

# ***Interactive comment on “On the segregation of chemical species in a clear boundary layer over heterogeneous land surfaces” by H. G. Ouwersloot et al.***

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Ian Faloon has risen very interesting points in his evaluation. Below we respond to them, including the modifications in the revised manuscript.

As stressed by the referee, the applied chemical scheme does not capture all atmospheric chemistry, but is stripped to the essentials (Vilà-Guerau de Arellano et al., 2011). This scheme was chosen to balance computational costs with a characteristic representation of chemistry in a prototype convective boundary layer. Related to the influence of NO<sub>x</sub> regimes, sensitivity analyses have been performed for the current

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set of chemical reactions and initial concentration profiles by changing the surface emissions of NO by a factor of 10 and changing the emission patterns. These tests indicated that the role of NO was small for the chosen set of (low NO<sub>x</sub>) conditions. This information will be included in the revised manuscript. However, we do acknowledge that for different emission strengths and/or different initial concentrations of the chemical species, NO<sub>x</sub> can be very important for the segregation between OH and hydrocarbons like isoprene. This especially holds true for entrainment of NO<sub>x</sub> from the free troposphere. In order to tackle this issue, we have performed two extra simulations and will include the main findings and discussion in an extra section.

The referee would like to put our findings on the effect of segregation in perspective of present uncertainties while evaluating chemical box model results. Compared to these sources of uncertainty the effect of segregation on atmospheric chemistry is less relevant from a practical point of view.

We do agree with the referee, which is the reason we recommend that “no parameterizations for the intensity of segregation need to be implemented for the sub-grid scale processes in chemistry-transport to help reconcile observations with modelling”. The knowledge of the intensity of segregation can be used after the understanding of the photochemical system has increased. We will include these sources of uncertainty in the last paragraph of the conclusions (p. 18956, l. 19), supporting the recommendation not to solely focus on the intensity of segregation to reconcile boundary layer chemistry observations with modelling.

#### More specific comments

The reference to the work of Sykes et al. (1992) concerns the intensity of segregation in case of a reacting plume, but will be included since it provides some extra background knowledge.

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The uncharacteristically low level of water vapor needed to prevent cloud formation does indeed influence the atmospheric chemistry by lowering the chemical production rate of  $\text{HO}_x$ . Unfortunately, this has a significant impact on the intensity of segregation under study, which is tested by doubling the reaction rate constant for reaction R2. However, these numerical experiments also showed that the intensities of segregation were affected similarly for all numerical experiments (approximately enhanced by a factor 1.3), which indicates that the dependences of the intensity of segregation are not affected that much. A caveat for the effect on the atmospheric chemistry will be included at the introduction of these low values of the specific humidity.

To avoid confusion in lines 1-2 on page 18943, the word “smaller” shall be removed. What was meant by this sentence is that the changes at 1350 m, which are smaller than the changes at 1600 m, are due to entrained air. The changes are smaller at 1350 m height, since the relatively warm and dry entrained air is distributed over the whole boundary layer, while the relatively cold and moist intruding air is not spread throughout the free troposphere, but located below the elevated inversion layer over the savannah.

The referee pointed out that the described distribution of isoprene in lines 10-11 on page 18943 is only valid above the surface layer. This clarification will be added.

The stated 30 minutes of transport time that does not match the dynamical time scale was calculated by using the vertical velocity output and considering transport from 50 m height to the boundary layer top. In light of the previous comment that the vertical transport starts above the surface layer, this time is recalculated. The resulting transport time is 15 minutes, which is similar to the dynamical time scale. This value will be updated.

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At page 18944, the word savannah is accidentally used instead of forested. This will be rectified.

Mixing is less effective over the forest than over the savannah. This is confirmed by examining the  $y$  averaged TKE. During the fourth hour of simulated time the TKE is approximately four times higher over the savannah than over the forest. This suppression of turbulence is in accordance with previous studies (e.g., Van Heerwaarden and Vilà-Guerau de Arellano (2008)).

The entrainment rate is not of importance for the main message of lines 8-12 of page 18944. The boundary layer averaged concentrations of isoprene and OH have risen, but the chemical production rate of reaction R9 has decreased. This chemical production rate is dependent on the reactant concentrations, so this decrease can only be explained by the intensity of segregation. This paragraph does not try to explain why these concentrations are altered, which is indeed influenced by both segregation effects (increasing the concentrations) and entrainment (lowering the concentrations). The meaning will be made clearer by adding the word “resulting” before “average chemical reaction rate”.

The drop off in isoprene concentrations in the lowest 50 m of Figure 4 is comparable to the change in concentration in Figure 3 of Vilà-Guerau de Arellano et al. (2011). The magnitude of this gradient is governed by the (similar) surface emissions and turbulence. Visually, the drop is smaller in the paper of Vilà-Guerau de Arellano et al. (2011), because of the higher boundary layer concentration of isoprene, which is caused by the longer time of integration, the lower boundary layer height and the higher isoprene concentration in the free troposphere. While the isoprene concentration gradient in the surface layer is strong, it is similar to observational data shown by Zimmerman et al. (1988).

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The low free tropospheric OH concentrations do indeed result from the absence of NO<sub>x</sub> in that region. Increasing the concentrations of NO<sub>x</sub> in the free troposphere can actually have a large impact on the boundary layer chemistry due to entrainment. The impact of NO<sub>x</sub> entrainment will be shown in a separate section.

Because of the low NO<sub>x</sub> conditions, the impact of NO on the OH in the surface layer is very low. The NO is also quite well mixed within the boundary layer, due to the relatively low NO emissions. Therefore, the NO gradient near the surface does not contribute to the OH variability under the chosen set of conditions.

In the Large Eddy Simulation model, the sub grid contribution to the intensity of segregation is not parameterized. However, sensitivity analyses have been performed on the grid resolution. These tests showed that intensities of segregation are not significantly affected by imposing a finer grid.

The impact of the wind on the intensity of segregation is actually not that surprising when considering Figure 7. Wind that is aligned parallel to the patch borders enhances the mesoscale structures. Therefore the effect of heterogeneity on the intensity of segregation (making it more negative) is enhanced.

We agree with the referee that the calculation of the second moment covariance budget equation is an interesting approach to identify contributions to segregation. However, it would not be straight forward for the case under study. For example, in our case the mesoscale circulation is a more important contribution to transport than the turbulent transport quantified by  $w_*$ . The chemistry would also complicate the evaluation. Since such an analysis, if performed properly, requires a large amount of research and explanation, we will not include it in this article. However, we do think that a budget approach to the intensity of segregation would be valuable for future analyses.

From the (not shown)  $y$  averaged profiles of the numerical experiments we found out that the entrainment reaches a limit for increasing length scales of heterogeneity. The

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fact that there is a limit seems natural, as it is expected that the entrainment over the savannah patch will not exceed the entrainment over a homogeneous savannah domain.

The initial temperature profile over the forested areas indeed has a distinct temperature inversion at the top of the boundary layer. However, because of the entrained warm air that is transported to the top of the boundary layer over the forest, the air above the forest is not heated uniformly. Since the strongest heating source is located at the top of the boundary layer and no direct entrainment takes place above the forest, the thermal inversion rapidly disappears. In Figure 2a the red line shows this process occurs within the first 30 minutes of the simulation. We will improve the explanation as follows: the word “encroachment” will be replaced by “generating a more gradual transition to the free tropospheric lapse rate”.

On page 18952 we conclude that the hydroxyl radical is impacted less than isoprene by the segregation based on the fact that instantaneous OH concentrations are governed by chemical reaction rates, while isoprene is a longer lived species. However, this explanation is not clear. It does not explain why the increase of isoprene almost entirely compensates the increased effect of segregation. A better explanation would be that while the chemical production rate is lowered by the increased segregation, the lifetime of isoprene is increased proportionally, since isoprene is only consumed by the isoprene-OH reaction. However, OH still has a short life time without this specific chemical reaction. Therefore the concentration of isoprene will build up more than that of OH. We therefore propose to include the following modification: “Since the concentration of the short-lived hydroxyl radical is governed by the chemical reaction rates (Krol et al., 2000), OH is also affected much less (only 0.5 %).” will become “While the lifetime of isoprene is inverse proportionally to the decrease in chemical production rate by segregation, the lifetime of the short-lived hydroxyl radical remains short due to other chemical reactions. Therefore the concentration of isoprene will

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increase more compared to OH and consequently OH is also affected much less (only 0.5 %).”

If a thinner region of the (lower part of the) boundary layer over the forest is characterized by high isoprene concentrations, more isoprene is transported from the forest to the savannah boundary layer by the induced mesoscale flow. As a consequence, more isoprene is present over the savannah. In response, the OH concentration decreases over the savannah and increases over the forest. This distribution corresponds to a more negative horizontal segregation. To avoid confusion, the word strong will be replaced with more negative.

#### Technical corrections

All technical corrections proposed by Ian Faloona will be applied.

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