## New measurements reveal a large contribution of nitrogenous molecules to ambient organic aerosol

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Nitrogen is a significant element that constitutes ambient organic aerosol. Individual N-containing organic molecules are known to have both natural and anthropogenic sources and implicated in a wide-ranging health and environmental effects. Yet, unlike carbon (C), the total quantity of aerosol organic nitrogen (ON) remains largely unquantified, hindering a quantitative understanding of their major sources and impacts. Here, for the first time, aerosol ON is quantified in hundreds of aerosol filter samples collected from sites of varying urban influence in China using our recently developed method that permits simple, and yet sensitive, simultaneous detection of inorganic and organic nitrogen. Annual average ON concentration was in the range of 0.4-1.4 µg N m<sup>-3</sup>, representing 17-31% of aerosol total nitrogen. Monte Carlo simulations constrained by paired ON and organic carbon (OC) measurements suggest N-containing organic molecules contributed 20-80% (with a mean of 43%) of ambient organic aerosols. Source apportionment analysis reveals that biomass burning and secondary formation are dominant ON sources, accounting for 21-24% and ~30% of ON, respectively. Primary biological aerosol is also a significant source of ON (7-18%), with its contribution more prominent in non-urban atmospheres. The revelation of the large presence of nitrogenous molecules in atmospheric aerosols implies their role in carbon chemistry has likely been under-appreciated in the past. This new compositional insight, we anticipate, would bring forth a breakthrough in our ability to describe and model organic aerosols and to rethink the focus of studying and modeling OA.