measurements suggest that emissions have started to rise again (Dlugokencky & Bruhwiler, 2009).

The secondary nature of ozone as a pollutant, the transboundary nature of ozone and its precursors, the complex atmospheric chemistry and the multitude of precursor sources makes the management of ozone challenging. In general, the north-western high-emission area of Europe is VOC sensitive and the southern areas are NO_x sensitive. However, large deviations from this overall rule are seen on the local scale. For example, the high NOx emission areas in Europe in the north-western part are also NO_x sensitive with respect to ozone production from NO_x titration. Furthermore, the relationship between the NO_x and/or VOC emissions and ozone concentrations is in general highly nonlinear (Bastrup-Birk et al., 1997). Since the chemical regimes have changed in Europe over the last 1-2 decades due to changed emission patterns and magnitude, it is very difficult to identify the current impact from emission changes on the NO_x and/or VOC sensitive regions based on the sensitivity studies from the 1990ties and therefore on the future ozone concentrations.

2.4 This study and the structure of the report

Any revision of existing EU legislation will need to carefully evaluate its effectiveness and make recommendations for improvements. Any review and revision of the target values for ozone must consider the existing air quality situation and the potential for reductions of emissions and concentrations in the Member States, as well as the overall progress made in the implementation of EU air pollution reduction measures.

This project “Services to assess the reasons for non-compliance of ozone target value set by Directive 2008/50/EC and potential for air quality improvements in relation to ozone pollution” is a project running from December 2011 commissioned by the General Directorate (DG) Environment of the European Commission to support the review of the implementation of the Directive 2008/50/EC, focusing specifically on ozone.

In addition to providing the overall situation for ozone there as a need for a better understanding of the exceedences of target values, the current overall ozone exposure as well as the environmental and health impacts across the EU, as well as reasons for Member States' difficulties or failure to comply with the ozone standard. Further, it was important to indicate how the compliance situation may likely evolve over time and discuss measures that would be important to implement in order to meet both the target values as well as the long term objectives.

The study was structured in 6 tasks:

- Task 1: Assessment of current air quality situation in the Member States and Croatia for ozone
- Task 2: Review of assessment methodologies for ozone and its precursors, with particular emphasis on measurement and modelling practices
- Task 3: Information on Member States' responses to ozone exceedences on their territory
- Task 4: Assessment of the factors responsible for any projected non-compliance with air quality target values for ozone (O_3) by the dates foreseen in Directive 2008/50/EC
- Task 5: Comparison with international assessment and management regimes for ozone (O_3)
- Task 6: Developing future objective(s) for ozone (O_3)

Task 1 provided inputs and information to several other tasks in particular Task 3 on ozone exceedences, Task 4 on factors for non-compliance and Task 6 on future objectives. The Tasks 1-5 are fundamental for Task 6.

3 Assessment of current air quality situation in the Member States for ozone

3.1 Objective and method

The primary aim of Task 1 was to undertake an assessment of the current air quality situation in the Member States for ozone including its effects on environment and health. This Task includes the following outputs:

- An evaluation of the historical trends in the main sources of emission of ozone precursors for all zones and Member States;
- A determination of historical trends in concentrations of ozone and ozone precursors for all defined ozone management zones in the Member States;
- An evaluation of the levels of ozone and ozone precursors in ambient air to identify if, and why, there is current and projected non-compliance with the target values;
- An estimation of ozone exposure for urban, suburban and rural populations in Member States and the enlarged EU to concentrations exceeding the long term objectives, the target and alert values.

Analysis of ozone precursor emissions

The trend analysis for the emissions of ozone precursors covers all EU Member States. It analyses the trend in emissions of methane (CH_4), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOC) and carbon monoxide (CO). Emissions of these pollutants are analysed for the time period 1990-2010. In addition to the overall trend analysis, the most important source sectors (e.g. road transport, enteric fermentation etc.) are identified and the trends in these subsectors are analysed and discussed separately.

Historical trends in ozone and ozone precursor concentrations

The analysis of monitoring data for ozone and nitrogen oxides considers measurements available in the AirBase database (v6 to the end of 2010 http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-6). A dataset was established considering data quality (missing data, step changes, negative/extreme values), data coverage (at least 10 years of data) and selecting only rural/background stations in order not to get interference from local conditions e.g. high, local emissions of NO_x. This resulted in selection of 286 rural background monitoring stations across 22 countries, over a 15 year period between 1996 and 2010. The methodology used for estimating trends is based on that used by Wilson et al. (2012) and uses the Openair software package (Carslaw and Ropkins, 2012a and 2012b). Openair is based upon the R statistical platform package (www.r-project.org) and employs methods which are statistically more advanced than the standard linear regression commonly used. In particular, Openair provides the functionality to 'deseasonalise' the data to account for differences between summer and winter concentrations.

The analysis was based on an aggregation of data to monthly averages, and only accounted for those months where there was >75% data capture. The trend analyses were undertaken for monthly mean, 95th percentile and 5th percentile concentrations, the latter being indicative of peak and 'background' concentrations respectively (the latter being a term used by Wilson et al. 2012). Significance values were calculated for all trends at individual monitoring stations, and results have

been marked as significant where p<0.1. Mean trends across all monitoring stations within each of the countries have also been calculated.

An assessment has been carried out of trends in the other key ozone precursors concentrations, carbon monoxide (CO) and volatile organic compounds (VOCs). Unlike with nitrogen oxides, there is little relationship between the monitoring sites reporting these and those rural background sites that have been used for the ozone, with the majority of sites for these pollutants in Airbase (v6) being classed as 'urban, 'traffic' or 'industrial' sites. Due to the small number of sites available with long-term datasets, trend analysis has only been undertaken for the 10-year period from 2001 to 2010.

Analysis of monitoring data with respect to objectives, target values, and thresholds

Analyses on the 10-year ozone datasets for 2001-2010 at the 286 rural background monitoring stations considered trends in the following statistics calculated on an annual basis:

- Number of days with maximum daily 8-hour mean exceeding 120 µg/m^3 (target value for protection of human health)
- Number of days where one or more hours exceeds the information threshold (180 µg/m^3)
- Number of hours exceeding the information threshold (180 µg/m^3)
- Number of days where one or more hours exceeds the alert threshold (240 µg/m^3)
- Number of hours exceeding the information threshold (240 µg/m^3)
- AOT40 (Accumulated Ozone exposure above a Threshold of 40ppb (80 µg/m^3) between the hours of 08:00 and 20:00 between May and July)
- Number of hours above the 40ppb (80 µg/m^3) threshold between the hours of 08:00 and 20:00 between May and July

The analyses are presented in the Task 1 report as time series plots showing the total number of days and hours where the target, information, alert and AOT40 thresholds were exceeded and grouped into two sets. The first is showing patterns over the 15-year period from 1996-2010 (based on 138 sites with limited spatial representation), and the second showing patterns over the larger set of 286 sites for the 10-year period from 2001 to 2010. These time series provide some indication of changing patterns in elevated ozone concentrations over the whole of Europe. Maps are shown indicating the trends for exceedences of the various thresholds for the individual sites. These have only been provided for the 2001-10 period due to the better spatial coverage of the sites.

Exposure assessment

The exposure assessment of the population across Europe to exceedences of a range of ozone thresholds (information threshold; alert threshold; long-term objective; target value) is based on three principal data sources:

- Assessment of exceedences, based on hourly model output from the 50 km x 50 km resolution EMEP ozone chemistry model for 2004-2010
- 1 km x 1 km resolution gridded population data from Geostat representing 2006
- Eurostat 'Degree of Urbanisation' (DegUrba) classifications for LAU2 administrative areas for EU-27.

These datasets have been combined in order to estimate the overall exposure to the various exceedence thresholds across Europe, as well as a country-by-country breakdown, and by whether the exposed population can be classified as urban, suburban or rural (based on the Eurostat DegUrba classification). Within the analysis both population and 'Degree of Urbanisation' have been kept constant, so that any variation in exposure from year to year can be attributed to variation in (modelled) ozone concentrations. The assessment has been undertaken for 31

countries that are covered by all 3 datasets (population, urbanisation, and modelled ozone). The 31 countries are: Andorra, Austria, Belgium, Bulgaria, Switzerland, Czech Rep., Germany, Denmark, Estonia, Spain, Finland, France, Greece, Hungary, Ireland, Iceland, Italy, Lichtenstein, Lithuania, Luxembourg, Latvia, Malta, Netherlands, Norway, Poland, Portugal, Romania, Sweden, Slovenia, Slovakia and the UK.

3.2 Trend analysis for ozone precursor emissions

The overall trend in ozone precursors for EU27 (Table1) shows that the main reductions in emissions happened in the first part of the time series while the rate of decrease slowed considerably in the latest years of the time series.

Table 1: Total CH_4, NO_x, NMVOC and CO emissions (Gg) from the EU27

EU27	1990	1995	2000	2005	2006	2007	2008	2009	2010	Reduc-tion 1990-2010	Reduc-tion 2000-2010	Reduc-tion 2005-2010
CH_4	28 112	25 422	22 870	20 502	20 188	19 858	19 653	19 242	19 081	32.1 %	16.6 %	6.9 %
NO_x	17 143	14 712	12 644	11 587	11 275	11 006	10 136	9 293	9 162	46.6 %	27.5 %	20.9 %
NMVOC	16 740	13 211	10 698	8 831	8 591	8 312	7 913	7 378	7 412	55.7 %	30.7 %	16.1 %
CO	65 670	50 677	38 928	29 683	28 478	27 576	26 631	24 091	24 908	62.1 %	36.0 %	16.1 %

For **NMVOC and CO** the transport sector has achieved such large reductions that the share of total emissions has been greatly diminished. Even though emission reductions in the transport sector will continue, measures will have to be implemented in other source categories e.g. small combustion for NMVOC and CO and solvent use for NMVOC if the rate of decrease for the total of all sectors is to be maintained. The share of total NMVOC emissions for small combustion doubled from 1990 to 2010. Also, the share for solvent use increased by almost 50 %. For CO, the largest source by far is now small combustion, where emissions have only been reduced by 13 % from 1990 to 2010 compared to the 79 % reduction achieved for the transport sector.

For **CH_4** the agricultural sector has increased its importance due to a decrease in the emissions from coal mining and handling. The dominant source categories therefore continue to be agriculture (enteric fermentation and manure management) and solid waste disposal on land. In many countries measures are enacted to limit (or even banning) the landfilling of organic degradable waste. That combined with increased attention on recovering the CH_4 and either flaring it or using it for energy production will lead to a continued decrease in the emissions from solid waste disposal on land. The reduction in other sectors will probably mean that agriculture will have an even higher share of emissions in future years despite any reduction measures implemented in the sector.

For **NO_x** the sectoral distribution is largely consistent between 1990 and 2010. This is the case for energy industries, manufacturing industries and transport. This combined with the significant reduction in emissions indicates that efficient measures have been implemented for all these categories, such as low-NO_x burners, DeNO_x and three-way catalysts. It is expected that these measures to a large degree have been implemented and therefore will provide a more limited reduction potential in future years.

3.3 Analysis of trends in monitoring data for ozone

The findings of the initial analysis of trends in monitoring data for ozone are based on an analysis of 286 rural background monitoring stations covering the 10 year period 2001-2010 and 138 of these stations covering an additional 5 years covering in total 15 years from 1996 to 2010.

In general there are no clear patterns across the whole of Europe that are observable over the full 15 years covered by this study and there is no clear, single trend in ozone concentrations. There are however a number of features that can be identified. These include:

- Overall trends over the 15 year period 1996-2010 are somewhat unclear. However, concentrations in north-west Europe (UK, Germany, Netherlands) have tended to increase, whilst concentrations in central Europe and Scandinavia (Czech Republic, Sweden, Finland) have tended to decrease.
- De-seasonalised monthly mean concentrations tended to increase over the period 1996-2005, whilst decreasing 2001-2010. When the period is split into two (overlapping) decadal periods, 1996-2005 and 2001-2010, a clear difference in observable trends is revealed, with the earlier period showing a general tendency for concentrations to increase, whilst the latter shows a general tendency for concentrations to decrease. There is a possibility that this is an effect generated by the very high concentrations experienced in 2003. A sensitivity test has been carried out with respect to this, and it appears the effect of high concentrations in this year tend to considerably increase trends for the 1996-2005 period, but have much less effect on the second 2001-2010 period. Discounting 2003, there is a general pattern for sites that have an upward trend in the first period to also have an upward trend in the second, and those with a downward trend in the first to also have a downward trend in the second.
- During this latter period, de-seasonalised monthly mean and 5th percentile (background) concentrations tended to reduce, but widespread reduction was less clearly demonstrated for 95th percentile (peak) concentrations.
- Monitoring stations recording the highest 10/15 year mean ozone concentrations (>60 $\mu g/m^3$) tended to show the greatest downward trends over the whole 15 year period.
- Trends are not consistent over the whole of Europe, and there is a tendency for trends to have increased in the areas of lowest 10/15 year mean concentration (northwest Europe), and to have decreased in the areas of highest 10/15 year mean concentration (southeast Europe). There are notable exceptions to the general downward trend from 2001 to 2010, which include Spain, the UK, northern Germany and northern Italy.
- Highest mean concentrations over the 10/15 years period tend to be in southern and eastern Europe, with the continent divided by a line running approximately from northwest Poland to the western end of the Pyrenees on the basis of mean concentrations and trends. However, highest maximum 1-hour and 8-hour concentrations tend to show a different pattern, and are orientated along a perpendicular line from the south coast of the UK and the Netherlands, down towards Italy. Generally trends in mean and maximum concentrations tend to be inversely related with mean concentrations increasing where maximum concentrations are decreasing.

3.4 Analysis of monitoring data with respect to objectives, target values, and thresholds

When moving beyond analysis of simple concentrations into exceedence statistics, patterns become much more complex and there is a wider range of criteria for judgement. Some of the patterns, which can be identified from the analyses, include the following:

- Whilst there is a discernable downward trend in some of the exceedence statistics (especially the target value and AOT40), this appears to be relatively insignificant in the context of year-to-year variations due to changing meteorological conditions between years.
- The extent to which trends are evident is highly dependent on the chosen metric, e.g. average concentrations or exceedences of particular thresholds.

- Whilst most of the time series plots show 2003 as the most extreme year, it is notable that in certain cases 2006 appears worse. This is particularly the case with respect to exceedences of the alert threshold (at 286 sites) and the AOT40 concentrations, but not for days where maximum 8-hour mean exceeds target value, exceedences of information threshold and exceedences of the alert threshold (at 138 sites). The difference between the pattern in alert thresholds when considering the 286 and 138 site groups highlights the care that needs to be taken with regard to the representativity of any of the samples.
- Whilst there has been a general downward trend across most of Europe in terms of both exceedences of the target value and AOT40 concentrations, the pattern is much less strong for exceedences of the information threshold.

It should also be noted that whilst 2003 and 2006 clearly appear to be unusually high years (due primarily to meteorology), it should also be considered that there will similarly be unusually low years. Taking this into consideration, it could be possible that 2007, 2008 and 2009 represent low years due to meteorological conditions rather than any reduction in emissions.

3.5 Population exposure assessment

The analysis showed that there is a significant level of ozone exposure across Europe, with two thirds of the European population being exposed to exceedences of the information threshold, and over 99% of population exposed to exceedences of the long-term objective. About 12% of the population across Europe has been exposed to exceedences of the target value in a year in the 2001-2010 period.

Urban exposure in terms of number of people exposed to exceedences is higher for all four metrics than for rural population, and particularly if urban and suburban areas are grouped, it is clear that in terms of human health, ozone pollution poses a disproportionate burden on 'non-rural' populations considering the number of people affected. This is found even though the rural concentration level in general is the highest.

The year 2006 was the worst year for the numbers of people affected by exceedences of the information threshold for all three Urban/Suburban/Rural (USR) classifications. The same was the case for exceedences of the alert threshold, both overall and in urban and suburban areas, but not in rural areas, where exceedences affected more people in 2005 and 2008.

Romania is the main country affected by exceedences of the alert threshold, but Switzerland, Portugal, Romania, Bulgaria, Italy and France also have notable populations exposed to exceedences of the alert threshold.

The difference in having the permitted exceedences days (<=25 exceedences averaged over 3 years) between the long-term objective and the target value has a very significant impact on exposure – from almost universal exposure to exceedences of the long-term objective, to comparatively very low exposure to exceedences of the target value, except for Italy and to a lesser extent Switzerland, Slovenia, France, Greece, Austria and Malta.

3.6 Conclusion

The EU total emissions of ozone precursors have decreased substantially in the range of 30-60% from 1990 to 2010. The main reductions happened in the first decade of this period, while the rate of decrease slowed considerably in the later years. The distribution between source sectors has

changed for some of the ozone precursors. In order to achieve further reductions, additional measures will have to be implemented in sectors other than those traditionally regulated.

Mean concentrations (2001-2010) tend to be highest in southern and eastern Europe, whereas the highest 1-hour and 8-hour mean concentrations tend to show a different pattern with the highest concentrations mainly lying in a north-south band across central Europe.

There is no clear single trend in ozone concentrations for all considered metrics from 1996-2010 across Europe, though concentrations in north-west Europe have tended to increase, and in central-eastern Europe and Scandinavia to decrease. There is a tendency for ozone concentrations to have increased in areas of lowest mean concentration (north-west Europe) and decreased in areas of highest mean concentration (southeast Europe).

When also considering exceedence statistics, patterns of trends become even more complex. There is a discernible downward trend in some exceedence statistics (particularly target value and AOT40), but year-to-year variations in meteorological conditions disturb this picture. The years 2003 and 2006 appear as unusually high years primarily due to meteorology those years, but similarly there are also unusually low years, which might be due to opposite meteorological factors e.g. 2007, 2008 and 2009.

Analyses covering the period 2004-2010 estimate that around 12 % of the European population is exposed to exceedences of the ozone target value in force since 2010, two thirds to exceedences of the information threshold, and over 99% of the population exposed to exceedences of the long-term objective. In terms of number of people exposed to exceedences, urban exposure is higher for all considered metrics than for the rural population. Therefore, although rural concentrations are, in general, the higher than urban concentrations, fewer people are exposed to levels considered in the Directive.

4 Assessment methodologies for ozone and its precursors - measurements and modelling practices

4.1 Objective and methods

The primary aim of Task 2 was to undertake a review of assessment methodologies for ozone and its precursors, with particular emphasis on measurement and modelling practices. This Task includes the following outputs:

- A review of information provided by Member States in their annual reporting on current levels of ground-level ozone and precursors.
- An evaluation if the Data Quality Objectives (DQO) for monitoring and modelling are met for ground-level ozone.
- An assessment if the DQO are sufficient and sound for the correct implementation of the ozone provisions of the AQ Directives.
- A compilation and analysis of the difficulties encountered by the Member States with regard to siting criteria, number of stations, quality assurance and quality control.
- An analysis of the weaknesses and strengths of the current reference methods.
- An assessment of the continued relevance of reference measurement techniques in relation to requirements for near real time public dissemination of air quality information.

Compliance with DQO was evaluated on the basis of statistics derived from the measurements stored in Airbase database (v6 to the end of 2010 http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-6), and the results of this analysis were combined with information obtained from what the Member State reported in a questionnaire survey.

Monitoring of ozone levels are mainly carried out on half-hourly or hourly time resolution at rural locations as well as in urban background. In order to assure appropriate and harmonized monitoring of ozone levels, as well as in order to properly support the understanding of its sources and sinks, EU Member States are obliged to apply the Directive 2008/50/EC on ambient air quality and cleaner air for Europe. For a forthcoming revision of the Directive, it would be important to evaluate to what extent the particular objectives of this Directive have been fulfilled including the relevant annexes.

In the work presented in this report, this was evaluated by means of:

- Information obtained from Airbase, the EU database on air quality. Data capture and related parameters were calculated from extracted data. The data capture analyses have been performed on all data available in Airbase 6. Furthermore, a data gap analysis was carried out on basis of full datasets for specific measurement stations selected to be representative for southern, central and northern parts of EU.
- A literature survey of measurements techniques including reference methods was conducted for ozone and ozone-related NO, NO_2, and organic ozone precursors VOCs.
- A Questionnaire distributed to the Member States was prepared aiming at obtaining information concerning their experienced difficulties with siting criteria, zones and number of sites, data quality objectives (DQO), and their experience with the use of modelling results for

ozone, ozone-related NO_x and organic precursors. The survey was intended to complement the information derived from Airbase and the literature survey. The questionnaire was distributed to the point of contacts in the Ambient Air Quality Committee representing the EU Member States in March, 2012. About 60% of the questionnaires were returned.

The effect of data gaps on ozone annual mean, AOT40 and 8-hour running average was evaluated by use of rural background data from Denmark, Austria and Spain. The selection of the three stations aimed at representing the ozone situation in South, Central and North European rural background. The measurement stations selected for the gap analysis were characterised by data captures around 95%. In order to study the effect of imposed data gaps, it was necessary to fill the gaps prior to introducing synthetic data gaps of 5, 10, 15, 20 % and whole months of no data. By use of this gap-filling procedure, three complete datasets were created. In the following step, random data gaps were introduced in the datasets according to three different scenarios for data loss. First February, then July data was removed from the datasets to evaluate the effect of extreme data loss, i.e. a month with low and high ozone concentrations. In the next two scenarios, one-hour values and whole weeks were omitted corresponding to 5, 10, 15 and 20% data loss. Three repetitions of this calculation procedure were performed to evaluate the variation in the modelled outcomes.

4.2 Compliance with data quality objectives

The analysis on Airbase extractions indicated that a data capture of 90% was not met at all sites. However, in this analysis it was not possible to accurately determine the number of measurement stations not in compliance with the required data capture for the following reasons:

1) Information about the time spent on calibration and routine maintenance, which the directives allow to compensate for when calculating data capture, was not available for the analysis;
2) No information was available on the identity of measurement stations, which were discontinued or established in 2010.

However, the analysis showed that on average 81% and 77% of the EU measurement sites for ozone and NO_2 fulfilled the requirement of 90% data capture. Data capture for organic precursors of ozone is not specified in the Directive as it only provides a list of recommended compounds to be measured. Only 39% of the measurement sites met the requirements 90% data capture for benzene, which is a regulated carcinogenic substance in addition to being an organic precursor of ozone that the Member States are obliged to measure. The corresponding number for the organic ozone precursor isoprene was 7%. Organic precursors of ozone are poorly covered and in general characterized by a low data capture. This situation could probably be greatly improved in case the list of recommended organic ozone-precursors in Directive was replaced at least partly by a mandatory list.

The Airbase analysis had limitations due to the lack of a notation key for precise characterisation of the type of measurement site for the stations in the data-base. The lack of a key that could be used to separate the stations into different measurement site categories meant that this analysis had to be performed with all available sites included in one single batch. Furthermore, the tools in Airbase did not provide options for extraction of data separated into monthly or season-wise datasets. For this reason, it was not possible to account for the fact that the Directive specifies lower data capture requirements for winter seasons. The analysis of extractions from the Airbase has thus been performed for overall data coverage regarding the entire year and for all types of measurement sites.

In the questionnaire survey, the Member States were requested to specify wishes in relation to the revision of the Directive. According to the survey, only a few of the Member States have experienced difficulties with siting criteria. Among the improvements requested by Member States was a more clear definition of exactly where and when model calculations could substitute fixed site measurements in air quality assessment. Furthermore Member States suggested that modelling should be accepted as a substitute for the high number of fixed measurement sites in large and sparsely populated areas.

4.3 Use of air quality modelling

Only seven Member States reported that they are using modelling data for (at least) ozone. A conclusion supported by the Member States is that the use of model results as a substitute for fixed site measurements may be justified in large areas with low population density and fairly uniform ozone concentrations. This is especially expected to work well in areas with rather uniform emissions and meteorological conditions. This is suggested as a way to reduce costs for currently required measurements.

Concerning use of air quality models, the Directive states that: “The results of modelling and/or indicative measurements shall be taken into account for the assessment of air quality with respect to limit values”. According to the FAIRMODE (Forum for air quality modelling in Europe) community, this formulation is “rather vague”. The role of computational models in air quality assessment is thus not very clear in the Directive, and this may be one of the main reasons why only few Member States are making use of modelling data in their air quality assessment reports (http://fairmode.ew.eea.europa.eu/).

In general the evaluation of the uncertainty associated with the use of modelling results appears to be at a preliminary stage. The accuracy of model calculations depends heavily on the available input of relevant measurements of atmospheric species. One prerequisite for more accurate model calculations could be extended measurements of organic precursors of ozone. The observed poor coverage as well as poor data capture is, however, a problem in this regard. For this reason, improved characterization as well as quantification of emissions, including those related to natural sources from e.g. vegetation and wildfires, is strongly encouraged.

A conclusion that can be drawn from reviewing several modelling studies is that the regional models may serve particularly useful in assessment of the highest exposures to ozone concentrations for which models show the best accuracy, as well as in source apportionment and impact assessment of reduction strategies. Naturally models serve as strong tools in assessment of ozone levels where measurements are not available. Ensemble models are likely to play stronger roles in the future and so is the use of data assimilation.

4.4 Measurement uncertainty and data gaps

Regarding uncertainty values for ozone measurements in the EU derived from the available ‘Intercomparison Exercise’ reports for the time period 2007-11, it can be concluded that there are no significant problems in terms of meeting the requirements for ground-level ozone. For benzene and isoprene, expanded measurement uncertainties were evaluated based on the returned questionnaires. All responding Member States reported uncertainties lower than or equal to 25%, but the actual number of responses is insufficient to conclude upon in a general EU wide perspective.

An analysis of how data gaps may affect the annual average ozone concentration showed that the annual average, 8-hour running average and AOT40 were only to a minor degree affected by data loss up to 20%. In contrast to this, most parameters changed significantly if the data loss corresponded to complete absence of data in July, where the highest ozone concentrations were observed. It should here be noted that the Directive accounts for this issue by specifying higher data capture during summer. The way data loss is compensated for in the calculation of AOT40, may furthermore increase the reported AOT40 during periods characterised by low ozone concentration.

Under the assumption that data loss is mostly in the form of shorter time duration, and unlikely to be dominated by whole weeks, the majority of observed data captures for ozone are not expected to significantly affect annual averaged concentrations, ozone-related 8-hour continuous average and AOT40. The same is true for the average concentrations of NO and NO_2. As long as the annual data capture is better than 80% and better than 90% during the summer month, measures related to NO, NO_2 and ozone are unlikely to be affected. On the other hand, the poor data capture, which in general is reported for organic precursors of ozone, are most likely to affect the annual averages.

4.5 Methods and reference methods

For the measurement of ozone and NO_x about 90% of the instrumentation is based on the reference method.

In connection with warnings of the public during episodes of high ozone concentrations it is important that the method used can provide data fast enough. It is estimated for 2010 that for ozone 93% of the measurements in EU are based on the reference method. In principle the application of the reference method for ozone by ultraviolet photometry at monitoring stations can provide fast results for the purpose of warning the public. The required calibrations can be performed automatically and the measurement data can be transferred electronically directly to the different public authorities at best within a few minutes after the actual measurement has been taken. These quickly transferred data have often not undergone full QA/QC procedures, which are also part of the reference method. Therefore these data are typically presented to the public with higher uncertainties than data where all the QA/QC and post processing have been performed. Adjustments of data for automatic (zero and span) calibrations will however typically only cause minor corrections (typically within 5% estimated from experiences in the Danish AQ monitoring programme) for the final ozone data as ozone monitors based on ultraviolet photometry in general are very stable. This means that the use of the reference method for ozone measurements with regard to near real time dissemination of data to the public during episodes of high ozone concentrations (according to the Directive 2008/50/EC article 26 and annex XVI) can relatively easily be achieved. It is therefore recommended to use the reference method for this purpose.

While no reference methods are specified for measurements of organic precursors of ozone, no less than four reference methods are available for the measurement of benzene, as this is a regulated carcinogenic compound. Dynamic sampling followed by thermal desorption or solvent extraction, either automated or off-line analysis with a time resolution not exceeding 24 hours, or PTR-MS (Proton Transfer Reaction mass spectrometry) are therefore recommended for analysis of organic precursors of ozone.

An analytical system for monitoring VOCs relevant as ozone precursors must meet low detection limits, high accuracy and precision in addition to being reliable for daily usage. Furthermore, the required time resolution dictates whether automated systems or off-line systems are applicable. Based on these considerations, systems with detection limits exceeding 0.1-1 $\mu g/m^3$ appear to be

of limited use. Furthermore, passive sampling techniques with a time resolution exceeding 24 hours may be adequate for benzene in some cases, but insufficient when it comes to monitoring ozone organic precursors and their trends for checking the consistencies of emission inventories and the efficiency of emission reduction strategies. With respect to the latter issue, VOCs should be differentiated into single species and not integrated measures such as non-methane VOC (NMVOC) or total VOC (TVOC).

4.6 General recommendations

The use of model calculations should be encouraged, and within this context it should be considered to what extent fixed site measurements could be substituted by modelling results.

The poorly covered organic precursors of ozone, for which the current analysis has shown measurements to be characterised by a low data capture, are identified as a major weakness in fulfilling the ozone organic precursor monitoring provisions. Such monitoring is central in the understanding of ozone formation in urban and suburban environments as well as in the background atmosphere.

Furthermore, a better spatial and temporal coverage of emission and concentration of organic precursors of ozone is a prerequisite for accurate air quality modelling of ozone.

A weakness in the directive appears to be that, except for benzene, monitoring of listed organic precursors of ozone is only recommended, and not mandatory. In this study, it is recommended to at least partly replace the recommended list of organic precursors of ozone in Directive 2008/50/EC by a mandatory list of compounds to be measured: i.e. the C5-C9 substances, which can be analysed using the automated GC (“BTEX monitors”) or laboratory GC-MS analysis of adsorbed substances. Other species with high ozone formation potential, e.g. short-chain hydrocarbons and oxygenated substances such as formaldehyde, are suggested to be made mandatory at a later stage.

5 Information on Member States' response to ozone exceedences on their territory

5.1 Objective and methods

The primary aim was to assess available information on Member States' response to ozone exceedences on their territory. This Task includes the following outputs:

- An assessment of measures taken by Member States to reduce ozone concentrations, including implementation of programmes prepared pursuant to Article 6 of Directive 2001/81/EC or air quality plans under Directive 2008/50/EC.
- Consideration of compliance with the obligation in Article 19 of Directive 2008/50/EC on information of the public in case of exceedence of the information and alert thresholds specified in Annex XII of the Directive, together with an assessment of best practice in information.
- Consideration of compliance with the obligations in Article 24 of Directive 2008/50/EC on short-term action plans in case of exceedence of the alert thresholds specified in Annex XII of the Directive.
- An assessment of the number and nature of initiatives on transboundary co-operation pursuant to Article 25 of Directive 2008/50/EC.

Through a search of the EIONET Central Data Repository, reports held by the Commission, internet searches, and direct communication with Member States, a total of 51 reports were identified for review covering nine Member States. Seven of the reports were deemed of definite interest, 25 of possible interest and the remainder unlikely to be of interest. All reports deemed definitely likely to contain action plan measures, and all English reports that were of possible interest, were appraised using an information pro forma checklist, and information relevant to the contract was extracted into a database. Non-English reports were translated with the assistance of online translation tools, however there were problems encountered where file sizes and types precluded translation in this way. Non-English language reports that were not able to be translated (two Belgian, two Spanish and one Slovak) were sent to linguists at Milieu and DCE in order to complete the checklists. Information from all of the checklists was then collated into a database and a summary document for each Member State produced. Out of the nine Member States for which reports were had been acquired, only seven reports were identified as being considered relevant for to ozone (Belgium, France, Germany, Malta, Slovakia, Spain, Sweden). The reports were analysed in terms of Measures taken, Long-term effectiveness, Short-term effectiveness, Background effectiveness, Hotspot (urban) effectiveness, Estimated cost, Public alerts, Short-term action plans, and Transboundary cooperation.

5.2 Reports and information

The reports obtained via EIONET, the Commission, direct contact with Member States and internet searches represent a sample of just 26% of the EU-28 Member States, but with a widespread geographical coverage. It is not clear whether the low number of Member States represented is due to the lack of accessibility of reports, their non-existence, or the failure of Member States to cooperate in sending requested information. However, either way, caution must be used in drawing representative conclusions from this small sample.

Overall there was found to be little consistency in the existence or content of reports across Member States, which may be due to the absence of coherent reporting requirements or assessment and follow-up by the Commission where reports are submitted. Findings of the review suggest a division of opinion between Member States as to the significance and nature of ozone problems. At one extreme there are Member States that do not currently experience significant exceedences and who believe that the requirements of the National Emission Ceilings Directive are likely to maintain this situation; at the other there are those that experience regular exceedences and attribute these, at least in part, to both extreme meteorological conditions (in terms of temperature and insolation) and exposure to significant transboundary transport of pollution (either ozone or its precursors).

5.3 Measures, actions and plans

There is little evidence for the existence of sophisticated management strategies for ozone. Most reported actions rely on the reduction of precursors (either in the short-term or long-term) with no significant analysis of the effects. This focus on precursors appears in many cases to lead to potential or actual increases in local ozone concentrations, particularly in urban locations.

Of the reports reviewed most measures were traffic-related and based simply on reduction of overall ozone precursors with little or no consideration of chemistry or source attribution. The reports covered a wide range of dates (2002-2011) making it difficult to draw comparisons between their reported temporal and spatial effectiveness. Information on the costs of measures was extremely limited with three Member States providing partial or aggregated data only.

The role of transboundary pollution is strongly recognised by Member States, and intentions to cooperate in this area were widely stated. However, little evidence was identified to suggest that much significant activity was actually occurring.

Most of the Member States included in this study reported moderate short-term effectiveness of measures to reduce ozone. However, there appeared to be a strong opinion from Member States that long-term targets (up to 2020) would not be achievable within the current legislation due to the transboundary nature of ozone and its precursors and increasing hemispheric background concentrations. In urban areas, however, the focus on reducing emissions of ozone precursors was reported as resulting in an upward trend in ozone due to the reduction in NO from traffic.
The response to the requirement to instigate public alerts and short-term plans in the event of alert threshold exceedences varied widely across the Member States reviewed. Belgium and France (e.g. Strasbourg) reported stringent measures that would be implemented in the event of a severe ozone episode. However, Germany and Spain have not developed any plans as they assume that the risk of exceedences of the alert threshold is low, and that measures already implemented in response to the NEC Directive will maintain this situation (in contrast to exceedences of the target value and long-term objective).

5.4 Conclusion

In summary, this review found that there is little consistent or comparable information on Member States' responses to ozone exceedences. The policy focus on reducing precursors to manage ozone appears to be inadequate taking into consideration the complex nature of ozone chemistry in urban areas, which suggests evidence that measures to achieve the NEC Directive may even result in leading to increases in urban ozone concentrations (where public exposure is greatest). Member States also appear to experience that national measures are not sufficient to tackle these

precursors given their transboundary effect, though international co-operation efforts were cited, and that efforts to reduce background ozone are hampered by increasing hemispheric concentrations.

6 Assessment of the factors responsible for any projected non-compliance

6.1 Objectives and methods

The primary objective of this Task is to identify, describe and where possible quantify the reasons or factors responsible for projected non-compliance with air quality target values for ozone. This task considered technical issues and factors such as:

- Failures of source-based emission control legislation;
- Structural weaknesses in air quality legislation; and
- Structural deficiencies in Member States approaches to air quality management

The Task includes the following outputs:

- An assessment of whether the target values and long-term objectives are technically attainable without disproportionate action costs.
- Consideration of any systematic short-comings in the Directive, which lead air quality managers and/or competent authorities to be poorly prepared.
- Consideration of any systematic short-comings in how Member States organise their administrative management structures which have led to insufficient action being taken.
- An assessment of any short-comings in EU source legislation to deliver the expected levels of air pollutant emissions reductions.
- An assessment of the short-comings in international conventions to deliver the required levels of air pollutant emissions reductions.

The task has been delivered using a research triangulation methodology utilising the expertise of the Project Team, a literature review specifically focussed on the task objectives and six Member State case studies to understand the reasons or factors responsible for non-compliance. The observations of the Project Team and the findings of this literature review are considered against the outcomes from Tasks 1, 2 and 3 and the case study interviews undertaken with selected Member States. The case studies were chosen to identify any systematic differences between Member States' experiences and therefore it was important to ensure that the selected case studies were:

a) Geographically diverse;
b) Influenced differently by transboundary ozone and precursors; and
c) Had differing levels of capacity, capabilities and air quality management frameworks.

The six case study Member States chosen include:

- Western Europe – Ireland (IE)
- Northern Europe – Sweden (SE) and Lithuania (LT)
- Southern Europe – Malta (MT) and Croatia (HR)
- Central Europe – Austria (AT)

The following information has been used in performing this Task:

- Air quality plans and programmes prepared by Member States and those involved in air quality management in the Member States (Task 3);

- The UNEP integrated assessment of black carbon and ozone;
- Documentation on ozone produced by the Task Force on Hemispheric Transport of Air Pollution under the Long Range and Transboundary Air Pollution Convention;
- The UK Royal Society Report on Ozone (Royal Society, 2008);
- Relevant peer-reviewed and "grey" literature;
- Contractor's expertise and familiarity with EU ambient air quality and source legislation and Member States management practices; and
- Consultations with relevant stakeholders (including those involved in air quality management) and open literature.

6.2 Failures of source based emission control legislation

Emission control legislation in Europe has achieved substantial reductions in ozone precursors over the last two decades but the issues of non-compliance with the target value among Member States continue. The key observations and factors for non-compliance are as follows.

The local / regional management of precursor emissions appear to have resulted in a reduction in the magnitude and frequency of peak ozone episodes across Europe. However, the influence of baseline hemispheric ozone and the transboundary nature of ozone and its precursors has resulted in mean and background ozone remaining constant or in some cases increasing across Europe hence the continued exceedences of target value.

The increasing transboundary background ozone concentrations can be approaching or exceeding the target value but mitigation is beyond the control of Member States.

In conclusion there is a need for improved global/hemispheric co-operation between countries and within regions for widespread implementation of precursor emission management measures.

6.3 Structural weakness in air quality legislation in relation to Ozone

Existing legislation is focussed on addressing peak ozone episodes with less emphasis on policy measures to address hemispheric baseline concentrations.

Ozone is regulated by a non-statutory target value and therefore the level of priority it is given by Member States may not have been sufficient to ensure significant action. The implementation of a binding limit value would give more focus to Member States activities but a binding limit value is not seen as possible to achieve or desirable by many Member States.

Given the complex nature of ozone as a secondary pollutant and the influence of transboundary baseline ozone concentrations, it needs to be considered if there are any actual benefits to the development of Short Term Action Plans, if their implementation actually results in the reducing of the ozone exceedence risk or if they are just burden on Member States that actually chose to develop them? There is no evidence of Short Term Action Plans developed so far of being successful in the mitigation of ozone and therefore could be considered a legislative weakness. The local/regional management has as stated above resulted in ozone precursor emission reductions, but there is no evidence that this can be attributed to Short Term Action Plans.

Given the nature of ozone pollution, the race by Member States to manage NO_2 concentrations (via NO_x emissions) may have a detrimental effect on mean and background ozone concentrations in urban areas where large populations are exposed. NO_x emission control needs to be

counterbalanced by sufficient VOC emission control. This does not appear to be currently happening.

Evidence from the Task 4 review indicates that the ozone target value and long-term objective are not believed by Member States to be technically attainable without disproportionate action costs at a national level (see case study interviews in Task 4 report).

6.4 Structural deficiencies in Member State approaches to air quality management in relation of Ozone

The investigations and analyses did not identify any structural deficiencies in Member States' approaches in relation to management of ozone, However, this is based from indicative evidence on a limited number of case studies interviews across 6 Member Statements.

6.5 Conclusions

Emission control legislation in Europe has achieved substantial reductions in ozone precursors over the last two decades, but the issues of exceeding target value among Member States continue. The key observations and factors are as follows.

- The local / regional management of precursor emissions appears to have resulted in a reduction in the magnitude and frequency of peak ozone episodes across Europe. However, the influence of background hemispheric ozone and the transboundary nature of ozone and its precursors has resulted in mean and background ozone remaining constant or in some cases increasing across Europe hence the continued exceedences of target value.
- The increasing transboundary background ozone concentrations can be approaching or exceeding the target value but mitigation is beyond the control of Member States.
- Evidence suggests that there is a need for improved global/hemispheric co-operation between countries and within regions for widespread implementation of precursor emission management measures.
- Existing legislation is focused on addressing peak ozone episodes with less emphasis on policy measures to address hemispheric background concentrations.
- Ozone is regulated by a non-statutory target value and therefore the level of priority it is given by Member States may not be sufficient. The implementation of a binding limit value would give more focus to Member States activities but a binding limit value is not seen as possible to achieve or desirable by Member States.
- Given the complex nature of ozone as a secondary pollutant and the influence of transboundary baseline ozone concentrations, it needs to be considered if there is any actual benefits to the development of Short Term Action Plans, if their implementation actually results in the reducing of the ozone exceedence risk or if they are just a burden on Member States that actually chose to develop them. There is no evidence of Short Term Action Plans developed so far of being successful in the mitigation of ozone and therefore could be considered a legislative weakness and a distraction from more long-term actions.
- Given the nature of ozone pollution, the race by Member States to manage NO_2 concentrations (via NOx emissions) may be having a detrimental effect on mean and background ozone concentrations in urban areas where large populations are exposed. NOx emission control needs to be counterbalanced by sufficient VOC emission control. This does not appear to be currently happening.
- Evidence indicates that the ozone target value and long-term objective are not technically attainable without disproportionate action costs at a Member State level.

7 International comparison

7.1 Objective and methods

The objective of this task was to prepare overviews of ozone management regimes in four countries outside of Europe: China, India, Japan and the United States. The case studies address the following topics:

- Legislative framework for assessment and management of ozone;
- Current status of air quality on ozone;
- Reasons for compliance/non-compliance;
- Trends in policy on ozone;
- Lessons to be learned for EU policy.

Information was gathered through desk research, together with phone interviews of experts for each country. At least two interviews were sought for each of the four countries: one official in a national authority and one representative of a stakeholder, such as an NGO or research institute working on ozone issues.

Several issues were encountered in the preparation of the case studies. For China and Japan, it was difficult to identify national officials and experts who could be interviewed in English. As a result, both interviews for China and one interview for Japan were made with experts from other countries who studied air pollution in China or Japan. For India, although several experts and officials were identified in the inception phase, it was not possible to obtain their assent to interviews; as a result, this case study in particular has suffered from a lack of information. In contrast, the US case study faced the opposite problem – a large number of reports are available, and information gathering tried to focus on those that appeared most valuable.

The results are synthesised in four country case studies, found in sections 2 to 5 below. Each of these sections provide a brief summary of each case study, and then presents a common set of conclusions regarding lessons learned for EU policy.

7.2 China

The legislative framework to address air pollution in general and ground-level ozone in particular is under development. China's 1995 law on air pollution does not refer to ozone; new air quality standards for ozone were introduced in February 2012. These standards are being phased in, first in major urban/industrial areas and are accompanied by plans to develop ozone monitoring system all over China. Other recent legislation seeks to reduce emissions of ozone precursors, in particular nitrogen oxides (NO_x) from industrial facilities and motor vehicles. Air pollution, though not ozone specifically, is also addressed in the country's 12th five-year plan, which sets a target to cut national NO_x emissions by 10%, from 2010 to 2015 (reduction targets are also set for SO_2 and NH_3).

A comprehensive overview is not yet available for the current air quality status concerning ozone levels, as monitoring for this pollutant is incomplete. Scientific studies have nonetheless identified ozone as a major concern in the highly populated and industrialised Eastern areas of China.

While information is not currently available to determine compliance with the newly introduced ozone standards, scientific studies identify industrial and transport emissions of NO_x as the major

sources of ozone, in particular as both sectors have grown rapidly in recent years. Assessments and controls on VOCs are an area where policy attention is only starting.

Overall, in terms of policy trends, a framework for ozone management is being put in place now. In general, air pollution policy in recent years has focused priority attention on 'key' cities and regions that face high air pollution levels.

7.3 India

India has not put in place a comprehensive ozone management policy or legislative framework. The Air Prevention and Control of Pollution Act, adopted in 1981 and last amended in 1987, sets general requirements and measures to reduce air pollution. In 2009, national air quality standards were adopted for several pollutants, including ozone. Some measures have been taken to reduce emissions of ozone precursors, in particular at local level.

India has not set in place a monitoring system for ground-level ozone; as a result, a comprehensive overview of the current air quality status concerning ozone is not available. Nonetheless, from the available monitoring data it can be concluded that the air quality in many urban areas is poor. Furthermore, international studies underline that the Indian sub-continent may suffer significant increases of ozone concentrations with impacts on health and food security, if current policy trends are not changed and ozone pollution not more effectively tackled.

7.4 Japan

Japan has a comprehensive legislative framework, built on the Air Pollution Control Law enacted in 1968 and developed extensively in the subsequent decades. Japan's legislation sets a country-wide air quality standard for photochemical oxidants (including ozone) as well as monitoring requirements and emissions controls for ozone precursors (including NO_x and VOCs) from fixed installations and transport. Voluntary agreements between government and industry are a hallmark of policy making in Japan: recent agreements have included actions to reduce industrial VOC emissions.

Despite a range of measures that have reduced emissions of ozone precursors, air quality status for ozone in Japan has not improved in recent decades and indeed has seen a worsening trend. The reasons for this trend, and recent compliance problems, are not fully clear. Scientific studies and modelling suggest that a share of current ozone concentrations in Japan is due to rising ozone transport from East Asia, in particular China.

Recent policy trends include the strengthening of the monitoring station network to determine the mechanism of formation of photochemical pollutants and to identify its transboundary movement as well as greater attention to industrial emissions of VOCs. Japan is also considering further research on photochemical oxidants and their precursors, including VOC sources not yet identified as well as non-anthropogenic VOCs.

7.5 United States

The United States has a comprehensive legislative framework to address air pollution, centred on the Clean Air Act (CAA), originally passed in 1970 and since amended several times. Moreover, the CAA is implemented through extensive secondary legislation: detailed 'rules' prepared by the Environmental Protection Agency (EPA). These include restrictions on emissions of ozone precursors from industrial and transport sources. Since air quality standards were first introduced in

1971, the United States has steadily set more stringent limit values on ozone levels; in 2010, a further change was proposed. A key element of the US policy framework is that the states are required to address air pollution issues in their territories through State Implementation Plans (SIPs). While the Federal government cannot fine states for not attaining national air quality standards, it can impose sanctions (in particular, withdrawing grants for highway construction) if a SIP does not adequately address the problems.

In terms of air quality, ozone concentrations in the main monitoring sites (in particular urban areas) have steadily decreased over the past three decades. These improvements are tied to the fall in emissions of major ozone precursors: carbon monoxide (CO), NO_x and VOCs. Nonetheless, ozone concentrations remain high in several large urban areas.

Reasons for non-compliance include motor vehicle emissions and VOC emissions. In addition, transport among the states is an issue, and hemispheric transport from Asia may have an influence, in particular on the Pacific Coast.

Recent policy trends include more stringent requirements for motor vehicle emissions, as well as stronger national approaches to address cross-state transport of air pollution: since 1990, an Ozone Transport Commission made up of state representatives advises the EPA, and a 2011 EPA rule set requirements on states to reduce emissions that contributed to non-attainment of air quality standards in other standards (the rule is being revised following an adverse court judgement). The EPA has also introduced a system of voluntary agreements with states that have attained existing standards: the goal is to support further improvements in air quality, in anticipation of more stringent future standards for ozone.

7.6 Key lessons and opportunities for EU policy

The US and Japan case studies provide the clearest lessons for EU policy. Fewer lessons are seen in China and India, where air pollution measures are still in development. The US sets strict requirements for the contents of State Implementation Plans (SIPs), and this approach may be considered as a model for more stringent EU requirements for planning to address zones that do not meet air quality standards. The enforcement mechanism between the federal government and states used in the US (withholding transport infrastructure grants) also deserves attention; though it may not fit in the EU budget practices, this form of conditionality between air quality measures and funding could be a valuable lever for action also in the EU. The US also has a voluntary approach to encourage states to go beyond attainment of air quality standards, also in preparation of more stringent future standards, and a similar initiative might be considered for the EU. The US provisions to address cross-state ozone transport, while under revision, also bear close attention as cross-border transport is an important issue in parts of the EU.

One lesson for consideration from the Chinese approach is that standards are implemented in a step-wise fashion – first addressing large urban and industrial areas where air pollution problems are the most serious. Such a priority setting approach could be considered in the EU for non-legislative programmes.

In addressing emissions from sources, Japan uses a voluntary approach to encourage the reduction of VOC emissions from industry, supplementing legal requirements. This programme is articulated at regional and local levels. While this approach also reflects a specific Japanese policy context, it could be considered as a supplement to EU legal mechanisms.

Japan and the US both have stringent standards on precursor emissions from motor vehicles – in the US, there are now standards also for low emissions vehicles – and these bear attention in setting EU standards of similar nature.

Japan and the US are also strongly focusing on research, including the development of new modelling systems to understand the formation of ozone and its transboundary movements, in order to take the adequate counter measures on precursors. The EU might consider similar investment in ozone modelling and in cooperating with research in Japan and the US in this field. In general, the four case studies highlight the importance of monitoring and scientific assessments of ozone trends and transport. Japan's plans to further assess non-anthropogenic sources of VOCs and non-identified VOCs may have lessons for the air quality management in other parts of the world, including the EU.

8 Reasons for increasing background ozone concentrations and recommendations for future objectives for ozone

8.1 Objectives and methods

The first objective for Task 6 is to explain the reasons for increasing background ozone concentrations based on a literature study and to make recommendations for future objectives for abatement strategies related to ozone pollution in the troposphere (the lower part of the atmosphere).

The background for the objective is related to answering the following question; why is it so that while ozone precursor emissions have decreased in Europe, the background ambient air ozone concentrations have increased, diminishing the expected positive effects on the ozone level of the existing policy on reduction of precursor emissions in Europe?

The challenge lies within finding the optimal policy options that will ensure abatement of the increasing background ozone concentrations in a situation where the chemical response to emission reductions has changed since the 1990's. This requires more knowledge about the impacts from changed emissions of the precursors (VOC, NO_x, CO and CH_4), both in Europe and on the entire Northern Hemisphere.

While results for Europe can provide running emission reduction scenarios, the increase in background concentrations is very complex, and more research is unquestionably needed in this area.

This is also the main conclusion from the Task Force on Hemispheric Transport of Air Pollution, acting within the framework of the Convention on Long-range Transboundary Air Pollution. This Task Force has for nearly a decade worked on improving the understanding of the intercontinental transport of air pollutants across the Northern Hemisphere. Following several years of science cooperation of experts across the northern hemisphere the report "Hemispheric Transport of Air Pollution 2010, Part A: Ozone and Particulate Matter, Air Pollution studies No. 17, was published (HTAP, 2010). It contains a state-of-science assessment with respect to intercontinental transport of ozone and particulate matter and to some extent tries to quantify possible reasons for increasing background ozone concentrations.

The present project provides a summary of the synthesis of the reasons for increasing background ozone concentrations and recommendations for future objectives for ozone based on a review of existing literature, review papers and results identified in the HTAP report is provided below.

Our synthesis is structured by considering 7 central hypotheses related to what may be the main reasons for increasing background levels of ozone:

1. Increased global emissions of the precursor CH_4.
2. Increased global anthropogenic precursor emissions of VOC/NO_x.
3. Increased global natural pre-cursor emissions of VOC (vegetation) / NO_x (soil) e.g. due to climate change (increased temperature).

4. Changes in the tropospheric chemistry e.g. due to changes in global radiation, temperature or humidity.
5. The reductions in European urban NO_x emissions have led to increased ozone levels due to reductions in ozone titration by NO ($NO/NO_2/O_3$ chemistry). This can lead to higher ozone concentrations locally in urbanized areas but lower ozone levels in more remote areas, depending on the chemical regime.
6. Increased ozone input to the troposphere from the stratosphere.
7. Changes in dry deposition of ozone due to changed land cover (less vegetation) or humidity.

The outcome is an overview of present knowledge related to the above mentioned topics.

A second objective is to outline policy options for how to best address the long term challenge of increasing ozone background levels through strengthening the existing strategy to reduce the precursor emissions (VOC, NO_x and CH_4), on the main presumption that the status and value of the existing ozone air quality standard will not change. The work was based on studies carried out at IIASA/JRC/EMEP. Results from these institutes/organizations are included directly in this final report.

A third objective is to propose measures to improve the monitoring networks, siting criteria and siting density to better detect long and short term changes and trends in ozone concentrations.

The work addressing the third objective was partly based on discussions and brainstorming at a partner workshop and partly on a literature study to ensure that all kinds of measures are considered. The workshop focused on proposing measures to improve the monitoring networks, siting criteria and siting density to better detect long and short term changes and trends in ozone concentrations in Europe. Experts in monitoring of ozone as well as experts in atmospheric modeling of ozone were invited. In particular harmonisation in siting, frequency and coverage and how this may be considered to improve the performance to detect trends and assess ozone exposure (both to humans and ecosystems) was assessed. Considerations towards the use of data assimilation through combinations of models and monitoring will be included.

8.2 Global anthropogenic emissions

The background tropospheric ozone concentrations at northern mid-latitudes have increased about 1% per year (see also chapter 4). This is also supported by Parrish et al. (2012) who concludes that ozone has increased at all background sites in the northern hemisphere in all seasons at approximately 1% per year relative to the site's 2000 year.

It is striking that background measurements of tropospheric ozone around the world show approximately the same rate of increase. This is a strong indication that the increase in ozone background concentrations is a global phenomenon.

On a global scale, ozone precursor emissions have since the year 2000 increased for NO_X (2.3 %/year), NMVOC (1.6 %/year), CH_4 (2.5 %/year), and CO (2.5 %/year). These global increases can easily explain the general increase in ozone background concentrations of 1% per year.

According to HTAP (2010), the transport of ozone from other source regions (North America, South Asia and East Asia) contributes together some 43% to ozone levels in Europe. The contributions

from North America, Asia and Europe observed in Europe are expected to have nearly equal weight. However, the increase in ozone pre-cursor emissions is strongest in Asia and especially the increases in NO_X emissions from Asia have potentially the strongest influence on the increasing ozone background levels in the Northern Hemisphere.

8.3 Changes related to the atmospheric chemistry of ozone

Tropospheric ozone depends on a number of parameters related to the chemistry and physics of ozone production, which may increase or decrease the future background ozone concentration. Not all these effects are known or fully understood, and in particular how these parameters influence each other, i.e. negative and positive feed-back. For example, CO and some volatile organic compounds have decreased significantly in USA and Europe over the past years, while CH_4 has increased. From rural and remote monitoring stations in Denmark and Switzerland, NO_2 appears to have decreased over the last two decades, which is most likely due to mitigation initiatives. However, global NO_x emissions have increased. In particular, NO_x has increased dramatically over industrialized regions in China up to at least 2005.

On a global scale, the chemistry which leads to ozone formation is mostly expected to be NO_x sensitive, for which reason increasing NO_x emissions are expected to exert a substantial impact on tropospheric ozone formation. In addition, NO_x emitted from microorganisms is also expected to increase in a future warmer climate (see e.g. Brasseur et al., 2006). An increase in the NO_x background concentration will generally have a larger effect on ozone formation than an equal increase in the VOC concentration due to the already large natural emission of VOC's and CH_4 as well as CO. Furthermore, an increase in the background NO_x may shift the chemical regime from being ozone-degrading to ozone-forming.

8.4 Changes related to chemical regimes in Europe

Ozone concentrations are strongly linked to NO_x concentrations. In urban streets and urban background the recent year's reductions in NO_x emissions from mainly road traffic have led to slight increases in annual mean ozone concentrations, although these tendencies at least for Danish conditions are not yet statistically significant. Both Danish and German measurements show that rural background annual mean ozone concentrations are slightly increasing although also these are not statistically significant. Reducing NO_x emissions in Europe can result in increased ozone concentrations in the highly urbanized areas of the central and northwestern parts of Europe (the VOC sensitive areas), including e.g. Germany, The Netherlands, Belgium and the UK.
More information is needed concerning the present chemical regimes in Europe with respect to NO_X and VOC sensitive areas. The latest larger studies of this were carried out in the mid-nineties. Since then, the concentration levels over Europe have changed and therefore also a change in the location of the chemical regimes – e.g. NO_x or VOC sensitive regions. It was recently found that a future decrease in anthropogenic emissions in Europe assuming the RCP4.5 scenario leads to an increase of up to 10% in ozone concentrations in the NO_X limited areas of Europe (central and western parts of Europe) (Hedegaard et al., 2013).

8.5 Changes related to stratospheric tropospheric exchange

Both measurement- and model-based studies points towards an impact on tropospheric ozone levels both regarding inter-annual variability and long terms trends from the stratosphere. Both a recovering of ozone levels in the lower stratosphere as well as an increased flux from the stratosphere to the troposphere possibly due to changes in the general circulation patterns, can

explain an increased input from the stratosphere. The impact also seems to be significant in the European area.

Direct quantifications of the impact is hard to estimate due to the complexity of the involved process ranging from mesoscale transport processes to hemispheric scale circulations as well as photochemistry. Recently published work used a climate model to estimate an increase in the ozone input of 50 (+/- 6 standard error) Tg/yr for the years 2000-2035, which is an increase of ~10% compared to the total estimated transport from the stratosphere of 552 +/- 168 Tg/yr.

8.6 Changes related to dry deposition of ozone

Dry deposition is a main removal process for ozone in the lower troposphere. Changes in land cover, especially deforestation of areas to agricultural or urbanised application and changes in meteorological conditions over the years can be an important factor for changes in ozone concentrations. However, the changes in land cover as well as the governing processes are relatively slow, and it is not likely that a change in dry deposition of ozone over Europe or globally is the main driver for increasing ozone background concentrations.

8.7 Modelling of the observed ozone trends, source allocation and future scenarios

Global climate models seem to better match observed data when looking at the variability (seasonal and regional) under investigation. The models have problems to foresee the surface ozone concentrations on small temporal and spatial scales. This means that formation processes on local scales are not well represented in the models. The models also have substantial problems to match the observed data especially in remote locations where they (largely) underestimate the measured values. Global models are not well suited yet to quantify trends or source allocations for ozone at regional or local scales.

There is evidence that human activities are a major reason for the increasing trend for surface ozone, but the source attribution of these increases remains very uncertain. Furthermore, further research using chemistry transport models is needed in order to enhance the understanding of the governing processes for the observed ozone trends.

It is estimated that over 50% of tropospheric ozone may be attributed to anthropogenic sources. About half of this anthropogenic component originates from sources over the region itself and the other half is transported from sources outside the region. Another 20-25% of ground-level ozone originates from the stratosphere, and a similar proportion is formed from natural precursor sources: lightning, soils, vegetation and fire, and from oxidation of natural hydrocarbons. Modeling indicates that emissions changes in the other source regions are about half as important as the same domestic emissions change. Furthermore, controlling CH_4 is suggested of major importance in limiting increases in baseline surface ozone, and has additional benefit for climate.

Langner et al. (2012) found a robust pattern among five CTMs with an increase in ozone levels in southern Europe towards 2050. However, the size of the projected change in ozone only due to climate change differed by up to a factor of two between the models. It turned out that this difference mainly was related to differences in the emissions of isoprene and differences in the sensitivity of these to changes in climate. Recent modelling studies on future ozone trends found that natural emissions of isoprene are crucial for the models in terms of ozone and the sensitivity of these emissions to changes in the forcing parameters needs to be better understood. Also,

modelling studies found that at surface level the ozone concentration is projected to increase due to climate change in the areas where substantial amounts of ozone precursors are emitted. Elsewhere a significant decrease is projected at the surface. In the free troposphere a general increase is found in the entire Northern Hemisphere except in the tropics, where the ozone concentration is decreasing. In the Arctic the ozone concentration will increase in the entire air column, which most likely is due to changes in atmospheric transport.

Changes in temperature, humidity and the naturally emitted VOCs are governing the changes in ozone. A modeling study, where the relative importance of the climate signal and the signal from changes in anthropogenic emissions were studied found that in large parts of Europe both future climate change and future reductions in anthropogenic ozone precursors (especially NO_X) contributes to increases in future ozone levels in Northwestern Europe (Hedegaard et al., 2013).

8.8 Policy options for the long term challenge of increasing background levels

The objective of Priority 2 was to identify options and recommendations for how to best address the long term challenge of tackling increased background ozone concentrations by strengthening the existing EU strategy of reducing emissions of precursors NO_x, VOC and CH_4 etc., in order to meet the current ozone target values. Of particular interest is to learn more about possible measures on CH_4 and CO emissions. The work in priority 2 is based on a review of two reports made available by IIASA and Metno/EMEP.

The results in the Metno/EMEP report includes some emission reduction scenarios where all anthropogenic emissions are reduced by 20% and the impacts on maximum daily ozone concentrations are assessed. As the emission reduction scenarios are not divided into the individual ozone precursors, it is not possible to distinguish the impacts from possible measures on CH_4 and CO emissions separately. Neither is it possible to assess the impacts from individual NO_x or VOC emission reductions as all reductions are conducted simultaneously. The report mainly contains a preparatory study with focus on the TF HTAP work, which will be conducted in 2013, with focus on the intercontinental transport between Europe, North America and South East Asia.

The IIASA (TSAP #10) report includes studies on future emission scenarios with focus on the impacts from the MTFR and CLE scenario in 2025 compared to the year 2005. The impact with respect to ozone (p. 24) is described by the number of premature deaths related to ozone estimated for the two scenarios. A decrease is predicted from app. 24600 cases of premature deaths in the year 2005 to app. 18200 and app. 15400 for the CLE and MTFR scenarios, respectively. The report does not contain further information applicable to identify options and recommendations for how to best address the long term challenge of tackling increased background ozone concentrations by strengthening the existing EU strategy of reducing emissions of precursors NO_x, VOC, CO and CH_4.

8.9 Measures to improve the monitoring networks, siting criteria and siting density

A workshop was conducted to address these issues and the following the prioritized list of recommendations was agreed:

1. To continue the measurements at sites already representing long time series. The value of the individual measurement stations increases with the age of the station. Furthermore, to continue the quality assurance of measurements following the CEN standards.
2. To conduct an analysis of the representativeness of all measurement sites in Europe considering issues such as the individual foot prints (meaning the extent of the

geographical area, which the specific measurement station represents) of the measurement sites, their location within the zones and chemical regimes the sites are representing, whether there are any systematic changes over time that can be explained by changes in local conditions, the density of the network, the use of super sites in the network, etc. The analysis should be divided into the zones and agglomerations already defined in the air quality directive, but also dividing Europe into overall zones depending on the chemical regimes (NO_x/VOC sensitive areas). One result from this analysis is defining the optimal networks for detecting trends and assessing exposure, based on subsets of the existing network for the specific purpose (e.g. trends in the rural background or human exposure in VOC limited areas).

3. To develop an overall European strategy for the number and density of supersites, measuring both ozone and precursors and to formalize the use of super sites in the monitoring network is recommended, including an analysis of the optimal density of the super sites in a European perspective and with a detailed description of minimum chemical and physical parameters measured. The strategy should include a common analysis of the optimized number of super sites versus the need for ordinary monitoring stations, measuring ozone only. A better understanding of chemical and physical processes important for ozone could decrease the need for a dense ozone monitoring network.
4. To develop an overall European strategy for measuring ozone precursors and especially VOCs is needed. Considerations could be given to have a higher density of measurement stations of VOCs in VOC limited areas, as e.g. in Southern Europe. To extend the requirements for measuring VOCs not only in urban and suburban areas but also in the rural background.
5. To include the expertise of the AQUILA and FAIRMODE networks for optimizing the monitoring networks e.g. for further development and evaluation of air pollution models.
6. To make it mandatory to conduct integrated monitoring combining both measurements and models results for understanding the chemical and physical processes governing trends and exposure (Hertel et al., 2007). The use of integrated monitoring has the potential for decreasing the need for ordinary ozone monitoring stations significantly.
7. To initiate the use of chemical data assimilation to make analysed fields (combined measurements and model results) covering Europe over a long time period (> 20 years). The analysed fields are optimal for assessing exposure to humans and ecosystems, and are an important supplement for detecting and understanding trends. The analysed fields could be produced by an extension of the MACC project (Monitoring Atmospheric Composition and Climate; http://www.gmes-atmosphere.eu/) with all the major modelling groups in Europe, representing state-of-the-art modelling systems, including data assimilation.
8. To introduce formalized measurements of vertical profiles of ozone and precursors, to increase the understanding of processes contributing to ozone formation.
9. To improve the emission inventories for ozone precursors, with special focus on the VOCs – both anthropogenic and natural emissions. It is recommended to establish a European working group (with experts within monitoring, modelling and emissions) to identify which species to measure to improve emission inventories and modelling of natural emissions.
10. To further exploit the results from an optimized network, it is important to have focused research projects with the purpose of increase the understanding of chemical and physical processes leading to ozone formation and loss, understanding the role of the individual precursors separately and together in different parts of Europe, and identifying the different chemical and physical regimes in Europe, as the different regimes require different regulatory actions.

8.10 Reasons for increasing ozone background concentrations

The work in this report is based on a literature study reviewing the recent work of the TF HTAP and open literature on the topic. The conclusions in the report show that it is presently not possible to provide firm and conclusive answers to the main reasons for increasing background levels of ozone. According to the results obtained within the Task Force (HTAP, 2010), there is not an unequivocal reason for the increased background levels and the Task Force stresses that more research is needed in this area.

According to TF HTAP (2010) over 50% of the ozone background concentrations in the HTAP regions may be attributed to anthropogenic sources and the other half to natural sources. About half of the anthropogenic component originates from sources over the region itself and the other half is transported from sources outside the region. Nearly half of the natural sources originate from the stratosphere, and the other half is formed from natural precursor sources: lightning, soils, vegetation and fire, and from oxidation of natural hydrocarbons.

The work in this report addressing the seven hypotheses points towards the following major reasons for increasing ozone background concentrations, since the year 2000 in the European area:

1. Increasing global emissions of ozone pre-cursors. Especially the increase in NO_X emissions, which to a large extend takes place in Asia. The lifetime for ozone in the background is sufficiently long for the air masses in the whole Northern Hemisphere can be long-range transported to Europe.
2. On a global scale, the chemistry which leads to ozone formation is mostly expected to be NO_X sensitive, for which reason increasing NO_X emissions are expected to exert a substantial impact on tropospheric ozone formation.
3. The reductions of NO_X emissions in Europe since the year 2000 can explain some of the increases of ozone levels in the central and north western parts of Europe.
4. There are indications that the input from the stratosphere is increasing, both due to the recovering ozone layer and due to climate change, increasing the flux from the stratosphere to the troposphere. However, over a ten year period, this process is assessed to have minor impact on the observed ozone trends.
5. The increase in CH_4 concentrations also contribute to the increasing ozone levels, According to HTAP (2010), the anthropogenic sources of CH_4 are estimated to contribute to about 5 ppbv to global mean surface ozone. However, from the present literature, it is impossible to quantify the importance of the increasing background CH_4 concentrations.
6. Future scenarios point towards an increase in ozone concentrations due to climate change (the climate penalty), which has to be taken into account in future regulations.

8.11 Conclusions

EU has decreased overall emissions of ozone precursors since 1990. The impact on ozone concentrations is especially due to reductions in NO_x and VOC, since these species are relatively short lived. Reducing emissions of these species in Europe has an effect within Europe before the "effect" is blown with the wind out of Europe. Methane and CO have relatively long life times – 9-10 years and 2-3 months, respectively. Their effect on ozone is therefore primary on the hemispheric scale and not on the European scale. Therefore, reducing emissions of CH_4 and CO in Europe will not have any immediate effect within Europe but rather globally.

Ozone should, in principle, have decreased in Europe due to the decreased emissions of NO_x and VOC in Europe. However, the expected effect in Europe is at the same time counterbalanced by the increased ozone concentrations in the hemispheric background.

The increase in the hemispheric background level of ozone concentration is most probably due to increased NO_x and VOC emissions on the hemispheric scale and this increase in emissions is especially seen in Asia. Since ozone has a lifetime in the background of ~21 days, the ozone produced anywhere in the Northern Hemisphere will be transported by the wind around the globe. In conclusion, to decrease hemispheric background concentrations of ozone, the greatest potential lies within emission reductions of NO_x and VOC in Asia.

The optimal strategy in Europe for reducing ozone concentrations in Europe is to continue reducing NO_x and VOC emissions. This has already had a significant impact during the last ten years. According to modeling studies, the ozone concentrations in Europe would have increased significantly instead of being approximately steady state or slightly increased, if these ozone precursor emissions had not decreased in Europe. However the long term objectives for ozone can't be met without additional action worldwide on VOCs and NOx and on the CO and CH_4 emissions.

Further reductions in NO_x and VOC in Europe towards 2020 assuming implementation of NEC-II will definitely have a positive effect in Europe with respect to the future ozone concentrations. Model studies have previously been conducted using the DEHM model (Brandt et al., 2012), comparing the ozone concentrations in the year 2005 (using EMEP emissions for 2005) with 2020 (using NEC-II emissions for 2020 from the Gothenburg Protocol) and these studies showed a significant and considerably decrease in ozone concentrations in 2020 over Europe.

In this context, it is important to improve the knowledge on which regions in Europe are NO_x or VOC sensitive (or sensitive to both) for optimizing an emission reduction strategy in Europe. In some, specific regions in Europe, the optimal strategy is to reduce VOCs as small or no effect may be seen from reducing NO_x and vice versa. In some regions reductions in both NO_x and VOC emission are required. If the opposite is conducted (e.g. reducing NO_x in VOC limited areas), there would be no or little effect on ozone (or even the opposite effect of increasing ozone concentrations). If information is missing on the exact locations of the NO_x and VOC sensitive areas in Europe, both precursors have to be reduced in the whole of Europe, which may not be cost effective.

The current modelling strategy used to identify optimal ozone precursor emission reduction strategies has some shortcomings. It is based on using the GAINS model to find the optimal reduction strategy for reducing VOC/NO_x emissions, since the VOC/NO_x/CH_4/CO/O_3 chemistry is highly non-linear and linear source-receptor relationships are assumed in GAINS. Even though polynomials are used for approximating the source-receptor relationships for ozone, they are still linearized assumptions of the highly non-linear problem. It can therefore only be applied to the specific scenarios, which they have been estimated for (e.g. a specific year though accounting or weather variations using five meteorological years, a specific chemical regime over Europe, a specific emission reduction from a single country) and are not applicable for multi-year, multi-species and multi-country emission reduction assessments. It is recommended that the emission reduction strategy is compared to other state-of-the-art regional air pollution models, in order to assess the robustness of the strategy and whether the expected and desirable effect from any emission reduction strategy is likely to be obtained.

The ideal solution for finding optimal and cost effective emission reductions strategy in Europe with respect to ozone is to identify the NOx and VOC sensitive areas and reduce emission of either NO_x or VOC or both species simultaneously in these areas in accordance with this. Since the NO_x/VOC sensitive regimes varies over Europe and evolves over time, it is most likely not an optimal strategy to reduce both VOC and NO_x in all regions. Reducing in a non-optimal way can result in increases in ozone concentrations. For the purpose of establishing a solid and robust ozone strategy, models will have to be used that allows both identification of the main options as well as the study of sensitivities. Model sensitivity studies should preferably use a number of different models conducting the same emission reduction scenarios to produce a kind of ensemble and thereby reducing, or at least clarifying, model differences and uncertainties. Emission reductions at the European level are very costly and therefore using state-of-the-art modelling to identify the NO_x and VOC sensitive areas and to calculate reliable impacts from proposed emission reductions is recommended.

The same applies for reductions of CO and CH_4. Model studies are needed in order to quantify the impacts from emission reductions of CO, CH_4, VOC and NO_x, both individually and simultaneously, in order to be able to quantify the impacts of emission reductions both on ozone mean concentrations as well as daily maximum concentrations.

Studies with global/hemispheric models are necessary to analyse emission reduction scenarios and study the important chemical and physical atmospheric processes to further understand the source allocation from both anthropogenic and natural sources and for understanding the sources and processes contributing to the positive trend in the ozone background concentrations on the hemispheric scale.

Studies on the European scale are necessary to understand the European ozone trends, which are very much dependent on the chemical regimes, whether it is VOC or NO_x sensitive. Due to the substantial non-linearity in ozone chemistry it is important to carry out new modelling studies to locate and quantify the VOC/NO_x sensitive regions in Europe for optimal regulation strategies. Furthermore, it is necessary to conduct emission reduction scenarios for CO and CH_4 in order to assess the sensibility of ozone concentrations to all ozone precursors.

9 References

Amann, M.(Ed.), 2013. Policy Scenarios for the Revision of the Thematic Strategy on Air Pollution. TSAP Report #10, Version 1.2. International Institute for Applied Systems Analysis IIASA.

Brandt, J., J. D. Silver, L. M. Frohn, C. Geels, A. Gross, A. B. Hansen, K. M. Hansen, G. B. Hedegaard, C. A. Skjøth, H. Villadsen, A. Zare, and J. H. Christensen, 2012: An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport. Atmospheric Environment, Volume 53, June 2012, pp. 156-176, doi:10.1016/j.atmosenv.2012.01.011

Brandt, J., J. D. Silver, J. H. Christensen, M. S. Andersen, J. Bønløkke, T. Sigsgaard, C. Geels, A. Gross, A. B. Hansen, K. M. Hansen, G. B. Hedegaard, E. Kaas and L. M. Frohn, 2013a: "Contribution from the ten major emission sectors in Europe to the Health-Cost Externalities of Air Pollution using the EVA Model System – an integrated modelling approach". Atmospheric Chemistry and Physics Discussion. 13, 5871–5922, 2013. www.atmos-chem-phys-discuss.net/13/1/2013/. doi:10.5194/acpd-13-5871-2013

Brandt, J., J. D. Silver, J. H. Christensen, M. S. Andersen, J. Bønløkke, T. Sigsgaard, C. Geels, A. Gross, A. B. Hansen, K. M. Hansen, G. B. Hedegaard, E. Kaas and L. M. Frohn, 2013b: "Assessment of Past, Present and Future Health-Cost Externalities of Air Pollution in Europe and the contribution from international ship traffic using the EVA Model System". Atmospheric Chemistry and Physics Discussions, 13, pp. 5923-5959, 2013. www.atmos-chem-phys-discuss.net/13/1/2013/, doi:10.5194/acpd-13-5923-2013

Brasseur, G.P., Schultz, M., Granier, C., Saunois, M., Diehl, T., Botzet, M., Roeckner, E., Walters, S., 2006. Impact of Climate Change on the Future Chemical Composition of the Global Troposphere. J. Climate, 19, 3932–3951.

Carslaw, D.C. and K. Ropkins, (2012a) openair --- an R package for air quality data analysis. Environmental Modelling & Software. Volume 27-28, 52-61.

Carslaw, D.C. and Ropkins, K. (2012b). openair: Open-source tools for the analysis of air pollution data. R package version 0.5-16.

Dlugokencky, E. J. and Bruhwiler, L., (2009), Observational constraints on recent increases in the atmospheric CH4 burden, Geophysical Research Letters, 36, L18803.

European Environment Agency (EEA), 2011, Air pollution by ozone across Europe during summer 2010,

European Environment Agency (EEA) (2012), Air Quality in Europe – 2012 Report. No4/2012 ISSN 1725-9177.

Hedegaard, G. B., J. H. Christensen, and J. Brandt, 2013: "The relative importance of impacts from climate change vs. emissions change on air pollution levels in the 21st century". *Atmospheric*

Chemistry and Physics, 13, 3569-3585, 2013. http://www.atmos-chem-phys.net/13/3569/2013/. doi:10.5194/acp-13-3569-2013.

Hertel, O. and Goodsite, M. E., 2009. Urban Air Pollution Climate Through out the World in: Air Quality in Urban Environments: Hester, R. E. and Harrison, R. M., Eds., RSC Publishing, Cambrigde, 1-22.

HTAP, 2010: Hemispheric transport of air pollution. Part A: Ozone and particulate matter Air pollution studies Nr. 17, Dentener, F., Keating, T., Akimoto, H (Eds.). United Nations Publication, New York and Geneva, 304 p.

Langner, J., Engardt, M., Baklanov, A., Christensen, J. H., Gauss, M., Geels, C., Hedegaard, G. B., Nuterman, R., Simpson, D., Soares, J., Sofiev, M., Wind, P., and Zakey, A., 2012. A multi-model study of impacts of climate change on surface ozone in Europe, Atmospheric Chemistry and Physics, 12, 10423-10440.

Mauzerall, D. L., Jacob, D. J., Fan, S. M., Bradshaw, J. D., Gregory, G. L., Sachse, G. W., and Blake, D. R., 1996. Origin of tropospheric ozone at remote high northern latitudes in summer, Journal of Geophysical Research-Atmospheres, 101, 4175-4188.

Parrish, D. D., K. S. Law, J. Staehelin, R. Derwent, O. R. Cooper,1, H. Tanimoto, A. Volz-Thomas, S. Gilge, H.-E. Scheel, M. Steinbacher, and E. Chan, 2012: Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes. Atmos. Chem. Phys., 12, 11485–11504, 2012. www.atmos-chem-phys.net/12/11485/2012/ doi:10.5194/acp-12-11485-2012 Royal Society (2008), Ground-level ozone in the 21st century: future trends, impacts and policy implications. London, UK, Science Policy Report 15/08, ISBN: 978-0-85403-713-1

Royal Society (2008),"Ground-level ozone in the 21st century: future trends, impacts and policy implications". London, UK, Science Policy Report 15/08, ISBN: 978-0-85403-713-1

Schulz, M., J.E. Jonson, J. Griesfeller, S. Valiyaveetil, M. Schultz, M. Decker, S. Waychal, M. Amann, Z. Klimont, C. Heyes, W. Schöpp, 2013. INTERIM REPORT. Service contract on "Assessment of hemispheric air pollution on EU air policy". Contract no. 7.0307/2011/605671/SER/C3. Metno/IIASA/Julich.

Sillman, S., 1999. The relation between ozone, NOX and hydrocarbons in urban and polluted rural environments. Atmospheric Environment 33, 1821-1845.

United Nations Environment Programme (UNEP) (2011), Integrated Assessment of Black Carbon and Tropospheric Ozone. ISBN: 92-807-3141-6

Wilson, R. C., Fleming, Z. L., Monks, P. S., Clain, G., Henne, S., Konovalov, I. B., Szopa, S., and Menut, L., 2012: Have primary emission reduction measures reduced ozone across Europe? An analysis of European rural background ozone trends 1996–2005, Atmos. Chem. Phys., 12, 437-454, doi:10.5194/acp-12-437-2012, 2012. http://www.atmos-chem-phys.net/12/437/2012/acp-12-437-2012.html

P.O. Box 4175
3006 AD Rotterdam
The Netherlands

Watermanweg 44
3067 GG Rotterdam
The Netherlands

T +31 (0)10 453 88 00
F +31 (0)10 453 07 68
E netherlands@ecorys.com

W www.ecorys.nl

Sound analysis, inspiring ideas