The following text, responding to the referee's comments of T.J. Dillon, was communicated to the authors of the manuscript by Jozef Peeters (Department of Chemistry, University of Leuven) and Jean-Francois Muller (BISA, Brussels).

1. The unimolecular rearrangement of ISOPOO

The referee states that "Peeters report a rate of around 1 s-1, and Crounse et al. derive a value of 0.002 s-1". However, two very different rate coefficients are being compared: our ~1 s-1 is the *isomer-specific* isomerisation rate coefficient of the *Z-delta*-OH-isoprenyl-peroxy isomers Z-ISOPAOO and Z-ISOPCOO, whereas Crounse's 0.002 s-1 is the 295 K *bulk* rate coefficient for formation of HPALDs from the ISOPOO *pool*. Our LIM0 prediction for the *bulk total* isomerisation rate at low NO can be found in Peeters and Muller 2010: 0.073 s-1 at 295 K, based on equilibrium *Z-delta*-OH-peroxy fractions of around 0.05 (see point 2 below). In a Sept. 2012 *Addition and Correction* to their 2011 paper, Crounse et al. stress the difference between *isomer-specific* and *bulk* rates, and moreover report observing other peroxy-isomerisation products than the HPALDs, and at least as important.
We should add that our more recent LIM1 computations at much higher levels of theory give *bulk* total isomerisation rates of ca. 0.01 s-1 at 295 K (presentations at AGU and ACM meetings of Dec. 2012), compatible with the Crounse results.

2. Effects of the *delta*-OH <=> *beta*-OH-peroxy interconversion on the MACR/MVK ratio.

We did indeed propose quasi-equilibration of the *delta*-OH-peroxys and *beta*-OH-peroxys for long enough ISOPOO life-times (Peeters et al. 2009), i.e. ISOPAOO <=> ISOPBOO on one hand, and ISOPCOO <=> ISOPDOO on the other hand; with our predicted *effective* conversion rates at 295 K of about 6.5 s-1 for ISOPAOO --> ISOPBOO, and 3 s-1 for ISOPCOO --> ISOPDOO, i.e. much faster than the total ISOPOO removal rates of ca. 0.25 s-1 of your work, equilibration should be established through the proposed mechanism. Yet, these equilibria should only slightly change the population fraction of ISOPBOO (in the total ISOPOO pool) from 0.44 (MCM-based *formation* fraction) to about 0.52 (LIM0 *equilibrium* fraction), and for ISOPDOO from 0.31 to around 0.36, such that the effect on the derived 15% HOx regeneration should also be limited. In any case, the *delta*- to *beta*- isomer interconversions we predict occur only for a given initial OH-adduct, i.e. A<=> B for the 1-OH-isoprene adducts and C<=> D for the 4-OH-isoprene adducts, but NOT between these two sub-pools. Therefore, the numbers the referee infers as resulting from the equilibration, i.e. "the percentages ISOPBOO (32%) and ISOPDOO (60%)", as well as "the factor of two larger MACR than MVK yields" are erroneous.

Crounse, J. D., Paulot, F., Kjaergaard, H. G., and Wennberg, P. O.: Peroxy radical isomerization in the oxidation of isoprene, Phys. Chem. Chem. Phys., 13, 13607-13613, 10.1039/c1cp21330j, 2011.

Peeters, J., Nguyen, T. L., and Vereecken, L.: HOx radical regeneration in the oxidation of isoprene, Phys. Chem. Chem. Phys., 11, 5935-5939, 10.1039/b908511d, 2009.

Peeters, J., and Muller, J.-F.: HO(x) radical regeneration in isoprene oxidation via peroxy radical isomerisations. II: experimental evidence and global impact, Phys. Chem. Chem. Phys., 12, 14227-14235, 10.1039/c0cp00811g, 2010.