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Segregation Effects on Chemical Reactions in Atmospheric Flows: A Comparison of Experimental and Modeling Results

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The chemical reaction cycle in the atmosphere can be influenced by the degree of inhomogeneous mixing (segregation) as shown by results from several modeling and field studies based on own measurements and data from literature. As a measure we compare the intensity of segregation I_S, the covariance of two reactants divided by the product of their means. Further on parameters influencing I_S (e.g. variances, correlation coefficients) and their dependance on meteorological and chemical conditions are considered. These results are compared with respect to the following reactions: Isoprene + OH, O_3 + NO and the photolysis of O_3. In addition some estimates are given for the reaction HO_2 + NO. The intensity of segregation I_S is mostly found to be negative in the range $-0.3 < I_S < 0$, which results in a reduced volume mean chemical removal rate compared to the homogeneously mixed case. But the modeled and experimental results may differ by the influence of the correlation coefficient between two reactants. For convective (i.e., unstable) conditions a tendency of increasing absolute values of $-I_S$ is found for bimolecular reactions (e.g. OH + isoprene) with increasing buoyant production. This effect is suppressed if - for example - significant chemical sinks for OH others than isoprene exist. Up to now the only case with positive I_S is evaluated for a strongly varying photolysis frequency of O_3 (as source of OH) for the reaction isoprene + OH.