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Comment

Interactive comment on “The balances of mixing ratios and segregation intensity: a case study from the field (ECHO 2003)” by R. Dlugi et al.

Anonymous Referee #1

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The research presented here studies the influence of atmospheric turbulence on the reactivity of isoprene and hydroxyl radical above a tall canopy. The quantification is carried out by analysing the intensity of segregation (Is) (namely the co-variance of isoprene and hydroxyl radical mixing ratio fluctuations made dimensionless by the mean concentration of isoprene and hydroxyl mixing ratio) and the main dynamic and chemistry contributors. The study is based on the estimation of Is and related terms by using meteorological and atmospheric chemistry observations of the co-variance equation of isoprene and the hydroxyl radical taken during the ECHO-2003 campaign. Previous research on Is was mainly based on large-eddy simulation or one-dimensional models coupled to high-order turbulent models. Selected findings of these numerical studies are used to compare and discuss the ECHO-2003 observations. In that respect, the

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authors, and in addition of their exhaustive analysis of the data, have done a very successful work by thoroughly discussing their results and integrating them with previous research based on numerical modelling. I have two main remarks: (1) The observational data used in the research is rather limited. I realized that this measurements are very difficult to be taken, and the data analysis and interpretation is very difficult. The study only used 22 block of 10 minutes. This insures a satisfactory quality to calculate mean statistics, but for higher-order moments is not enough to obtain robust statistics. Have they additional data set that can be used to further support their findings and obtain more reliable statistics? (2) I found the paper with more than 24 figures very lengthy and with some repetitions. I will recommend to the authors to shorten the paper and remove unnecessary figures. By so doing, the paper will be more accessible to potential readers.

Below I include more specific comments:

1- In the introduction there is some confusion on the definition of the Damkohler number first introduced as the time scale of a specific chemical reaction, but later on used as a time scale of a chemically active species. I recommend to clarify it.

2- Section 2. In Equation (1), and since they focus entirely their research on isoprene and the hydroxyl radical, it is perhaps handier and clearer to substitute c_i and c_j by iso and oh .

3- Section 2.1.(lines 17 on page 12920 to 3 on page 12921). Quoting them: 'variation of only 4.7 % in k_{ij} '. This leads to 4.7 % difference in chemical reaction rate. Therefore, the influence of this variable is of the same magnitude as the effect that is investigated (e.g. Fig. 22). This does not seem a valid justification to use a constant k_{ij} in the analysis.

4- Section 2.1. The flux divergences is calculated a residual term and therefore includes contributions of the horizontal advection and horizontal turbulent advection. In order to avoid misunderstandings I will recommend to name it in a different way.

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5- Section 2.1 (line 27 on page 12921 to lines 3 on page 12922): since the ‘flux divergence’ is actually a residual term, calculated with the mean reaction rate, it is not surprising that it has a linear relation with a high correlation coefficient. This could also indicate that the observations are wrong. For example, when in this analysis, the mean reaction rate would be multiplied by a factor 1000, the ‘flux divergence’ would be virtual equal to the negative of the mean reaction rate since it is the residual term. This would lead to a linear relation with correlation coefficient 1. The conclusion that ‘advection is not significant in our case’ can therefore not be drawn on this argument.

6- Section 3.2.1 (pages 12923, l 20-23) It will be interesting if a similar relation holds for isoprene.

7- Section 3.2.2. (lines 20-23) Related to the previous point, the authors seem to indicated that entrainment of isoprene (quoting them: “...some hundred meters above the surface...”) is an important processes influencing the reactivity behaviour. Van Stratum et al. (ACP 12, 5329-5341) (see Figure 6) presented a budget of reactants where entrainment was a relevant process (at least during the large ABL morning growth). Could they clarify and discuss the role of entrainment in the analysis of the observations?

8- Section 3.2.4 In the comparison with LES results, I was wondering how representative it is to compare surface layer measurements influenced by canopy effect with large-eddy simulations that are mainly representative for mixed-layer properties.

9- Section 4.1 (equation 5). The introduction of Equation (5) is very confusing. First they mentioned that the advections terms are neglected and thereafter are included (term $A2k$ for instance). I recommend to introduce it in a systematic and clearer way.

10- Equation 10. The use of C_{ij} can bring confusion when comparing with the concentration. I recommend to use another notation.

11- Section 4.1 (equation 5). The introduction of Equation (5) is very confusing. First they mentioned that the advections terms are neglected and thereafter are included

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(term A2k for instance). I recommend to introduce it in a systematic and clearer way.

12- Section 4.1 (equation 11) Equation (11) does not seem right: the + sign should be a minus. From Eq. (8) there is within \square basically $RES - k_{ij} (C_{ij} + c_j c_i'^2)$. This results, after division by $k_{ij} c_i$ in $- C_{ij}/c_i - c_j/c_i c_i'^2$. So, the + sign should be a minus. Could they check the consistency of the sign?

13- Section 4.2 (page 12934, line 20). A question that perhaps deserve some attention is if a radical fast compound like OH is transported vertically by turbulent motions. Has the OH-turbulent flux a physical meaning? What is the accuracy of the measurement of the OH-flux?

14- Section 4.2 (page 12935, line 20). Although the large-eddy simulation loses accuracy within the atmospheric surface layer, I think it can be interesting, at least qualitatively, (and for the sake of completeness of the discussion) to include a comparison with the co-variance budget presented by Vinuesa et al. (Tellus 55B, 935-949, 2003) for the NO-NO₂-O₃ triad to determine if the vertical distribution and the order of magnitude of the different contributions of the co-variance equation is similar.

15- Connected to the previous comment, the diffusion term seems to have a different meaning and order of magnitude in the research presented by Patton et al. (2001) and Vinuesa et al. (2003) (see for instance Figure 7). In the LES studies, the term is representing a dissipation at the smaller scales and it is comparable in order of magnitude to the other terms in the co-variance equation (gradient production, chemistry or turbulent transport). In turn, in this observational research, this term represent a molecular diffusion term and it is smaller compared to the other terms (see discussion at lines 23-25 at page 12935). Could the authors comment on this differences?

16- Section 5.2.2. In view of the completeness of the research and the temporal evolution of the atmospheric stability, it is perhaps interesting to show a figure of Is as a function of a stability parameter like z/L or the Richardson gradient number.

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17- General comment on figures and data treatment. In view of the uncertainties associated to the simultaneous measurements of turbulence and fast atmospheric compounds, I will recommend the use of error bars in all the figures.

18- Figure 22 is a very good summary of the observational results of Is combined with the buoyancy surface forcing. In order to complete and integrate with the numerical studies, it is perhaps interesting to include some representative results reported by Patton et al. (2003) and additional point for Is under homogeneous surface conditions. The Is value from Ouwersloot et al. (2011) in Fig. 22 and line 28 of page 12947 is indeed for a numerical simulation with $\Delta H_v = 0$, but this experiment considers still differences in isoprene emissions (0.7 ppb m/s) and is therefore NOT representative for homogeneous surface conditions. To truly compare for homogeneous conditions, use their HOM experiment: Is = -0.070. As can be expected, this result also fits better with the other values in Fig. 22. It is possible to combine both values, explaining the difference.

19- Appendix A: f is not the relative reaction rate for OH consumption by isoprene compared to total OH consumption, but compared to OH consumption by other compounds. However, it is stated (line 22 of p 12953) that f is an estimation to calculate the relative isoprene sink from the reaction with OH, so it would make more sense to calculate f as the relative reaction rate for OH consumption by isoprene compared to total OH consumption. Why this discrepancy?

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