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Interesting Scientific Questions Regarding Interactions in The Gas-Aerosol-Cloud System

Azadeh Tabazadeh

NASA Ames Res. Center, Earth Sciences Division, Moffett Field, CA

The growth of human population and their use of land, food and energy resources affect the Earth's atmosphere, biosphere and oceans in a complex manner. Many important questions in earth sciences today deal with issues regarding the impact of human activities on our immediate and future environment, ranging in scope from local (i.e. air pollution) to global (i.e. global warming) scale problems. Because the mass of the Earth's atmosphere is negligible compare to that found in the oceans and the biosphere, the atmosphere can respond quickly to natural and/or manmade perturbations. For example, seasonal "ozone hole" formation in the Antarctic is a result of manmade CFC emissions in just the last 40 years. Also, the observed rise in global temperatures (known as global warming) is linked to a rapid increase in carbon dioxide and other greenhouse gas concentrations (emitted primarily by combustion processes) over the last century.

The Earth's atmosphere is composed of a mixture of gases, aerosol and cloud particles. Natural and anthropogenic emissions of gases and aerosols affect the composition of the Earth's atmosphere. Changes in the chemical and physical makeup of the atmosphere can influence how the Earth will interact with the incoming solar radiation and the outgoing infrared radiation and vise versa. While, some perturbations are short-lived, others are long-lived and can affect the Earth's global climate and chemistry in many decades to come. In order to be able to separate the

natural effects from anthropogenic ones, it is essential that we understand the basic physics and chemistry of interactions in the gas-aerosol-cloud system in the Earth's atmosphere. During my presentation, I will briefly discuss the important physics and chemistry that takes place in the coupled gas-aerosol-cloud system as it relates to aircraft observations.

Another important point to consider is the realization that one cannot address all relevant and important scientific questions with a limited set of tools available in most field and/or spaceborne projects. For example, some important questions are better addressed by conducting laboratory investigations under controlled and known ambient conditions. However, data collected from in-situ and remote sensing platforms can provide valuable information to help in the design of more focused future laboratory investigations. Thus it is helpful to sort out early on what scientific questions can or cannot be addressed by the HIAPER project in order to have realistic expectations of what can be accomplished from possible future planned field programs. Also, it is worth exploring how the HIAPER project can make contributions to advance laboratory and theoretical research in atmospheric sciences.

Often, when one examines the scientific agenda of a planned field project, it is common to find3 or 4 (sometimes even more) pages of questions that the project is hoping to address. While, some questions posed in the planned agenda are feasible, many are not, at least in my opinion. Posing too many questions, knowing the limitations in the scope of the project, will only dilute the importance of questions that the project can successfully. Thus I am not planing to present a long list of burning questions in the area of aerosol chemistry and microphysics. Instead, during my presentation, I will suggest a few areas of research where I believe thoughtful, well-planned and focused aircraft projects can make important contributions

In the area of aerosol and cloud heterogeneous reactivity, I will highlight why heterogeneous chemical reaction rates in the coupled gas-aerosol-cloud system may yield products at different rates than those determined in the laboratory. Some aircraft data collected during the SAFFARI campaign will be shown to suggest possible rapid in situ conversion of methanol to formaldehyde occurring in clouds and/or biomass burning aerosols. Data from limited observations during a number of GTE missions also support in-situ conversion of methanol to formaldehyde occurring in clouds. To better understand how oxygenated organic molecules interact with aerosol and cloud particles in the atmosphere is certainly an area where future aircraft observations can make a huge contribution. Oxygenated organic molecules can not only affect the chemistry in the gas phase, but their interactions with cloud and aerosol particles may also play a critical role in producing condensable organic mass in tropospheric aerosols. As discussed below the accumulation of organic mass in tropospheric aerosols can alter their physical, chemical and radiative properties.

Heterogeneous reactivity on cirrus also merits further investigation because cirrus cloud surface areas are vast and widespread in the upper troposphere. Some new laboratory investigations show that both organic molecules and nitric acid can absorb onto cirrus cloud surfaces under ambient conditions. Thus it is interesting to explore whether such molecules in an absorbed state can yield products faster than their corresponding gas phase counterparts.

In the area of cloud processing, it is helpful to conduct investigations to explore how clouds alter the vertical structure of soluble and/or reactive species in the atmosphere. Changes in the vertical profiles of reactive gases will directly impact the gas phase chemistry in air. For example, it is currently assumed that all soluble species are basically rained out when precipitation forms in wet convective systems. However, in convective cloud systems many ice

particles at higher levels originate from lower altitudes through direct conversion of supercooled water cloud droplets into ice particles. Such ice particles may contain a sizeable fraction of soluble species such as nitric acid and soluble organic molecules. Upon ice particle evaporation, containing trapped soluble species, a fresh source of reactive species can be released directly into the upper tropospher, affecting gas phase chemistry in this region.

I will also highlight the need for quantification of tropospheric aerosol chemical composition. Providing quantitative (not qualitative) information on a single particle composition is extremely useful to both theory and laboratory researchers. Thus far the in-situ information obtained on particle composition is somewhat qualitative and biased, i.e. chemical composition of only a narrow size range is being measured at the molecular level. In general qualitative data on a single particle is not sufficient for an aerosol modeler or laboratory investigator to explore how composition may affect the chemical (i.e. heterogeneous reactivity) and physical properties (i.e. ice nucleating potential) of an aerosol particle. If such information is known for aerosol particles of all sizes, then one can start to address important questions such as which aerosol particles (with what chemical compositions and sizes) in the size distribution are good cloud condensation nuclei (CCN), and which CCNs (out of many) are good ice nuclei (IN). I will also discuss how the same chemical composition in different size aerosols can affect their physical, chemical and radiative properties to advocate a need for particle composition data over the entire range of size distributions and not just a limited size range.

The question of what are good IN in the atmosphere is still an open question. In fact we know very little about how ice particles nucleate in the atmosphere. I will discuss why the in-situ data collected on ice nuclei to date is not adequate to address the basic question of which particles in the atmosphere can nucleate ice and which ones cannot. This question is inherently

important because one needs to know whether anthropogenic aerosols are of the former type or of the latter type. If manmade induced aerosols are of the former type, then they can greatly influence the rate of ice particle production in the atmosphere. Rate of ice formation influences important cloud properties such as reflectively, absorption and precipitation. I will discuss a possible way of aerosol sampling by aircraft, which may help to identify ice-nucleating aerosols in the atmosphere. I will also address how accumulation of organic matter in tropospheric aerosols over time can affect their microphysical properties (CCN activation and ice nucleation).

Finally, to address issues on CCN activation, ice nucleation, radiative effects, and heterogeneous reactivity in the atmosphere one needs to have accurate and quantitative information on the chemical composition (and its physical distribution) within individual aerosol particles. For example, regarding heterogeneous reactivity, small amounts of some molecules in aerosols may dominate the heterogeneous reactivity of the whole particle if such molecules have surfactant type properties (i.e. molecular nitric acid and/or organic molecules in water-based aerosols can be considered as soluble surfactants). In addition, the composition and the physical distribution of organic matter (also black carbon) will also directly affect the chemical (i.e. heterogeneous reactivity) physical, (i.e. ice nucleating potential) and radiative (i.e. absorption) properties of tropospheric aerosols.

Overall, I recommend four main interrelated areas of research where future aircraft observations can help in advancing science in the field of heterogeneous chemistry, aerosol and cloud research. First, the ability to measure oxygenated organic molecules in the gas phase both inside and outside cloudy regions. It is also advantages to simultaneously measure the concentration of soluble organic species in aerosol particles and/or cloud droplets. Second, targeted experiments to explore whether or not cirrus should be considered as a reactive chemical

surface in the upper troposphere. Third, the investigation of how cloud processing may alter the vertical structure of soluble and/or reactive species and their impact on gas phase chemistry. Fourth, is the need to quantify the chemical composition of tropospheric aerosols since this factor directly influences the chemical, physical and radiative properties of tropospheric aerosols.

Some Related Science Questions

1. Do oxygenated organic molecules react on aerosol and cloud surfaces? If so, what are the primary and secondary gas phase products of such reactions?
2. Do reactions of oxygenated organic molecules in aerosol and cloud particles contribute to accumulation of condensable organic mass in tropospheric aerosols?
3. What is the chemical composition of organic matter in tropospheric aerosols over the entire range of size distributions? Also, what is the physical distribution of organic matter and black carbon (soot) in atmospheric aerosols?
4. How do organic molecules in tropospheric aerosols affect their chemical and physical properties? Do organic molecules make aerosol and cloud particles chemically more reactive or less reactive? Do organic molecules affect the CCN and IN potential of troposphere aerosols? If so, in what direction?
5. How do clouds alter the vertical profiles of soluble reactive species in the atmosphere and what is the effect of such redistribution on gas phase chemistry?
6. Are cirrus cloud surfaces chemically reactive in the upper troposphere? If so, what reactions should be targeted for future field investigations? Can day- and nighttime sampling of cirrus shed light on the feasibility of photochemistry (i.e. photolysis of nitric acid and organic molecules) occurring in-situ on cirrus cloud surfaces? Can adsorbed

species on ice react with gas phase OH or other reactive species to yield products at a faster rate than their corresponding gas phase counterparts?