Atmospheric alkaline gases: impacts, observation, and source apportionment

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Atmospheric alkaline gases, including NH₃ and amines, have been demonstrated to play pivotal roles in atmospheric chemical processes, from enhancing atmospheric nucleation to promoting secondary particulate matter formation. Particularly, aminecontaining critical nuclei have been detected during nucleation events, and the underlying formation mechanisms have been actively investigated. High time-resolution (within a few minutes) and high-sensitive (sub-ppbv levels) measurements of alkaline gases are essential to fully elucidate their roles in atmospheric chemistry. Such requirements present great challenges in analytical techniques. Recently, chemical ionization mass spectrometry has been developed to make in-situ observations of NH₃ and amines possible. NH₃ and amines are found to be ubiquitously present in the atmosphere. However, little is known about their specific source profiles and the corresponding source contributions. Although it is well accepted that agriculture-related emissions dominate the global NH₃ budget in the atmosphere, several recent field studies have demonstrated that non-agricultural emissions are the primary NH₃ sources in many urban areas in China. Therefore, atmospheric models based on emission inventories concentrated on agricultural emissions may not realistically simulate the atmospheric environment, particularly in the populated megacities of China. In this study, NH₃, amines, amides, and imines emission characteristics of motor vehicles have been determined explicitly through in situ roadside measurements. Based on constrained SoFi-PMF analysis, we have investigated the specific impacts of motor vehicle emissions on NH₃ and other alkaline gases in urban Beijing. It is found that motor vehicles can contribute a predominant portion of amines in the urban environment, particularly during the daytime. Hence, reducing on-road vehicular emissions can decrease primary emissions of criteria air pollutants and suppress secondary aerosol formation. Meanwhile, the septic system in the urban area is recognized as a significant contributor to background NH₃ and amines in urban Beijing. Motor vehicle emissions usually show less seasonal variability, and intense demands for central heating, power generation, and biofuel usage in suburban areas may significantly affect wintertime haze formation. These emission activities, however, may highly depend on environmental conditions, such as temperature and humidity, and their emission features are still critically needed to fully assess their impacts on the environment.