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Interactive comment on "Transport and chemical transformations influenced by shallow cumulus over land" *by* J. Vilà-Guerau de Arellano et al.

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Response to the interactive comment on "Transport and chemical transformations influenced by shallow cumulus over land" (Dr. Bruce Denby)

p1. par.1: The authors state that shallow cumulus forms under the same conditions as pollutants tend to accumulate. Can the authors be more specific? Pollutants accumulate with low wind speeds, low ABL heights, poor mixing and stable conditions. Are these the conditions for shallow cumulus convection?

Poor air quality events are frequently associated with benign weather and weak synoptic forcing, which are conditions favorable to the formation of shallow cumulus over land. In the manuscript, we have clarified this sentence to read:

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"Shallow cumulus clouds generally form in synoptically high pressure regions, which are conducive to the formation and accumulation of both passive and photochemically generated pollutants in the boundary layer because of the low wind speeds, the intensification of capping inversions, and high insolation."

p2 par 2: "increase/decrease" can you clarify this? Do you mean both or just a non-defined change.

The sentence should have stated only increase. It has been corrected.

p2 par.3: Last sentence: This is the first time such a study has been carried out with two models. Am I to infer that this sort of study has been carried out with one model? If so references to these here would be useful.

This is also the first time that a systematic study of turbulent reacting flows under the presence of shallow cumulus clouds has been carried out. As the referee mentioned, the additional value in this research is that two different LES codes are used to support better the results and the discussion. The text has been clarified and now states the following:

"Not only does this study provide a systematic study of turbulent photochemical reacting flows in the presence of shallow cumulus clouds, but it is the first time that an intercomparison of reactive chemistry in the LES framework is being conducted."

p.4 eq. (1) and (2): The reactive species being discussed clearly refers to the NOx, Ox reactions. Is there a particular reason why the authors refer to them as A, B and C rather than NO2, NO and O3? It does not make it any clearer to this reviewer. It may be useful for the reader to know the photostationary equilibrium of these species, given the standard clear sky disassociation rates and concentrations modelled. This would give the reader, especially in Section 4, a clearer idea of how different the LES results are from a simple well mixed boundary layer description.

As recommended by the reviewer, we have switched the terminology from the generic

A, B, and C nomenclature to NO2, NO, O3 nomenclature. We have also included the value of the photostationary state after discussing the reaction system on p. 8817-8818 and have included a brief discussion of the variation of the photostationary state values at section 5.

p.4: The surface fluxes used seem to indicate a rather high percentage of emitted NO2 for the total NOx emissions. Is there a reason for this? Is it already close to equilibrium?

The purpose to prescribe a high flux for NO2 is to quickly reach conditions that are close to chemical equilibrium. It is now mentioned in the text.

eq (6). Is there a reference to this equation or has it been derived?

The reference of Joseph et al (1976) to calculate the energy transmission coefficient tr has been added.

p8. par. 1: This paragraph, in a way, gives the conclusion of the first study. That being that it is very important to accurately describe the effect of shallow cumulus on boundary layer dynamics in large scale CTMs. This, in this reviewer's opinion, over emphasises that importance. Though one can point to a 50% difference in average concentration in the ABL, due to its increased extent, concentrations within the majority of the ABL do not actually vary so much at all, with a maximum decrease of 12%. The authors then go on to state that it is essential to accurately know various boundary conditions, e.g soil moisture, in such large scale models. This is an unrealistic request considering the sensitivity of the formation of Cu to the surface conditions. This reviewer would actually consider the most important effect of Cu, in regard to large scale models, to be the enhancement of the exchange of pollutants between the ABL with the free atmosphere. This is inferred to, when the night time residual layer is mentioned, but not discussed. (The final note in the conclusion actually refers to this as a future study, which this reviewer would strongly agree to).

The reviewer has made a good point. We have rewritten the paragraph to include a dis-

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cussion on the impact of the presence of shallow cumulus in the initial concentrations of the residual layer and their possible impact on the exchange of pollutants between the ABL and free troposphere.

p9 par 2: '... greater cloud cover ($<$=0.5; the simulation ...; ' This text is unclear to me. Does the simulation with greater cloud cover have a cloud cover less than 0.5?

It means maximum values between 20 and 22 UTC higher than 0.5. We have rewritten this part to provide additional information on the more cloudy simulation.

Sec 5. This section seems to mix two concepts. That being the effect of clouds on the photolysis rates, and hence on the concentration of the reactant species, and the effect of the Damkohler number. Never the less the authors try to use this to demonstrate that given particular sets of chemical reactions with Da$>$1 the effect of clouds on photolysis can be significant. It does not seem, however, suitable to dwell very long on the instantaneous results, rather than temporal and spatially averaged ones. The authors do not look at the instantaneous values of any other parameter, e.g. concentrations or vertical velocities, and the resulting discussion implies a significance that is truly misleading.

Our aim in showing the instantaneous variables (Figure 14) compared to the horizontal averages (Figure 9) and the volume average (Figure 15) is to demonstrate that spatial/temporal averaging can smooth out the effects of the turbulence induced by clouds and the UV perturbation above, in and below clouds. We realize that from a point of view of large scale modeling it is only important to show results in terms of spatial and temporal averages. However, one of the beauties of LES is that the 3D and time-dependent output allows us to show that boundary layer processes can produce large fluctuations in the reactant fields (depending on the Damköhler number) and consequently could lead to modifications in the reactivity. The instantaneous variables are more likely to be measured (depending on the time resolution of specific instruments) and therefore the interpretation of observations would be improved by the knowledge of the fluctuations occurring in the PBL. In section 4 and the conclusions, we have included a comment on the importance of instantaneous fields in analyzing observations.

Sec 6. Conclusions: There is a tendency in the conclusion to overstate the results of the study. The reviewer recommends these be reformulated to reflect the true situation, as a result of the study, rather than to try to exaggerate them. Firstly: the authors state '... that the presence of clouds could lead to a decrease of 50% of the reactant mixing ratio ...'. They neglect to point out that this is a boundary layer depth average (including the Cu convection region), not a below cloud average, which shows a significantly lesser decrease. Secondly: they state that mass-flux parameterisations underestimate the flux at cloud base by a factor of 2. In the text this is given as a factor of 1.5. Thirdly: They state that as a result of the perturbation of the photolysis rates, due to the presence of Cu that the instantaneous effect is of the order of 40%. They then state that these are smaller when averaged over time and space. In fact, they are significantly smaller. (See comment on Section 5)

Following the advice of the referee, we have modified the conclusions as follows: a) We now state that the presence of clouds leads to a decrease in PBL mixing ratios. b) We changed the mass-flux parameterization underestimation factor to 1.5. c) We now state that differences in concentrations caused by perturbations in photolysis rates are much smaller when averaged over time and space.

Technical corrections

We have substituted UTC to LT in the text and in the figures. All of the technical comments have been taken into account.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 8811, 2005.