REPORT

A comparison of emissions from ethanol and petrol fuelled cars

Health risk assessment for Västra Götaland

Erik Fridell* Marie Haeger-Eugensson* Jana Moldanova*
Lin Tang* Karin Sjöberg* Bertil Forsberg+

B1962
September 2010

* IVL Swedish Environmental Research Institute
+Umeå University, Occupational and Environmental Medicine

This report approved
2011-03-16
John Munthe
Scientific Director

IVL Swedish Environmental Research Institute

IVL Swedish Environmental Research Institute

Organization

IVL Swedish Environmental Research Institute Ltd.

Address

P.O. Box 5302
SE-400 14 Göteborg

Telephone

+46 (0)31-725 62 00

Report Summary

Project title
Emissions from ethanol cars - dispersion and impact

Project sponsors
Saab Automobile, The Swedish Transport Administration and The Foundation for the Swedish Environmental Research Institute

Author

Erik Fridell Marie Haeger Eugensson Jana Moldanova Lin Tang Karin Sjöberg Bertil Forsberg

Title and subtitle of the report

A comparison of emissions from ethanol and petrol fuelled cars
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Summary

See below

Keyword

emissions, dispersion modelling, health risks, E85, ethanol, petrol

Bibliographic data

IVL Report B1962

The report can be ordered via
Homepage: www.ivl.se, e-mail: publicationservice@ivl.se, fax+46 (0)8-598 563 90, or via IVL, P.O. Box 21060, SE-100 31 Stockholm Sweden

Abstract

Facing the problems with global warming and the diminishing supplies of oil, alternative fuels are becoming more and more important for road traffic. One fuel that has been used for several years is ethanol (E85). The main discussion points regarding the environmental performance for ethanol as a fuel are related to the production. However, there are also some notable differences in the emissions between E85 and petrol fuelled vehicles. This relates to some extent to the emissions of nitrogen oxides (NO_X) and particulate matter (PM) but mainly to the composition of the emitted organic compounds.

These differences in emissions will potentially give different impacts on health and on the environment. This can be both through risks linked to the primary emissions and to secondary products formed in the atmosphere. In order to assess the health risks it is necessary to calculate the emissions in space and time, describe the dispersion and chemical reactions taking place in the atmosphere and to calculate the exposure to humans.

In the present study two fuel scenarios for passenger cars are studied; one where the cars with Otto engines run on petrol and one where they run on E85. Two emission scenarios for 2020 are constructed and dispersion modelling is applied to obtain the human exposure to key pollutants. The dispersion modelling is performed with the EMEP model for extended Europe and the data obtained are used as boundary conditions for the model for the Västra Götaland Region. In the latter, detailed traffic and emissions scenarios are used together with the TAPM model to obtain concentration levels and population exposure. The differences in health impacts are then assessed.

The differences in emission factors reflect in differences in emissions. The emission calculations for all Swedish road traffic show a decrease for the E85 scenario relative to the petrol scenario of 6.5% for NO_X, 3.4% for $PM_{2.5}$, 67% for benzene. For acetaldehyde there is an increase of 770%.

The differences obtained from the TAPM modelling show decreased levels of NO_X, ozone and benzene with E85 and increased levels of acetaldehyde. For the latter the increase may be up to 80%, while NO_X and ozone show decreases of up to a few per cent and a few tenths of per cent, respectively.

The health risk assessment shows decreased health risks in the E85 scenario relative the all-petrol scenario, due to the decreased NO_X exposure, correlated with both preterm deaths and asthma. However, NO_X may be mainly an indicator of unmeasured causal exhaust components in the epidemiological studies and thus the exposure-response functions for NO_X may not be applicable in the present case where there is a difference in NO_X exposure but not necessarily a difference in exposure to other exhaust components normally associated with NO_X. Smaller effects are expected from the changes in ozone, acetaldehyde, $PM_{2.5}$ and benzene exposure. The overall difference is about 1.6 preterm deaths per year for the Västra Götaland Region, with lower values for the E85 scenario, when the uncertain differences due to the differences in NO_X exposure are not considered.

Content

1 Introduction

Facing the problems with global warming and the diminishing supplies of oil, alternative fuels are becoming more and more important for road traffic. Several alternative fuels are being tested such as biogas, alcohols and dimethyl ether (DME). One fuel that has been used for several years in Brazil and has become popular in, e.g. Sweden and the USA during the last ten years is ethanol. It is used both as a component (about 5 % vol.) in regular petrol and as E85 (85 % vol. ethanol, 15 % vol. petrol).

The main discussion points regarding the environmental performance for ethanol as a fuel are related to the production. In this study the production of ethanol fuel is not in focus but rather the impact from emissions stemming from E85 fuelled cars. Although the emissions from E85 cars are not as well characterised as the emissions from petrol cars, there are some notable differences in the emissions. This relates to some extent to the emissions of nitrogen oxides (NO_X) and particulate matter (PM) but mainly to the composition of the emitted organic compounds. The main differences here are that petrol generates relatively higher emissions of benzene while E85 generates relatively higher emissions of acetaldehyde and ethanol.

Ethanol has a reaction lifetime of several days and the major product of its OH-initiated degradation is acetaldehyde. The OH-initiated degradation of acetaldehyde gives upon further reaction with NO_2 peroxy-acetyl nitrate (PAN). Other reactions of acetaldehyde give mainly formaldehyde and CO_2. All of acetaldehyde, formaldehyde and PAN have negative effects on human health.

These differences in emissions will potentially give different impacts on health and the environment. This can be both through risks linked to the primary emissions and to the mentioned secondary products formed in the atmosphere. In order to assess the health risks it is necessary to calculate the emissions in space and time, describe the dispersion and chemical reactions taking place in the atmosphere and to calculate the exposure to humans.

Jacobson (2007) studied the impact on health risks of a change from petrol to E85 in a scenario analysis for the US in 2020. A greater risk was found for the E85 scenario compared to an all-petrol scenario due to the effects on the ozone concentration. However, the differences found were small and the conclusion made is that it is unlikely that the use of E85 fuel would improve air quality relative to petrol.

In the present study the change in impact on health risks between a scenario with all petrol and one with all E85 is studied for 2020. The fuel switch is applied to all positive ignition injection cars for the EMEP-model domain (covers Europe and surrounding parts of North Africa and Asia) and the health impact is studied for the region of Västra Götaland in Sweden. Two emission scenarios for 2020 are constructed and dispersion modelling is applied to obtain the human exposure to key pollutants in the region. The dispersion modelling is performed with the EMEP model for extended Europe and the data obtained are used as boundary conditions for the model for the Västra Götaland region. In the latter detailed traffic and emissions scenarios are used together with the TAPM model to obtain concentration levels and population exposure. The differences in health impacts are then assessed. The setup of the scenarios is similar to what is presented by Jacobson but of course applied to a different region with different climate as well as traffic and population density.

2 Methods

2.1 Overview

The overall objective of the present study is to compare the health impacts in the Västra Götaland region of emissions from petrol fuelled cars (i.e., cars with Otto engines and spark ignition) in two scenarios for 2020: one with all petrol and one with all E85. Thus in one scenario all these cars are assumed to be fuelled by petrol and in the second all these cars are assumed to be fuelled by E85. The 'all-petrol' scenario is called S1, the 'E85' scenario S2. The study comprises:

- emission calculations
- dispersion modelling
- population exposure analysis
- health impact assessment.

Two dispersion model runs for the Västra Götaland region (see Figure 1b) are performed with the TAPM model. In order to calculate the pollutants entering this region the larger scale EMEP (see Figure 1a) model is applied to the extended European region.

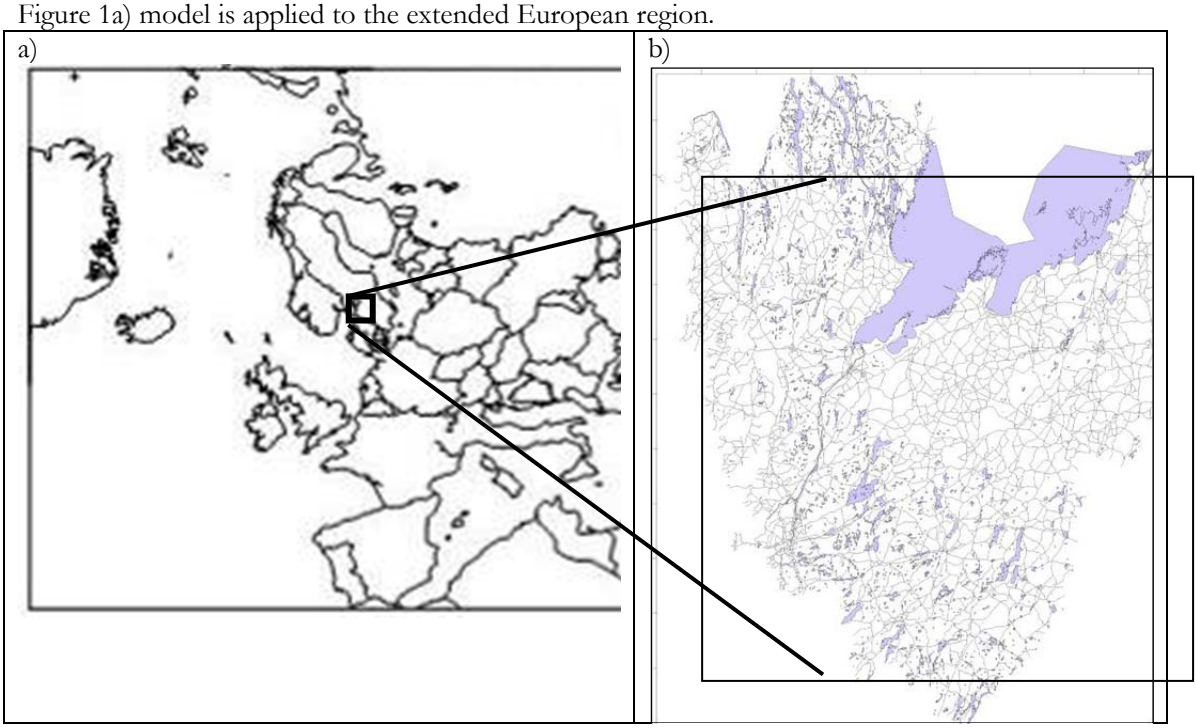

Figure 1. The model area for a) the EMEP model and b) the TAPM model. For the TAPM modelling the indicated square in b) is the model domain and the defined map is the Västra Götaland region.

2.2 Emission factors

The study starts with establishing emission factors for petrol- and E85-fuelled cars. This basically gives two sets of emission factors that can be applied to the projected traffic volumes. For the Västra Götaland region the emission factors used, with the exception of E85 cars are taken from

the Artemis model (Andre 2005). This is a detailed emission model used also in the Swedish reporting of emissions from road traffic and must be considered as the source with the most accurate emission factors for road traffic for Swedish conditions. Artemis contains emission factors for the year 2020 which are used in this study. Speciation of NMVOC emissions (except for E85 fuelled cars) come from Passant (2002).

The emission scenarios used for Europe are built on EMEP's 'Current Policy' (CP) scenario for the year 2020 (Vestreng et al., 2004). This scenario contains emissions and fuel consumption divided into different source sectors, where one is road traffic. In order to calculate the emissions for the two scenarios the relative change in emission factors for positive ignition injection cars between petrol and E85 are used.

The data available in the EMEP emission database are, however, not detailed enough for the purpose of this study. For example, detailed information on the emissions from different vehicle categories is needed. Further, division of emissions of hydrocarbons on exhaust emission and evaporative emissions are needed as well as a breakdown of PM emissions on exhaust particles and wear particles. Therefore a detailed partition of the traffic emissions in the gridded EMEP emission database is done for all countries in the EMEP model domain with help of the data on national emissions in the GAINS model (GAINS 2010) which includes a detailed breakdown of emissions to vehicle categories, fuels used etc. An exception is made for Sweden where the partition of emissions is calculated with the Artemis model (with the exception of the wear particles that are not covered by Artemis and are taken from GAINS). The GAINS model is an instrument used for policy scenario development and optimisation and contains data of activities (e.g. fuel and energy consumption), emission factors and emissions from different sources specified for detailed categories. This model has a two-way connection to EMEP program in general and to the EMEP model in particular as the transport, chemical conversion and deposition matrix for the pollutants emitted from individual countries in GAINS are pre-calculated by EMEP and the future policy scenarios in EMEP are generally consistent with GAINS scenarios as both tools are used for the LRTAP convention and the EU legislation work. In the scope of this project it is found that EMEP and GAINS emissions are not fully consistent, while the EMEP emission database is based on expert-assessed data reported by the member states the GAINS emissions are based on model calculations. The continental scale modelling was based on the EMEP emissions database since this is a gridded database commonly used in EMEP modelling which has its historical scenarios validated within the EMEP monitoring program.

While emission factors for petrol cars are relatively well established the data for E85 are much scarcer. The Artemis model does not contain detailed emission factors for E85 cars and the situation is similar for the EMEP model. The GAINS model considers only zero emissions of CO_2 while all other emissions are the same as for petrol. To obtain the emission factors for E85 cars data were sought in the open literature (see further below).

2.3 Traffic data

In order to calculate emissions, the emission factors must be applied to data on traffic volumes of the different vehicle categories. For Europe the mentioned EMEP policy scenario is used. Since the traffic volumes are not available in the EMEP database, the relative volumes of traffic in different vehicle categories relative to the total traffic volume are recalculated on national basis using the GAINS model.

For the whole Sweden the traffic prognosis and emissions obtained from the Artemis model are used. The results here are very similar to what can be found in the EMEP database, which is to be expected since Artemis is used in the Swedish reporting. Figure 2 shows the fleet composition in Sweden 2020 from the Artemis model.

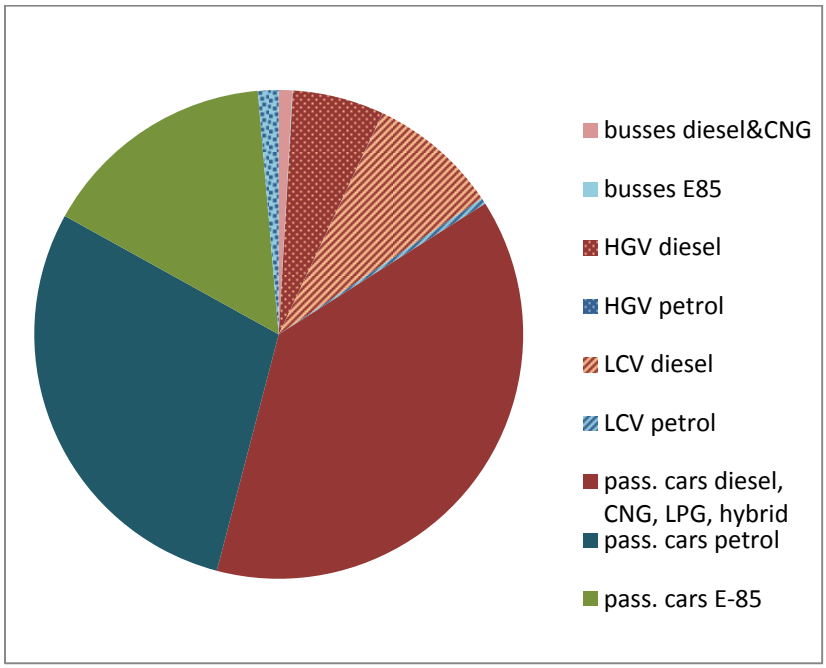

Figure 2. Contribution of vehicles and fuel categories to the total transport work (vehicle-kilometers/year) in 2020 in the Artemis model. S1-scenario: all categories with blue or green colour, except the E85 busses, use petrol. S2-scenario: all categories with blue or green colour use E85.

For the Västra Götaland region the traffic data for 2005 come from two sources. For the city of Gothenburg detailed traffic data are obtained from the local environmental agency. For the remaining Västra Götaland region traffic data from the Swedish Transport Administration are used.

The changes in all national Swedish emission sources in the EMEP model from year 2005 to 2020 are applied to the emission sources in the inventory for Västra Götaland to produce the 2020 emission scenario.

2.4 Emissions

The emission factors and traffic data are used to obtain emissions in two emissions scenarios: S1 where all ignition injection cars are assumed to use petrol and S2 where these cars all use E85. Note that all other road traffic sources, such as diesel cars, heavy duty vehicles etc., are treated in the same way in both scenarios. The emissions for Västra Götaland are also calculated for two base-scenarios, for 2005 and 2020, where the actual traffic and fuel composition (for 2005) and a prognosis on traffic and fuel composition (for 2020) from the Swedish Transport Administration are used. In the impact assessment, however, only the scenarios with all petrol or all E85 are used.

The input of emission data required by the EMEP model consists of annual national emissions. The species considered are sulphur dioxide (SO_2), nitrogen oxides ($NO_X = NO+NO_2$), ammonia (NH_3), non-methane volatile organic compounds (NMVOC), carbon monoxide (CO), and

particulate matter ($PM_{2.5}$, PM_{10}). These emissions are provided for ten anthropogenic source sectors, of which only emissions in the road traffic sector changes between the emissions scenarios calculated in this study. The emissions are also distributed to vary between months, weekdays and over the hours of the day. Distribution of the NMVOC emissions between different compounds which are specific to each emission sector and country are used. In this study this speciation will vary for road traffic between the scenarios as is described below.

For the Västra Götaland region the emissions from traffic and non-road machinery are distributed as 500 m x 500 m grids while emissions from industry and heating plants are treated as point sources (including all physical processes usually connected to that type of sources). Emissions from ships are described as line sources. The data are time resolved hourly over the day, over the days of the week for each month.

2.5 Dispersion modelling

Effects of the Europe-wide use of the E85 fuel on air pollution in Sweden are studied with two models on two different geographical scales. To capture the large-scale changes, especially those concerning ozone and other secondary air pollutants, the European-scale chemistry transport model EMEP is used (Simpson et al., 2003). The effects on exposure of people to air pollutants and consequent health effects are studied using air pollutant concentrations calculated with the small scale dispersion model TAPM which is nudged into EMEP air pollution fields (See Appendix 2 for details). In this way the large scale effects, such as formation of ozone, secondary PM and secondary aldehydes are accounted for also on the local scale. In the case of benzene, only the road traffic contribution is calculated due to lack of information on emissions from sources other than traffic.

2.5.1 Local scale modelling

The TAPM model (Hurley 2008) is used to calculate the local contribution to air pollutants in the Västra Götaland region in the two scenarios. The geographically distributed pollutants are then used for the exposure calculations and eventually the health risk assessment. TAPM is a three dimensional meteorological and chemical model for air pollution studies that includes topography as well as land-use. On the basis of this, the model generates the meteorological parameters required for the dispersion modelling, such as temperature layering (inversions) and three dimensional wind field, all based on daily synoptic input data (large scale meteorological re-analyse data –called GASP (Global AnalysiS and Prediction) from Australian Government Bureau of Meteorology (www.bom.gov.au). TAPM also includes chemistry such as NO/NO_2, ozone, SO_2 and particle transformation. Acetaldehyde and benzene are however not included in the chemical scheme of TAPM and these species are therefore simulated as reactive tracers in this study (see below).

In order to perform the calculations with TAPM a number of input data are required:

- Emission data for the Västra Götaland region as described above.
- Meteorological data.
- Levels of the relevant pollutants on the model domain boundaries for the two scenarios obtained from the EMEP model.
- A model describing the decay of benzene and acetaldehyde emitted from traffic.

Since acetaldehyde and benzene are not included in the chemical scheme of TAPM, their chemical lifetimes are set to be their rate constant with respect to the reaction with OH radicals multiplied by the OH mean concentrations obtained from the EMEP simulations (see below). Four different rates for the four seasons are used according to Table 1.

Table 1 Rate constant for reaction with OH and chemical lifetimes for acetaldehyde and benzene used in the TAPM simulations.

	k(OH)	Chemical lifetime (days)			
	$mol^{-1}* cm^3*s^{-1}$	Mar, Apr	May, Jun, Jul, Aug	Sep, Oct	Nov, Dec, Jan, Feb
Acetaldehyde	$1.5\ 10^{-11}$	1.2	0.4	1.3	15.5
Benzene	$1.2\ 10^{-12}$	14.8	4.9	15.9	193.7

2.5.2 EMEP modelling

Several EMEP models are developed and used for air quality policy work in Europe, mainly for the Convention on Long-range Trans boundary Air Pollution. The Unified EMEP model used in this study is designed to calculate concentrations in air and deposition patterns for major acidifying and eutrophying pollutants, photo-oxidants and particulate matter. In its standard version it has a 50 km x 50 km resolution in the horizontal direction and 20 layers distributed from the ground up to the pressure level of 100 hPa (c.a. 16 km) in the vertical direction. A detailed description is given in Simpson et al. (2003). The meteorological data used for the simulations, obtained from www.emep.int, are for the year 2005 and are of the same spatial resolution with a time resolution of three hours.

As already mentioned, the concentration fields generated by the EMEP model are used as an input to the TAPM model. Boundary concentration fields for TAPM are calculated from hourly concentrations in the grid cells surrounding the Västra Götaland region that are weighted together with wind field vectors on corresponding boundaries of the Västra Götaland grid cells. Since the chemistry of the organic compounds differs between the two models all organic compounds in EMEP are used and recalculated to the reactivity of organic compounds as used by TAPM.

While the role of NO_X, NMVOC and primary PM can be investigated directly by simulations of emission in scenarios S1 and S2, effects of E85 fuelled cars on concentrations of acetaldehyde, benzene and secondary PM are obtained from additional sensitivity runs. The contribution of the emissions from the use of E85 fuel on acetaldehyde concentrations can be modelled directly in the EMEP model, which includes also the formation and consumption of acetaldehyde in atmospheric reactions. However, TAPM does not include these processes and therefore the role of acetaldehyde formation is investigated further. Two additional EMEP simulations are run, one excluding primary acetaldehyde emissions and one excluding all NMVOC emissions from petrol and E85 fuelled vehicles. By relating the results of these runs to the results of S1 and S2 the contribution of primary and secondary acetaldehydes, respectively, can be deduced. The results from this analysis can be found in Appendix A2.3. The conclusion is that on the scale of the Västra Götaland region the formation of acetaldehydes in the atmosphere is not important and therefore the acetaldehyde concentration in TAPM can be described using the lifetimes given in Table 1. Further, also the impact of the NMHC emissions on the ozone levels is studied (Appendix 2.3) and found to be small.

Benzene is not included explicitly in the EMEP model. The benzene concentrations are therefore studied in sensitivity runs with benzene from road traffic added to the S1 and S2 scenarios as

methanol, which has similar atmospheric lifetime as benzene. The benzene concentration is then obtained by comparing the methanol concentration from the different runs.

The contribution of primary and secondary PM, respectively, is investigated by excluding the primary PM from petrol and E85 fuelled vehicles in separate runs of the EMEP model. The PM boundary values for the TAPM modelling are not separated into secondary and primary PM. The results from this analysis are used to draw general conclusions about the difference in secondary PM formation between the two scenarios.

2.6 Exposure

The human exposure of the annual mean concentrations of NO_X, NO_2, O_3, acetaldehyde and benzene are calculated based on the dispersion calculations of the Västra Götaland area in combination with the population density. The population density distribution for the region is obtained from EEA (EEA, 2009) and shown in Figure 3. The total population in the investigated area is 1 078 400.

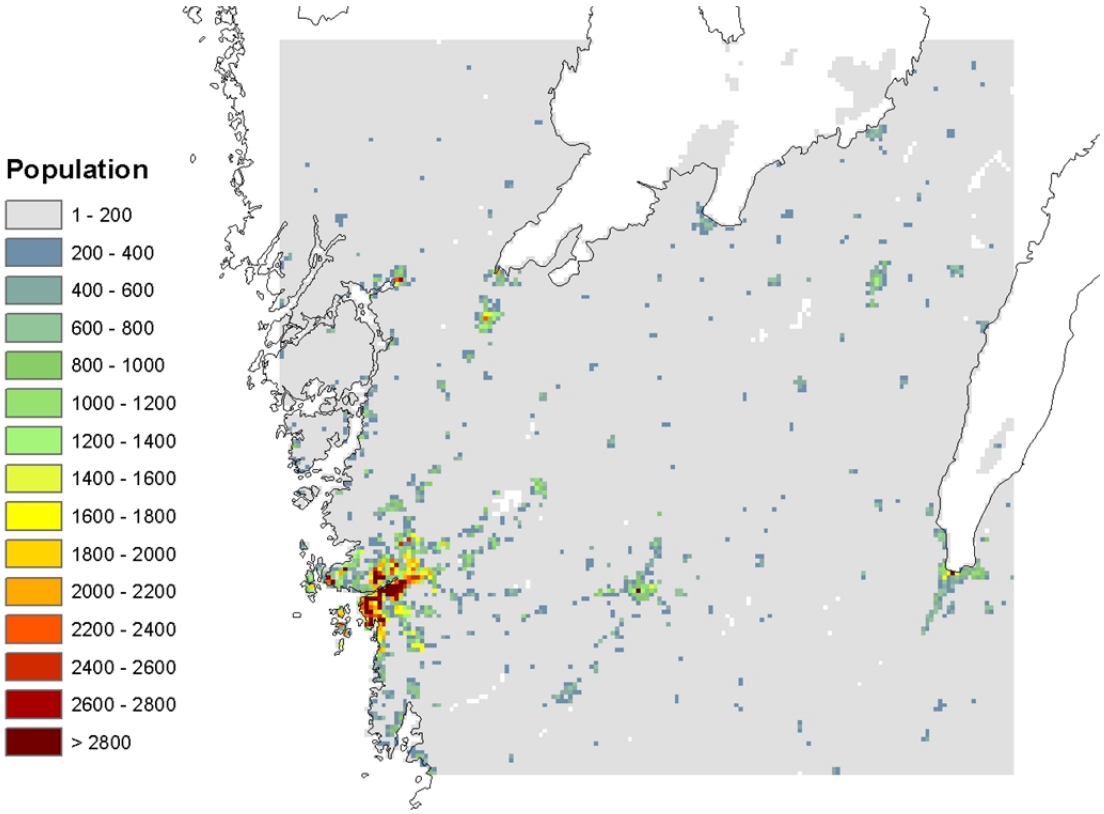

Figure 3. Population density distribution (number of people in a 1 km x 1 km grid for 2006) for the investigated area (according to Figure 1).

2.7 Health effects and risk assumptions

2.7.1 NO_X and NO_2

Using modelled levels of NO_X in the residential area as a vehicle exhaust marker, a Norwegian study of 16 000 men from Oslo, of whom 25 % died during the follow up, found a strong association (Nafstad et al., 2004). This cohort, with people of between 40-49 years of age at the start of the study, was followed from 1972/73 through 1998. NO_X was estimated in a model with 1000 m grids, and a street contribution added for the largest streets. When the median concentration of NO_X for 1974-78 was used (mean 10.7 μg/m³), the relative risk for total non-violent mortality was 8 % per 10 μg/m³. Nitrogen oxides do not have to be important as causal components behind this association.

2.7.2 Ozone

Short-term effects of ozone on the respiratory system and on daily number of deaths have been reported from a large number of studies, especially for summer concentrations, why a threshold has sometimes been assumed in impact assessments. Only recently has an effect of long-term exposure on mortality been reported from the very large American Cancer Society (ACS) Cohort Study (Jerret et al, 2009).

Jacobson (2007) reported that E85 would cause an increase in mortality but assumed an effect of short-term exposure only. The estimated impact was explained by the calculated ozone formation.

2.7.3 Acetaldehyde

An increased incidence of nasal tumours in rats and laryngeal tumours in hamsters were observed after inhalation of high concentrations of acetaldehyde (HEI, 2007). A few epidemiological studies of occupational exposure found associations but with a complex mixture of chemicals and have not been used in the cancer risk assessment for acetaldehyde. Acetaldehyde is classified by the National Institute of Occupational Safety and Health as "a potential human carcinogen", by the US EPA (1999) as "a probable human carcinogen", and by the IARC as "possibly carcinogenic to humans" (OEHHA, 1999; HEI, 2007).

2.7.4 Benzene

There is clear and widely accepted evidence from a variety of epidemiologic studies of occupational exposure that benzene increase the risks of acute myeloid leukaemia (HEI, 2007), but there is more uncertainty about other lymph hematopoietic cancers. Studies on traffic exposure and childhood leukaemia have produced mixed results with regard to associations.

2.7.5 Formaldehyde

IARC's Monograph 88 says that the evidence is sufficient to conclude that formaldehyde causes cancer in humans (IARC, 2006). This was because of evidence of nasopharyngeal cancer at concentrations historically encountered in industrial settings. However, this conclusion by IARC was largely based on the results from one occupational exposure site, and the results and conclusions have not been supportive in other reviews and more recent studies (Bosetti et al., 2006; Beane Freeman et al., 2009; Bachand et al., 2010). OEHHA (1999) uses a lifetime risk for cancer of 0.000006 per μg/m³ for formaldehyde, and US EPA (1999) a lifetime risk of 0.000013 per μg/m³.

For non-cancer effects the eye is the organ most sensitive to formaldehyde, subjective irritation has been reported at 300-500 μg/m^3, which is above the odour thresholds (Wolkoff & Nielsen, 2010). Effects in the eyes and in airways occur around 600–1000 μg/m^3. Effects on lung function have not been found in controlled human exposure studies below 1 mg/m^3. In epidemiological studies of associations between formaldehyde and exacerbation of asthma several co-pollutants make the findings difficult to interpret. An indoor air quality guideline of 100 μg/m^3 has been considered protective against both acute and chronic sensory irritation in the airways in the general population assuming a lognormal distribution of nasal sensory irritation (Wolkoff & Nielsen, 2010).

2.7.6 PM

In the studies of long-term exposure to $PM_{2.5}$ and mortality like the American Cancer Society (ACS) Cohort, the exposure contrast were mainly related to secondary PM, why the reported relative risk approx. 6 % increase in all-cause mortality (in adults) per 10 μg/m^3 is most relevant for background secondary PM concentrations (Pope et al, 1995).

2.7.7 Health Impact Assessment

To summarize, the types of pollutants that are found possible to include in the health impact assessment are NO_X and NO_2, benzene, acetaldehyde, formaldehyde, secondary PM and ozone. The assessment is restricted to the most important and well established effects of these pollutants. This means that not all types of potential health impacts are included. Results should however do for a comparison of the scenarios.

The impact on formaldehyde levels are not calculated why it is not included in the current assessment. Relative risks from epidemiological studies of vehicle exhaust using NO_X and NO_2 as exposure variables, may be influenced from effects of their co-pollutants, mainly ultrafine particles (elemental carbon, soot), CO and HCs such as formaldehyde. Exhaust particles are often introduced in a relatively high background of particles from other sources. Thus, exhaust particles were not measured directly in the important studies of long-term exposure and effects on mortality.

3 Results

3.1 Emission factors

The emission factors are selected from available published data, both for the exhaust and the evaporative emissions. In both cases values relative to petrol are used. The majority of the studies have been done with both E85 and petrol making this comparison more robust.

Westerholm et al. (2008) tested two flexi-fuel cars with E5, E70 and E85 in the European driving cycle NEDC and in the Artemis driving cycles. The former is used at two temperatures (22°C and -7°C). They report the regulated emissions, greenhouse gases, a number of organic compounds and particle number. Graham et al. (2008) present measurement results for regulated emissions, greenhouse gases and a number of organic compounds from four cars with E85 and regular petrol in the FTP cycle. They also made a compilation of literature data for ethanol cars. Yanowitz and McCormick (2009) present an analysis of US certification testing data in order to compare emissions for E85 and petrol.

The results from the three studies (for flexi fuel cars), and in addition the values used by Jacobson (2007), are given in Table 2.

Table 2. Emissions factors for E85 fuelled cars relative to the emission factors for petrol fuelled cars.

Relative change E85 vs. petrol (%)	Yanowitz and McCormick (2009) (95% conf)	Graham et al. (2008) (sd %)	Westerlund et al. (2008) (average for NEDC)	Jacobson (2007)
NO_X	-14 (-23 - -5)	-42 (15)	-38	-30
NMOG	26 (17 - 37)	12 (39)		
NMHC	-43 (-52 - -32)	-45 (24)		
THC	-14 (-24 - -4)		2	
TOG				22
CO	-19 (-25 - -12)	-5 (51)	1	5
PM			25	0
Formaldehyde	59 (32 - 91)	80 (44)	184	60
Acetaldehyde		2855 (1189)	1031	2000
1,3 Butadiene		-64 (31)	-74	-10
Benzene		-72 (15)	-71	-79
Ethanol			3950	from TOG
Methane			50	43

Table 2 shows lower emissions of NO_X for E85 fuelled cars relative to petrol fuelled cars from test cycles generally including cold starts. The emissions of hydrocarbons are higher for petrol while the emissions of organic gases are somewhat higher for E85. The emissions of aldehydes, especially acetaldehyde, are much higher for E85 as are the emissions of ethanol and methane. The emissions of benzene and 1,3-butadiene are higher with petrol. It should be noted that the data in Table 2 are in general for tests performed at ambient temperatures of around 20 °C. Westerlund et al (2008) report that for ambient temperatures of -7 °C the emissions of CO and NMHC may be significantly higher with E85 compared with petrol. For one vehicle they also report very high NO_X emissions with petrol at this lower temperature.

As seen in Table 2, there are a number of different quantities when it comes to the emission of organic compounds or unburnt fuel. From a chemistry point of view HC is no molecule but a notation for the sum of all compounds that are hydrocarbons, thus are of the form H_xC_y and not containing any elements but carbon and hydrogen. Total organic gases (TOG) include organic compounds containing oxygen such as alcohols and aldehydes (but not CO or CO_2). Volatile organic compounds (VOC) means carbon containing organic compounds present in the gaseous state at ambient temperature. Non-methane hydrocarbons (NMHC) means HC minus the methane mass. NMTOG and NMVOC are defined in the corresponding way[1].

[1] When it comes to exhaust gas regulations, the definitions differ somewhat. In the US, THC is defined as the signal obtained from a FID (Flame Ionisation Detector) instrument calibrated on propane. This means that alcohols and aldehydes contribute to THC but to a lower extent since their responses in a FID instrument are lower than for hydrocarbons. TOG is defined in the US regulations as THC + the mass of alcohols (e.g. methanol) and carbonyls (e.g. formaldehyde) measured separately. NMHC is equal to THC – r_{CH4}*CH_4 – r_{alc}*Alc where the CH_4-concentration is measured by gas chromatography and a FID calibrated with methane, Alc is the alcohol concentration and r_{CH4}¤ and r_{Alc} are the respective FIDresponse functions. NMOG means TOG - CH_4 and VOC is TOG - CH_4 - C_2H_6. In the regulations it is stated that also a number of other non-ozone forming organic compounds, e.g. acetone, should be subtracted but these are present in very low concentrations in engine exhaust. Note that there is actually a double counting of aldehydes in NMOG since they contribute to a certain extent to the NMHC value and then are added using values from separate measurements.

There will be emission limits for both THC and NMHC in also the future. For petrol cars, with low methane emissions, the challenge will be to meet the limit for NMHC. For E85 cars with higher methane emissions, the challenge will be to meet the THC limit. Thus it is reasonable that the emissions of NMHC from E85 cars will be lower than for petrol cars.

In the modelling presented here the emission factors from Graham et al (2008) are used with some exceptions as will be discussed. The reason is that that study comprises a compilation of a relatively large number of tests. For CO the emission factor is set to the same value for E85 and petrol since the differences are small and within the uncertainty. For particle emissions the studies cited in Table 2 give little information. Instead data from Ericsson et al. (2008) are used, indicating that the primary PM emission using E85 is about 65% of that when using petrol. For NO_X the emission for E85 is set to 58% of that for petrol.

The emission factors for NMHC (physical emission in g/km), including ethanol, are in this study set to the same value for E85 as for petrol. This is in line with the data for NMOG reported by Graham showing only small differences. It is also reasonable considering that the car producers optimize emissions to meet the future emission legislation in Europe for NMHC and THC, i. e the Euro 6 standards that apply from September 2014: For petrol, with low CH_4 emissions, the NMHC value will be limiting and this is on 68 mg/km. For E85, with higher CH_4 emissions, the THC value of 100 mg/km will be limiting. When considering the FID response as a measure of emission instead of physical emission, about 50% of the THC comes from NMHC for E85 cars. However, the FID response is low for ethanol which means that the physical NMHC emission is about 25% higher. Considering these factors, the emissions of NMHC (physical) for E85 can be expected to be around 50 mg/km * 1.25 = 62.5 mg/km which is about the same as for petrol. The methane emission for E85 is set to 1.42 times the value for petrol following Lipman and Delucchi (2002). Further, also the differences in the evaporate emissions are considered as is described in Appendix 1.

The emissions factors for petrol cars are taken from the Artemis model (André, 2005). The distribution of NMHC for petrol cars is from Passant (2002). The resulting emission factors used to obtain the emissions scenarios are presented in Table 3.

In Europe THC is defined as the signal from a propane calibrated FID and thus contains oxygen containing compounds, although the response factors for the latter are lower. The FID instrument gives the number of molecules and in order to calculate the mass the EU regulations use the densities obtained from assuming the molecular formulas $C_1H_{2.74}O_{0.385}$ (20.9 g/mole) for E85 and $C_1H_{1.89}O_{0.016}$ (14.15 g/mole) for petrol. This thus compensates for the lower response factor for ethanol and acetaldehyde. NMHC is obtained by subtracting the concentration of CH_4 (times the FID response factor) measured separately.

Table 3. Emission factors (g/km) for 2020 for passenger cars (including evaporative emissions but excluding wear particle emissions) used in the modelling

	Petrol	**E85**
NO_X	0.112	0.065
CO	1.79	1.74
PM	0.0013	0.0008
CH_4	0.013	0.018
NMHC	0.33	0.28
division of NMHC:		
Ethanol	0.0000	0.1898
Acetaldehyde	0.0025	0.0299
Ethene	0.0244	0.0237
Ethyne	0.0094	0.0011
Formaldehyde	0.0057	0.0072
Benzene	0.0187	0.0045
1,3 Butadiene	0.0030	0.0009
Ethane	0.0107	0.0009
Alkanes C3+	0.0928	0.0076
Alkenes C3+	0.0301	0.0025
Aromatics C7+	0.1297	0.0106
Aldehydes/ketones C3+	0.0051	0.0004
sum NMHC	*0.3321*	*0.2792*

3.2 Emissions

The emissions from road traffic vary between the two scenarios due to the differences in the emission factors. From the EMEP modelling the total emissions from road traffic in the different scenarios are calculated for the countries included. In Figure 4 emissions of NO_X, NMVOC and PM in the S2 (E-85) scenario relative to the S1 (all-petrol) scenario are shown for these countries. The relative change is larger for countries with a high fraction of petrol cars relative to diesel cars (since the latter are the same in the two scenarios) such as Russia and Ukraine. The relative change is smallest for PM emissions due to the very small contribution of the exhaust particles from petrol fuelled cars compared to the contribution from diesel vehicles and wear particles.

Figure 4. National emissions of NO_X, NMVOC and PM from the road traffic sector in E85 scenario (S2) relative to 'all-petrol' scenario (S1).

An important difference in the emissions between the scenarios lies in the speciation of the NMVOC emissions. Figure 5 shows this speciation of NMVOCs for all road traffic emission categories used in this study. The two rightmost bars in Figure 5 show the NMVOC speciation for Swedish road traffic emissions for the two scenarios S1 and S2. Comparison with Figure4 shows that the relative changes between the two scenarios are much larger for some individual NMVOC species that for the total NMVOC emissions. Since the NMVOC emissions from traffic in Sweden are dominated by the emissions from petrol cars (about 85% of the road traffic emissions of NMVOC comes from petrol cars), the variations in NMVOC speciation between petrol and E85

will have a significant impact on the total road traffic NMVOC speciation. As can be seen in Figure 5, scenario S1 is dominated by alkanes and aromatics while scenario S2 is dominated by ethanol. It can also be noted that S1 contains a significant emission of benzene which is much lower in S2, while the opposite is true for acetaldehyde.

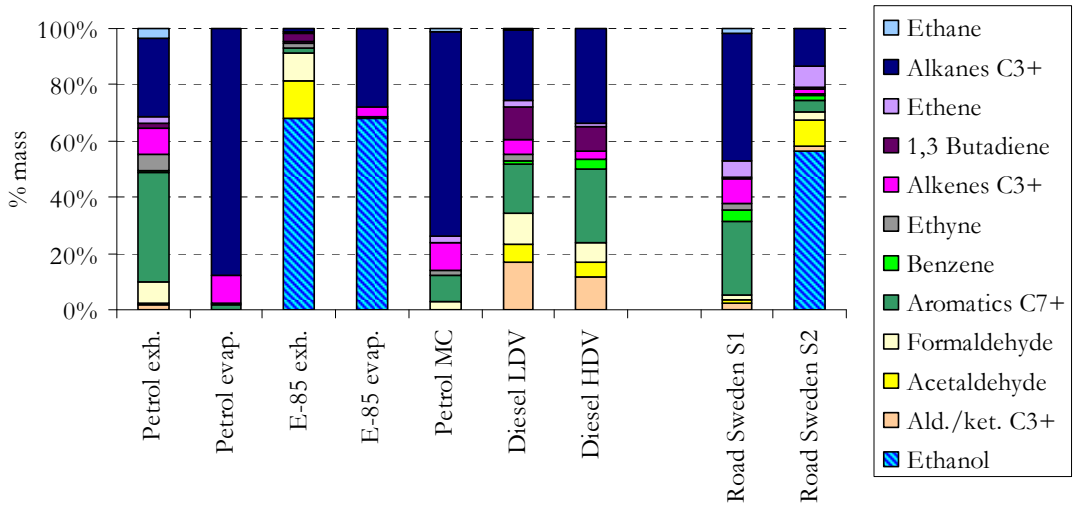

Figure 5. Distribution of hydrocarbon species in NMVOC emissions from individual road traffic emission categories as defined in emission scenarios in this study and from the whole road traffic emission sector in Sweden for scenarios S1 (All petrol) and S2 (All E85).

Figure 6 shows a map of the EMEP domain with emissions of acetaldehyde for the two studied scenarios as an example of the geographical distribution of emissions.

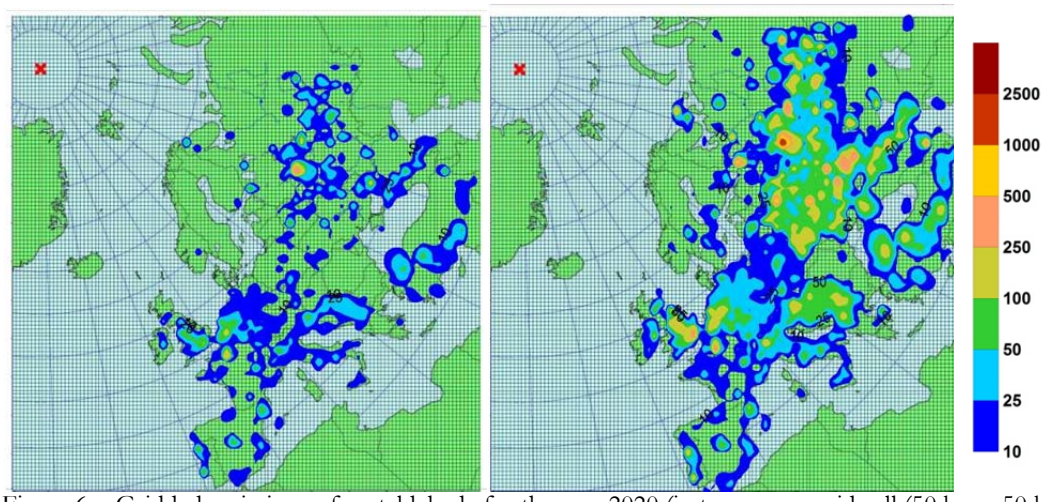

Figure 6. Gridded emissions of acetaldehyde for the year 2020 (in tonnes per grid cell (50 km x 50 km) and year). a – 'All petrol scenario, b – 'All E85 scenario'

As mentioned, the emissions from road traffic in the Västra Götaland region are calculated in a 500 m x 500 m grid. Figure 7 shows the emissions of NO_X for 2005, as obtained from the database, and the projected emissions for 2020 with a mixed use of E85 and petrol. This can be considered as the base case for the emissions in 2020. It can be seen that the emissions decrease over the period which is due to reduction in NO_X emissions factors as older vehicles are replaced by newer models (the traffic actually increases from a total of 75.4×10^9 vehicle-kilometres in 2005 to 93.7×10^9

vehicle-kilometres in 2020). The major roads in the region are clearly visible as well as the larger cities (especially Gothenburg).

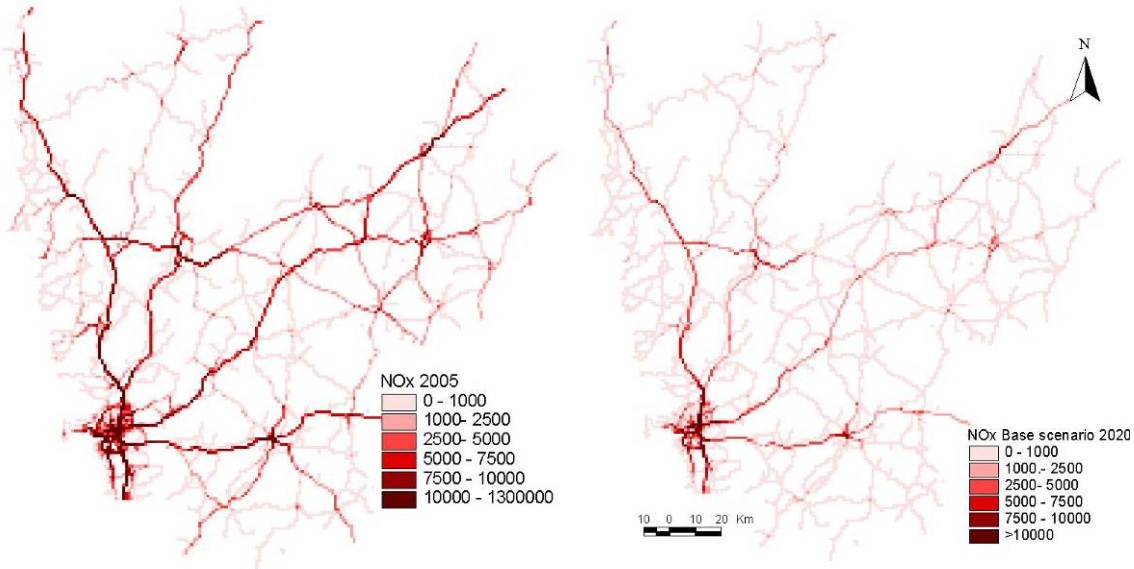

Figure 7. Emissions of NO_X (kg/year for a 500 m x 500 m grid cell) in the Västra Götaland region for 2005 and for 2020 for the base case.

In Table 4 the traffic emissions for NO_X, benzene, acetaldehyde, ethanol and PM_{10} for the Västra Götaland region are shown for the four different emissions scenarios. The emissions of NO_X and benzene are reduced between 2005 and 2020 for the base case while the emissions of ethanol and acetaldehyde increase due to an expected increase in the use of ethanol fuel. The NO_X emissions for S1 are somewhat higher than for S2 due to the higher NO_X emissions factor for petrol compared with E85.

Table 4. Emissions from traffic for the Västra Götaland region in four different emission scenarios (in tonnes/year).

Substance	base 2005	base 2020	S1	S2
NO_X	13 243	4 843	4 954	4 631
Benzene	2 833	927	1 201	386
Acetaldehyde	628	1 074	305	2 595
Ethanol	192	5 192	0	15 400
PM_{10}	1 324	486	483	476

3.3 Dispersion modelling

Resulting concentrations from calculations of the S1 and S2 scenarios with the EMEP model are used to provide input of primary and secondary air pollutants for the calculations with the TAPM model for the Västra Götaland region. Concentrations of NO_X, NMVOC, $PM_{2.5}$, PM_{10}, acetaldehyde, ethanol and benzene are therefore calculated as boundary conditions for the Västra Götaland region. In addition, sensitivity studies are performed to investigate the role of formation of secondary acetaldehyde and secondary particulate matter (see Appendix 2).
Time series analysis of the TAPM and EMEP model results of the S1 and S2 scenarios are presented in Figure 8.

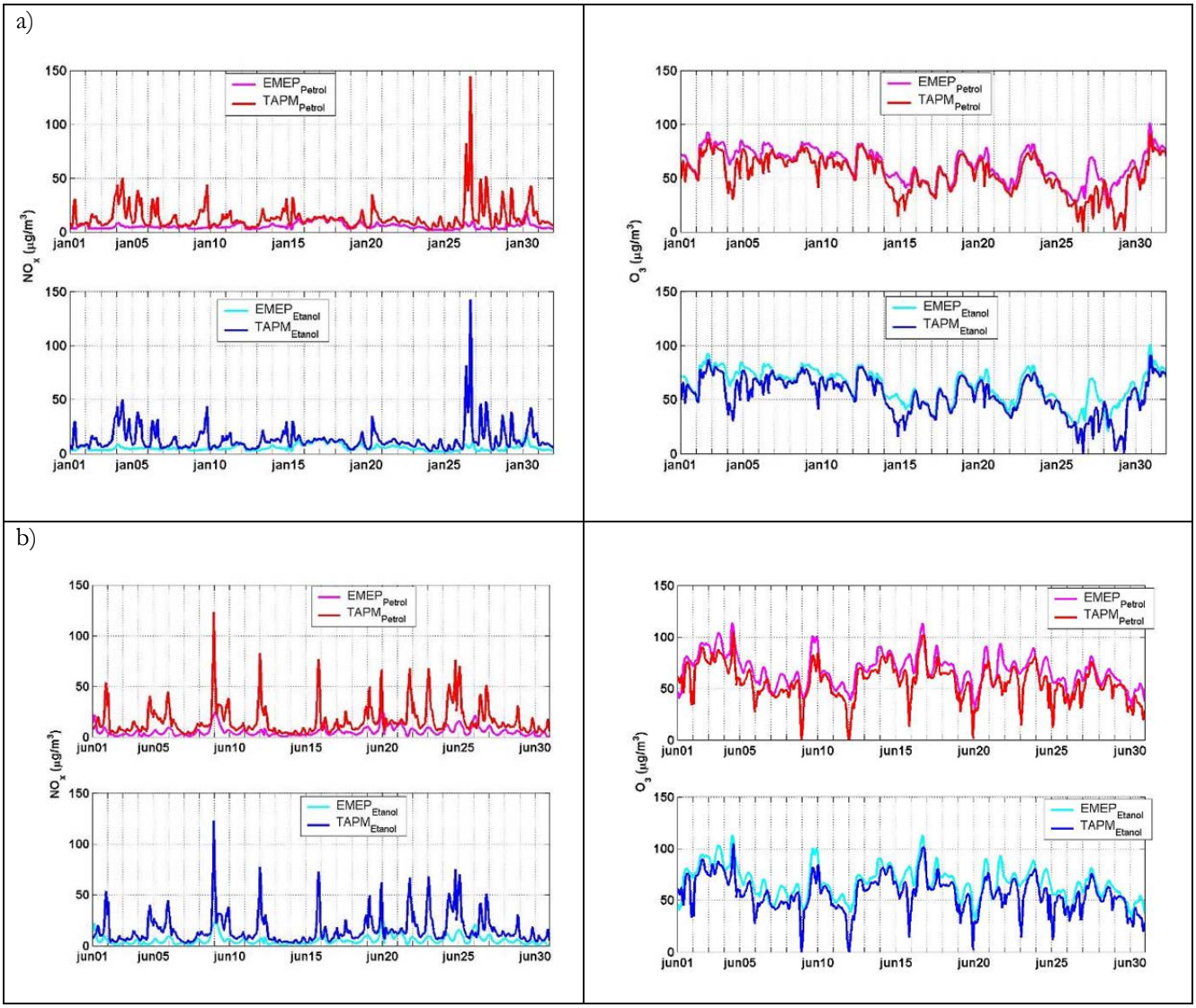

Figure 8. Time series analyses of S1 and S2 scenarios for the EMEP and TAPM modelled concentrations of NO_X and ozone at an urban background site in Gothenburg (TAPM) and in EMEP model grid cells including Gothenburg city (50 km x 50 km).

Concentrations of NO_X and ozone at an urban background site in Gothenburg (TAPM), and in the 50 km x 50 km EMEP grid cells including Gothenburg, are compared for one summer and one winter month. Due to the well-known process of ozone titration by freshly emitted nitrogen monoxide (Seinfeld and Pandis, 1998) the concentration of ozone often decreases in urban areas. This phenomenon is expected to be captured by the fine-gridded TAPM model but not necessarily by the courser scale of the EMEP model. Figure 8 shows the generally higher NO_X levels in the small grid cell of the TAPM model in urban background location comparing to those in the large EMEP grid cell while the opposite is observed for the ozone concentrations. (Remember that the boundary concentrations for TAPM are obtained from the EMEP model and they are thus the same in both cases.) This indeed shows that local scale emissions and processes contribute to locally increased levels of NO_X in urban areas, both for the summer and the winter cases. For ozone, where there of course are no primary emissions, the situation is the opposite where instead the result is decreased concentration levels at urban sites on a local scale. The influence of the

NMHC emissions from the passenger cars is studied in a sensitivity analysis (see Appendix 2.3) and it is found to have only a small influence on the ozone levels.

To illustrate the results of the different scenario calculations, the geographically distributed concentrations are presented as the difference in yearly average concentrations between the all E85 (S2) and all petrol (S1) scenarios, both as μg/m^3 and sometimes as percentage of the difference to scenario S1 (see Appendix 3).

The difference in yearly average concentrations between scenario S1 and S2 for acetaldehyde and benzene can be seen in Figures 9 and 10, respectively. The increase in acetaldehyde concentration resulting from a switch from petrol to E85 is up to about 0.5 μg/m^3 or 80% in the central parts of the largest urban area of the region, Gothenburg. Outside this area the relative increase is about 10 % for the other urban areas and less than that for the rural areas. This large relative increase (see map in Appendix 4) is a result of the relatively high emissions of acetaldehyde from E85 fuelled cars. For benzene the situation is opposite. In the scenarios for 2020 the concentration of benzene decreases by up to a few tenths of μg/m^3 as a result of a switch from petrol to E85. For benzene the relative change is not calculated.

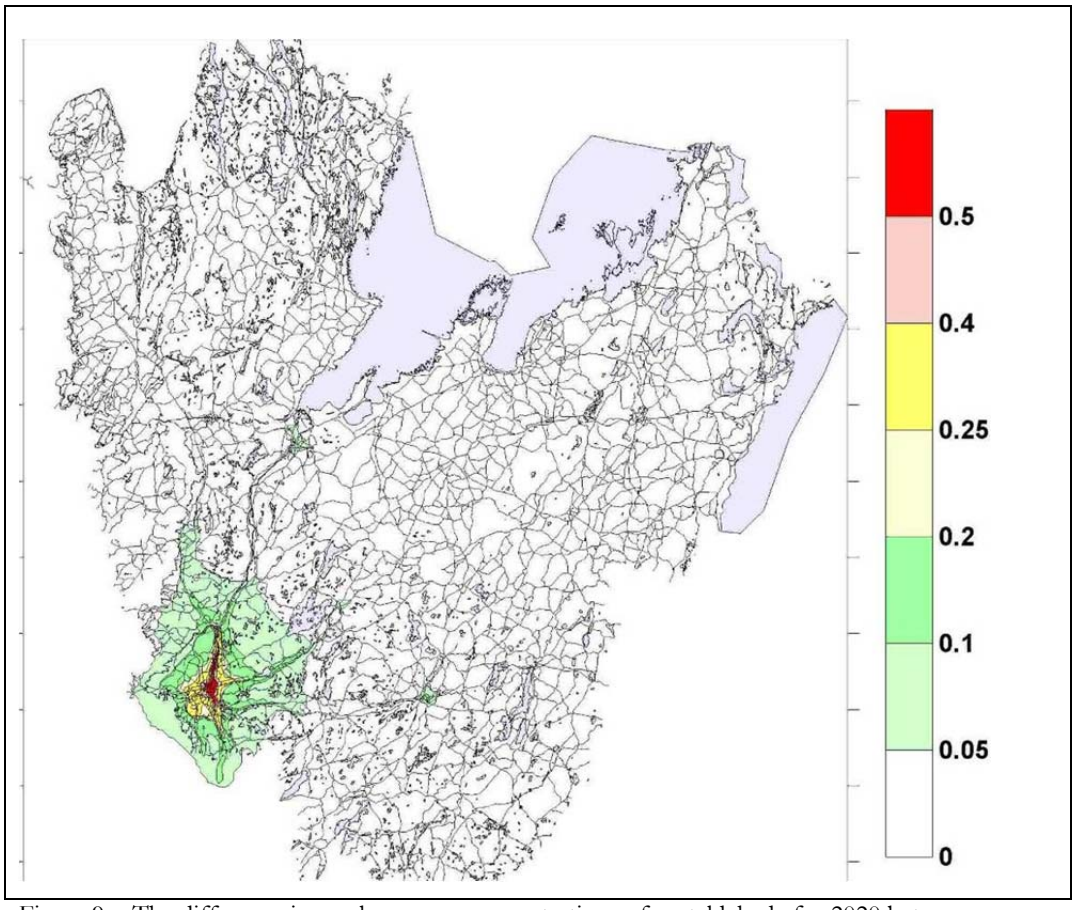

Figure 9. The difference in yearly average concentrations of acetaldehyde for 2020 between the all E85 (S2) and all petrol (S1) scenarios (μg/m^3).

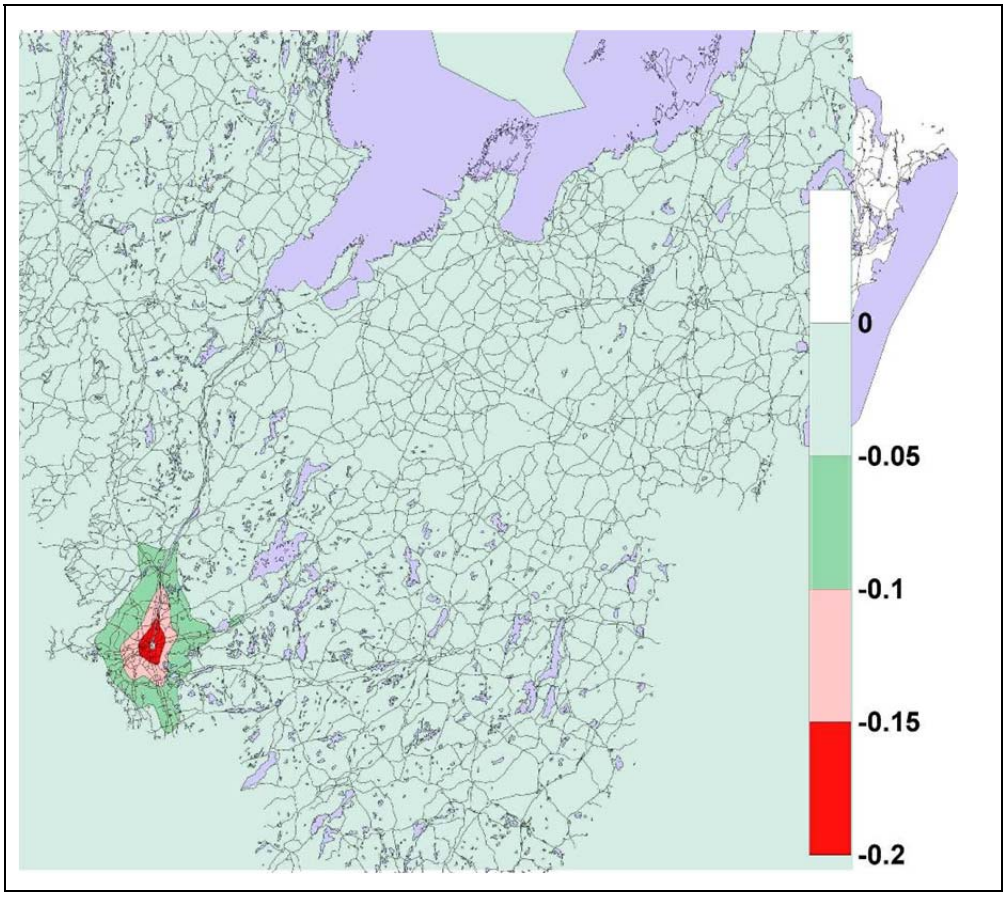

Figure 10. The difference in yearly average concentrations (μg/m^3) of benzene for 2020 between the all E85 (S2) and all petrol (S1) scenarios.

The difference in the calculated NO_X, NO_2 and ozone concentrations between the two scenarios are shown in Appendix 4. The change in NO_X concentration between the two scenarios only represents a small fraction of the total NO_X concentration. This is because the emissions of NO_X from petrol cars are relatively low compared with the contribution from diesel engines and other sources. Thus, the resulting decrease in the NO_X concentration with E85 is only up to 3%. For ozone there is a small increase of about 0.1 μg/m^3 of E85 in the central areas of Gothenburg where there is relatively high traffic intensity (see Appendix 4 Figure A9). In the rest of the region there is a small decrease in the ozone concentration in the E85 scenario.

3.4 Exposure

The resulting concentrations from the dispersion modelling together with the population density distribution are used to assess the exposure to acetaldehyde, benzene, NO_2, NO_X and ozone for the two scenarios S1 and S2. The exposure to $PM_{2.5}$ is very similar for the two scenarios (the difference in concentration between scenarios S2 and S1 is only about -0.02 μg/m^3. Table 5 shows the population weighted mean exposure both as total concentrations and as the difference between the two scenarios. Detailed exposure results can be found in Appendix 5. It can be seen that the exposure to all the pollutants in Table 5, with the exception of acetaldehyde, are somewhat lower for the all E85 scenario (S2) than for the all petrol scenario (S1). However the effect on health also

depends on the level of the dose-response functions. For example if the level of concentration is low in a populated area but the dose-response function is high, the effect of even a low concentration increase can become important. The increase in exposure from S1 to S2 (S2-S1) calculated as percentage of the population exposed to increased mean concentrations of pollutants is presented in Figure 11.

Table 5. Populated weighted mean exposure (μg/m³).

	S1	S2	S2-S1
Acetaldehyde	0.65	0.77	0.14
Benzene	0.1	0.05	-0.07
NO_X	8.5	8.4	-0.12
Ozone	57.0	56.9	-0.12
$PM_{2.5}$	2.70	2.68	-0.02

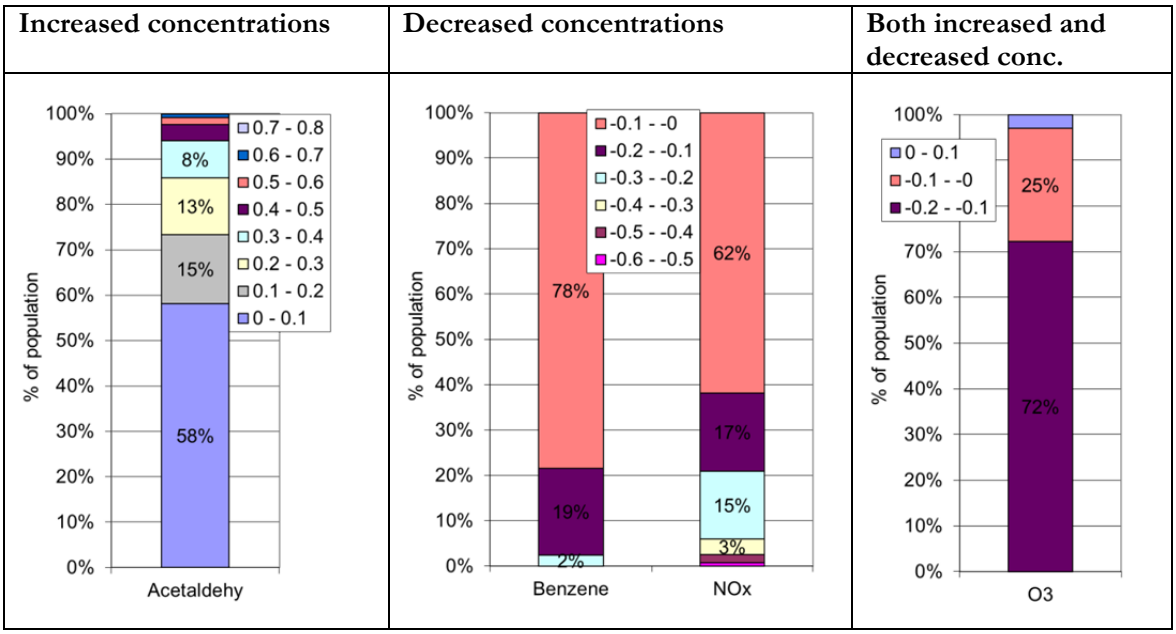

Figure 11. The relative exposure (S2-S1) calculated as percentage of the population.

In Figure 11 it is clear that the only pollutant that does not show an improvement regarding air quality and thus the health risks, when modelling a switch from petrol to E85, is acetaldehyde. For all pollutants, except for ozone, the changes of concentration levels observed between the scenarios are within the urban areas, but for ozone there are also differences in rural areas.

According to the data in Figure 11, about 14% of the population in the Västra Götaland region will be exposed to 0.3-0.8 μg/m³ higher concentration levels of acetaldehyde in scenario S2 relative to S1. From Figure 9 it can be seen that the majority of these people live within the Gothenburg area. Further, about 28% of the population will be exposed to increased levels of between 0.1 to 0.3 μg/m³, mainly in the suburban areas and rural surroundings of Gothenburg, but also in some of the smaller cities in the region. Outside the urban areas the concentration difference is lower than 0.1 μg/m³.

In the S2 scenario relative to the S1 scenario, the population is exposed to decreased concentration levels of benzene and NO_X. This is mainly true also for O_3 but with an exception for the central

parts of Gothenburg where the concentration actually increases (see Appendix 4 Figure 7A) resulting in that 3% (blue area in Figure 11 for O_3) of the population will be exposed to higher concentrations of O_3. For the rest of the area there are decreased concentrations of ozone upon a switch to E85.

The largest differences are again in the central parts of Gothenburg where about 20% of the population is being exposed to a lower benzene concentration of 0.1-0.3 μg/m³. This also means that, concerning benzene, the majority (78%) of the population have none or very low influence from a change to ethanol fuel.

The largest effect on exposure difference is seen for NO_X where just over 20% of the population has its exposure decreased with 0.3-0.5 μg/m³ (in the central parts of Gothenburg). In the suburbs the decrease is between 0.1-0.2 μg/m³ lower. Along the largest roads there is a small but visible improvement on a yearly basis of 0.1 μg/m³.

For $PM_{2.5}$ there is a difference in the populated weighted mean exposure of 0.02 μg/m³ between the scenarios, with a lower value for the all-E85 case (see also Appendix 5).

3.5 Impact on health in Västra Götaland

3.5.1 NO_2 and NO_X

The impact of long-term exposure on mortality was estimated using the relative risk from the Norwegian cohort study (Nafstad et al, 2004), reporting a relative risk of 0.8% increase per μg/m³ increase in NO_X, however without adjustment for particle concentrations. Assuming this effect on the total mortality and a typical baseline mortality of 1000 deaths per 100 000 person years, a decrease in the population mean exposure of 0.12 μg/m³ would correspond to approximately 10.4 less preterm deaths per year. However, for the present case this number is highly uncertain since the exposure to substances normally associated with changes in NO_X exposure may here change in different ways since the relation between different exhaust components varies between the scenarios.

The impact of long-term exposure on asthma incidence (new cases) in adults has been estimated using results from the Swedish RHINE Study Cohorts (Modig et al., 2009), with very similar results in the ECRHS Study, corresponding to approx. 4% increase per μg/m³ increase in NO_2. From the RHINE Study the incidence can be estimated to 0.3% per year (proportion of adults developing the disease), with children and elderly excluded this means around 1500 adult cases per year. A reduction of 0.09 μg/m³ corresponds to 5.4 avoided new cases of adult asthma per year. However, also NO_2 in the asthma study may have been mainly an indicator of vehicle exhaust.

3.5.2 Ozone

The impact of long-term exposure on respiratory mortality has been reported to be approx. 0.2% per μg/m³ increase in the summer concentration when studied in the large ACS Cohort (Jerret et al., 2009). Assuming this effect on the total population and a typical baseline respiratory mortality of 100 deaths per 100 000 person years, a decrease in the population mean exposure of 0.12 μg/m³ corresponds to approximately 0.3 less preterm deaths per year.

3.5.3 Acetaldehyde

According to US EPA (1999) a lifetime risk for cancer of 0.000002 per μg/m³ is to be assumed for this potential human carcinogen. An increase in the population mean exposure of 0.12 μg/m³ corresponds to 0.26 cases more per year in the current population.

3.5.4 Benzene

According to US EPA (1999) a lifetime risk for cancer (leukaemia) of 0.0000022-0.0000078 per μg/m³ is to be assumed for this human carcinogen. A decrease in the population mean exposure of 0.074 μg/m³ corresponds to approximately 0.3 cases less per year in the current population (assuming 0.000004 per μg/m³).

3.5.5 $PM_{2.5}$

In CAFÉ and other impact assessments a relative risk of 6% increase in all-cause mortality per 10 μg/m³ has usually been assumed. Assuming this effect on the total mortality and a typical baseline mortality of 1000 deaths per 100 000 person years, a decrease in the population mean exposure of 0.02 μg/m³ corresponds to approximately 1.3 less preterm deaths per year.

4 Discussion

In this study the health risks from a large introduction of E85 to replace petrol is analysed in scenario studies for 2020 for the Västra Götaland region. It is found that the change in pollutant concentration, exposure and health risks are small between the two scenarios. It should be emphasized that the study only considers vehicle emissions and does not take into account effects from the fuel production.

The emissions of NO_X in the model are somewhat lower from E85 fuelled cars than from petrol fuelled cars. This is in line with the emission factors used by Jacobson (2007). The nitrogen oxides are key substances in the atmospheric reactions determining the concentrations of ozone in air. NO_X is involved in the formation of ozone but close to the NO source the ozone concentration is lowered due to the rapid reactions between NO and O_3 forming NO_2. If NO is released in an environment with already high NO concentration, it may result in lowering of ozone levels on the regional scale. In the present study the ozone concentrations calculated are somewhat lower for the E85 scenario than for the petrol scenario.

The health risk analysis shows that the main difference between the scenarios is due to the lower concentration of NO_X. However, it may have been other exhaust components than NO_X, such as exhaust particles, that were behind the reported association with mortality. In cases when NO_X is not a good indicator of exhaust particle emissions and their toxicity, our results may be misleading. This should be studied further Neglecting the effects of the changes in NO_X exposure, the total difference between the scenarios sums up to 1.6 preterm deaths per year with lower values for the E85 scenario, while it would sum up to 12 less preterm deaths for the E85 scenario if the effect related to NO_X were included.. Since there are no similar studier for the effect on mortality among those younger than 30-50 years of age, we assume the same relative effect on mortality in all ages. This makes only a small difference since the large majority of deaths occur in older groups.

The opposite result for the ozone concentration was obtained by Jacobson. The reason for this discrepancy is that the Jacobson study concerns Los Angeles, a region with high concentrations of pollutants, while the study here concerns the Västra Götaland region with much less dense traffic and also lower concentrations of pollutants, importantly NO_X. It is not unlikely that the effect that Jacobson report also would be found for large parts of Europe with higher traffic densities than in the Västra Götaland region.

It is worth to emphasize that the major negative health effect in the Jacobson (2007) study is attributed to the increase in ozone concentration that is a result of the decrease of the NO emissions from E85 cars. Directly after the emission, NO reacts with ozone resulting in drop of the ozone concentration, the so-called ozone titration, commonly observed in many cities. This NO does, however, form even higher quantities of ozone further downwind contributing to the general increase in the background ozone levels. It is also precursor of NO_2, secondary PM and other toxic species, as e.g. PAN, and hence reductions in NO emissions are never regarded as disbeneficial from the environmental viewpoint.

There are of course several sources of uncertainties that influence the results of this study. Since the main results concern differences between the two scenarios, several of these factors loose on importance for the conclusions. This comprises uncertainties in meteorology, in future traffic volumes and in the population density distribution. However, there are some uncertainties that remain important. The emission factors for E85 fuelled cars are much less characterised than what they are for petrol cars. The emission of particles from E85 fuelled cars is especially uncertain with only very few published results. Also the emissions of NMHC are somewhat uncertain and may be higher for E85 at lower ambient temperatures. However, the sensitivity analysis performed shows that for the studied region the NMHC emissions only have a small influence on the ozone concentrations.

5 Conclusions

In this study the impact of tailpipe and evaporative emissions from E85 fuelled cars on air pollution and health risks are calculated and compared with petrol fuelled cars. The scenario calculations for the Västra Götaland Region give data on emissions, pollutant concentrations, exposure and a health risk evaluation.

The differences in emission factors between E85 and petrol lies in the emissions of PM and NO_X which are about 65% and 58% for E85 relative petrol, respectively. Also the speciation of the emitted hydrocarbons is significantly different with large emissions of acetaldehyde and ethanol from E85 and large emissions of benzene, higher aromatics, alkanes and alkenes from petrol.

These differences in emission factors reflect in differences in emissions. The emission calculations for all Swedish road traffic show a decrease for the E85 scenario relative to the petrol scenario of 6.5% for NO_X, 3.4% for $PM_{2.5}$, 67% for benzene. For acetaldehyde there is an increase of 770%.

The differences obtained from the TAPM modelling show decreased levels of NO_X, ozone and benzene with E85 and increased levels of acetaldehyde. For the latter the increase may be up to 80%, while NO_X and ozone show decreases of up to a few per cent and a few tenths of per cent, respectively.

The health risk assessment shows decreased health risks in the E85 scenario relative the all-petrol scenario, due to the increased NO_X exposure, correlated with both preterm deaths and asthma. However, NO_X may be mainly an exhaust indicator. Smaller effects are expected from the changes in ozone, acetaldehyde and benzene exposure. The overall difference considering the exposure to PM, acetaldehyde, benzene and ozone is 1.6 preterm deaths with lower values for the E85 scenario.

6 Acknowledgements

Saab Automobile, The Swedish Transport Administration and The Foundation for the Swedish Environmental Research Institute are gratefully acknowledged for financial contribution.

Magnus Ekström and Maria Holmström at Saab and Martin Juneholm at the Swedish Transport Administration are acknowledged for their most valuable input to the study.

David Simpson, Norwegian Meteorological Institute, EMEP MSC-W, Norway and Dept. Radio & Space Science, Chalmers Univ. Technol., Sweden is gratefully acknowledged for help with implementation of the Unified EMEP model.

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Appendix 1. Evaporate emissions of NMHC

The following assumptions are done for evaporative emissions:
The NMHC emissions from E85 are reduced relative to petrol by 45%. Methane emissions are zero and the split of NMHC species is 68% ethanol and the remaining 32% is divided in the same way as evaporative emissions for petrol. The estimates by Graham et al. (2008) are missing evaporative emission data, however these assumptions agrees very well with measurements of evaporative emissions due to fuel permeation reported by Haskew et al. (2006) where NMHC emissions are reported to be reduced by 50% for E85 fuel compared to petrol. Haskew et al. also presents the NMHC split to species with 60% ethanol. This number agrees quite well with our assumption of 68% NMHC being ethanol. NMHC in exhaust and evaporative emissions are thus divided as in Table A1.

Table A1. Division of NMHC in the exhaust and evaporative emissions for E85 and petrol, respectively (wt.%).

	E85 exhaust	Petrol exhaust	E85 evaporative	Petrol evaporative
Ethanol	68.0	0	68.0	0
Acetaldehyde	13.1	0.8	0.2	0
Ethene	9.9	7.3	2.4	0
Ethyne	0.3	2.8	0.9	0
Formaldehyde	3.1	1.7	0.6	0
Benzene	1.6	5.6	1.8	1.0
1,3 Butadiene	0.3	0.9	0.3	0
sum	96.3	19.1	74.1	1.0

The remaining NMHC emissions are divided as for petrol (ref) and listed in Table A2.

Table A2. Division of NMHC in the exhaust and evaporative emissions for E85 and petrol, respectively (wt.%).

	E85 exhaust	Petrol exhaust	E85 evaporative	Petrol evaporative
Ethane	0.2	3.2	1.0	0
Alkanes (C3+)	1.3	27.8	8.9	88.0
Alkenes (C3+)	0.4	9.0	2.9	9.5
Aromatics (C7+)	1.8	38.8	12.5	1.5
Aldehydes/ketones (C3+)	0.1	1.5	0.5	0

Appendix 2. Description of the model system

A2.1. Atmospheric lifetimes

Table A3 gives tropospheric lifetimes with regard to reaction of ethanol, formaldehyde, acetaldehyde and benzene (C_6H_6) with OH, nitrate radical (NO_3) and photolysis. The studied organic species are listed together with three species common in the gasoline exhaust (benzene, ethene and toluene) for comparison. Table 1.1 shows also photochemical ozone creation potential (POCP) of these species. POCP is a measure of amount of ozone that is formed from a certain VOC emission relatively to other emission source, which has been established to the same emission mass of ethene (Derwent et. al., 1996; Altenstedt et al., 2000) The range represents different scenarios for VOC and NO_X background concentrations. Derwent et al. (2007) used similar methodology to calculate POCP of different VOC sources and calculated POCP of mean road emission exhaust to be 69.

Table A3. Atmospheric lifetimes of studies NMVOC species with respect to the OH^- and NO_3^- initiated degradation τ(OH) and τ(NO_3) and with respect to the photolysis. (The lifetime of a chemical species which decays in a first-order process is the time needed for a concentration of this species to decrease to 1/e of its original value. Statistically, it represents the mean life expectancy of an excited species.) The OH concentration of 1×10^6 molecules/cm^3, typical for summer troposphere, was used for the lifetime calculation. The NO_3 concentrations of 5 and 100 ppt(V) were used for lifetime calculations as representative of rural and urban night-time 12 hrs. mean concentration. The studied organic species are listed together with 3 species common in the gasoline exhaust for comparison. The rates of photolysis were calculated with radiation transfer model for July, 58º latitude and integrated over 24 hours.

	$\tau_{(OH)}$	$\tau_{(NO3)}$ urban/rural	τ (photolysis)	POCP
	hr.	hr.	hr.	
Ethanol	85	56 / 1×10^3	-	44 - 63
Acetaldehyde	15	47 / 1×10^3	86	68 - 80
Formaldehyde	30	194 / 4×10^3	10	18 - 55
Benzene	214	-	-	23 - 47
Ethene	34	-	-	100
Toluene	50	-	-	30 - 67

A2.2 Defining of boundary concentrations for TAPM from EMEP fields

Calculation of initial concentration field for TAPM from EMEP concentration field of NO_2 (as example) and EMEP mean wind vector components u_mid and v_mid. Two EMEP model gridcells (i=56, j=60 and i=56, j=61) represent the TAPM model domain and concentrations from the surrounding EMEP gridcells SE-S, SE-N, NW-S, NW-N, NE, NW (Figure A1) are used for calculation of boundary concentration NO_2(bc) according to the following code:

```
if
  u_mid <= 0 then ( isite = SE-S and jsite = SE-N)
else
  isite = NW-S and jsite = NW-N)
endif
```

if
  v_mid <= 0 then (ksite = NE)
else
  ksite = SW
endif

$NO_2(bc) = [(NO_2(isite)*u\_mid(isite) + NO_2(jsite)*u\_mid(jsite))/2. +$
$NO_2(ksite)*v\_mid(ksite)] / [(u\_mid(isite) + u\_mid(jsite))/2 + v\_mid(ksite)]$

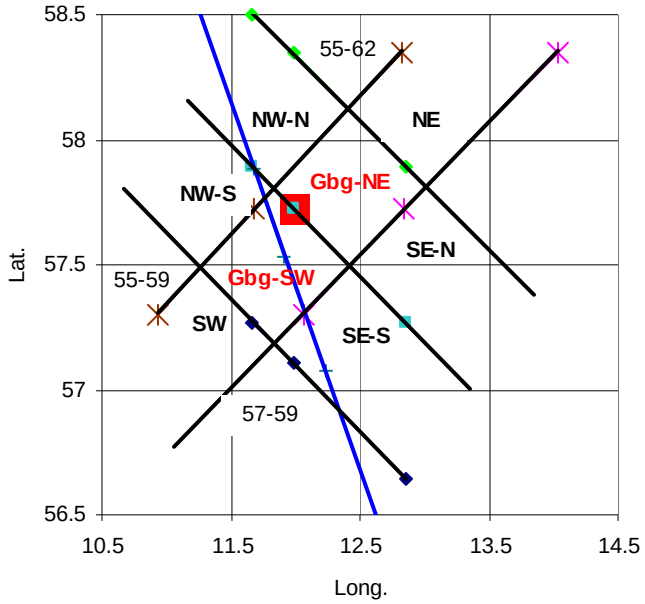

Figure A1. Gridcells covering Gothenburg domain and its boundary in EMEP model plotted on latitude-longitude coordinates. The red square represents centre of Gothenburg and the blue line the coastline. EMEP i and j coordinates for the boundary gridcells are: SE-S = 57-60, SE-N = 57-61, NW-S = 55-60, NW-N = 55-61, NE = 56-62, NW = 56-59 and for Gothenburg grid cells are: Gbg-NE = 56-61, Gbg-SW = 56-60.

A2.3 Atmospheric chemistry

The role of secondary chemistry in for the concentration of acetaldehyde are studied by first excluding the primary acetaldehyde emissions from the EMEP model simulations of scenarios S1 and S2 and then by excluding all NMVOC emissions from the car category with spark ignition engines. The contribution from the primary acetaldehyde emissions as well as the contribution of secondary acetaldehyde, formed from other NMVOC, to the total acetaldehyde concentration is then obtained. However, the secondary chemistry is highly nonlinear due to the interactions of NMVOCs with ozone and OH, and thus the results for the contribution of secondary acetaldehyde are somewhat uncertain. Differences in acetaldehyde concentrations between S1 and S2 scenarios are studied with respect to contribution of primary and secondary acetaldehyde: It is observed that in summer background air the monthly mean contribution of the secondary acetaldehyde is of the same magnitude as the contribution of the primary acetaldehyde to the difference between S1 and

S2. In the winter and in more polluted atmosphere (Gothenburg grid cells) the contribution from secondary acetaldehyde to the concentration difference appears to be negative due to the chemistry feedback, i.e. the E-85 scenario has less secondary acetaldehyde than the all-petrol scenario.

A comparison of S2-S1 differences in primary and secondary acetaldehyde concentrations in the background (=upwind) and Gothenburg grid cells show no increase in the secondary acetaldehyde from upwind to the Gothenburg grid cell. It can thus be concluded that there is no significant contribution from secondary aldehydes on the small scale (tenths of km). Conclusion from this sensitivity study is that the secondary formaldehyde can be of importance under summer in the clean background air but that formation on the local scale is unimportant. Using EMEP simulated boundary concentrations of acetaldehyde in the local scale model and simulating the primary acetaldehyde emissions will capture the most important features of acetaldehyde dispersion and chemistry in Västra Götaland.

In the EMEP model, the tail-pipe PM emissions are specified as primary particles. Emissions of NO_X and VOCs (and NH_3, SO_2) are oxidised in the atmosphere and a part is then transferred through the gas-to-particle transport to the PM as secondary PM. The petrol fuelled cars emit both more primary PM and more NO_X compared to the E85 cars. If we compare differences in primary and secondary PM between the S1 and S2 scenarios simulated by EMEP, we can directly see what part of the difference is from the tail-pipe emissions of the primary particles and what part is from the formation of the secondary PM in atmosphere (mostly from higher emissions of NO_X). Table A4 shows the differences between S1 and S2 (S1-S2) of monthly PM averages for January and June (two months with low and high photochemical activities, respectively). It can be seen that the higher formation of secondary PM in the S1 (all-petrol) scenario is more important than the higher tail-pipe emission of primary PM by factor 10 - 20.

When we compare simulated PM differences between S1-S2 for the background air (=upwind) grid cells and for grid cells including Gothenburg (50 km x 50 km scale), we can see significant increase in primary PM in the S1 scenario from the upwind grid cells to Gothenburg (74.6% in winter, 132% in summer) while no systematic increase can be seen for the secondary PM (-4.2% in winter, +2.4% in summer). From this comparison it can be concluded that no systematic increase in secondary PM can be seen (in difference from the primary PM) on the scale of 50-100 km and we can neglect this process on the small scale. However, the overall change in PM concentrations due to the switch of the fuel is very small, both on the large scale and on the small scale as can be seen when the differences in PM concentrations between S1 and S2 are compared to absolute PM concentrations in S1 presented in the last column of Table A4.

Table A4. Differences in primary and secondary PM concentrations between the 'All-petrol' (S1) and 'E-85' (S2) scenarios simulated in gridcells upwind Gothenburg and in gridcells including Gothenburg (2 50 km x 50 km gridcells)

		S1-S2 Background µg/m^3	S1-S2 Gothenburg µg/m^3	Increase S1-S2 Gothenburg µg/m^3	% bkgnd	S1 Background µg/m^3
Prim. PM10	Jan	0.0008	0.0014	0.0006	74.6%	0.84
	Jun	0.0008	0.0018	0.0010	131.9%	0.84
Sec. PM10	Jan	0.0155	0.0148	-0.0006	-4.2%	1.57
	Jun	0.0080	0.0081	0.0002	2.4%	2.13

The sensitivity of the used emission factors for NMHC with respect to the ozone formation in the region of the Swedish West Coast can be analysed with the help of results from studying the

formation of secondary acetaldehyde. In this study first all primary acetaldehyde emitted by the E85 fleet are omitted in a model simulation (S2_nopAA) and secondly all NMHC emissions from the E-85 fleet are switched off (S2_noVOC). Acetaldehyde has a somewhat higher, though still comparable, photochemical ozone creation potential compared to ethanol (ethanol: 44-63, acetaldehyde: 68-80; Altenstedt and Pleijel, 2000) and this simulation will thus also give information on the sensitivity of the calculations to uncertainties in the ethanol emissions. Setting the emission factor for NMHC to zero gives the sensitivity to the emission factor of total NMHC. Figure A2 shows the differences in ozone concentration as a result of acetaldehyde only and the total NMHC from the E85 car fleet (S2 scenario). We can see that the effect on ozone formation is very small for the Swedish West coast. The reason is that the ozone formation in this area is controlled by the availability of NO_X while NMHC, or rather their degradation products the peroxy radicals, that serve as a 'fuel' to ozone formation, are usually in excess here and further VOC emissions do not increase ozone concentrations significantly. The situation looks different in areas that are more heavily polluted with NO_X and/or which is limited in the availability of peroxy radicals. The results from this sensitivity study are fully consistent with the observed increase in ozone concentrations as a result of lower NO emissions (the so-called ozone titration) only very close to the major roads but no larger-scale decrease in ozone concentrations in the Gothenburg area or in any other communities.

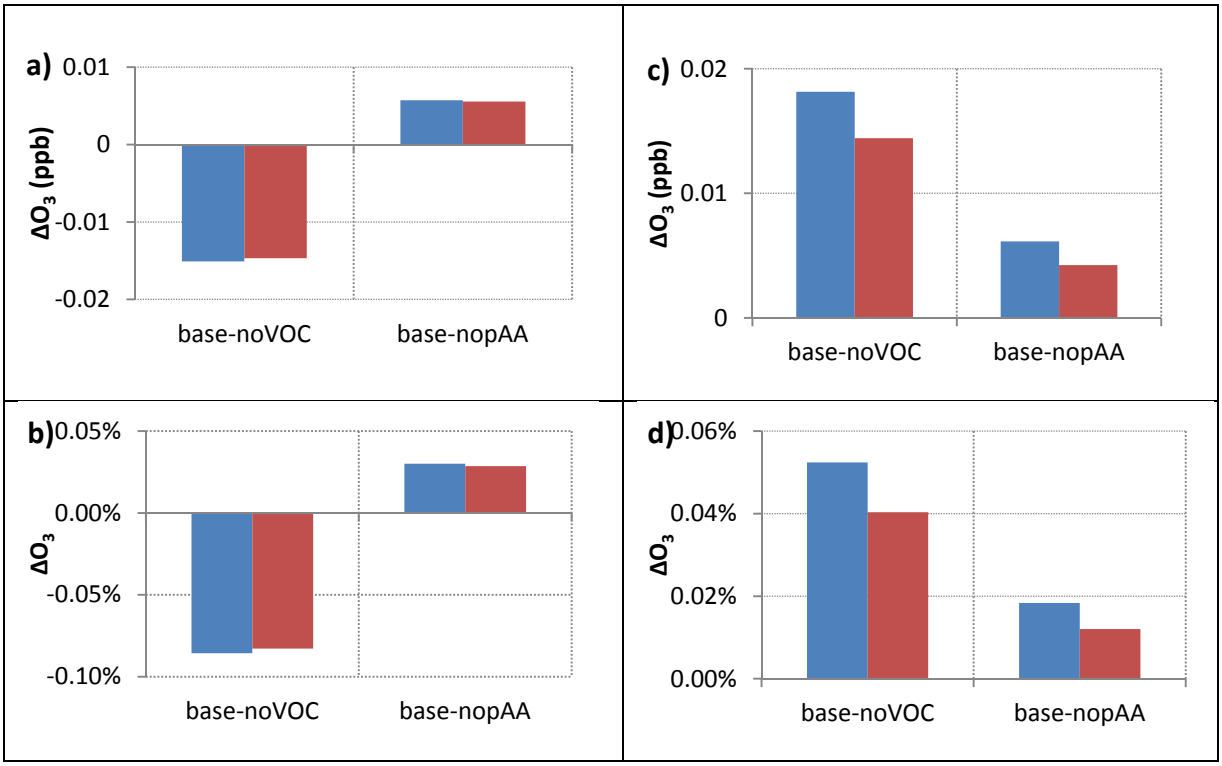

Figure A2. Sensitivity of the ozone formation in the 2 EMEP model grid cells covering Gothenburg (blue) and in the 6 surrounding grid cells (red, see Fig. A1) to changes in NMHC emission factor of the E85 fuelled cars. 'base' is simulation S2 with EF for NMHC as in Table 3, 'noVOC' is S2 with EF for all NMHC = 0, 'nopAA' is S2 with EF for acetaldehyde = 0. a – Mean sensitivity in January, absolute difference in ozone formation in ppb(V) as a result of NMHC (base-noVOC) and acetaldehyde (base-nopAA) emissions, b – Sensitivity relative to ozone concentration in the S2-base simulation, January, c – same as a) but June, d – same as b) but June.

Appendix 3. Emission results

The geographically distributed NO_X and acetaldehyde emissions for the Västra Götaland region are presented in Figures A3 and A4, respectively.

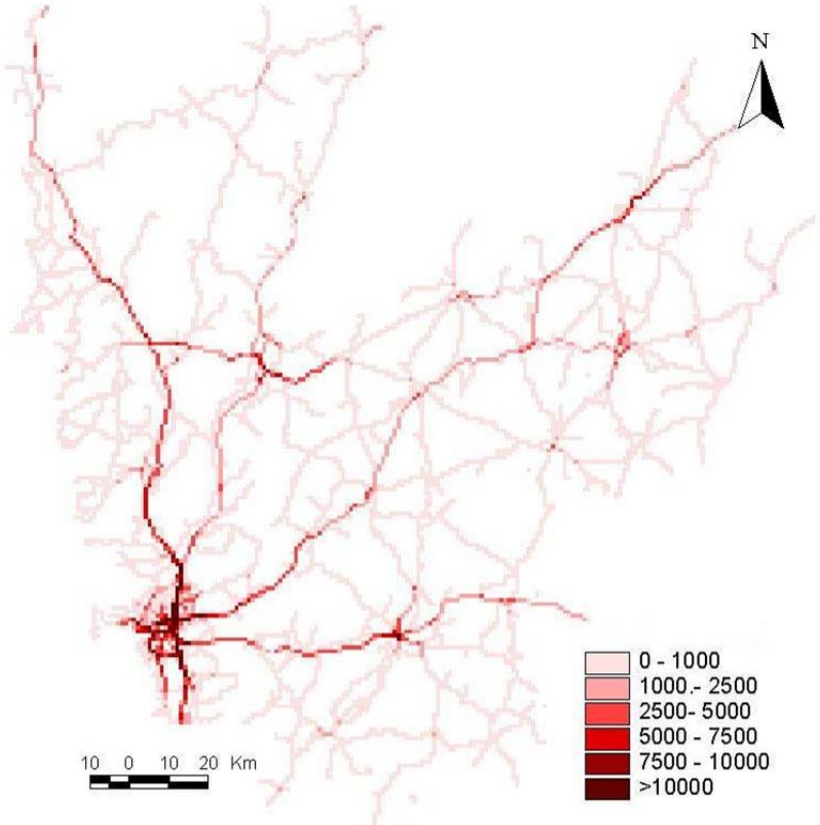

Figure A3. NO_X emissions for the Västra Götaland region for the All Petrol scenario.

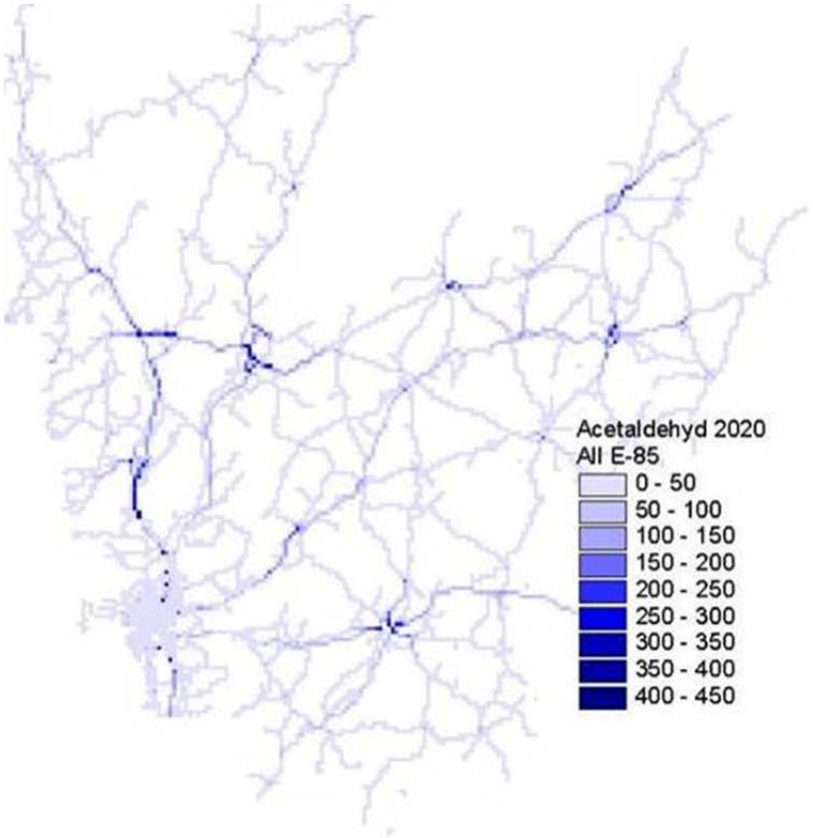

Figure A4. Acetaldehyde emissions for the Västra Götaland region for the All Ethanol scenario.

Appendix 4. Dispersion modelling results

The difference in yearly average concentration between scenario S1 and S2 for acetaldehyde and benzene can be seen in Figure A5 and Figure A6, respectively.

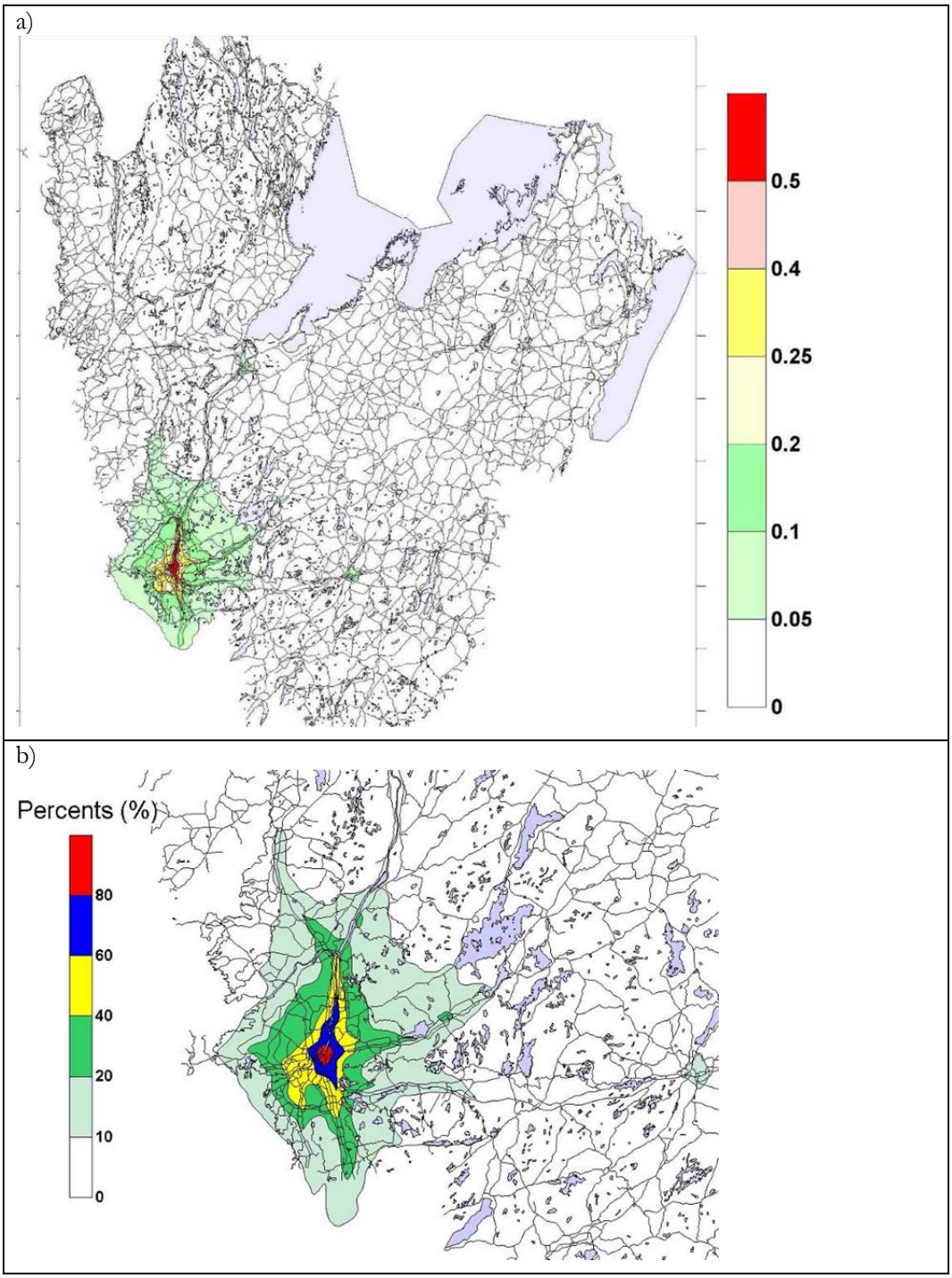

Figure A5. The difference in yearly average concentrations of acetaldehyde for 2020 between the all E85 (S2) and all petrol (S1) scenarios presented as a) µg/m^3 and as b) percentage for the Gothenburg area.

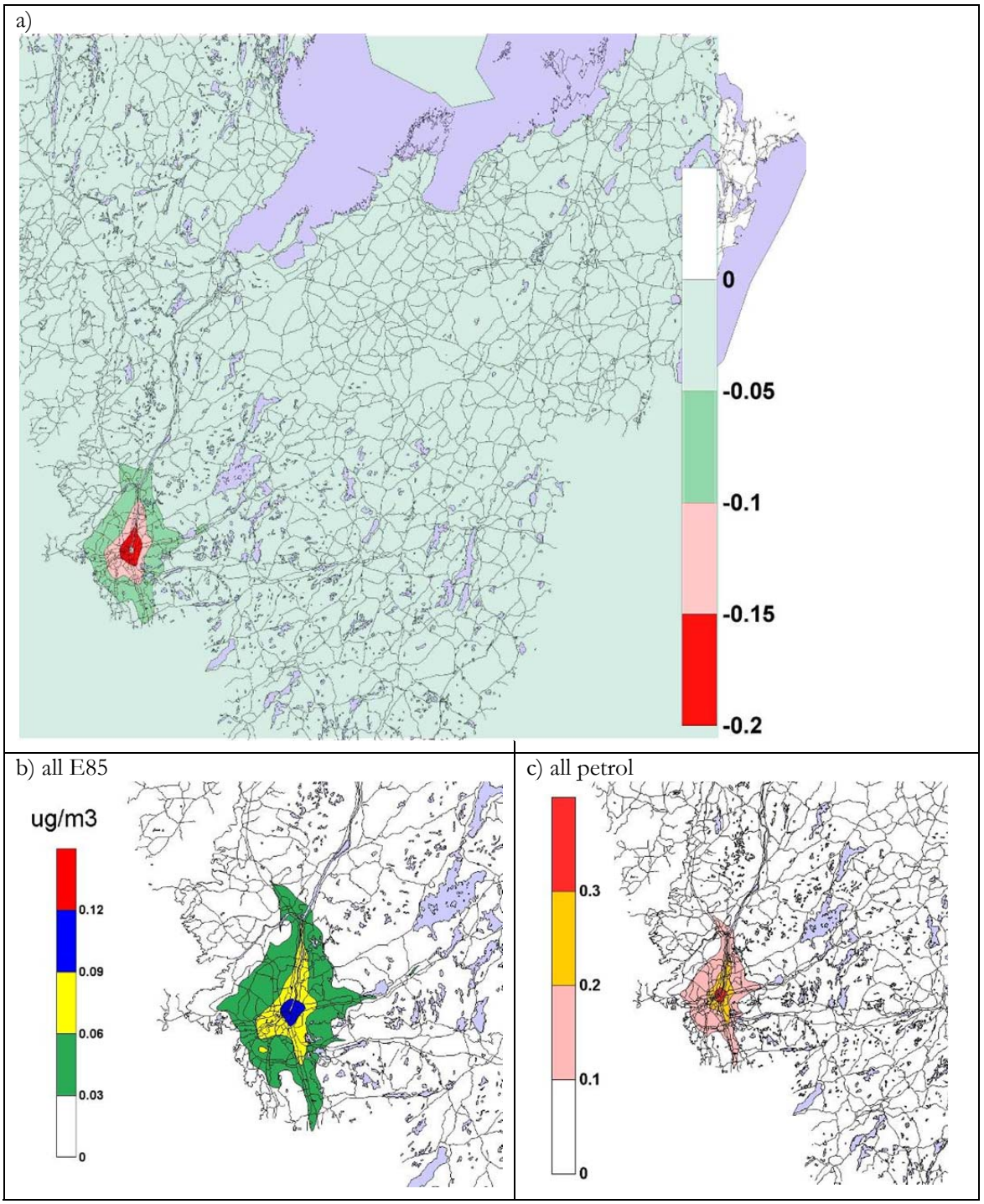

Figure A6. a) the difference in yearly average concentrations (µg/m³) of benzene for 2020 between the all E85 (S2) and all petrol (S1) scenarios, yearly means for b) all E85 and c) all petrol (note the different scales)

The difference in the calculated NO_X, NO_2 and ozone concentrations between the two scenarios are shown in Figures A7, A8 and A9.

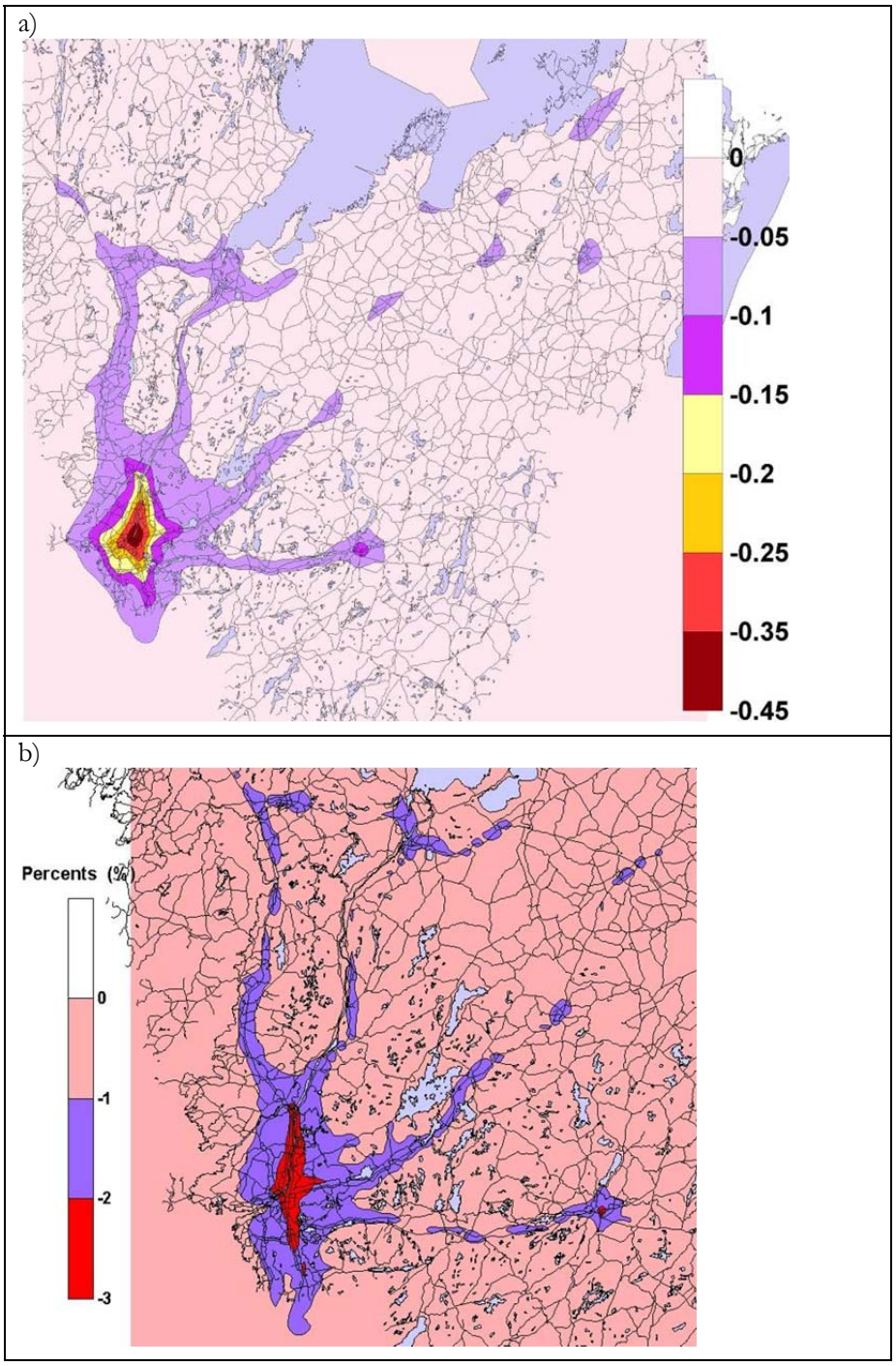

Figure A7. The difference in yearly average concentrations of NO_X for 2020 between the all E85 (S2) and all petrol (S1) scenarios presented as a) µg/m³ and as b) percentage for the Gothenburg area.

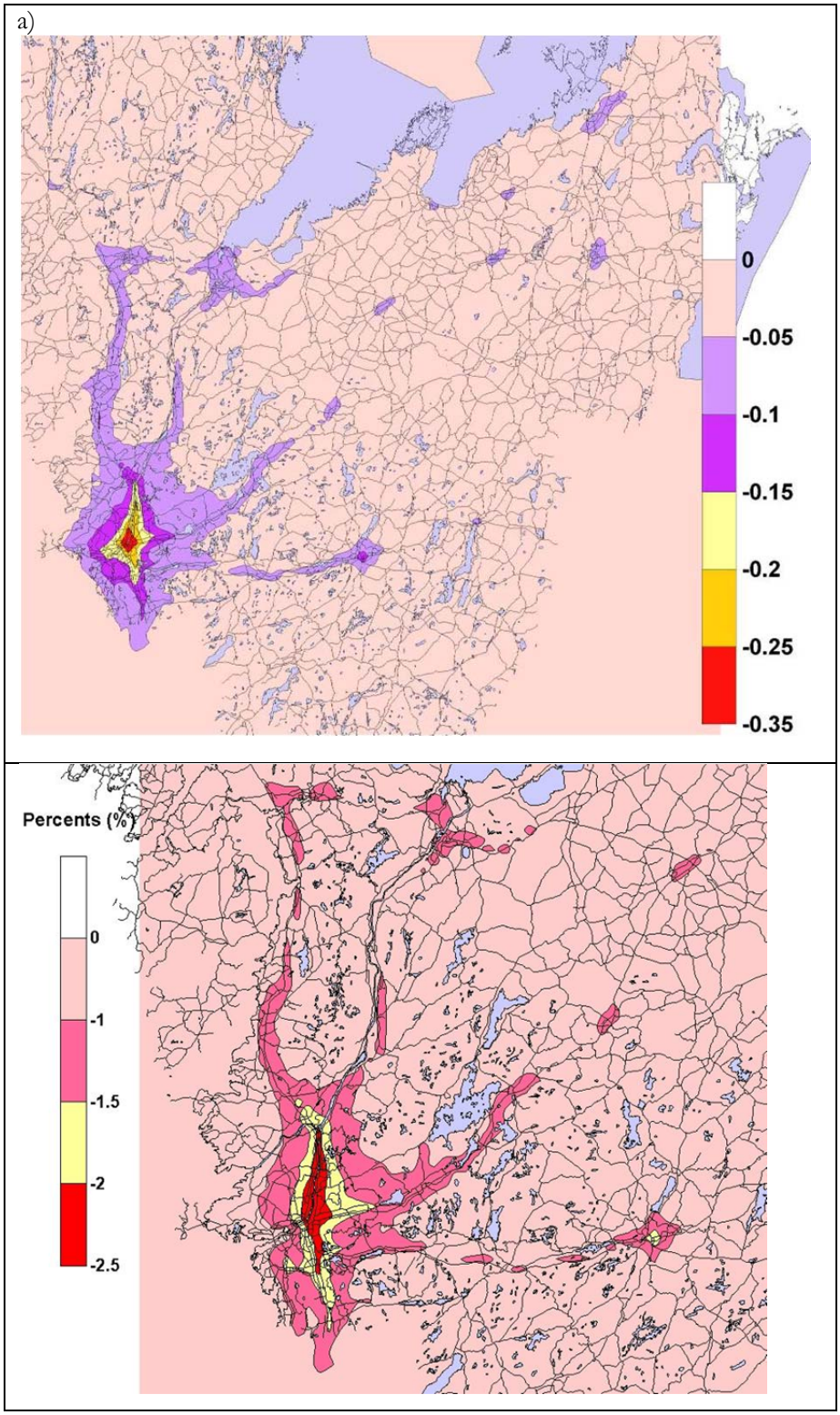

Figure A8. The difference in yearly average concentrations of NO_2 for 2020 between the all E85 (S2) and all petrol (S1) scenarios presented as a) µg/m^3 and as b) percentage for the Gothenburg area.

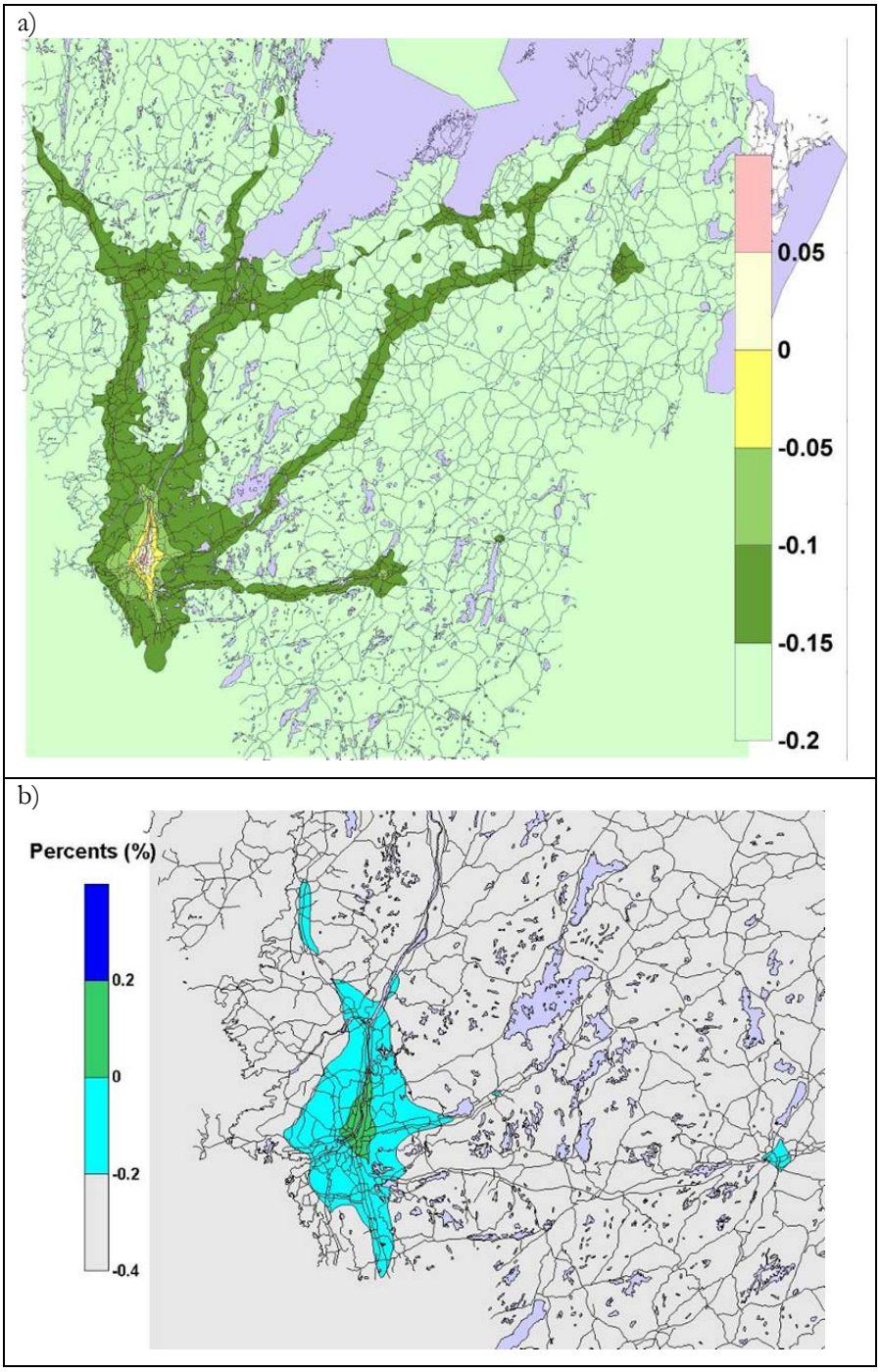

Figure A9. The difference in yearly average concentrations of ozone for 2020 between the all E85 (S2) and all petrol (S1) scenarios presented as a) µg/m^3 and as b) percentage for mainly the Gothenburg area.

Appendix 5. Detailed Exposure data

Detailed exposure data for the difference between the all E85 (S2) and the all petrol (S1) scenarios can be found in Table A5.

Table A5. Exposure data for the difference between the all E85 (S2) and the all petrol (S1) scenarios.

Acetaldehyde diff E-P (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0 - 0.1		627198	58.2%
0.1 - 0.2		163394	15.2%
0.2 - 0.3		134879	12.5%
0.3 - 0.4		88099	8.2%
0.4 - 0.5		38686	3.6%
0.5 - 0.6		16880	1.6%
0.6 - 0.7		7983	0.7%
0.7 - 0.8		1248	0.1%
Tot	***0.14***	***1078367***	

Benzene diff E-P (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
-0.3 - -0.2		26680	2.5%
-0.2 - -0.1		205757	19.1%
-0.1 - -0		845930	78.4%
Tot	***-0.074***	***1078367***	

NO_2 diff E-P (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
-0.4 - -0.3		7698	0.7%
-0.3 - -0.2		50131	4.6%
-0.2 - -0.1		296288	27.5%
-0.1 - -0		724250	67.2%
Tot	-0.09	1078367	

NO_X diff E-P (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
-0.6 - -0.5		7698	
-0.5 - -0.4		19639	1.8%
-0.4 - -0.3		36626	3.4%
-0.3 - -0.2		161685	15.0%
-0.2 - -0.1		186500	17.3%
-0.1 - -0		666219	61.8%
Tot	-0.12	1078367	

O_3 diff E-P (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
-0.2 - -0.1		779132	72.3%
-0.1 - -0		266417	24.7%
0 - 0.1		32818	3.0%
Tot	-0.12	1078367	

Exposure data for the total levels in the two scenarios can be found in Table A6.

Table A6. Exposure data for the total levels in the two scenarios.

Acetaldehyde tot All ethanol (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0.6 - 0.7		568825	52.7%
0.7 - 0.8		171845	15.9%
0.8 - 0.9		124551	11.5%
0.9 - 1.0		100946	9.4%
1.0 - 1.1		59235	5.5%
1.1 - 1.2		30559	2.8%
1.2 - 1.3		13175	1.2%
1.3 - 1.4		7983	0.7%
1.4 - 1.5		1248	0.1%
Tot	***0.77***	***1078367***	

Acetaldehyde tot All petrol (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0.5 - 0.6		45111	4.2%
0.6 - 0.7		1032008	95.7%
0.7 - 0.8		1248	0.1%
Tot	***0.65***	***1078367***	

Benzene tot All ethanol (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0 - 0.1		1055961	98%
0.1 - 0.2		22406	2%
Tot	**0.05**	**1078367**	

Benzene tot All petrol (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0 - 0.1		729093	67.6%
0.1 - 0.2		232961	21.6%
0.2 - 0.3		94042	8.7%
0.3 - 0.4		21023	1.9%
0.4 - 0.5		1248	0.1%
Tot	***0.10***	***1078367***	

NO_x tot All ethanol (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0-10		779108	72.2%
10-20		254658	23.6%
20-30		28683	2.7%
30-40		11070	1.0%
40-50		3426	0.3%
50-60		0	0.0%
60-70		0	0.0%
70-80		639	0.1%
80-90		0	0.0%
90-100		0	0.0%
100-110		783	0.1%
Tot	***8.4***	***1078367***	

NO_x tot All petrol (µg/m³)	**Populated weighted mean**	**Number of people**	**Percentage of population**
0-10		769557	71.4%
10-20		264209	24.5%
20-30		28683	2.7%
30-40		11070	1.0%
40-50		3426	0.3%
50-60		0	0.0%

60-70		0	0.0%
70-80		639	0.1%
80-90		0	0.0%
90-100		0	0.0%
100-110		783	0.1%
Tot	***8.5***	***1078367***	

O_3 tot All ethanol ($\mu g/m^3$)	**Populated weighted mean**	**Number of people**	**Percentage of population**
40-45		1422	0.1%
45-50		26487	2.5%
50-55		176636	16.4%
55-60		770370	71.4%
60-65		103452	9.6%
Tot	***56.9***	***1078367***	

O_3 tot All petrol ($\mu g/m^3$)	**Populated weighted mean**	**Number of people**	**Percentage of population**
40-45		1422	0.1%
45-50		20229	1.9%
50-55		181876	16.9%
55-60		763361	70.8%
60-65		111479	10.3%
Tot	***57.0***	***1078367***	

$PM_{2.5}$ tot All ethanol ($\mu g/m^3$)	**Populated weighted mean**	**Number of people**	**Percentage of population**
1-2		0	0.0%
2-3		885325	82.1%
3-4		192259	17.8%
4-5		783	0.1%
Tot	***2.68***	***1078367***	

$PM_{2.5}$ tot All petrol ($\mu g/m^3$)	**Populated weighted mean**	**Number of people**	**Percentage of population**
1-2		0	0.0%
2-3		868773	80.6%
3-4		208811	19.4%
4-5		783	0.1%
Tot	***2.70***	***1078367***	