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Impact of bioethanol fuel implementation in transport based on modelled acetaldehyde concentration in the urban environment

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HIGHLIGHTS

- Acetaldehyde emissions increase about 233% for the E85-fleet scenario.
- Acetaldehyde levels increase up to 650% in the urban area for the E85-fleet scenario.
- Full implementation of bioethanol as fuel for transport impacts urban air quality.

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ABSTRACT

This study shows the results obtained from emission and air dispersion modelling of acetaldehyde in the city of Oslo and associated with the circulation of bioethanol vehicles. Two scenarios of bioethanol implementation, both realistic and hypothetical, have been considered under winter conditions; 1) realistic baseline scenario, which corresponds to the current situation in Oslo where one bus line is running with bioethanol (E95; 95% ethanol–5% petrol) among petrol and diesel vehicles; and 2) a hypothetical scenario characterized by a full implementation of high-blend bioethanol (i.e. E85) as fuel for transportation, and thus an entire bioethanol fleet. The results indicate that a full implementation of bioethanol will have a certain impact on urban air quality due to direct emissions of acetaldehyde. Acetaldehyde emissions are estimated to increase by 233% and concentration levels increase up to 650% with regard to the baseline.

1. Introduction

Climate change is one of the largest challenges at present and will be in the future. Reducing CO_2 and other greenhouse gas (GHG) emissions from transport is crucial and the use of biofuels is an alternative to emission reductions. The European Renewable Energy Directive aims to achieve 20% renewable energy by 2020 and a minimum target of 10% of the energy to be used for transport. Further on, the Fuel Quality Directive (FQD) evokes the life cycle reduction of GHG emissions for all fuels with target of 6% cut. This could be done in several ways, but the use of more biofuels is pervasive. The sustainability criteria in the FQD will encourage in addition that these biofuels are produced in sustainable manners and once this criteria is met, the target value will likely be increased. Most EU member states already have some percentage of biodiesel and/or bioethanol in diesel or petrol fuels. In Norway for instance standard 95 octane petrol contains up to 7% bioethanol and for diesel the share is up to 10% biodiesel blend.

The use of bioethanol as fuel for transportation became popular around the 1980s in many countries such as Brazil, United States or Sweden. Its production has increased worldwide in the last decade from 17.3 billion litres in 2000 to over 46 billion litres in 2007, and a production over 125 billion litres is estimated for 2020 (Balat and Balat, 2009). Bioethanol can be a first or second generation product depending on the feedstock used, which ranges from the fermentation of raw materials such as starch-containing (e.g. corn, wheat), sugar-containing (e.g. beet and cane) to cellulose-containing (e.g. wood) plants. When used as fuel for transportation, bioethanol is commonly blended with petrol in different proportions, from 90% petrol and 10% ethanol, referred to as E10, to 15% petrol and 85% ethanol (i.e. E85), in which case modifications need to be applied to the vehicle (i.e. Flexi-Fuel Vehicles; FFV).

The advantages and disadvantages associated with the production and combustion of bioethanol have been addressed by several studies, and many uncertainties are still not solved. Among the advantages, reduction of GHG emissions and sustainability (Scacchi et al., 2010), and enhancement of the agriculture sector in specific areas have been highlighted. However, disadvantages have also been pointed out, such as the controversy associated with the use of food-based bioethanol,

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or the relatively high emissions of hazardous air pollutants (HAPs; Gaffney and Marley, 2009; Anderson, 2009). One of the main issues associated with alcohol fuels is that they oxidise to aldehydes, hazardous pollutants and precursors of strong oxidants such as ozone and peroxyacetyl nitrate (PAN). Thereby the ethanol molecule (C_2H_5OH) oxidises to acetaldehyde (CH_3CHO) while methanol will oxidise to formaldehyde. Thus, bioethanol fuelled vehicles may contribute to urban pollution through emissions of unburned ethanol, acetaldehyde and acetic acid (CH_3COOH) as an oxidation product of the former one. A previous study carried out in Oslo showed that the use of high bioethanol blend (E95) as fuel for buses can have an adverse impact on urban air quality (López-Aparicio and Hak, 2013). Acetaldehyde and acetic acid were identified as of concern based on ambient and on-line measurements during driving conditions of an E95 bus. Acetaldehyde was 1) measured at very high levels in the exhaust of an E95 bus (>150 ppm), 2) measured at higher ambient concentrations at locations exposed to the E95 buses than those not exposed, and 3) estimated to be above the threshold limit value at a close distance to the bus. Although these results contributed to the understanding of emissions from bioethanol combustion, additional research is still needed to establish the potential impact at urban scale, especially by means of modelling techniques.

To our knowledge very few studies modelled air pollutant concentration associated with emissions from alternative fuels such as bioethanol. Jacobson (2007), for instance, modelled ozone exposure associated with a change scenario from petrol to E85 in the United States in 2020, and concluded that ozone-related mortality would increase by 9% in Los Angeles and by 4% in the Unites States. Likewise, Cook et al. (2011) concluded that ozone would increase over much of the United States because of ethanol use, and increase dramatically under winter conditions. Fridell et al. (2010) estimated emissions considering two fuel scenarios, petrol vehicles versus E85 vehicles, in a Swedish county (i.e. Västra Götaland) and concluded that the health risk decreases in the E85 scenario compared to petrol due to decreasing NOx exposure, although emissions of acetaldehyde were estimated to increase by 770%. Ginnebaugh and Jacobson (2012) studied the impact associated with a substitution of petrol vehicles by E85 on ozone production under winter and summer conditions. They emphasized the pollution enhancement associated with E85 at low temperature.

Most of the modelling studies have been performed at regional scale and focussed on the end product of bioethanol emissions as increased ozone concentration, and possible health risk consequences therein. However, emissions and concentration of acetaldehyde need to be evaluated as it is suspected carcinogenic and respiratory toxic in addition to contributing to the formation of ozone and PAN. Acetaldehyde is relatively stable under winter conditions and cold climate; the tropospheric lifetime of acetaldehyde has been reported to be of about 11 h, 17 days and 5 days in the presence of OH radicals, NO_3 radicals and sunlight, respectively (Seinfeld and Pandis, 2000). In US cities for instance the residence time of acetaldehyde is reported to be on the order of hundreds to thousand hours in winter and tens of hours in summer (Ligocki and Whitten, 1991). Similarly, acetaldehyde lifetimes of about 15 days in the winter months and less than half a day in the summer were considered by Fridell et al. (2010) based on the reaction with OH radicals using OH mean concentrations from EMEP simulations (The European Monitoring and Evaluation Programme).

Our study evaluates the results obtained by means of air dispersion modelling at urban scale (i.e. Oslo), which has been applied to estimate acetaldehyde emissions and concentration in the urban area and associated with combustion from bioethanol vehicles. Two different scenarios have been considered under winter conditions; 1) a baseline case which represents the real fleet situation (i.e. petrol and diesel vehicles, and one bus line which runs with E95); and 2) scenario characterized by a full implementation of high blend bioethanol as fuel for transportation (i.e. light and heavy duty vehicles running on E85; 85% bioethanol and 15% petrol), and from now on named E85-fleet scenario in this study.

2. Methodology

2.1. Scenario description

The air dispersion modelling has been carried out for the two scenarios (i.e. baseline and E85-fleet scenarios) under winter conditions, and covering Oslo city and part of the municipality of Bærum. The baseline represents the current situation with updated information on vehicle fleet and traffic numbers. Regarding the light duty vehicles (LDVs), about 60% are running on diesel and the rest on petrol. Concerning bus transport, one bus line runs with high-blend bioethanol (i.e. E95) and over 70% of the buses in Oslo are running on standard or B7 diesel (i.e. up to 7% biodiesel) all year around. During summer season, 22% of the buses run on B30 or B100, whereas they run on B7 during winter season. Hence, over 92% of the buses are running on B7 during winter conditions. In the baseline scenario we have therefore assumed all buses, except the bioethanol (E95) bus line, to run on normal diesel fuel just as other heavy duty vehicles (HDVs) do. The E85-fleet scenario involves that all vehicles, both LDV and HDV, run on a high blend bioethanol fuel (i.e. E85).

Winter season has been selected as it represents the time period when pollutant concentrations are highest in Oslo due to unfavourable dispersion conditions, especially during cold periods with thermal inversion and stable atmosphere. Moreover, significantly higher emissions of acetaldehyde and other compounds have been reported for E85 than for petrol at low temperature conditions (Whitney and Fernandez, 2007; Westerholm et al., 2008; Ginnebaugh et al., 2010), thus the largest possible effect on urban air quality is expected to be found in winter. Additional emission and dispersion calculations have been performed using the meteorological conditions corresponding to May 2010, for 1) comparison with indicative measurements published elsewhere (López-Aparicio and Hak, 2013) and 2) discussion purposes.

2.2. Dispersion and emission modelling

Acetaldehyde has been assumed to behave as an inert component inside our model domain based on its lifetime at winter temperature conditions, and therefore implications for ozone and/or PAN formation are not considered in this study.

An air quality integrated modelling system (AirQUIS; Oftedal et al., 2004) developed by NILU — Norwegian Institute for Air Research, was selected for the modelling activities. It uses meteorological field determined using observed meteorology combined with the mass conserving wind field interpolation model MC-WIND. In our study the meteorology input data is from the observation site Valle Hovin for the year 2009. The dispersion model used is EPISODE on a 1×1 km^2 grid resolution, and calculates emissions, meteorology and concentrations in this grid region (Slørdal et al., 2013). The model domain spans 28 km in the East–West direction and 22 km in the North–South direction, covering most of Oslo municipality, as well as part of the neighbouring municipality (i.e. Bærum; Fig. 1). The dispersion model EPISODE has, in addition, a sub-grid model which treats roads as line sources giving an extra contribution along the road network and capturing the high concentration gradient near the source. The EPISODE dispersion model has been used for a large number of applications, including forecasting systems for five city regions in Norway (e.g. Ødegaard et al., 2013; Bedre byluft, 2014).

Traffic emissions are generated using traffic information on all roads in Oslo from the Norwegian road administration's database (NVDB), which contains information on the annual average daily traffic (ADT), the speed limit, the heavy duty fraction of the traffic and information about the roads (e.g. slope and location). The ADT is used, in combination with a daily cycle of traffic, to generate hourly traffic numbers for all roads. For verifying the ADT, specific traffic-counting was performed at the road side in this study, and led to minor changes for several road links. The main discrepancies were related to the city streets for the total ADT and the HDV fraction. The traffic-counting indicated that the

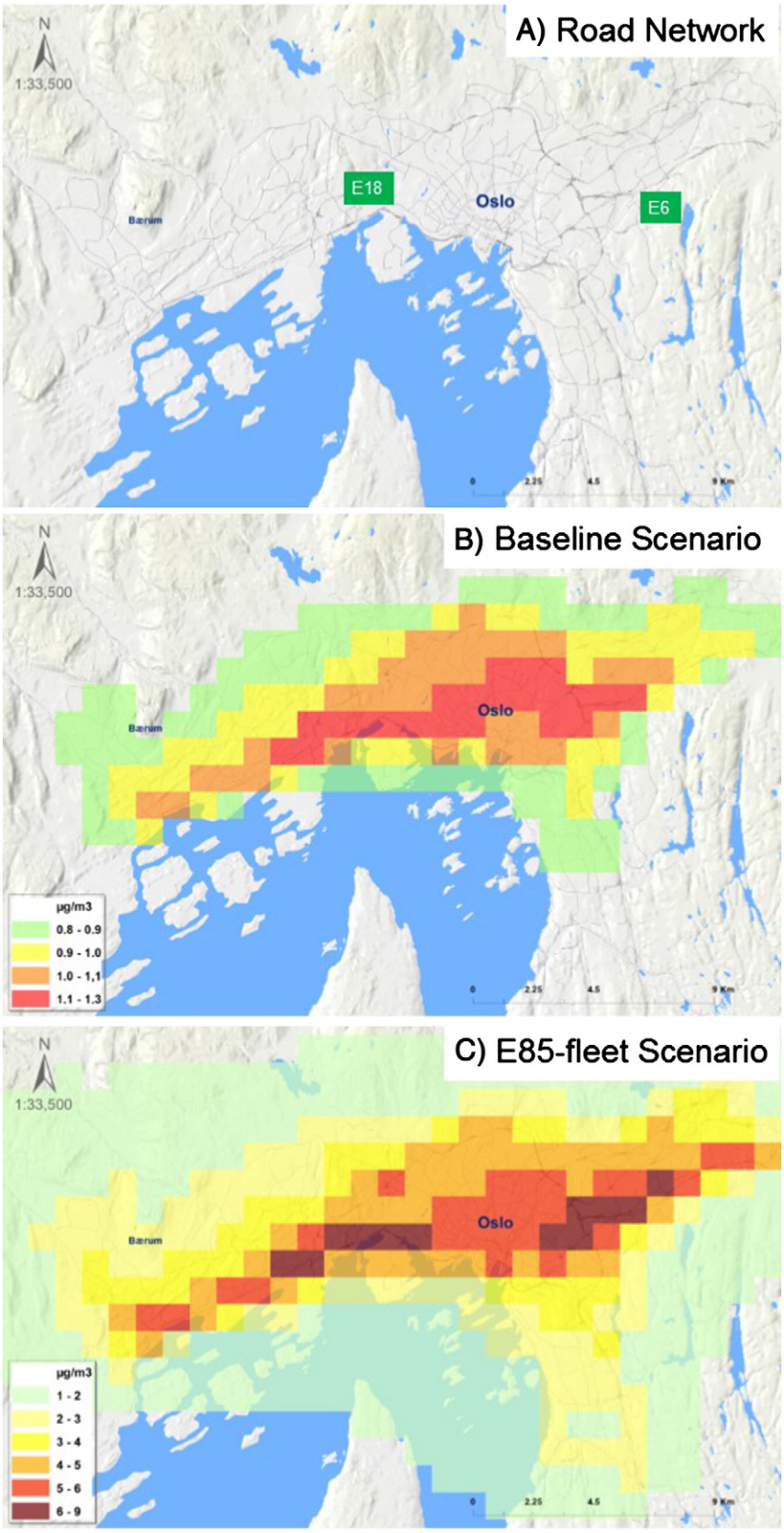

Fig. 1. Model domain and modelled average acetaldehyde concentration fields under winter conditions (October 2009 to March 2010) in the area of Oslo, where A) Road Network, (B) Baseline Scenario and C) E85-fleet scenario are shown.

available traffic input data were relatively good for the modelling purpose. Usually, detailed information on speed and slope as well as vehicle fleet, such as fraction in each emission class (i.e. EURO classification) and fuel type, are used together with EFs and the hourly fractions of ADT to estimate exhaust emissions on each road segment. However, for acetaldehyde, the existing limitations concerning available information on the EFs has led us to only use the ADT and the heavy duty fraction and to consider the division in 2 main vehicle groups: 1) Passenger cars and light duty vehicles and 2) heavy duty vehicles and buses. The existing E95 bus line has been treated separately.

The main known source of acetaldehyde in Oslo is traffic, however emissions from wood burning has also been included in the modelling. The wood burning EF for acetaldehyde was assumed to be 0.34 g kg^{-1} wood which is about 1/3 of the EF used for NOx (McCrillis, 2000; McDonald et al., 2000). Wood consumption data is available, spatially distributed on small administrative areas, and with a time variation of week, weekday and hour. For Oslo these data are also divided into consumption according to different technologies such as newer clean-burning stoves, older stoves and open fireplaces. The consumption data are based on questionnaires and studies performed by Statistics Norway (SSB) and are used with the EF to get hourly wood burning emissions as gridded data.

For background concentrations, the annual acetaldehyde average concentration (0.6 μg m^{-3}) from a regional background station situated in the south of Norway (i.e. Birkenes) is used. This is likely to be an underestimation of the background concentration for the model domain, as activities in the larger Oslo region will likely contribute to ambient acetaldehyde concentrations entering at the border of the modelled area.

2.3. Emission factors for vehicles

To our knowledge, relatively few emission studies exist in the scientific literature looking into bioethanol emissions, and those existing only include a few vehicle types, leading to high uncertainties and, for some components (e.g. NOx), even inconsistencies (Kousoulidou et al., 2008). Studies addressing emissions of acetaldehyde associated with bioethanol combustion are even fewer. However, enough studies have been performed to show that acetaldehyde emissions statistically increase when introducing ethanol in petrol, and values up to 500% increase for E85 have been reported (Reading et al., 2002; Graham et al., 2008).

Different acetaldehyde EFs for winter conditions have been selected in our study for LDV and HDV running on petrol, diesel and E85-bioethanol (Table 1). Since Oslo has quite cold winter conditions, the EF for petrol and diesel vehicles in the baseline scenario is assumed to be 10 times higher than that for summer conditions, including also urban and more congested driving. Based on the evaluation of available literature we decided to use the same acetaldehyde EF for diesel and petrol LDVs and passenger cars to be around 0.005 g km^{-1} (Table 1), independent of age, mileage, or Euro classification. This EF is twice as high than the one used by Fridell et al. (2010) for petrol vehicles (i.e. 0.0025 g km^{-1}) and year round, independent of summer or winter conditions, and it is three times lower than the acetaldehyde EF for diesel vehicles reported by Kristensson et al. (2004), which is of about 0.015 g km^{-1}. Even though higher acetaldehyde EFs exist in the literature for petrol and diesel vehicles, we consider that our baseline scenario may be slightly overestimated. On the other hand, the E85-fleet scenario has been designed following a conservative approach and therefore underestimated results are expected.

Table 1
Acetaldehyde emission factors (EFs) selected for the modelling activities. E95 bus* represents the range of emission factors estimated from the measurements carried out under driving conditions.

Vehicle	Fuel	Acetaldehyde EF (g km^{-1})
Light duty vehicle (LDV)	Petrol	0.005
	Diesel	0.005
	E85	0.03
Heavy duty vehicle (HDV)	Diesel	0.03
	E95 bus*	0.9–6.5
	E85	0.8

For the E85-fleet scenario, characterized for a complete vehicle fleet running on E85-bioethanol blend, EFs for LDVs running on E85 were considered based on factors published in the literature (e.g. 0.008 g km^{-1} by Karman, 2003; 0.03 g km^{-1} by Fridell et al., 2010) and the ratio found between emissions from E0 and E85 fuels. Several studies reported acetaldehyde emissions for E85 more than 40 times higher than for E0 (Karman, 2003; Graham et al., 2008; Fridell et al., 2010). Based on this ratio, and taking into account that the EF for petrol vehicles was selected to be around 0.005 g km^{-1}, the result would be an acetaldehyde EF of about 0.2 g km^{-1}. As indicated in Table 1, an acetaldehyde EF of about 0.03 g/km was selected for LDVs running on E85, in accordance with the acetaldehyde EF used in other modelling studies (e.g. Fridell et al., 2010).

For HDVs, even scarcer data are to be found, but they show naturally larger emissions than for light vehicles. For instance Ban-Weiss et al. (2008) reported about 6 times larger emissions for HDV compared to LDV. Hence, in the lack of more information on acetaldehyde emissions from HDV, we have used the relationship established by Ban-Weiss et al (2008) to obtain EFs for all heavy duty vehicles including buses running on standard diesel. The acetaldehyde EF for HDV running on diesels was then assumed to be 0.03 g km^{-1} (Table 1) based on a factor of 6 over the EF selected for LDV (i.e. 0.005 g km^{-1}).

For the high blend bioethanol fuelled buses there are no published measurements for acetaldehyde emissions. Taking into account the E85 to E0 emission ratio for LDV from Ban-Weiss et al. (2008) and Karman (2003), and the factor of 6 for HDV emissions regarding those from LDV, EFs for HDV tend to vary between 0.08 and 0.13 g km^{-1}. Additionally, acetaldehyde EF for the E95 bioethanol bus and relative to CO_2 was estimated based on the online monitoring carried out with a proton-transfer-reaction time-of-flight (PTR-TOF) mass spectrometer during driving conditions and published elsewhere (López-Aparicio and Hak, 2013). Time-integrated emissions of acetaldehyde and CO_2 (ppm $*$ min) were derived for each 5 minute period of the total driving (i.e. 1 h and 20 min) and the acetaldehyde to CO_2 emission ratio was determined (i.e. $\Delta VOC/\Delta CO_2$, g/g). EFs for acetaldehyde were estimated, based on a published CO_2 EF (i.e. 1 090 g km^{-1}; from TRANSPHORM project, 2014) and the derived emission ratio ($\Delta VOC/\Delta CO_2$), to range between 0.9 and 6.5 g/km, depending on driving conditions (e.g. free flow, idling conditions). An acetaldehyde EF of about 0.8 g km^{-1} has been considered in our study (Table 1) for the dispersion modelling of acetaldehyde emitted from E95 bioethanol buses, and therefore for all HDV running on E85 in the E85-fleet scenario. This value is in the lower range of the EFs estimated from the measurements. Contrary to the baseline scenario and the EF for petrol and diesel fuels, we do not increase the emissions and thus the EF due to cold outdoor temperatures for high bioethanol fuel, neither for light or heavy vehicles. This is done mainly because of the lack of studies looking into this aspect as well as the mentioned conservative approach.

3. Results and discussion

3.1. Emissions

The modelling results are highly dependent on the input data and especially on the emission factors. To compare the scenarios defined in our study and to discuss uncertainties we have estimated total acetaldehyde emissions from all sources.

Emissions of acetaldehyde in the baseline scenario were estimated and the contributions of different sources established; LDV running on both petrol and diesel was found to be the main contributor with 42% to total emissions of acetaldehyde in Oslo area (Fig. 2), closely followed

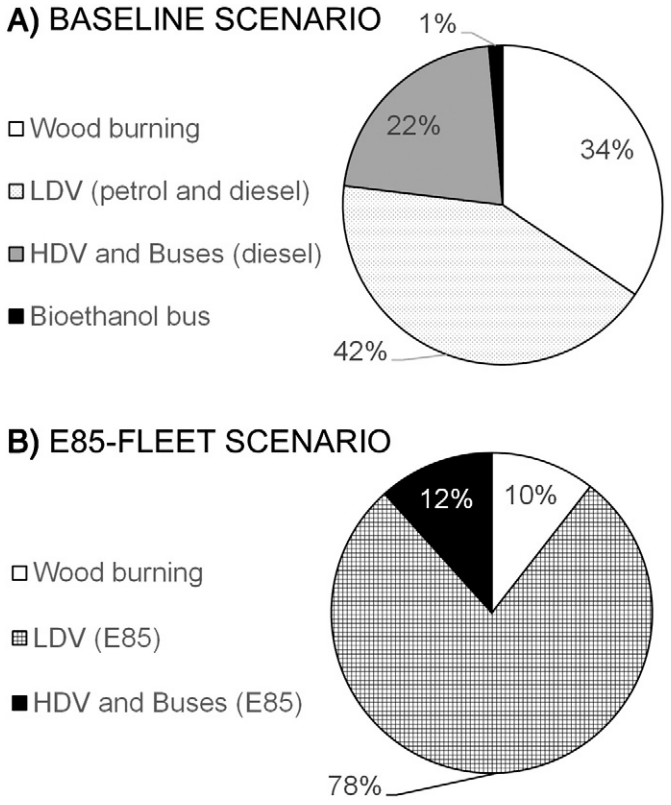

Fig. 2. Contribution of acetaldehyde emission sources to total emissions for the baseline (A) and E85-fleet scenarios (B). LDV: light duty vehicle; HDV: heavy duty vehicle.

by wood burning with 34% of total emissions (Fig. 2) and HDVs running on diesel (22%). The E95 bioethanol bus line barely contributes 1% to total acetaldehyde emissions, which is understandable as the E95 bus line in Oslo consists of merely 21 buses. With a full implementation of a high-ethanol blend, LDVs are also the main contributor to total emissions of acetaldehyde, although this time the LDV contribution reaches about 78% of total emissions. HDVs running with bioethanol and wood burning contribute similarly 12 and 10%, respectively, to total emissions of acetaldehyde. The emission calculation shows an increase of about 233% for acetaldehyde for the E85-fleet scenario (i.e. 60 tons in 6 months) relative to the baseline (i.e. 18 tons in 6 months) representing the current situation. The acetaldehyde emission increase is lower than that reported by Fridell et al. (2010) who established an increase of 770% for acetaldehyde from a petrol scenario to an E85 scenario. The result from our study is obtained by means of a conservative approach, which is defined by an EF for acetaldehyde for HDV of about 0.8 g km^{-1}. The EF estimated from the measurements carried out from an E95 bus during driving conditions ranged from 0.9 and 6.9 g km^{-1}. Taking into account 6.9 g km^{-1} as EF for acetaldehyde emissions for HDV and thus defining an extreme emission scenario, total acetaldehyde emissions reach 108 tons in 6 months, involving an increase of about 500% regarding the baseline scenario. This result is closer to that reported by Fridell et al. (2010) and supports a previous study where 500% increase of acetaldehyde emissions was reported for fuel blends over 70% ethanol (Reading et al., 2002).

3.2. Acetaldehyde concentrations

Results from the dispersion calculations for the baseline and E85-fleet scenarios are shown in Fig. 1, along with the model domain with the road network. Acetaldehyde concentration levels for the baseline scenario, representing the current situation in Oslo, are generally low, and close to the background level of about 0.6 μg m^{-3} in a large part of the model domain. Areas in the city centre, close to the E95 bus line (i.e. at that time line 21) and roads with large traffic density (i.e. E6 and E18; Fig. 1), show higher concentrations and the highest grid value is about 1.2 μg m^{-3} for 6 month average and 10.4 μg m^{-3} as the maximum acetaldehyde hourly value (Table 2). Using gridded population information according to home addresses, the population weighted average concentration is about 0.95 μg m^{-3}.

At the roadsides, concentrations are higher due to closeness to the source. For instance for the baseline scenario, hourly modelled acetaldehyde concentration at roadside for a receptor point at a street (i.e. Sannergata) located along the E95 bus line is shown in Fig. 3. The modelled concentration has a period average of about 1.3 μg m^{-3} and a maximum hourly value of about 8.4 μg m^{-3}, and grid values covering the same location reach 1.16 μg m^{-3} and 8.23 μg m^{-3}, respectively (Table 2). The road side increment, estimated as grid average versus road side, for Sannergata is hence 12% for the average and 2% for the maximum hourly value. Sannergata is situated in the inner city and with a registered ADT of 10 000 vehicles. For other locations along side roads in less polluted areas, this increment is relatively larger.

A full implementation of bioethanol as fuel (E85-fleet scenario) involves higher modelled acetaldehyde concentration values than for the baseline, reaching a maximum grid value of about 8.04 μg m^{-3} as winter average (Fig. 1 and Table 2). The population weighted gridded average concentration is 3.80 μg m^{-3}, which means a 300% increase regarding the baseline. This may be of certain concern for human health as at average ambient concentration of 5 μg m^{-3}, the increased chance of developing cancer due to exposure to acetaldehyde is 1 in 100 000 (US EPA, 1999). Based on our results, 28% of the urban population in Oslo would be living in an area with winter average concentration above 5 μg m^{-3} under full implementation of bioethanol as fuel for transport.

In the E85-fleet scenario, the period average and maximum hourly acetaldehyde levels for the roadside location Sannergata are of about 5.97 μg m^{-3} and 61.67 μg m^{-3}, respectively (Table 2), demonstrating a 350% and 650% increase regarding the baseline scenario. The concentration increases for a full implementation of a high-bioethanol blend are exceptionally high compared with other studies. Fridell et al. (2010) obtained an increase of acetaldehyde levels up to 80% in the central part of the urban area in Gothenburg, 10% for other urban areas and less than 10% for rural areas. These ambient concentration increases seem very low considering the acetaldehyde emissions increase of about 770% reported by the same authors (Fridell et al., 2010). In our total model domain and under E85-fleet conditions, the highest grid value is found to be of about 8.04 μg m^{-3} and the maximum hourly acetaldehyde is modelled to be of about 116.28 μg m^{-3} (Table 2). Exceptionally high levels of acetaldehyde have also been recorded at locations where the use of ethanol vehicles is widespread. Brazil, for instance, is one of these locations (Anderson, 2009). Over 4 million LDVs in some Brazilian sites use hydrated ethanol. Ambient levels of acetaldehyde have been reported to be between 4 and 82 μg m^{-3} in Rio de

Table 2
Summary of modelled acetaldehyde concentration (μg m^{-3}) obtained for the total model domain and in a receptor point for both scenarios.

	Scenarios	
Modelled values (μg m^{-3})	Baseline	E85-fleet
Total model domain		
Maximum hourly value	10.4	116.28
Highest grid value	1.2	8.04
Receptor point (Sannergata)		
Maximum hourly value	8.4	61.67
Maximum grid value	8.23	61.02
Average grid	1.16	5.45
Winter average	1.3	5.97

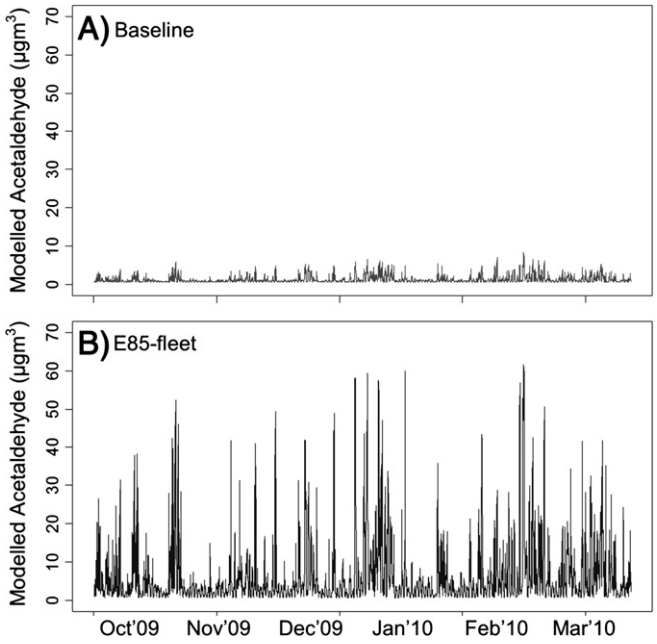

Fig. 3. Modelled hourly acetaldehyde concentration at a roadside in Oslo (i.e. Sannergata) in the baseline (A) and E85-fleet (B) scenarios.

Janeiro and are attributed to direct emissions of the vehicular fleet, in addition to the photochemical oxidation of organic compounds (Corrêa et al., 2003).

Model validations are difficult for acetaldehyde as there are no available continuous measurements for this compound. However in López-Aparicio and Hak (2013), some indicative measurements of ambient acetaldehyde concentrations were performed. The measurements represent 1.5 hour averages at six sites, four exposed and two non-exposed to the circulation of E95-bioethanol buses, obtaining higher concentrations at the locations exposed to the E95 bus-line. The acetaldehyde average concentration of the two non-exposed sites is about 6.9 µg m^{-3}, while the average of the four exposed sites is 9.6 µg m^3, involving around 2.7 µg m^{-3} increment, nearly 40% increase. If we compare this increment with that observed from the modelling results to evaluate the road contribution (i.e. grid average versus road side), it seems that the model results show lower increases, for instance at the receptor point in Sannergata where an increase of about 12% is observed. However, acetaldehyde concentration levels are similar at the same street, where the measured value was about 8.7 µg m^{-3} and the maximum modelled hourly values at the baseline scenario are about 8.4 µg m^{-3}.

Differences between modelled results and measurement results may be expected as model scenarios were designed for winter conditions and the measurements were carried out in May, when temperatures are higher and the contribution from wood burning is not existing. Based on these differences, we would expect modelled concentrations during winter to show larger values than the indicative measurements showed, however this is not the case. In order to shed light on the understanding of this difference, a one month model calculation was performed for May 2011, when measurements took place. The model calculation was performed with the same EFs as the winter calculations for both scenarios, resulting in month average concentrations at Sannergata of about 0.93 µg m^{-3} and 2.86 µg m^{-3}, respectively for May 2011. These values represent approximately half of the average concentration levels obtained for winter and mainly due to the meteorology with less stable conditions. The maximum hourly acetaldehyde concentration at Sannergata for the baseline scenario and May dispersion conditions is about 3.09 µg m^{-3}, less than 1/3 of the indicative measurements (i.e. 8.7 µg m^{-3}). Even though one cannot put too much emphasis on these comparisons due to the fact that very few measurements are available, and due to differences in time and duration, these comparisons do raise some questions. For instance, if there are additional acetaldehyde sources which were not considered in the modelling activities, such as acetaldehyde from the oxidation of evaporative emissions of ethanol or as results of photochemical oxidation of organic compounds, or the background levels considered in this study are slightly low. Moreover, acetaldehyde EFs reported in the literature are scarce, adding additional uncertainties. The emission measurements performed in López-Aparicio and Hak (2013) did indicate larger EF for the E95 bioethanol bus (i.e. 0.9–6.5 µg m^{-3}) than the one applied here (i.e. EF = 0.8 µg m^{-3}) for HDVs. In this study, we would like to emphasise the need of research for the determination of emission factors for non-regulated compounds in order to contribute to a better understanding of their emissions from biofuel combustion, and reduce uncertainties as it has been previously pointed out by other authors (Cook et al., 2011).

4. Conclusions

The results obtained in our study show that the use of alternative fuels such as bioethanol needs to be evaluated with respect to the potential effects on urban air quality. The main outcome from air dispersion modelling shows that a full implementation of high blended bioethanol (E85) as fuel for transportation involves an acetaldehyde emission increase of about 233% and 500%, based on a conservative approach (i.e. acetaldehyde EF = 0.8 g km^{-1}) or extreme emission approach (i.e. acetaldehyde EF = 6.9 g km^{-1}), respectively. By shifting fuels from the current situation characterized by petrol and diesel vehicles to an E85-fleet scenario, the acetaldehyde ambient concentration increases by up to 650%, involving that around 28% of the urban population will be exposed to a winter concentration level above 5 µg m^{-3}, implicating a certain risk for human health. From a methodological point of view, the E85-fleet scenario developed in this study may be considered conservative, giving rise to potentially underestimated results. Uncertainties associated with the emission factors exist and the need for additional research is emphasized.

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