Geophysical Research Abstracts
Vol. 21, EGU2019-3634, 2019
EGU General Assembly 2019

Relative importance of gas uptake by ground and aerosols in the planetary boundary layer

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Gas uptake by ground and aerosol surfaces represent important multiphase processes in the atmosphere. Quantifying the relative contribution of gas uptake on ground versus aerosol surfaces is important to determine which process should be included or simplified in atmospheric models. The purpose of this study is to identify the aerosol uptake processes that is equally or more important than the dry deposition on ground surfaces but has not been adequately addressed. Gas uptake by ground is often described by the deposition velocity (V_d) while uptake by aerosols is mostly characterized by the effective uptake coefficient (γ_{eff}). The different formulations make it difficult to directly compare their contributions. In this study, we compared the uptake fluxes of several reactive gases on both ground and aerosol surfaces to identify the dominant multiphase processes especially within the planetary boundary layer (PBL). We introduced a parameter γ_{eqv}, the equivalent reactive uptake coefficient at which the aerosols have the same uptake fluxes as the ground surfaces. If $\gamma_{eff} > \gamma_{eqv}$, the aerosol surfaces are more important than the ground surfaces regarding gas uptake and vice versa. A series of γ_{eqv} have been determined for different gas species (O_3, NO_2, SO_2, N_2O_5, HNO_3, H_2O_2), aerosol types (mineral dust, soot, organic aerosol, sea salt aerosol), land use categories (urban, agricultural land, Amazon forest, water), seasons and mixing heights. We show that for all investigated chemical species, γ_{eqv} is lowest in the urban area, in the range of $10^{-5} \sim 10^{-4}$, and highest in the Amazon forest, reaching up to $10^{-3} \sim 10^{-2}$. In urban, the aerosol uptake can play key roles for all investigated species and therefore should be considered in models. For other land use types (Agricultural land, Amazon forest, water body), NO_2, SO_2 and O_3 tend to deposit on ground surfaces ($\gamma_{eff} < \gamma_{eqv}$), except for NO_2 and SO_2 on sea salt aerosols, and O_3 on liquid organic aerosols ($\gamma_{eff} > \gamma_{eqv}$). The uptakes of N_2O_5, HNO_3 and H_2O_2 by mineral dust are similar or even more important than those on the ground ($\gamma_{eff} > \gamma_{eqv}$). Given the fact that most models have considered their uptakes on the ground surface, we suggest also considering the N_2O_5 uptake by all types of aerosols, HNO_3 and H_2O_2 uptakes by mineral dust, O_3 uptake by liquid organic aerosols and NO_2, SO_2 and HNO_3 uptakes by sea salt aerosols in atmospheric models because of their higher or comparable importance.