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Vertical profiles of O_3 and NO_x chemistry in the polluted nocturnal boundary layer in Phoenix, AZ: I. Field observations by long-path DOAS

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Abstract. Nocturnal chemistry in the atmospheric boundary layer plays a key role in determining the initial chemical conditions for photochemistry during the following morning as well as influencing the budgets of O_3 and NO_2. Despite its importance, chemistry in the nocturnal boundary layer (NBL), especially in heavily polluted urban areas, has received little attention so far, which greatly limits the current understanding of the processes involved. In particular, the influence of vertical mixing on chemical processes gives rise to complex vertical profiles of various reactive trace gases and makes nocturnal chemistry altitude-dependent. The processing of pollutants is thus driven by a complicated, and not well quantified, interplay between chemistry and vertical mixing. In order to gain a better understanding of the altitude-dependent nocturnal chemistry in the polluted urban environment, a field study was carried out in the downtown area of Phoenix, AZ, in summer 2001. Vertical profiles of reactive species, such as O_3, NO_2, and NO_3, were observed in the lowest 140 m of the troposphere throughout the night. The disappearance of these trace gas vertical gradients during the morning coincided with the morning transition from a stable NBL to a well-mixed convective layer. The vertical gradients of trace gas levels were found to be dependent on both surface NO_x emission strength and the vertical stability of the NBL. The vertical gradients of O_x, the sum of O_3 and NO_2, were found to be much smaller than those of O_3 and NO_2, revealing the dominant role of NO emissions followed by the O_3+NO reaction for the altitude-dependence of nocturnal chemistry in urban areas. Dry deposition, direct emissions, and other chemical production pathways of NO_2 also play a role for the O_x distribution. Strong positive vertical gradients of NO_3, that are predominantly determined by NO_3 loss near the ground, were observed. The vertical profiles of NO_3 and the calculated vertical profiles of its reservoir species (N_2O_5) confirm earlier model results suggesting complex vertical distributions of atmospheric denoxification processes during the night.

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