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Missing gas-phase source of HONO inferred from Zeppelin measurements in the troposphere

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Nitrous acid (HONO) is an important trace gas in the atmosphere due to its contribution to the cycles of nitrogen oxides (NO_X) and hydrogen oxides (HO_X). In the past decades, ground-based observations of HONO around the world showed much higher daytime concentrations than can be explained by the known gas-phase chemistry (i.e., HONO + $h\nu \rightarrow$ OH + NO, HONO + OH $\rightarrow$ NO_2 + H_2O, OH + NO + M $\rightarrow$ HONO + M). Different light-dependent reactions on ground surfaces have been proposed as additional daytime HONO sources. However, due to lack of measurements, little is known about the concentrations of HONO and its sources in the planetary boundary layer (PBL) at higher altitudes above the earth's surface.

The airship Zeppelin NT is an ideal platform to investigate the chemistry and dynamics of the PBL. During the PEGASOS field campaigns in 2012 and 2013, HONO and its gas-phase sources and sinks were measured simultaneously on-board the airship Zeppelin NT, for the lowest 1 km of the PBL. In the upper part of the altitude range during morning hours, when the airmass is still isolated from processes at the earth's surface by the remaining nocturnal boundary layer, we find unexpectedly large concentrations of HONO which can neither be explained by heterogeneous reactions on aerosol and ground surfaces, nor by known gas-phase reactions. Our observations show evidence for an unknown gas-phase light-dependent HONO production which dominates the overall HONO formation in the lower troposphere. This new HONO source requires NO_X and possibly OH or HO_2 radicals. As a result, the general impact of HONO on the OH formation is likely overestimated.