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Observations of HNO_3, Σalkyl nitrates, Σperoxy nitrates and NO_2 fluxes: Mechanisms controlling exchange over a ponderosa pine forest

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Measurements of exchange of reactive nitrogen oxides between the atmosphere and a ponderosa pine forest in the Sierra Nevada Mountains are reported. During winter, we observe upward fluxes of NO_2, and downward fluxes of total peroxy and peroxy acyl nitrates (ΣPNs), total alkyl and multifunctional alkyl nitrates (ΣANs), and the sum of gaseous HNO_3 and semi-volatile NO_3^- aerosol ($HNO_{3(g+p)}$). The signs and magnitudes of these wintertime individual and ΣNO_{yi} fluxes are in the range of prior measurements and indicate net ΣNO_{yi} deposition. However, during summer, we observe downward fluxes only of ΣANs, and upward fluxes of HNO_3, ΣPNs and NO_2 with signs and magnitudes that are unlike most, if not all, previous observations and analyses of fluxes of individual nitrogen oxides. The results imply that the mechanisms contributing to NO_y fluxes, at least at this site, are much more complex than previously recognized. We show that the observations of upward fluxes of HNO_3 and ΣPNs, and downward fluxes of ΣANs, during summer are consistent with oxidation of NO_2 and acetaldehyde by elevated OH within the forest canopy. We discuss the implications of elevated HO_x, and explore the relative importance of deposition, canopy chemistry, and ecosystem emissions in controlling biosphere-atmosphere exchange of reactive nitrogen oxides.