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# *Interactive comment on* “Impacts of using reformulated and oxygenated fuel blends on the regional air quality of the upper Rhine valley” *by* J.-F. Vinuesa et al.

**J.-F. Vinuesa et al.**

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### Answer to both reviewers

The use of bio-fuels has been identified as one of the main solutions to improve air quality. Air quality, and more generally the chemical composition of the atmosphere, depends on both the emission of chemical compounds and the dynamics of the atmosphere. Thus, the appropriate evaluation of the impact of the modifications of the emission requires the use of chemical transport models (CTM) in order to account for the chemistry and the dynamics. Unfortunately, the core of the research in this area is mainly composed of evaluating the modification of the composition of the emission by the use of such fuels. As far as we know, only one study has been devoted to the effect of using bio-fuels including ethyl tertio butyl ether or ETBE on air quality (Vinuesa et al., Atmos. Env. 37, 1757-1774, 2003). This study is devoted only to the local scale of a European urban area and consequently do not take into account the regional transport of pollutants. In this paper, we are extending the analysis performed by Vinuesa et al. (2003) to the regional scale.

For the first time, real emission factors obtained by an independent laboratory are used to assess the impact of fuel modification on air quality. Since air quality does not reduce to ozone concentrations, we also include the effect of the use of the bio-fuels on NO, NO2, and VOCs. We present for the first time a quantitative analysis using a state-of-the-art methodology for emission calculations (see the references included in the manuscript). To quantify the effects of the bio-fuels, we choose to focus on two quantities that are the maximum and the daily average and we present the results on the maximum using composite maps that indicate, in each grid cell, the maximum or the changes of this quantity obtained during the whole period of simulation. In addition, since the fuel reformulation/oxygenation results in both changes in total emission amounts and chemical characteristics of the emission, a sensitivity analysis based on modifying only the amount of the emissions is also performed.

## General comments

*Boundary conditions and modeling methods:*

As mentioned by both referees, lateral boundary conditions are of importance in air quality studies. In particular, when the effect of emission changes on air pollutant concentrations are investigated, one has to make sure that the choice of the boundary conditions allows a good simulation of the base case or reference case. As can be seen in Figures 2 and 3, the lateral boundary conditions allowed simulating the reference case both the meteorological fields and the ozone concentrations with a very good agreement with measurements. The set of the lateral boundary conditions is described in the manuscript (see pages 12075 and 12076).

The top of the domain is set at 6000 meters (as mentioned in the manuscript), meaning that the top boundary condition has no impact on the simulations of the atmospheric boundary layer and that a discussion on these top boundary conditions is irrelevant in this work. In addition and as mentioned in the manuscript, the highest point of the domain is The Feldberg (1493 m) located in the Black Forest. The bottom of the valley is in the north-east region of the domain and it shows an altitude of around 150 meters. The area of interest has been extensively studied in the past (see the list of references in the manuscript). These studies have shown that the dynamical characteristics of the upper Rhine valley do not allow intrusion of free tropospheric ozone as it is observed in Alpine deep valleys especially during summer pollution episodes where the atmospheric boundary layer is strongly convective and well developed.

*Emission characteristics*

The use of real measured emission factors for bio-fuels involves changes in emission amounts and VOC compositions. Since these emission factors have been already published in detail elsewhere (Lopez de Rodas, B., and Marduel, J.-L.: Influence sur la nature et le niveau des émissions à l'échappement, d'une incorporation de composés oxygénés dans les essences, PV n°97/00581, U.T.A.C., BP212, 91311 Montlhery, France, 1997 or in the European DGTREN report untitled "Non Technical Barriers to the development of Liquid Biofuels" as part of the ALTENER program), we have chosen not to show them in this paper. Actually only aggregated emission factors by type of roads were shown in Table 4.

However and following the suggestions of referee 2, we have extracted total scenario-wide emissions that are relevant in this work. In the following table (Table A1), the total scenario-wide changes in NOx and total VOCs emissions due to the fuel-GPC fleet are presented. In the same table, the total scenario-wide VOC emissions (in kg) are given.

Table A1: Total daily scenario-wide emissions of the NOx, total VOCs and main VOC splitted compounds (in kg) when 100% of the fuel-GPC fleet is using the fuel blends REF, ETBE1, ETBE2 and R2.

| | REF | ETBE1 | ETBE2 | R2 |
|---|---|---|---|---|
| NOx | 123.9 | 137.9 | 133.4 | 114.7 |
| Total VOCs | 523.5 | 416.3 | 442.8 | 403.1 |
| Methane | 33.1 | 31.8 | 36.2 | 26.8 |
| Ethane | 3.8 | 3.9 | 4.2 | 4.6 |
| Propane | 0.2 | 0.2 | 0.2 | 0.2 |
| Alkanes | 168.2 | 118.4 | 151.0 | 158.6 |
| Ethene | 20.1 | 19.0 | 20.0 | 18.9 |
| Propene | 10.7 | 9.9 | 10.9 | 18.8 |
| Alkenes | 21.7 | 24.3 | 28.3 | 22.3 |
| Acetylene | 37.3 | 36.5 | 33.9 | 26.4 |
| Alkynes | 2.5 | 2.8 | 3.4 | 3.2 |
| Esters | 0.0 | 22.8 | 24.1 | 0.0 |
| Benzene | 10.0 | 7.7 | 7.2 | 6.9 |
| Aromatics | 188.3 | 119.0 | 104.0 | 97.2 |
| Styrenes | 0.4 | 0.4 | 0.2 | 0.1 |
| Formaldehyde | 2.0 | 1.9 | 1.9 | 1.8 |
| Other aldehydes | 3.7 | 5.6 | 5.1 | 3.5 |
| Acetone | 0.2 | 0.9 | 1.0 | 0.6 |
| Ketones | 0.1 | 0.0 | 0.1 | 0.1 |
| Non identified | 21.1 | 11.2 | 11.3 | 12.9 |

As can be seen in Table A1, both ETBE and R2 fuels decrease the VOC emissions levels while only R2 allows, in addition, a decrease of the emissions of NOx. Alkanes and aromatics are the VOCs for which the emissions are the most affected by the use of alternate fuels. In the case of the use of ETBE, aldehydes, alkenes and ethers also show modifications to certain extend.

*Interpretation of the results*

To answer the specific comment of reviewer 1 about the metrics used in this paper, we should mentioned that these metrics are not new and have shown to be good indicators to assess emission effects on secondary pollutants at various scales. For instance and apart from the references already listed in the manuscript, some recent researches using this kind of metrics and composite maps are presented in Pison and Menut, Atmos. Env. 38, 971-983, 2004 (for their use to assess the impact on air traffic on ozone levels at regional scale) or Ma and Van Ardenne, Atmos. Chem. Phys., 4, 877-887, 2004 (for quantifying the impact of emission inventory uncertainties at continental scale).

Both the uses of ETBE1 and ETBE2 enhance the emissions of NOx and reduce the total VOC emission leading to a reduction of ozone levels. Similar results are found in the sensitivity analysis where only the total amount of NOx and VOC are changed. However, ETBE1 is the fuel that gives the better improvement with respect to ozone levels (both peaks and background concentrations). The difference between ETBE1 and ETBE2 can be mainly explained by the differences in the VOC exhaust speciation (since NOx emission levels are quite similar see Table A1). The use of ETBE1 implies the emission of more aromatics than ETBE2 which result in enhanced ozone reactivity.

Table 6 of the manuscript shows that the most affected levels (peaks and background concentrations) are the one of highly reactive alkanes and aromatics with scenario-wide reductions between 12 and 25% for both RIM and RIA. This table also shows that ETBE2 allow a bigger reduction of those levels than ETBE1. Actually the highest reductions are obtained for R2. These levels of highly reactive alkanes and aromatics can be directly correlated with the emission levels shown in Table A1. The impacts on

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the levels of formaldehydes and aldehydes obtained by the use of ETBE1 and ETBE2 as shown in Table 6 are quite similar and relatively small.

If ETBE based-fuels and R2 had similar NOx emissions and since the sensitivity analysis revealed that an improvement of regional ozone levels can be obtained by increasing NOx emissions or reducing total VOC emissions, one can guess that the reduction of ozone levels induced by the use of R2 would have been enhanced. It is more difficult to quantify this reduction with respect to ETBE ones since the enhancement of ozone reduction would have also benefit from the levels of VOC emission induced by R2 (that show the lowest emission levels) but disbenefit from the levels of aromatics emissions.

*Sensitivity analysis*

The sensitivity analysis has been performed in order to help quantifying how the characteristics of the emission (amount or reactivity) are driving the results. As mentioned in the manuscript, the analysis is based on systematic reductions or increases of road traffic emissions: increase/decrease of 10% of NOx emissions from the GPC fleet (SCE1+/SCE1-) and increase/decrease of 10% of VOC emissions from the GPC fleet (SCE2+/SCE2-). Note that the speciation of the VOC is kept similar to the one of the reference case (REF). The results show that an improvement of regional ozone levels can be obtained by increasing NOx emissions or reducing total VOC emissions (that is basically what the use of ETBE1 and ETBE2 implies). In the manuscript, we concluded that the impact of alternative fuels on lowering the ozone levels is not only related to the reduction of the emissions, but also to the modification of the composition of the emissions as well as the VOC speciation.

As mentioned in the manuscript, the upper Rhine valley region is a high-populated area and most of it can be considered as urban or semi-rural area meaning that urban or regional impacts are following the same trends: negative effect on NOx levels and

VOC sensitive. With that respect it can be hardly compared with regional scenarios in the United States where the regional scales are mainly composed of rural areas.

*Comparison with previous study at local scale*

In Vinuesa et al. (2003), the REKLIP 1992 emission inventory is used and has been updated for the period of interest, i.e. 9-15 May 1998 and for the region of interest. The INTERREG II 1998 emission inventory used here is the updated and extended version of the REKLIP one. In addition, we used the same methodology as Vinuesa et al. (2003) to calculate the emission scenario. The only difference remains in the spatial resolution of the emission inventories (1x1 km2 and 4 x 4 km2). As mentioned in the paper, artificial dilution effects due to different averaging procedure can induce some difference in concentrations however since we are interested in relative impact, we assume that this dilution effect has no effect of the trends of the results. In addition and to support our assumption, we found comparable results while simulating the base case and also a good agreement with measurements (see Figure 3 of the manuscript and Figure 5 of Vinuesa et al. (2003)).

## Specific comments and revision of the manuscript

In the section related to the emission inventories, we have included a short description on the topography of the domain to answer the concerns of both referees. The new sentences read: *"The highest peaks are the Feldberg (1,493 meters) and the Grand Ballon (1,425 meters) in the southern parts of respectively the Black Forest and Vosges. The bottom of the valley is very flat and is in the range of 120 to 180 meters from north to south. This configuration have large impact on the dynamics of the low atmosphere and it leads to large periods of weak wind with temperature inversion and even if the topography favors the emergence of local valley and mountains breezes, polluted air masses remain over the valley and are hardly dispersed at regional scale."*

As suggested by reviewer 2, we have included a new table (Table A1) together with discussions on the emission levels (total amount and reactivity differences in the VOC speciation). This new paragraph reads: *"In order to emphasize the differences of the emissions involved by the use of the various fuel-blends, the daily total emission changes when the whole GPC fleet is using such fuels is presented in Table 5. Both ETBE and R2 fuels decrease total VOC emission levels while only R2 allows a decrease of the emissions of NOx. Alkanes and aromatics are the VOCs for which the emissions are the most affected by the use of alternate fuels. In the case of the use of ETBE, aldehydes, alkenes and ethers also show modifications to certain extend."* We have also correlated mass emission and reactivity changes on air quality in several places in the manuscript.

For the sake of clarity and taking into account both reviewers' comments, the short description of ozone chemistry has been removed.

The definition of the RIA and RIM implies that negative numbers indicate increases and positive numbers indicate decreases. The goal of this paper is to determine the impact of using alternative fuels on the air quality and such a goal implies that a positive impact refers to a reduction of the concentrations (decreases of the RIA and RIM) and a negative impact refers to an increase of the concentrations (increases of the RIA and RIM). In addition and as mentioned previously, this paper is a extension of Vinuesa et al. (2003) to the regional scale therefore the same definition of RIA and RIM should be use to allow a better/easier comparison of the results. We have added the following sentence: *"Note that this definition allow to clearly distinguish between improvement (positive values) and degradation (negative values) of the air quality"*.

Also to clarify the interpretation of the indicators, we emphasized their 'composite aspect' by adding *"and that HMC map is a so-called composite map since the concentration peaks can be reached at different times"* while presenting the HMC and DAC maps for the first time in the manuscript.

In order to improve readability, we have followed the referee 2's suggestions and modified Tables 5, 6 and 8 (respectively Tables 6, 7 and 9 in the revised manuscript) taking into account the near-linear effects of the results. The following comment has been added: *"Since the impacts show a near-linear effect with respect to fractional change in the fleet penetration, only the results concerning 50% and 100% of the GPC fleet using alternative fuels are shown in Tables 6 and 7"*. However and since air quality does not reduce to ozone concentrations, we included the effect of the use of the bio-fuels on NOx and VOCs.

As suggested by referee 2, we have enhanced the discussion of the sensitivity analysis by including the benefits of using such an analysis to assess which characteristics of the emission (amount or reactivity) has more impact on the results. The new first line of the paragraph reads *"In order to estimate the sensitivity of the results presented previously and to distinguish which characteristic of the emission (amount or reactivity) is driving these results, we have defined 4 extra emission scenarios."* The end of the paragraph now reads: *"Since the use of both ETBE fuel-blends reduce the total scenario wide emissions of VOC but increase the ones of NOx (Table 5), these scenarios can be considered as scenario-like combination of SCE1+ and SCE2-. The corresponding scenario-like for R2 combines SCE1- and SCE2-. Since the sensitivity analysis shows that an improvement of regional ozone levels can be obtained by increasing NOx emissions or reducing total VOC emissions, the main differences in the results obtained between R2 and ETBE based scenario is due to the level of NOx emissions. However, the differences between the impact of using ETBE1 and ETBE2 are clearly related to the changes of reactivity of the VOC emissions. Therefore, the impact of alternative fuels on lowering the ozone levels is not only related to the reduction of the emissions, but also to the modification of the composition of the emissions in particular the VOC speciation."*

In addition, we strengthen the argumentation about the validity of our assumption in the comparison with Vinuesa et al. (2003) results by emphasizing the fact that the methodologies used in both studies are the same. In particular the first paragraph of the corresponding section now reads: *"As mentioned previously, V2003 studied the effect of using the same alternative gasoline fuel blends, i.e. ETBE1, ETBE2 and R2, on the air quality of an urban area. Since we are using the same methodology, the same meteorological fields and constant boundary conditions (i.e., in space and time) for the pollutant concentrations that allowed reproducing the base case, i.e. the ozone pollution episode of May 1998 with a very good agreement with measurements, it is reasonable to compare the results obtained at local scale (V2003) with those obtained from the present study by focusing on the Strasbourg area."*

Finally, we emphasized the two effects (amount of emissions and reactivity of VOC) of using alternate fuels in the concluding remarks by adding *"The sensitivity analysis revealed that the impact of alternative fuels on lowering the ozone levels is not only related to the reduction of the emissions, but also to the modification of the reactivity of the VOCs."* to the paragraph concerning ozone levels.

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