Outstanding Questions in Atmospheric Composition, Chemistry, Dynamics and Radiation for the Coming Decade

Proceedings of a Workshop held at NASA Ames Research Center in May 2014

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0.0 Executive Summary

This document is the product of a workshop organized by the Atmospheric Composition Focus Area of NASA Headquarters' Earth Science Division and held at the NASA Ames Research Center in May 2014. Seven experts in various research areas of interest to NASA were asked to present an overview of the state-of-the-art in their particular area with an emphasis on identifying outstanding questions for the coming decade. Discussion groups followed the overviews. Seven rapporteurs were asked to capture the high points of the discussions. The results of those discussions are summarized in the sections below.

Tropospheric gases [Section 2.1]

To address the questions connected with tropospheric composition and chemistry we must continue to measure numerous trace gas and aerosol species on a variety of scales from both satellite and suborbital platforms. Models must continue to play an important role in the analyses. Understanding changes to tropospheric composition requires a commitment to determining sources, fates and long-term trends of a number of species, including: O_3, CH_4, NOx, CO, PAN, NH_3, N_2O, HNO_3, VOCs, and radical oxidants. The connections between gaseous constituents and aerosols must be understood and measured. The effects of boundary layer dynamics, including convection, precipitation scavenging, and surface exchange on composition and chemistry, transport and lifetimes should be studied and understood. The impact of human activity and a changing climate on the atmospheric nitrogen cycle should be investigated. Critical questions include:

2.1.1.1 What are the major pollution sources in the developing world and how can we improve projections of future air quality and global composition?
2.1.1.4 How will changing patterns of energy-related and agricultural emissions affect global tropospheric composition?
2.1.2.1 What are the processes, source types and fluxes responsible for methane emissions and their trends?
2.1.3.1 How are water-soluble species transported and transformed in different types of deep convective systems?
2.1.3.3 How does convection produce chemically distinct layers in the troposphere, why do these layers persist, and what are the implications of these layers?
2.1.4.2 What is the full oxidation cascade of VOCs across NOx regimes and how can this be represented in models?
2.1.4.3 What is the fate of VOC oxidation products? What is the importance of wet and dry removal? How do we relate VOC emissions to the formation of organic aerosols?
2.1.5.1 Will methylchloroform continue as a reliable long-term record of global OH concentration? If not, what can serve as a substitute proxy?
2.1.5.2 What are the distributions and drivers of the most important radical oxidants (OH, NO_3, halogens) and their reservoirs (e.g., peroxides) over a spectrum of spatial and temporal scales?
2.1.7.1 How is tropospheric ozone changing globally and regionally, and what drives these long-term trends?

Stratospheric gases [Section 2.2]

To answer outstanding questions associated with stratospheric composition and chemistry we must address issues of composition change and its climatic impacts and feedbacks. This includes research to increase the understanding of 1) perturbations to the ozone layer driven by the decline in the abundance of ozone depleting substances (ODSs) and the rise in greenhouse gases (GHGs), as well as future trends of water vapor, 2) natural and climate change forced perturbations on exchange between stratosphere and troposphere and 3) the fundamentals that control the stratospheric aerosol layer and how it could change through volcanic or human intervention. Answering these questions requires measurement of an array of coupled tracers, chemically reactive intermediates, and radicals. These observations can be obtained by a synergistic deployment of orbital and sub-orbital measurement platforms, and ground-based remote sensing observations. Additionally, a suite of models of varying complexity are needed to fully understand measurements and trends. Critical questions include:

2.2.1.1 How will stratospheric H_2O and O_3 and associated chemical processes evolve in a climate with increased GHGs and changing ODSs?

2.2.1.2 How do stratospheric temperatures respond to changes in O_3 and the major GHGs? How do we quantify impacts of temperature changes on stratospheric chemistry, dynamics, and radiation?

2.2.2.2 What are the relative roles of various stratosphere-troposphere exchange (STE) mechanisms (Brewer-Dobson Circulation, convection, monsoon transport, isentropic transport across the subtropical jet, Pyro Cbs) in establishing the composition of the lower stratosphere. How will those roles change in an evolving climate, assuming the Coupled Model Intercomparison Project (CMIP) view of future climate?

2.2.3.1 What controls the basic evolution of aerosols in the Junge layer under non-volcanic background conditions or with only small volcanic input?

Clouds [Section 2.3]

The inadequate treatment of clouds in atmospheric models is a major obstacle to improved understanding. Six broad topical areas represent the foremost questions relating cloud processes to climate prediction. These questions are pertinent and remain unanswered largely because of uncertainties in our predictive skill that in turn are due to a scarcity of relevant observations. A number of concepts are common among the outstanding questions. These are: the role of microphysical processes, process rates, and the coupling of those processes to atmospheric motions; the role of aerosols in cloud processes; ice nucleation and collection processes; the spectrum of vertical motion, from the turbulent to the grid scale, is especially critical because of the role of vertical motions in vertically advecting condensate and/or in making water vapor available for condensation. Observations must be combined with modeling at the process level as an integral component to addressing each question. The critical questions include:

2.3.1 What is the sensitivity of the climate system to low-level cloud structure and variability over the middle and high latitude oceans?

2.3.2 How will the radiative balance over sea ice and permafrost change as the polar regions warm? What are the important feedback mechanisms between clouds, precipitation, and surface processes in the high latitudes?
2.3.3. How will shortwave cloud forcing change as the climate warms?
2.3.4. How will long-wave cloud forcing change as climate warms?
2.3.5 How do clouds respond to perturbations in aerosols Cloud Condensation Nuclei (CCN) and Ice Nuclei (IN)?
2.3.6 What is the role of (mixed phase) snow and rain in cloud processes and how is this modulated by aerosols and dynamics?

Aerosols [Section 2.4]

Aerosol science is an inherently interdisciplinary field, crossing the boundaries of most core earth science disciplines. Important feedbacks occur between cloud and aerosol life cycles, between atmospheric temperature and secondary aerosol formation, and between wind, surface moisture, and soil dust mobilization. To meet the demands of this interdisciplinary field, aerosol data has to evolve into a more integrated system of satellite data and network observations combined with models, and supplemented with targeted field campaigns. Aerosol particles are ubiquitous in the atmosphere and have strong relationships to such overarching climate themes as radiation, circulation, thermodynamics, hydrology, cryosphere, biosphere, bio-geochemical cycles, composition and chemical processes**.** Understanding aerosol impacts on climate is predicated on the quantification of the physical processes and short-term weather relationships within the aerosol system. Aerosol lifecycle (including sources, transformation, transport and scavenging/fate), especially in the troposphere happens fundamentally on short time scales and must be understood. Aerosols have a significant impact on the radiation budget, which affects boundary layer dynamics and chemistry, and thus the air quality, human health, biological productivity and visibility. Critical questions include:

2.4.1.1 How are natural and anthropogenic aerosols and their impact on the radiation budget at the top, within, and at the bottom of the atmosphere changing in response to a warming climate, and how to these changes feed back to the climate system?
2.4.1.2 How will projected changes in atmospheric circulation affect aerosols within the climate system through generation, transport, and deposition mechanisms and through their impact on biogeochemical cycles?
2.4.1.4 What regions are most susceptible to aerosol phenomena, how are different aerosol types affecting regional brightening or dimming and what impacts are regional pollution controls and emissions changes having?
2.4.1.5 What are the connections between forcing in one region and impact in another and what are the underlying transport mechanisms of different aerosol types?
2.4.2.1 What are the meteorological processes that control the distribution, transport, and deposition of aerosols?
2.4.2.2 Clouds as transformative and removal processes.
2.4.2.3 Vertical redistribution, PBL entrainment/detrainment

2.4.2.5 What are the important underlying mechanisms through which aerosols impact the meteorological cycle? What are the relative magnitudes of the dynamic, radiative, microphysical, and thermodynamic effects, and when and where are these important?
2.4.2.6 At what temporal and spatial scales and at what loadings do different aerosol types impact the meteorological cycle, especially through their activation as cloud droplets?
2.4.2.7 How do biogenic emissions regulate secondary aerosol formation and how is this process influenced by cloud processing?
2.4.2.14 What is the vertically resolved distribution of atmospheric heating due to the presence of different types of aerosols in clear sky, partly cloudy, and cloudy conditions?
2.4.2.15 How do aerosol changes affect the radiation budget through the diurnal cycle?
2.4.3.1 How do different aerosol types, stratified by size and composition, from natural and anthropogenic sources affect air quality in the PBL and surface layer?

Atmospheric radiation [Section 3.0]

Outstanding questions in the area of atmospheric radiation stand on their own, but also are clearly connected to cloud, aerosol, convection, atmospheric circulation, and atmosphere-surface exchange questions. While radiation science questions tend to be large time and space scale questions related to climate change, the related cloud and aerosol questions contain an additional focus on smaller time/space scale processes. Solving climate change requires both of these perspectives. Process studies are critical to improving physical processes in climate models, while long time/space scale observations are key to testing the ability of climate models to accurately predict climate change. The radiation science questions contain some common themes. The most obvious of these is the need for higher accuracy in a wide range of satellite observations: top of the atmosphere (TOA) and surface radiative fluxes, passive and active cloud properties, passive and active aerosol properties, total and spectral solar irradiance, precipitation, as well as in-situ ocean heat storage observations and boundary layer temperature/humidity profiles. New observations of far-infrared spectra also are key to certain questions. Finally, the potential use of systematic aircraft flights to achieve sufficient statistical sampling of cloud and radiation processes is a common theme. Critical questions include:

3.1.1.1 What are the cloud and radiation properties, with sufficient interannual and decadal climate change accuracy, necessary to reduce cloud feedback uncertainty by at least a factor of 2?
3.1.2.1 What observations and modeling are required to reduce uncertainty in anthropogenic aerosol by at least a factor of 2 relative to the AR5 IPCC estimate?
3.1.3.1 What is the annual net radiation (1σ) and uncertainty in interannual variations to 0.1 Wm^{-2} (1σ)?
3.1.4.1 What observations are required to close the surface and atmosphere net energy budget to within less than 5 Wm^{-2} ?
3.1.5.1 Do clouds in the arctic increase or reduce the rapid warming there? Determine the arctic cloud feedback to within 25% of the sea ice extent driven surface albedo feedback.
3.1.6.1 What is the far-infrared absorption spectrum at the spectral resolution, coverage, and accuracy sufficient for verification of the water vapor greenhouse effect and water vapor feedback in climate models?

Circulation [Section 4.1]

Outstanding questions related to the circulation of the atmosphere breakdown into a number of specific but connected questions.

The Brewer-Dobson circulation (BDC) is critical to understanding the evolution of stratospheric ozone as well as gases relevant to the overall climate, such as water vapor. The fundamental theoretical framework for understanding the BDC is well-established, but important details, including the role of unresolved gravity waves, are not well-quantified.

Long-term stratospheric temperature trends will occur most prominently in the upper stratosphere as a result of GHG changes. The issue is crucially important because of its direct impact on the photochemistry of ozone in the stratosphere. As CO_2 increases, stratospheric temperature decreases, and ozone increases.

There are significant uncertainties in quantifying convective influences on the lower stratosphere. Entrainment and detrainment throughout the full vertical profile is likely important for maritime convection but is poorly understood.

Monsoon circulations are a fundamental aspect of the large-scale circulation in the tropics and subtropics. Monsoons exhibit a high degree of dynamic variability, reflected in constituent behavior that is poorly understood.

Questions related to interhemispheric and extra-tropical to tropical transport are directly related to composition questions through atmospheric oxidation chemistry (lifetime) and vertical transport into the stratosphere, both of which are dominated by processes in the tropics.

Gravity waves are ubiquitous in the atmosphere and can propagate in any stably stratified region, thus almost everywhere. While the most prominent effects of GWs are their influences on the mean winds and overturning circulations, especially in the middle atmosphere, GWs also produce rapidly fluctuating vertical velocities and temperatures.

Critical questions include:

4.1.1 To what degree is the Brewer-Dobson stratospheric circulation accelerating?
4.1.2 How is the mid-to-upper stratosphere temperature responding to GHG increases?
4.1.3 What are the transport pathways from the PBL to the stratosphere?
4.1.4 How does deep convection contribute to troposphere-stratosphere coupling?
4.1.5 How do monsoon circulations contribute to troposphere-stratosphere coupling?
4.1.6 What are the processes controlling atmospheric transport between the tropics and extra-tropics and between hemispheres, and how are they changing?
4.1.7 What role do gravity waves (GWs) play in driving the large-scale circulation?

Convection [Section 4.2]

Atmospheric convection exists in a wide variety of forms and in a broad range of scales. Each form has its unique characteristics and circulation features. It is rarely in a steady state, so that observations are needed during the growing, mature, and dissipating states of the convection, it is highly turbulent so that observations are needed on the sub-convective scale, and both warm and

cold-cloud microphysics require data down to the micron scale. A full understanding of convection or any of the roles it plays in the atmosphere requires attention to the specific form taken by the convection as well as all of its internal properties. The interactions of convection with the wide variety of aerosol and important trace constituents of the atmosphere are among the factors that need to be specified in reference to the particular form of convection. Outstanding questions are generally related to the situation that: basic in-cloud properties are poorly known; the relationship of deep convective clouds and mesoscale convective systems to the humidity field is very poorly understood; why and how mesoscale systems evolve is still under study; understanding of multiscale diurnal behaviors related to underlying surface conditions has not been achieved; aerosol environment effects on convection are not understood; the role of cold pools is a focus in efforts to understand how convective populations develop; and one of the primary inhibitors in understanding how convective processes vary around the globe is the lack of time resolution in observations from space. Critical questions/tasks include:

4.2.2.1.1 Quantify how different forms of convection affect lightning-generated NOx production and subsequent ozone production, namely, ordinary thunderstorms, supercell convection, and mesoscale convective systems.

4.2.2.2.1 Processes need to be examined on various timescales, different latitudes, and between land and ocean.

4.2.2.2.2 Collect measurements of chemical species in cloud top regions.

4.2.2.3.1 Address aerosol effects on convection from space platforms in order to capture the global variability.

4.2.2.3.2 Quantify the transformation of all types of aerosols (dust, black carbon, sulfate, and nitrate) in the context of different forms, scales, and strengths of convection.

4.2.2.3.3 Enhance ground-based networks to sample the full vertical profile of aerosol through both the PBL and free atmosphere.

4.2.2.3.5 Create a model that treats processes in a unified and consistent manner, e.g. cloud particle microphysics and transformation.

1.0 Introduction

This document is the product of a workshop organized by the Atmospheric Composition Focus Area of NASA Headquarters' Earth Science Division and held at the NASA Ames Research Center in May 2014. Seven experts in various research areas of interest to NASA were asked to present an overview of the state-of-the-art in their particular area with an emphasis on identifying outstanding questions for the coming decade. Discussion groups followed the overviews. Seven rapporteurs were asked to capture the high points of the discussions. The surveyors and rapporteurs, and their topics were:

Topic	Surveyor	Rapporteur
Tropospheric composition	D. Jacob Harvard University	E. Fischer Colorado State University
Stratospheric composition	R. Gao NOAA	K. Rosenlof NOAA
Clouds	J. Mace University of Utah	S. Massie NCAR
Aerosols	J. Reid NRL	C. Dutcher University of Minnesota
Radiation	B. Wielicki NASA LaRC	P. Zuidema University of Miami
Circulation	P. Newman NASA GSFC	W. Robinson N. Carolina State University
Convection	R. Houze University of Washington	M. Barth NCAR

A list of attendees at the workshop is contained in Appendix A. Discussions were specifically guided away from identifying particular instrument developments or favorite field programs or missions. Rather, there was an emphasis on identifying specific observations that could be made to help answer outstanding questions. Reports from the seven groups were collected and revised to a uniform format.

Critical, Very Important and Important questions are identified in each section.

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2.0 Atmospheric Composition and Chemistry

2.1 Tropospheric gases

This section describes the priority questions related to tropospheric gases. To address the questions we must continue to measure numerous trace gas and aerosol species on a variety of scales from both satellite and suborbital platforms. Models will continue to play an important role in the analyses. Understanding changes to tropospheric composition requires a commitment to long-term measurements, and to measurements that aid in the interpretation of target species.

2.1.1 What is the future of air quality in a changing world?

The developing world is experiencing serious and worsening air quality problems as rapid industrialization takes place in combination with high population density and limited emission controls [*Cohen et al.*, 2005]. China and India have tremendous air quality problems and other countries in Southeast Asia and Africa (such as Nigeria and South Africa) are following suit [*Kan et al.*, 2009; *Tawari and Abowei*, 2012; *Zhu et al.,* 2012; *Liousse et al.,* 2014]. Smoke from biomass burning in the tropics is also becoming a major public health issue as it affects population centers [*Pavagadhi et al.*, 2013]. Relevant observations include near surface and profiles of particulate matter and O_3 at very high spatial and temporal resolution, and measurements of NO_x, CO, and VOC oxidation products. Priority research questions include:

2.1.1.1 What are the major pollution sources in the developing world and how can we improve projections of future air quality and global composition **(Critical)**?

2.1.1.2 Can we develop a space-based observing system for global air quality with spatial and temporal resolution relevant for public exposure (PM and O_3) that can be used to issue air quality warnings **(Very Important)** ?

2.1.1.3 How do extreme air quality events form and evolve in understudied regions and how do they affect global composition **(Important)** ?

2.1.1.4 How will changing patterns of energy-related and agricultural emissions affect global tropospheric composition **(Critical)** ?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.1.1	Satellite and in situ observations of AOD, AAOD, tropospheric ozone, CO, NO_2, HCHO, SO_2, CHOCHO,NH_3	Hourly 1x1 km^2 (aerosol), 4x4 km^2 (gases)	See specifications for GEO-CAPE geostationary mission [Fishman et al., 2012]	
2.1.1.2	Ibid.	Ibid.	Ibid.	
2.1.1.3	Ibid.	Ibid.	Ibid.	
2.1.1.4	Ibid.	Ibid.	Ibid.	

2.1.2 How are the sources of methane changing?

Radiative forcing by methane emissions since pre-industrial times amounts to more than 50% of that by CO_2 [*IPCC*, 2013]. Methane is an important driver of tropospheric ozone, a pollutant and GHG, and tropospheric OH, the primary atmospheric oxidant. Methane is a near-term climate forcer that is as important as CO_2 on a 20-year horizon and is thus a critical component of climate policy. However, the sources responsible for the methane trends over the past decades are poorly understood [*Kirschke et al.*,2013]. Satellite observations of methane combined with in situ observations of fluxes can make a unique contribution for quantifying anthropogenic emissions (fossil fuel extraction and use, agriculture, landfills) and natural emissions (in particular wetlands. Better validated space-based methane measurements could constrain top-down approaches to deriving emissions. Satellite observations of CH_4 will need support from sub-orbital measurements, including co-emitted species (e.g. hydrocarbon ratios, and agricultural tracers) and isotopic methane. Priority research questions include:

2.1.2.1 What are the processes, source types and fluxes responsible for methane emissions and their trends **(Critical)** ?

2.1.2.2 How do we integrate top-down and bottom-up approaches towards a better understanding of the processes controlling methane sources **(Very Important)**?

2.1.2.3 What role will methyl hydrates play as sources of methane in the future **(Important)**?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.2.1	Geostationary and low-Earth orbit observations of CH_4	Hourly (geo), daily (LEO), 5x5 km^2	0.5%	0.5%
2.1.2.2	Ibid + in situ field campaigns including isotopic information	1-min, landscape	0.5%	0.5%
2.1.2.3	Ibid + in situ field campaigns including isotopic information	1-min, landscape	0.5%	0.5%

2.1.3 How do convection and precipitation transform and redistribute trace species?

Convection plays a dominant role in the vertical redistribution of chemicals in the troposphere with implications for radiation, long-range transport, and chemical aging *[Pickering et al.,* 1996*; Apel et al.*, 2012; *Bertram et al.*, 2007]. Transformation of gases and aerosols during convective transport from the boundary layer is critical in understanding the free troposphere and global atmospheric composition [*Pickering et al.,* 2001; *Tost et al.*, 2010]. The research community needs a combination of remote sensing, ground-based lidar, sondes and in

situ (aircraft) observations integrating cloud dynamics, microphysics, and chemical properties. Priority research questions include:

2.1.3.1 How are water-soluble species transported and transformed in different types of deep convective systems **(Critical)**?

2.1.3.2 How do different precipitation forms transform and redistribute water-soluble species **(Important)**?

2.1.3.3 How does convection produce chemically distinct layers in the troposphere, why do these layers persist, and what are the implications of these layers **(Critical)** ?

2.1.3.4 How do pollution and deep convection interact to deliver pollutants to the upper troposphere **(Very Important)**?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.3.1	Multi-aircraft campaigns focused on sampling deep convective events and including extensive payloads for gases, aerosol, cloud microphysics	Cloud scale	See specifications for SEAC^{4}RS campaign; add NH_3 to payload (accuracy and precision 100 pptv)	
2.1.3.2	Ibid. but for different kinds of precipitation systems including shallow convection and frontal lifting	Cloud scale	Ibid	ibid
2.1.3.3	Satellite and aircraft observations of persistent layers – geostationary and LEO lidar aerosol observations, aircraft campaigns	Hourly on synoptic to global scale, 1 km	0.05 AOD for lidar, other specs as above	0.05 AOD for lidar, other specs as above
2.1.3.4	Same as 2.1.3.1	Same as 2.1.3.1	Same as 2.1.3.1	Same as 2.1.3.1

2.1.4 How do biogenic, anthropogenic and pyrogenic organics affect oxidants and aerosols?

The terrestrial biosphere is the largest source of volatile organic compounds (VOCs) to the atmosphere. Anthropogenic and pyrogenic VOCs are also important in urban and biomass burning regions respectively. The atmospheric chemistry of these VOCs has important implications for oxidants and aerosol formation but is very poorly understood, particularly in the low-NO_x regime prevalent in the tropics (and, increasingly, in North America) [*Paulot et al.*,

2012]. Observations over the remote oceans indicate that the oceans may also play a major role in controlling atmospheric abundances of oxygenated VOCs [*Carpenter et al.*, 2012], with possibly important implications for global atmospheric chemistry. We also understand little about the lifecycles and importance of marine organic aerosol [*Gantt and Meskhidze*, 2013]. Answering the questions below will require combining in situ measurements and satellite observations of VOCs, their oxidation productions and land surface properties (e.g. Leaf Area Index, LAI and Net Primary Productivity, NPP). Priority research questions include:

2.1.4.1 What are VOC emissions and their trends on land and over the oceans **(Very Important)**?

2.1.4.2 What is the full oxidation cascade of VOCs across NO_x regimes and how can this be represented in models **(Critical)** ?

2.1.4.3 What is the fate of VOC oxidation products? What is the importance of wet and dry removal? How do we relate VOC emissions to the formation of organic aerosols **(Critical)** ?

2.1.4.4 How will biogenic and biomass burning VOC emissions respond to changes in climate, land use, and atmospheric composition **(Important)**?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.4.1	Geostationary and LEO observations of HCHO, CHOCHO, CH_3OH, HCOOH, together with surface properties	Hourly (geo), daily (LEO), 4x4 km^2	$1x10^{15}$ molecules cm^{-2}	$1x10^{15}$ molecules cm^{-2}
2.1.4.2	Laboratory studies of VOC oxidation with particular focus on low-NO_x regime	N/A	N/A	N/A
2.1.4.3	Aircraft campaigns with extensive gas an aerosol payload and focus on tropics	1-minute, 1-km	SEAC^{4}RS payload requirements	
2.1.4.4	Same as 2.1.4.1	Same as 2.1.4.1	Same as	Same as 2.1.4.1

2.1.5 What controls the concentrations of radical oxidants?

Inferences of the global OH concentration from measurements of the methylchloroform proxy have played a critical role in the past 30 years to constrain the atmospheric lifetime of methane and other gases such as HCFCs and HFCs. A recent analysis by *Montzka et al.* [2011] shows no trend in global OH for the past 30 years and an interannual variability (IAV) of only 2%, consistent with models though not necessarily for the right reasons. The models show strong potential for future increases or decreases in OH concentrations due to changes in emissions and

climate [*Voulgarakis et al.*, 2013]. However, the continued value of the methylchloroform proxy is called into question as its concentrations decay. At a process level we need to better understand the factors controlling local OH concentrations, particularly the dependence on NO_x and VOCs, but are presently limited by confidence in OH measurements. A research priority is to improve, intercompare and evaluate (via related species) in situ measurements of OH concentrations. We also need better measurements and understanding of halogen radical oxidants (ClO_x, BrO_x, IO_x) and their implications for the budgets of O_3, NO_x, and mercury in the different tropospheric domains where halogens play an important role [*Saiz-Lopez and von Glasow*, 2012]. Priority research questions include:

2.1.5.1 What atmospheric constituents can effectively serve as substitute proxies for global, regional, and vertical distributions of OH concentrations? **(Critical)** ?

2.1.5.2 What are the distributions and drivers of the most important radical oxidants (OH, NO_3, halogens) and their reservoirs (e.g., peroxides) over a spectrum of spatial and temporal scales **(Critical)** ?

2.1.5.3 What are the mechanisms that determine lightning NO_x emissions? How are they coupled to deep convective transport and outflow chemistry **(Very Important)**?

2.1.5.4 What is the role of halogen radicals in the budgets of tropospheric O_3, NO_x, and mercury **(Very Important)**?

2.1.5.5 What is the role of nighttime radical chemistry with respect to sulfur, nitrogen, halogen and aerosol chemical cycles [*Brown and Stutz*, 2012] **(Important)**?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.5.1	Surface measurement of global OH proxy	Global	1%	1%
2.1.5.2	Aircraft measurements of OH, HO_x and reservoirs, halogen radicals and reservoirs, NO_3	1-min, 1-km	10%	10%
2.1.5.3	Focused aircraft campaign including measurements of NO_x and reservoirs, together with surface measurements of lightning flashes and energies	Cloud scale	10%	10%
2.1.5.4	Improved satellite retrievals of tropospheric halogen radicals, aircraft campaigns with payloads including halogen radicals and reservoirs, HO_x, NO_x and reservoirs, Hg(0)/Hg(II)	Synoptic to global scales	10%	10%

2.1.5.5	Aircraft campaign focused on nighttime radical chemistry including measurements of NO_3, N_2O_5, other NO_y species, ozone, VOCs, organic nitrates, halogen radicals and their reservoirs, DMS	1-min, 1-km	10%	10%

2.1.6 How are humans and climate perturbing the nitrogen cycle?

Global fixation of nitrogen by human activity, principally for food production and with also some contribution from fossil fuel combustion, now greatly dominates over natural fixation. The resulting emissions of NH_3 and NO_x have important implications for aerosols and oxidants. Atmospheric transport delivers this nitrogen to remote ecosystems through deposition, resulting in inadvertent fertilization that affects ecosystem function and carbon uptake. Improving our understanding of these processes will require taking advantage of NH_3 observations from space, and integrating bottom-up and top-down approaches to constrain emissions [*Paulot et al.*, 2014; *Van Damme et al.*, 2014].
We also need observations of the broadest suite possible of oxidized nitrogen species (NO_x, PAN, N_2O, HNO_3, etc) over land and ocean through in situ measurements and satellites where possible. Priority research questions include:

2.1.6.1 What are typical vertical distribution of NH_3, what controls these, and what is the role/response of gas-aerosol partitioning **(Important)** ?

2.1.6.2 What is the quantity and seasonality of ammonia emissions over local to global scales **(Very Important)** ?

2.1.6.3 How is nitrogen deposited and cycled through the terrestrial and marine ecosystems, and re-emitted, resulting in a "grasshopper effect" **(Very Important)**?

2.1.6.4 How are organic nitrates formed, what is their fate, how do they contribute to nitrogen deposition and what is their impact on O_3 and secondary organic aerosol [*Perring et al.,* 2013] **(Important)** ?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.6.1	NH_3, aerosol composition	Vertical profiling with 0.1 km	10%	10%
2.1.6.2	Geostationary and LEO satellite observations validated with aircraft campaigns	Hourly (geo), daily (LEO), 1-min (aircraft), 4x4 km^2	10%	10%

2.1.6.3	Field studies of N biogeochemistry in different terrestrial ecosystems and including atmospheric deposition and emission measurements,	Hourly, landscape-scale	10%	10%
2.1.6.4	Aircraft campaign with SEAC^{4}RS-type payload in the tropics	1-min, landscape to synoptic scale	10%	10%

2.1.7 What drives long-term trends in tropospheric ozone?

Observations generally show a steady increase of baseline O_3 in the remote troposphere over the past century, particularly at northern mid-latitudes; although long-term records for Europe show that ozone has either leveled off or is decreasing since 2000 [*Cooper et al.,* 2014]. Observed increases are larger than simulated in models [*Parrish et al.*, 2014], suggesting that Intergovernmental Panel on Climate Change (IPCC) estimates of radiative forcing by tropospheric O_3 may be too low and calling into question the ability of models to capture the sensitivity of O_3 production to future changes in emissions. Changes in the O_3 baseline have increasing implications for surface air quality as current air quality standards to protect public health are being ratcheted down. Thus O_3 pollution is transitioning from an urban/regional problem to a global problem [*NRC*, 2010; *UNEP*, 2011], and better understanding of the linkage of air quality and climate is required. Understanding these changes will require continuation and interpretation (models) of satellite, sonde, lidar and in situ records of O_3 and its precursors. The development of capabilities for multi-constituent chemical data assimilation to integrate all available observations of tropospheric O_3 and related species may benefit these research questions. A priority research questions is:

2.1.7.1 How is tropospheric ozone changing globally and regionally, and what drives these long-term trends? **(Critical)** ?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.7.1	Development of a global multi-constituent data assimilation system and the measurements necessary to produce a meaningful tropospheric ozone product	Global, multi-decadal	5 ppb (ozone)	5 ppb (ozone)

2.1.8 How do boundary layer dynamics, surface exchange and chemistry interact to determine the vertical distribution and evolution of trace species?

Boundary layer mixing and venting is critical to understanding the impact of emissions on air quality and global atmospheric composition. Observations show boundary layer structure that is far more complex than is commonly represented in models, and this may have important consequences for chemical evolution and transport. Cloud cycling in the boundary layer may be a major driver for chemical and aerosol evolution through aqueous-phase processes. Better understanding of vertical gradients in the boundary layer is essential for improving UV and thermal infrared (TIR) retrievals of atmospheric composition from space, and for relating satellite measurements of tropospheric columns to surface concentrations. Thus a key task is to characterize lower atmosphere vertical structure for the benefit of satellite retrievals and for relating satellite retrievals to surface processes. The scale of this problem points toward more ground based and in situ measurements. Priority research questions include:

2.1.8.1 How does the coupling of chemical and dynamical timescales determine the vertical structure of trace species in the lower troposphere **(Very Important)**?

2.1.8.2 What is the role of boundary layer cloud processes for chemical and aerosol evolution, and for venting to the free troposphere **(Very Important)**?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.8.1	Ground-based lidar observations and in situ profiles of chemical and dynamical variables	1-min, 100-m (vertical)	10%	10%
2.1.8.2	Observations of chemical, cloud, and dynamical properties from aircraft	1-min, 100-m (vertical)	10%	10%

2.1.9 What controls background ozone concentrations in surface air?

Surface air observations in high-elevation areas show elevated O_3 background concentrations that are problematic for meeting air quality standards. Understanding the factors determining this elevated background and its variability is essential for air quality policy. The related processes are not well understood [*Lin et al.*, 2012a; *Lin et al.*, 2012b]. Satellites have a critical role to play in observing the long-range transport of O_3 in the free troposphere and its implications for surface air. Ozone profiles with high vertical resolution and correlative observations (such as CO and PAN) and information on lightning strikes and fire activity may be of high utility. Priority research questions include:

2.1.9.1 What is the variability of background surface ozone in response to biogenic emissions, air mass history, meteorology, stratospheric intrusions, deep convection, lightning, and fires **(Very Important)**?

2.1.9.2 Can we develop a space-based capability for diagnosing and forecasting high-ozone background events **(Important)**?

Question	Observation	Resolution time and space	Accuracy	Precision
2.1.9.1	Geostationary observation of O_3 and CO with sensitivity to boundary layer Ground based and in situ observations of ozone profile	Hourly 4x4 km^2 Vertical resolution: (lidar) ~700m (sonde) ~100m	10 ppb O_3, 20ppb CO	10 ppb O_3, 20ppb CO
2.1.9.2	Ozone data assimilation system using geostationary data	Hourly 4x4 km^2	10 ppb	10 ppb

References (2.1):

Apel, E. C., J. R. Olson, J. H. Crawford, R. S. Hornbrook, A. J. Hills, C. A. Cantrell, L. K. Emmons, D. J. Knapp, S. Hall, R. L. Mauldin Iii, A. J. Weinheimer, A. Fried, D. R. Blake, J. D. Crounse, J. M. S. Clair, P. O. Wennberg, G. S. Diskin, H. E. Fuelberg, A. Wisthaler, T. Mikoviny, W. Brune, and D. D. Riemer (2012), Impact of the deep convection of isoprene and other reactive trace species on radicals and ozone in the upper troposphere, *Atmos. Chem. Phys.*, *12*(2), 1135-1150,10.5194/acp-12-1135-2012.

Bertram, T. H., A. E. Perring, P. J. Wooldridge, J. D. Crounse, A. J. Kwan, P. O. Wennberg, E. Scheuer, J. Dibb, M. Avery, G. Sachse, S. A. Vay, J. H. Crawford, C. S. McNaughton, A. Clarke, K. E. Pickering, H. Fuelberg, G. Huey, D. R. Blake, H. B. Singh, S. R. Hall, R. E. Shetter, A. Fried, B. G. Heikes, and R. C. Cohen (2007), Direct Measurements of the Convective Recycling of the Upper Troposphere, *Science*, *315*(5813), 816-820.

Brown, S. S., and J. Stutz (2012), Nighttime radical observations and chemistry, *Chemical Society Reviews*, *41*(19), 6405-6447,10.1039/C2CS35181A.

Carpenter, L. J., S. D. Archer, and R. Beale (2012), Ocean-atmosphere trace gas exchange, *Chemical Society Reviews*, *41*(19), 6473-6506,10.1039/C2CS35121H.

Cohen, A. J., H. Ross Anderson, B. Ostro, K. D. Pandey, M. Krzyzanowski, N. Künzli, K. Gutschmidt, A. Pope, I. Romieu, J. M. Samet, and K. Smith (2005), The Global Burden of Disease Due to Outdoor Air Pollution, *Journal of Toxicology and Environmental Health, Part A*, *68*(13-14), 1301-1307,10.1080/15287390590936166.

Cooper, O. R., D. D. Parrish, J. Ziemke, N. V. Balashov, M. Cupeiro, I. E. Galbally, S. Gilge, L. Horowitz, N. R. Jensen, J.-F. Lamarque, V. Naik, S. J. Oltmans, J. Schwab, D. T. Shindell, A. M. Thompson, V. Thouret, Y. Wang, R. M. Zbinden (2014), Global distribution and trends of tropospheric ozone: An observation-based review, *Elementa: Science of the Anthropocene, 2*, 000029, doi: 10.12952/journal.elementa.000029

Fishman, J., L. et al., The United States' next generation of atmospheric composition and coastal ecosystem measurements: NASA's Geostationary Coastal and Air Pollution Events (GEO-CAPE) Mission, *BAMS,* doi: 10.1175/BAMS-D-11-00201.1, 2012.

Gantt, B., and N. Meskhidze (2013), The physical and chemical characteristics of marine primary organic aerosol: a review, *Atmos. Chem. Phys.*, *13*(8), 3979-3996,10.5194/acp-13-3979- 2013.

IPCC (2013), Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, 1535 pp, Cambridge University Press, Cambridge, U. K. and New York, NY, USA.

Kan, H., B. Chen, and C. Hong (2009), Health impact of outdoor air pollution in China: Current knowledge and future research needs, *Environmental Health Perspectives*, *117*(5), A187.

Kirschke, S., P. Bousquet, P. Ciais, M. Saunois, J. G. Canadell, E. J. Dlugokencky, P. Bergamaschi, D. Bergmann, D. R. Blake, L. Bruhwiler, P. Cameron-Smith, S. Castaldi, F. Chevallier, L. Feng, A. Fraser, M. Heimann, E. L. Hodson, S. Houweling, B. Josse, P. J. Fraser, P. B. Krummel, J.-F. Lamarque, R. L. Langenfelds, C. Le Quere, V. Naik, S. O'Doherty, P. I. Palmer, I. Pison, D. Plummer, B. Poulter, R. G. Prinn, M. Rigby, B. Ringeval, M. Santini, M. Schmidt, D. T. Shindell, I. J. Simpson, R. Spahni, L. P. Steele, S. A. Strode, K. Sudo, S. Szopa, G. R. van der Werf, A. Voulgarakis, M. van Weele, R. F. Weiss, J. E. Williams, and G. Zeng (2013), Three decades of global methane sources and sinks, *Nature Geosci*, *6*(10), 813-823,10.1038/ngeo1955 http://www.nature.com/ngeo/journal/v6/n10/abs/ngeo1955.html

Lin, M., A. M. Fiore, O. R. Cooper, L. W. Horowitz, A. O. Langford, H. Levy, B. J. Johnson, V. Naik, S. J. Oltmans, and C. J. Senff (2012a), Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions, *Journal of Geophysical Research: Atmospheres*, *117*(D21), D00V22,10.1029/2012JD018151.

Lin, M., A. M. Fiore, L. W. Horowitz, O. R. Cooper, V. Naik, J. Holloway, B. J. Johnson, A. M. Middlebrook, S. J. Oltmans, I. B. Pollack, T. B. Ryerson, J. X. Warner, C. Wiedinmyer, J. Wilson, and B. Wyman (2012b), Transport of Asian ozone pollution into surface air over the western United States in spring, *Journal of Geophysical Research: Atmospheres*, *117*(D21), D00V07,10.1029/2011JD016961.

Liousse, C., E. Assamoi, P. Criqui, C. Granier and R. Rosset, Explosive growth in African combustion emissions from 2005 to 2030, *Environ. Res. Lett*., 9, doi:10.1088/1748-9326/9/3/035003,2014.

Montzka, S. A., M. Krol, E. Dlugokencky, B. Hall, P. Jöckel, and J. Lelieveld (2011), Small Interannual Variability of Global Atmospheric Hydroxyl, *Science*, *331*(6013), 67-69.

NRC (2010), Global Sources of Local Pollution: An Assessment of Long-Range Transport of Key Air Pollutants to and from the United States, National Academies, Washington, DC.

Parrish, D. D., J. F. Lamarque, V. Naik, L. Horowitz, D. T. Shindell, J. Staehelin, R. Derwent, O. R. Cooper, H. Tanimoto, A. Volz-Thomas, S. Gilge, H. E. Scheel, M. Steinbacher, and M. Fröhlich (2014), Long-term changes in lower tropospheric baseline ozone concentrations: Comparing chemistry-climate models and observations at northern midlatitudes, *Journal of Geophysical Research: Atmospheres*, 2013JD021435,10.1002/2013JD021435.

Paulot, F., D. K. Henze, and P. O. Wennberg (2012), Impact of the isoprene photochemical cascade on tropical ozone, *Atmos. Chem. Phys.*, *12*(3), 1307-1325,10.5194/acp-12-1307-2012.

Paulot, F., D. J. Jacob, R. W. Pinder, J. O. Bash, K. Travis, and D. K. Henze (2014), Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural

emissions inventory (MASAGE_NH3), *Journal of Geophysical Research: Atmospheres*, *119*(7), 4343-4364,10.1002/2013JD021130.

Pavagadhi, S., R. Betha, S. Venkatesan, R. Balasubramanian, and M. Hande (2013), Physicochemical and toxicological characteristics of urban aerosols during a recent Indonesian biomass burning episode, *Environmental Science and Pollution Research*, *20*(4), 2569-2578,10.1007/s11356-012-1157-9.

Perring, A. E., S. E. Pusede, and R. C. Cohen (2013), An Observational Perspective on the Atmospheric Impacts of Alkyl and Multifunctional Nitrates on Ozone and Secondary Organic Aerosol, *Chemical Reviews*, *113*(8), 5848-5870,10.1021/cr300520x.

Pickering, K.E., et al. (1996) Convective transport of biomass burning emissions over Brazil during TRACE-A, *J. Geophys. Res*. 101, 23,993-24012.

Pickering, K.E., et al. (2001) Trace gas transport and scavenging in PEM-Tropics B South Pacific Convergence Zone convection, *J. Geophys. Res*. 106, 32,591-32,602.

Saiz‐Lopez, A., and R. von Glasow (2012), Reactive halogen chemistry in the troposphere, *Chemical Society Reviews*, *41*(19), 6448‐6472,10.1039/C2CS35208G.

Tawari, C. C., and J. F. N. Abowei (2012), Air Pollution in the Niger Delta Area of Nigeria, *International Journal of Fisheries and Aquatic Sciences*, *1*(2), 94-117.

Tost, H., M. G. Lawrence, C. Brühl, P. Jöckel, G. T. The, and S.-O. D. A. T. The (2010), Uncertainties in atmospheric chemistry modelling due to convection parameterisations and subsequent scavenging, *Atmos. Chem. Phys.*, *10*(4), 1931-1951,10.5194/acp-10-1931-2010.

United Nations Environment Program (2011), Near–term Climate Protection and Air Quality Benefits: Actions for controlling Short-lived Climate Forcers, UNEP, Nairobi, Kenya.

Van Damme, M., L. Clarisse, C. L. Heald, D. Hurtmans, Y. Ngadi, C. Clerbaux, A. J. Dolman, J. W. Erisman, and P. F. Coheur (2014), Global distributions, time series and error characterization of atmospheric ammonia (NH3) from IASI satellite observations, *Atmos. Chem. Phys.*, *14*(6), 2905-2922,10.5194/acp-14-2905-2014.

Voulgarakis, A., V. Naik, J. F. Lamarque, D. T. Shindell, P. J. Young, M. J. Prather, O. Wild, R. D. Field, D. Bergmann, P. Cameron-Smith, I. Cionni, W. J. Collins, S. B. Dalsøren, R. M. Doherty, V. Eyring, G. Faluvegi, G. A. Folberth, L. W. Horowitz, B. Josse, I. A. MacKenzie, T. Nagashima, D. A. Plummer, M. Righi, S. T. Rumbold, D. S. Stevenson, S. A. Strode, K. Sudo, S. Szopa, and G. Zeng (2013), Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, *Atmos. Chem. Phys.*, *13*(5), 2563-2587,10.5194/acp-13-2563-2013.

Zhu, T., M. L. Melamed, D. Parrish, M. Gauss, L. Gallardo Klenner, M. Lawrence, A. Konare and C. Liousse, WMO/IGAC Impacts of Megacities on Air Pollution and Climate, WMO/GAW Report No. 205, 2012.

2.2 Stratospheric gases

The Stratospheric Composition (hereafter SC) community has a fundamental responsibility to address issues of stratospheric composition change and its climatic impacts and feedbacks. This includes research to increase the understanding of 1) perturbations to the ozone layer driven by the decline in the abundance of ozone depleting substances (ODSs) and the rise in greenhouse gases (GHGs), as well as future trends of water vapor, 2) natural and climate change forced perturbations on exchange between stratosphere and troposphere and 3) the fundamentals that control the stratospheric aerosol layer and how it could change through volcanic or human intervention. These issues require a deeper understanding than we currently have of feedbacks between stratospheric processes and tropospheric climate. Variations in ODSs and GHGs alter stratospheric ozone, which in turn affect surface climate. The variability of stratospheric ozone is difficult to accurately forecast, as exemplified by the unusually large amount of Arctic ozone depletion experienced during winter 2011. These issues have a direct bearing on public health, given that changes in stratospheric ozone will alter UV exposure at the surface, affecting large parts of the earth's ecosystem. A 5% decrease in column ozone in the summer over the US translates into an increase of approximately 350,000 skin cancer cases a year. This quantitatively defines the degree to which we must understand the coupling between forcing of the climate and changes to the column concentration of ozone.

Significant work has been conducted to quantify ozone depletion due to anthropogenic perturbations in the stratospheric halogen budget and forecast ozone recovery in response to controls established by the Montreal Protocol and its subsequent amendments. Stratospheric water vapor changes also impact surface climate, as do stratospheric aerosol changes, although there is a recent study that questions the impact of stratospheric sulfur on surface climate. Predicted GHG-induced increases in the strength of the Brewer Dobson Circulation, which drives key components of the variation in stratospheric ozone, differ by large amounts between various climate models, but have not been verified by observation. Methane (CH_4) and nitrous oxide (N_2O) are a special class of GHGs, as these compounds have both radiative and direct chemical effects on stratospheric composition. The response of stratospheric O_3 to rising N_2O is sensitive to future stratospheric temperature, which in turn is difficult to accurately forecast given our present state of knowledge. Finally, NASA satellite and balloon data show evidence that ocean biota are presently supplying a considerable portion of the total bromine loading to the stratosphere, in the form of so-called very short lived (VSL) compounds. It has been suggested that increased tropical maritime convection could result in more efficient future delivery to the stratosphere of halocarbons produced by oceanic biota.

Recent work indicates that the exchange between the troposphere and the stratosphere is not as simple as previously thought. Missing from the accepted conceptual view is the influence of midlatitude convection on the lower stratosphere. Recent work has shown evidence that water vapor enters the stratosphere via convective activity over North America in summer. One study using NASA aircraft observations [*Anderson, et al.,* 2012] suggested major ozone loss could result, while a second study [*Schwartz, et al.,* 2013] using NASA satellite data reported that in today's climate, convective injection of H_2O does not appear to enhance halogen-driven, mid-latitude ozone loss. Finally, changes in the latitudinal extent of the tropics caused by changes in

lower stratospheric midlatitude water vapor has been postulated; this needs to be further explored.

To detect trends in climate-relevant stratospheric ozone, water, aerosols, and temperature, it is critical that observations of profiles of these species continue with at least current spatial and temporal resolutions. Measurements of chemical constituents (nitrogen, hydrogen, chlorine, and bromine oxides) are needed to empirically quantify a number of emerging issues, such as the relative roles of transport and photochemistry in Arctic ozone loss during particularly cold winters, the effect of future changes in stratospheric H_2O, CH_4, and N_2O on ozone photochemistry, and the role of biogenic halocarbons in stratospheric ozone chemistry. Additionally, in anticipation of the possible use of Solar Radiation Management (SRM) for climate change mitigation, a solid understanding of the stratospheric aerosol (Junge) layer is needed. Many models handle this region very simplistically; a more thorough scientific understanding of the current layer is needed to properly assess how SRM may alter stratospheric constituent distributions. Recent work shows that there may be a tipping point associated with biogenic halocarbons, which may not be accurately represented in models, that controls whether surface UV will rise, or fall, if SRM via stratospheric sulfate injection is pursued.

Coupled with the continuation of stratospheric profile measurements from satellite and ground-based networks, it is also important that work continues to allow the merging of new data records with existing time series for those species/parameters in a well-documented manner that allows a realistic estimation of uncertainties. Significant work is ongoing on these issues, but it should not be considered a solved issue and requires additional research support. Merging data sets of stratospheric ozone, water and temperature has not proven to be easy or straightforward; significant problems remain. These need to be solved to improve our ability to detect and attribute trends.

Meeting the SC science goals requires measurement of an array of coupled tracers, chemically reactive intermediates, and radicals. These observations can be obtained by a synergistic deployment of orbital, sub-orbital measurement platforms, and ground-based remote sensing observations. Additionally, a suite of models of varying complexity are needed to fully understand measurements and trends. Questions noted below are split into three science topics. These include 1) Stratospheric Ozone and Water – Drivers and Impacts; 2) Stratosphere-Troposphere Exchange: How will climate change alter transport and links between the stratosphere and troposphere? And 3) Stratospheric Aerosols: Linkages between their chemical and radiative impacts and the temperature and species distribution of the stratosphere. Measurements needed for each question are detailed in Tables 2.2.1-3.

2.2.1 Stratospheric Ozone and Water – Drivers and Impacts

2.2.1.1 How will stratospheric H_2O and O_3 and associated chemical processes evolve in a climate with increased GHGs and changing ODSs? **(Critical)**

2.2.1.1.1 Water vapor has been observed to increase in the mid-latitude stratosphere since 1980. Why? How much is natural variability and how much is forced? How have individual pathways evolved over the past decades to change the stratospheric entry of water and how will that be affected by climate change?

2.2.1.1.2 How does O_3 and H_2O transport change with increased GHGs? (This directly links to the Atmospheric Circulation question on "How is the Brewer Dobson circulation (BDC) changing?")

2.2.1.1.3 What is the sensitivity of O_3 to CH_4 and N_2O, which are chemically as well as radiatively active, and will it become a larger driver of the abundance of radicals that control both photochemical loss and production of O_3?

2.2.1.1.4 What role does continental convection play in altering H_2O and O_3? Does summertime injection of H_2O over the US produce chlorine activation? Quantify transport of water and other short-lived tropospheric species due to direct injection at mid latitudes in summer as well as their ultimate mixing out into the background lower stratosphere.

2.2.1.2 How do stratospheric temperatures respond to changes in O_3 and the major GHGs? How do we quantify impacts of temperature changes on stratospheric chemistry, dynamics, and radiation? **(Critical)**

2.2.1.3 How do very short-lived (VSL) halocarbons get into the stratosphere? How much and in what form are they getting in now? How will that change in the future? What are the emission regions from which transport is most effective? **(Very important)**

2.2.1.4 How can we improve our ability to model denitrification and springtime chlorine activation/re-activation? How much do these processes change ozone in springtime? **(Important)**

2.2.2 Stratosphere-Troposphere Exchange (STE): Impact of climate change on transport and links between the stratosphere and troposphere

2.2.2.1 What is the current contribution of stratospheric ozone on the tropospheric O_3 budget? How will transport of O_3 from the stratosphere change with climate change? **(Very important)**

2.2.2.2 What are the relative roles of various STE mechanisms (BDC, convection, monsoon transport, isentropic transport across the subtropical jet, PyroCbs) in establishing the composition of the lower stratosphere. How will those roles change in an evolving climate, assuming the Coupled Model Intercomparison Project (CMIP) view of future climate? **(Critical)**

2.2.2.3 What is the effect of future changes of stratospheric composition (ozone, water, and aerosol loading) on surface temperature/precipitation? **(Very important)**

2.2.2.3.1 What is the radiative effect of stratospheric aerosol loading on surface climate:

i.e., does longwave trapping of heat largely offset reflection of solar radiation, as has been suggested?

2.2.2.3.2 How do atmospheric climate modes such as the decadal time scale variability in the strength of the Atlantic Meridional Overturning Circulation (AMOC), which is not well represented in climate models and which happens by chance to be aliased with stratospheric aerosol loading, complicate proper association of stratospheric drivers and surface response?

2.2.2.4 How do the budgets of chemically reactive greenhouse gases (CH_4, N_2O) and O_3 depleting gases evolve with climate change? (answering this question requires models as well as measurements) **(Very important)**

2.2.3 Stratospheric Aerosols: Linkages between their chemical and radiative impacts and the temperature and species distribution of the stratosphere.

2.2.3.1 What controls the basic evolution of aerosols in the Junge layer under non-volcanic background conditions or with only small volcanic input? **(Critical)**

2.2.3.1.1 What are the relative roles of natural and anthropogenic sources in determining the background stratospheric sulfate aerosol layer?

2.2.3.1.2 What controls the presence of the Asian Tropopause Aerosol Layer (ATAL) and North American Tropopause Aerosol Layer (NATAL)?

2.2.3.2 What are the relative contributions of sulfate aerosols, PSCs and cirrus clouds to chlorine activation and how will these factors impact ozone depletion in a changing climate? **(Important)**

2.2.3.3 How does a major volcanic eruption impact the earth's climate? **(Important)**

When there is a major volcanic eruption, we need to be prepared to quickly respond and make direct measurements and those that can be used as surrogates for studying the radiative and chemical effects due to possible large anthropogenic perturbations to the Junge layer and additional understand the perturbation on the key stratospheric variables. Measurements should encompass:

• Short-wave and long-wave direct radiative forcing of the volcanic aerosols integrated over the time of largest forcing.

• Rate of change of tropical upwelling; and any other major dynamical shifts (beyond climatological variability) such as tropopause height.

• Regions where ozone photochemistry appears to have changed in response to the aerosols; and (2^{nd} tier) if there appears to be enhanced chlorine activation and ozone loss.

References (2.2):

Anderson, J. G., D. M. Wilmouth, J. B. Smith, and D. S. Sayres, (2012), UV Dosage Levels in Summer: Increased Risk of Ozone Loss from Convectively Injected Water Vapor, *Science*, **337**, 835-839, DOI: 10.1126/science.1222978.

Schwartz, M. J., W. G. Read, M. L. Santee, N. J. Livesey, L. Froidevaux, A. Lambert, and G. L. Manney (2013), Convectively injected water vapor in the North American summer lowermost stratosphere, *Geophys. Res. Lett.*, **40**, 2316–2321, doi:10.1002/grl.50421

Table 2.2.1 Satellite measurements needed for answering Stratospheric Composition Questions. C = Critical; VI = Very Important; I = Important

Rank	Q 2.2.x.x	Measurement	Rationale	Sampling		Resolution		Precision
				Spatial	Temporal	Horizontal (lat. x long.)	Vertical	
C C VI VI	1.1 1.2 2.3 2.4	Total Column Ozone	a) A crucial factor for determining the amount of UV reaching the ground. b) Document spatial coverage of polar ozone losses.	Global	Weekly for surface UV, daily for process studies	1° x 2°	N/A	3 DU
C C VI C VI VI I I	1.1 1.2 2.1 2.2 2.3 2.4 3.2 3.3	Ozone profiles	a) Separate photochemical changes due to chlorine change from photochemical and transport changes due to climate change b) Separate stratospheric and tropospheric ozone changes	Global	Weekly for surface UV, daily for process studies	5° x 10° for UV, 1° x 2° for process studies	2 km for UV, 0.5 km for process studies	0.1 ppmv *(should be valid in cases of high aerosol loading)*
C C C VI VI I I	1.1 1.2 2.2 2.3 2.4 3.2 3.3	Temperature Profiles	a) Determine kinetic rates for ozone removal b) Temperature trends (see below).	Global	Weekly for trends, daily for process studies	5° x 10° for trends, 1° x 2° for process studies	2 km for trends, 0.5 km for process studies	2K/decade *(Long-term precision critical; should be valid in cases of high aerosol loading)*
C C I C VI VI C I I	1.1 1.2 1.4 2.2 2.3 2.4 3.1 3.2 3.3	H_2O	Quantify processes affecting water vapor and its impact on climate and ozone chemistry	Global	Weekly for radiative forcing, daily for process studies	5° x 10° for climate impact, 1° x 2° for process studies	2 km	0.5 ppmv *(should be valid in cases of high aerosol loading)*
C VI I C	1.1 1.3 1.4 2.2	Sulfate aerosols	Radiation, separation of natural variability from chlorine-caused	Global	Weekly	5° x 10°	2 km	10%/decade

C I I	3.1 3.2 3.3		changes					
VI C C I	1.3 2.2 3.1 3.3	SO_2	Major sulfur uncertainty	Global	Monthly	5° x 10°	1 km	10 ppt
I I I	1.4 3.2 3.3	Polar stratospheric clouds, HNO_3	Attribute polar ozone change to Montreal Protocol	Polar (>50°)	Daily	1° x 2°	1 km	10%/decade
C C VI I C VI I I	1.1 1.2 1.3 1.4 2.2 2.4 3.2 3.3	N_2O, CH_4, CFCs	O_3 chemistry, GHGs, dynamics	Global	Monthly, daily for polar vortex	10° x 10°, 1° x 2° for polar vortex	4 km, 1 km for polar vortex	20%/decade
C VI I I I	1.1 1.3 1.4 3.2 3.3	NO, NO_2, $ClONO_2$, HNO_3, ClO, HCl, BrO	Separate ozone loss due to non-chlorine catalytic processes Document temporal change in relative balance of ozone loss processes	Global	Monthly to seasonal, daily for polar vortex	10° x 10°, 1° x 2° for polar vortex	2 km, 1 km for polar vortex	20%/decade
C	1.1 1.2	CO_2	GHG	Global	Seasonal	10° x 10°	4 km	10%/decade
C VI VI C	1.1 1.3 2.3 3.1	HDO, HCN	Convection tracers	Global	Weekly	1° x 2°	0.5 km	□D < 20 parts per thousand
C VI VI C C	1.1 1.3 2.1 2.2 3.1	CO	Tropospheric and mesospheric air tracer	Global	Weekly	1° x 2°	4 km, 0.5 km for UTLS	10 ppb
VI	2.3	Precipitation	Target species	Global	Weekly	10° x 10°	N/A	10%/decade

Table 2.2.2 Aircraft in situ measurement requirements

Rank	Q 2.2.x.x	Measurement	Rationale	Detection limit	Accuracy	Time resolution
C VI I VI C VI I I	1.1 1.3 1.4 2.1 2.2 2.4 3.2 3.3	O_3	Target gas, transport tracer	1 ppb	5%	1 Hz
C VI I VI I I	1.1 1.3 1.4 2.1 2.2 2.4 3.2 3.3	H_2O	Target gas, transport tracer	100 ppb	10%	1 Hz
C VI I C I	1.1 1.3 1.4 2.2 3.2	Ice	Dehydration	0.1 liter^{-1}	20%	1 Hz
C VI VI C	1.1 1.3 2.1 2.2	CO	UTLS tracer		10%	1 Hz
C C VI	1.1 2.2 2.4	CH_4	Target gas, transport tracer, O_3 chemistry	1 ppb	5%	0.01 Hz
C VI C VI	1.1 2.1 2.2 2.4	N_2O	Target gas, transport tracer, O_3 chemistry	1 ppb	5%	0.01 Hz
C C VI	1.1 2.2 2.4	SF_6	Transport tracer	< 1 ppt	10%	0.01 Hz
C C VI	1.1 2.2 2.4	CO_2	Target gas, transport tracer	100 ppb	< 1 %	1 Hz
C VI C VI	1.1 1.3 2.2 2.4	Halocarbons (CFCs, HCFCs, solvents, also including VSL ODSs)	Target gas, transport tracer, O_3 chemistry	< 1 ppt	10%	0.01 Hz
VI C C I	1.3 2.2 3.1 3.3	SO_2	Major sulfur uncertainty	< 10 ppt	10%	0.01 Hz
C I	3.1 3.3	OCS	Major sulfur species	10 ppt	10%	0.01 Hz
C C C I I	1.1 2.2 3.1 3.2 3.3	Aerosol ND, SAD, composition	Target species	Size < 100 nm	20%	0.01 Hz
C VI C C I	1.1 1.3 2.2 3.1 3.3	Organics with various lifetime	UTLS tracers	1 ppt	10%	0.01 Hz
C VI C C	1.1 1.3 2.2 3.1	HDO	Convection diagnostic species	□D < 10 parts per thousand	10%	0.1 Hz

C	1.1 1.3 1.4 3.2 3.3	OH	O_3 chemistry, chlorine activation	< 1 ppt	20%	0.1 Hz
VI		HO_2		5 ppt	20%	0.1 Hz
I		NO		5 ppt	20%	0.1 Hz
I		NO_2		1 ppt	20%	0.1 Hz
I		NO_y or HNO_3		20 ppt	20%	0.1 Hz
		ClO		1 ppt	20%	0.1 Hz
		BrO		1 ppt	20%	0.1 Hz
		$ClONO_2$		20 ppt	20%	0.1 Hz
		HCl		20 ppt	20%	0.1 Hz
C	All	T, p, winds	Chemistry, transport, dynamics	< 1 hPa, 0.3 K, w <0.1 m s^{-1}, u,v < 1 m s^{-1}	< 1%	1 Hz

Table 2.2.3 Balloon, remote, and ground-based in situ measurement requirements

Rank	Q	Measurement	Rationale	Detection limit	Accuracy	Time resolution
C VI I VI C C I I	1.1 1.3 1.4 2.1 2.2 3.1 3.2 3.3	Temperature profile (aircraft)	Tropopause locator	100 m vertically	0.5 K	0.01 Hz
C C VI I	1.1 1.2 2.1 3.3	O_3 column (ground-base network)	Long-term trend, satellite validation	N/A	< 3 DU	0.01 Hz
C C VI VI C	1.1 1.2 2.1 2.4 3.1	CO_2, CH_4, N_2O, SF_6, sulfur species, halocarbons (ground-base network)	Source stratosphere gases	See Table 2.2.2	See Table 2.2.2	Daily
C C VI	1.1 1.2 2.4	O_3, H_2O, T profiles (balloons)	Target gases	See Table 2.2.2	See Table 2.2.2	Monthly
C	1.1	SF_6, N_2O, CH_4 profiles (balloons)	Target gases, age of air determination	See Table 2.2.2	See Table 2.2.2	Seasonally
VI	2.1	O_3 profiles for STE and above ~30km (lidar)	Track of stratospheric intrusion	1 ppb, 100 m vertically	1 ppb precision	0.01 Hz
VI C	2.1 2.2	UTLS H_2O (lidar)	Radiatively important	500 m vertically	1 ppm	0.01 Hz
C I VI VI I I	1.1 1.4 2.1 2.4 3.2 3.3	O_3, HNO_3, HCl, HF, NO_2, ClO, $ClONO_2$ (ground-base network FTIR, MW, UV)	O_3 chemistry, chlorine activation	4-8km	See Table 2.2.2	Daily
C VI I	1.1 2.1 3.3	O_3, HNO_3, HCl, CO, HDO, CH_4, N_2O (ground-base FTIR network)	Long-term trend, satellite validation	4-8km	See Table 2.2.2	Daily
C C VI I	1.1 1.2 2.3 3.3	H_2O and O_3 (microwave)	Long-term trend, satellite validation	10 km vertically	1%	Weekly

2.3 Clouds

Improvements in representing hydrological processes in models have been difficult to realize. *Knutti et al.,* (2013) show that the current generation of climate models in the most recent Coupled Model Intercomparison Project (CMIP5) exhibit modest improvements in reducing biases in temperature and precipitation fields relative to observations. *Klein et al* (2013) demonstrate that cloud properties in current models also exhibit only incremental improvements.

The physical processes that drive the forcing and feedback mechanisms that are the root of the biases noted by *Klein et al.* and *Knutti et al.* occur within circulation regimes that inhabit the largest spatial scales of the climate system. However, the physical processes that drive cloud and precipitation properties proceed on the microphysical scale. Until recently, these processes have been represented in global models as statistical representations that attempted to represent cloud effects based on the spatial scales resolved by the models (*Randall et al.,* 2003). Global cloud resolving models are now being developed (*Satoh et al.,* 2008) and cloud parameterizations are being abandoned or modified extensively (*Larson et al.,* 2012). Explicitly representing microphysical processes or their statistics on small scales has emerged as the fundamental weak link in climate change predictions because of our limited understanding of the important processes that link cloud microphysics to atmospheric motions at the finer grid scales of present day models.

Clouds tend to form in small-scale updrafts while precipitation grows by accreting condensate through differential sedimentation. Within any cloudy volume, both precipitation and cloud droplets (often of different phases) can, and typically do, coexist. It is the very coexistence of different droplet modes that cause certain microphysical processes to proceed. Therefore, to fully understand cloud processes, the properties of coexisting clouds and precipitation and their dynamical environment must be known. *This degree of complexity demands that multiple measurements that independently constrain different aspects of the vertically resolved particle continuum must be observed simultaneously to diagnose and ultimately understand the dominant microphysical processes.*

We discuss six broad topical areas that are among the foremost questions relating cloud processes to climate prediction. These questions are pertinent and remain unanswered largely because of uncertainties in our predictive skill that in turn are due to a scarcity of relevant observations. The approach we take is to describe an issue briefly and then discuss the geophysical parameters, required observations, and obstacles associated with each question. We finish with a discussion of issues that cut across all the questions and how these overarching issues might be dealt with.

2.3.1 What is the sensitivity of the climate system to low-level cloud structure and variability over the middle and high latitude oceans?

While the storm tracks of the middle latitudes are typically known for deep frontal cloud systems associated with migratory cyclonic disturbances, the maritime regions are

characterized by a background state of *shallow convective clouds* (*Mace,* 2010; *Haynes et al.,* 2011). The large cloud coverage of these low-level clouds significantly increase the albedo of oceanic regions resulting in a net negative cloud radiative effect at the surface that acts to maintain meridional temperature gradients – particularly in the summer (*Mace,* 2010). *Trenberth and Fasullo*, (2010) and others (*Bodas-Salcedo et al*, 2012; *Naud et al.,* 2014; *Williams et al.,* 2013) note that model biases in these regions, *particularly the Southern Oceans*, are large and lead to a significant bias in absorbed sunlight at the ocean surface in the present generation of climate models. Beginning with anomalously low cloud coverage, anomalously high absorbed energy at the surface, and therefore weak poleward heat transport, climate models tend to respond to increased greenhouse gasses by dramatically *increasing* cloud cover in this region as the climate changes. However, such a change is untenable as noted by *Trenberth and Fasullo* because observed cloud coverage in these regions is now near a maximum. Therefore, the actual response of these vast oceanic regions under climate change is not known.

To address this problem, it is necessary to understand the lifecycle of shallow convective clouds in these regions. This understanding needs to be linked to precipitation processes in clouds that in turn are coupled to boundary layer dynamics. Ultimately, knowing the relationship between accretion and autoconversion in the growth of the precipitation mode is also necessary (*Feingold et al.,* 2013).

Question 2.3.1: High Latitude Boundary Layer Clouds

Geophysical Quantities	Observations	Resolution
• Aerosol • Cloud and Precip Liquid water contents • Cloud and precip phase, size distribution • Vertical motion spectra	• Multi-Frequency Doppler Radar • Low frequency microwave • Visible, IR and TOA radiation • Polarimetry • Lidar	• Cloud-scale measurements are needed for process studies. • Larger basin and global scale statistics are crucial for model validation

2.3.2 How will the radiative balance over sea ice and permafrost change as the polar regions warm? What are the important feedback mechanisms between clouds, precipitation, and surface processes in the high latitudes?

One of the robust features that emerged in early climate model predictions was an amplification of projected global warming in the high latitudes (i.e. *Manabe and Weatherald,* (1967), *Weatherald and Manabe,* (1988)). However, the scenarios now playing out in the Polar Latitudes are complex and have the potential for feedbacks that have global consequences (*Comiso and Hall,* 2014). These feedbacks are linked fundamentally to precipitation and the surface radiation budget that depends largely on clouds.

A lack of knowledge regarding clouds and precipitation significantly hinders our ability to understand high latitude surface processes. For instance, the problem of sea ice retreat in the Arctic is well known (*Stroeve et al.,* 2012; *Kattsov et al.,* 2011) with summer minimum ice extent declining at 11% per decade over the past 40 years. The decline in summer ice cover has been linked to seasonal variability in cloud cover (*Kay et al.,* 2008 2009) via surface radiative fluxes.

Cloudy sky surface fluxes have also been noted as one of the fundamental unknowns regarding the melting of permafrost. It is thought that the upper 3 m of Arctic permafrost that comprise nearly a quarter of the Earth's northern hemisphere surface area contain as much as 1000 petagrams of carbon (*Cosimo and Hall,* 2014). The variables involved in this process are complex yet increasing periods of snow free surfaces over the northern continents amplify the snow-albedo feedback (*Stieglitz et al.,* 2003; *Dery and Brown,* 2007) linking this problem to surface radiative fluxes and precipitation.

Critically, while Arctic clouds are often shallow stratiform or strato-cumulus clouds, they are not the same as their low latitude dynamical cousins, because they often contain ice as well as liquid. Many of the important processes also occur in conditions where liquid phase supercooled cloud produces solid precipitation. For instance, stratiform mixed phase clouds often exist over large spatial scales and produce light ice precipitation (*De Boer et al.,* 2009) over long periods of time where ice nucleating aerosols are seemingly absent (*Morrison et al.,* 2012).

Question 2.3.2: High Latitude Radiative Forcing

Geophysical Quantities	Observations	Resolution
• Cloud occurrence and vertical structure • Phase and Particle Habit • Vertical Motion spectra • CCN (especially ice nuclei) • Surface Fluxes	• Multi-Frequency Doppler Radar • Passive microwave at high frequencies • Visible, IR include far IR fluxes • IR and Far IR Spectra • In Situ measurements of particle habit, sizes, and aerosol • Surface Polarization Radar	• Process Studies require airborne and collaboration with surface sites • Global satellite measurements are crucial because of remoteness of high latitude regions

2.3.3. How will shortwave cloud forcing change as the climate warms?

The predicted change of top of atmosphere radiative forcing per unit change in global temperature due to clouds (i.e. the cloud feedback) is the primary contributor to uncertainty in climate change predictions (*Dufresene and Bony,* 2008). It has been shown that large regions of shallow cumulus or stratocumulus clouds that occur over the sub-tropical oceans are the principal sources of this variability in cloud feedback among climate models

(*Dufresene and Bony,* 2008; *Soden and Vechi,* 2011). These clouds, while coupled to the surface via fluxes of heat, water, and aerosol, are also sensitive to the strength of the marine inversion, water vapor in the free troposphere, and to subsidence above the boundary layer (*Wood,* 2012; *Stevens and Feingold,* 2009). Individual cloud elements go through lifecycles that depend to a great extent on precipitation processes, which in turn are sensitive to the portion of the aerosol distribution that serves as cloud condensation nuclei, on cloud depth, and vertical motion (*Feingold et al.,* 2013). The overall cloud fraction in a region is therefore a complicated balance between the lifecycles of individual cloud elements and the meso- and larger scale dynamics and thermodynamics. Small changes in cloud condensation nuclei (CCN) over clean oceans can have large effects on cloud albedo and precipitation, and hence have a large effect on climate (*Twomey,* 1977; *Albrecht et al.,* 1989).

The principal interaction between marine boundary layer clouds and the climate system is through their regional-scale albedo. Given the low albedo and therefore highly absorbing nature of the ocean, the amount of shortwave radiation reflected by these cloud systems imposes large controls on climate sensitivity. Observations are needed to provide constraints on many aspects of high-resolution cloud resolving models and to provide global statistics from which global models can be evaluated.

Question 2.3.3: Shortwave Cloud Forcing

Geophysical Quantities	Observations	Resolution
• Boundary Layer Cloud occurrence and vertical structure • Cloud and Precipitation droplet spectra • Vertical Motion spectra • CCN • Surface Fluxes of water, heat, chemistry	• Doppler Cloud Radar • Passive microwave sensitive to liquid • High Spectral Resolution Lidar • Polarimetry • In situ measurements of aerosol, cloud and precipitation mode size distributions • Aerosol composition • Boundary Layer Thermodynamics and inversion strength	• Measurements for process studies must resolve cloud vertical and horizontal structure (10's of meters) on timescales critical to cloud element lifecycles (minutes). • Global satellite measurements are crucial to observe relationships with larger scale dynamics. • Global measurements for model diagnostics

2.3.4. How will long-wave cloud forcing change as climate warms?

Cloud feedbacks are consistently positive in the leading climate models (*Soden and Vechi*, 2011). While the uncertainty (intermodel variability) of this feedback is primarily

determined by marine boundary layer clouds, the positive sign in the feedback is determined by tropical cirrus. As shown by *Zelinka and Hartmann,* (2010), this positive feedback arises because as the climate warms, the troposphere deepens and convective clouds are predicted to detrain at colder temperatures. While models tend to agree in this prediction, their mean ice water paths differ by enormous amounts with each other and with observations. It is not known why this is so. What is certain is that tropical cirrus clouds are fundamental to the response of the climate system to changes in greenhouse gasses. Being able to faithfully simulate tropical cirrus is of first order importance to understanding climate change.

High-level cirrus heat the upper troposphere and, thereby, influence the stability of the troposphere. Both the cirrus themselves and the associated water vapor perturbations have strong impacts on the longwave radiation budget (*Liou,* 1986; *Forster and Shine,* 2002; *Solomon et al.,* 2010). Basic uncertainties in physical processes such as ice nucleation, aggregation, and entrainment have limited our ability to represent cirrus even in high-resolution detailed models. The relative importance of heterogeneous nucleation on insoluble particles (with implications for anthropogenic perturbations to cirrus) has not been fully determined.

Progress on these issues requires a better quantification and understanding of temperature variability driven by mesoscale waves and radiatively-driven turbulence in the upper troposphere and lower stratosphere. The compositions, sources, concentrations, and regional variations of heterogeneous ice nuclei need to be better quantified, as well as the impact of nucleation mechanisms on overall cirrus lifecycle and radiative effects. Quantification of crystal aggregation efficiency at low temperatures and the corresponding impact on development of cirrus ice crystal size distributions and habits is required.

Question 2.3.4: Longwave Cloud Forcing

Geophysical Quantities	Observations	Resolution
• Ice water content vertical profiles • Particle size and number concentration vertical profiles • Ice crystal habit • Vertical motion • Ice Water Path • Radiative Heating Profiles	• Lidar attenuated backscatter and depolarization ratio profiles • Doppler cloud radar vertical profiles • Aerosol – IN measurements • Vertical motion spectra • Water vapor supersaturation • In situ crystal Habit, particle spectra and bulk mass.	• The nature of extended cirrus clouds require a collaboration between remote sensing and in situ airborne assets. • Global satellite data sets are crucial to observe cirrus over remote tropical regions and under conditions that aircraft have difficulty reaching routinely. • Global satellite data sets remain crucial for model diagnostics and validation.

2.3.5 How do clouds respond to perturbations in aerosols (CCN/IN)?

Human activities give rise to significant emissions of aerosols and their chemical precursors and the anthropogenic source of aerosols significantly modifies the distribution of natural aerosol in many regions on earth. Since aerosols, in the form of Cloud Condensation Nuclei (CCN) and Ice Nuclei (IN), govern the drop and crystal size distributions, knowledge of them are critical for understanding many cloud processes. Aerosol effects can be complex, by both increasing cloud albedo (with more and smaller liquid drops, e.g. *Twomey,* (1977)) and changing cloud element lifetime via modulation of precipitation processes (*Albrecht*, 1989). But even given a CCN or IN concentration, we do not fully understand the microphysical responses that are possible. Understanding the details of autoconversion and accretion rates and the details of CCN activation in a turbulent vertical motion field is fundamental to understanding aerosol-cloud interactions.

With respect to aerosols, it is expected that aerosols of different composition types, with differing scattering and absorption characteristics will have different impacts on cloud development. Absorbing aerosol can alter the temperature profile in the first few kilometers above the surface, and have a stabilizing effect on the temperature profile and convection (*Ramanathan et al.* 2007). Conversely, an increase in aerosol is thought to alter the vertical profile of latent heat release, leading to an "invigoration" of cloud development (*Rosenfeld et al.,* 2008). However, these processes are complex, and often competing, resulting in counter intuitive outcomes. Of particular importance is how the aerosol are vertically distributed – a diagnostic that is nearly impossible to know from passive satellite data.

One reason for difficulty in making conclusive headway in aerosol-cloud research is that the physics is governed by a multitude of variables, only several of which have been adequately incorporated in analyses. As stated in the recent comprehensive review of *Tao et al.,* (2012) "Despite ever-escalating efforts and the virtually exponential increase in published studies concerning aerosol-cloud-precipitation interactions, we are still puzzled by many seemingly contradictory findings as reviewed above, although they really attest to the complexity of the problem. Disentangling meteorological and aerosol effects remains a daunting task."

There is a need to obtain a better understanding of fundamental process rates that convert one cloud particle type to another in the presence of different CCN backgrounds. These are inherently dependent on vertical motion statistics at multiple scales that ultimately control supersaturation. Also, knowledge of the characteristics and vertical distribution of the aerosol and cloud particle size distributions, habit, composition, etc. are necessarily needed to improve understanding.

Question 2.3.5: Aerosol-Cloud-Precipitation Interaction

Geophysical Quantities	Observations	Resolution and Scale of Measurements
• Aerosol Extinction, single scattering albedo,	• Vertical distribution of aerosol Size Distribution, speciation, CCN, IN	• Process studies will require airborne measurements, preferably with multiple

asymmetry parameter vertical profile • Vertical profiles of Cloud and Precipitation Liquid water, size distribution, phase, particle habit • Vertical motion spectra from cloud-scale to mesoscale	• Aerosol precursor gasses vertical profiles • High Spectral resolution lidar • Vertically pointing Doppler radar measurements (multiple freq in precip) • Polarized weather radar for precip • Passive microwave • thermodynamics	aircraft (in situ and remote sensing) • Global satellite measurements of a selected subset of the observations are needed for statistical regional and global studies of aerosol indirect effects.

2.3.6 What is the role of (Mixed Phase) snow and rain in cloud processes and how is this modulated by aerosols and dynamics?

Most precipitation on Earth starts as ice in synoptic systems (fronts), shallow convection, and stratiform anvils. The process of initial freezing is complex, and the subsequent evolution of the frozen condensate includes interactions with ice and liquid in clouds. These processes include riming, aggregation and ice multiplication that modulate the ice phase precipitation. These processes determine many of the resulting cloud properties, and the resultant mass and intensity of precipitation from clouds. Understanding these cold phase processes are critical for assessing cloud and condensate dynamics.

This question shares many of the same challenges expressed in questions 2.3.1 and 2.3.2. Both of these questions address mixed phase precipitation from cumuliform and stratiform clouds. Of particular emphasis here are the processes that control ice crystal properties such as shape, degree of riming, and aggregation. These ice crystal properties can be reduced to several measurable parameters such as bulk ice density and the internal distribution of ice within crystals as a function of particle size at various locations within the ice-producing cloud system. Knowledge of these parametric relationships are at a rudimentary level and cause significant uncertainties in both measurements and modeling.

Question 2.3.6: Mixed Phase Snow

Geophysical Quantities	Observations	Resolution
• Liquid and Ice water content vertical profiles • Particle size and number concentration	• Multiple frequency Doppler cloud radar vertical profiles • Polarization scanning cm radar measurements	• Process studies require a collaboration between airborne and ground-based measurements, preferably with multiple aircraft (in situ and

vertical profiles of liquid and ice phases • Ice crystal habit • Vertical motion • Ice and liquid Water Paths • Vertical profiles of Particle habit and bulk density as a function of size	• Vertical motion spectra • Water vapor supersaturation as a function of height • In situ crystal Habit, particle spectra and bulk mass.	remote sensing) • Global satellite measurements of a selected subset of the observations are needed for statistical regional and global studies especially over remote oceanic and high latitude regions.

3. **Overarching Issues**

Ideally, formal Science Traceability Matrices would be developed to address the questions listed above. Such a set of matrices would allow for a natural flow of understanding from the broad questions to what geophysical parameters must be known, what measurements are necessary to derive the geophysical parameters, what instruments are necessary to make the measurements, and a strategy for collecting the appropriate information.

A number of concepts emerge that are common among the questions outlined above. These are:

- The role of microphysical processes, process rates, and the coupling of those processes to atmospheric motions must be better understood.
- The role of aerosols in cloud processes is largely unknown and central to understanding.
- The spectrum of vertical motion, from the turbulent to the grid scale, is especially critical because of the role of vertical motions in either vertically advecting droplets and ice crystals and/or in making water vapor available for condensation.
- The need for modeling at various scales as an integral component in addressing each question.

Several cross cutting geophysical quantities are common among the questions. The commonality of these are largely derived from the need to understand basic processes – the processes that convert aerosols to cloud mode droplets and ice crystals through nucleation, and that convert cloud droplets to precipitation.

- Cloud-mode particle size distributions: liquid and ice water contents, effective particle size, total particle concentration, phase
- Precipitation-mode particle size distributions: liquid and ice water content, effective particle size, total particle concentration, and phase.

- The spectrum of vertical motion and humidity. Particularly pertinent are the statistics of vertical motion on the 1 to 10 km scale that would tend to drive modern GCM parameterizations.
- Aerosol properties, their sources, sinks, and composition below, near, and within clouds.
- The presence and properties of ice nuclei are fundamental to several of the questions.
- The physical properties of ice crystals such as the particle mass as a function of particle maximum dimension, the particle cross sectional area as a function of particle maximum dimension and the distribution function of mass within the ice crystal volume among others. Not only are these issues important for models in representing ice microphysical processes, but uncertainty in ice crystal properties drive errors in remote sensing retrievals to unacceptable levels. Not knowing the statistics of the ice crystal properties cause bias errors in retrievals to remain unknown.

The general nature of these questions results in a number of required cross cutting measurements.

- Many of the cloud types associated with the questions listed above are optically thick in the traditional visible and infrared observing bands. These clouds tend to be optically thin in the microwave and submillimeter wavelengths bands. Therefore, passive and active (including Doppler) measurements in the microwave and submillimeter wavelengths are fundamentally important.
- Optical lidar, including high spectral resolution (HSRL) technology, is critically important to characterizing the aerosol concentrations and composition and the properties of thin ice clouds.
- In situ measurements are fundamental. In particular, in situ aircraft flying in tandem with remote sensing aircraft is a powerful combination and highly synergistic.
- Most of the required geophysical parameters cannot be derived from a single remote sensing measurement or instrument. Measurement synergy is required that simultaneously combines various active remote sensing measurements with vertically integrated constraints such as microwave brightness temperatures and visible reflectances at various wavelengths.

Multi-sensor retrieval algorithms can provide information that goes beyond what is possible of single sensor retrievals or even from what has been accomplished to date with data from the A-Train (*Delanoë and Hogan,* 2010) or from airborne platforms. In each of the questions listed above, we have envisioned that multiple radar frequencies with Doppler velocity, lidar backscatter, passive microwave brightness temperatures and visible and IR radiances can provide a comprehensive set of measurements from which the necessary geophysical parameters can be retrieved.

References (2.3):

Albrecht, B. A, 1989: Aerosols, Cloud Microphysics and Fractional Cloudiness. Science, 245, 1227-1230.

Bodas-Salcedo, A., K D. Williams, P. R. Field, and A. P. Lock, 2012: The surface downwelling solar radiation surplus over the Southern Ocean in the Met Office Model: The role of midlatitude cyclone clouds. J. Climate, 25, 7467-2498.

Comiso, J. C. and D. K. Hall, 2014: Climate trends in the Arctic as observed from space. WIREs Clim. Change, 5:389-409. Doi: 10.1002/wcc.277.

Delanoë, J., and R. J. Hogan, 2010: Combined CloudSat-CALIPSO-MODIS retrievals of the properties of ice clouds, J. Geophys. Res., 115, D00H29, doi:10.1029/2009JD012346.

De Boer, G. E. W. Eloranta, M. D. Shupe, 2009: Arctic mixed-phase stratiform cloud properties from multiple years of surface-based measurements at two high-latitude locations. J. Atmos. Sci., 66, 2874-2888.

Dery, S. J. and R. D. Brown, 2007: Recent Northern Hemisphere snow cover extent trends and implications for the snow-albedo feedback. Geophys. Res. Lett., 34, doi: 10.1029/2007GL031474.

Dufresne, J-L., and S. Bony, 2008: An Assessment of the primary sources of spread of global warming estimates from coupled atmosphere-ocean models. J. Climate, 21, 5135-5144.

Feingold, G. A. McComiskey, D. Rosenfeld, and A. Sorooshian, 2013: On the relationship between cloud contact time and precipitation susceptibility to aerosol. J. Geophys. Res., 118, 10544-10554, doi: 10.1002/jgrd50819.

Forster, P. M. de F., and K. P. Shine, 2002: Assessing the climate impact of trends in stratospheric water vapor. Geophys. Res. Lett., 29, No. 6, 1086, 10.1029/2001GL013909.

Haynes, J. M., C. Jakob, W. B. Rossow, G. Tselioudis, and J. Brown, 2011: Major characteristics of Southern Ocean cloud regimes and their effects on the energy budget. J. Climate, 24, 5061-5080.

Jiang, J. H., et al. (2012), Evaluation of cloud and water vapor simulations in CMIP5 climate models using NASA "A-Train" satellite observations, J. Geophys. Res., 117, doi:10.1029/2011JD017237.

Kay, J. E., and A. Gettelman, 2009: Cloud influence on and response to seasonal Arctic sea ice loss, J. Geophys. Res., 114, D18204, doi:10.1029/2009JD011773.

Kattsov, V. M., V. E. Ryabinin, J. E. Overland, M. C. Serreze, M. Visbek, J. E. Walsh, W. Meier, X. Zhang, 2011: Arctic sea-ice change: A grand challenge of climate science. Journal of Glaciology, 56, 1115-1121.

Klein, S.A., Y. Zhang, M.D. Zelinka, R. Pincus, J. Boyle, and P. J. Gleckler, 2013: Are climate model simulations of clouds improving? An evaluation using the ISCCP simulator, J. Geophys. Res. Atmos., 118, 1329–1342, doi:10.1002/jgrd.50141.

Knutti, R., D. Masson, and A. Gettelman, 2013: Climate model genealogy: generation CMIP5 and how we got there. Geophys. Res. Lett., 40, 1194-1199, doi:10.1002/grl.50256.

Larson, V. E., D. P. Schanen, M. Wang, M. Ovchinnikov, S. Ghan, 2012: PDF parameterization of boundary layer clouds in models with horizontal grid spacings from 2 to 16 km. Mon. Wea. Rev., 140, 285-306.

Liou, Kuo-Nan, 1986: Influence of Cirrus Clouds on Weather and Climate Processes: A Global Perspective. Mon. Wea. Rev., 114, 1167–1199.
doi: http://dx.doi.org/10.1175/1520-0493(1986)114<1167:IOCCOW>2.0.CO;2

Mace, G. G., 2010: Cloud properties and radiative forcing over the maritime storm tracks of the North Atlantic and Southern Ocean as derived from A-Train. J. Geophys. Res., 115, D10201, doi:10.1029/2009JD012517

Manabe, S., and R. T. Wetherald, 1967: Thermal equilibrium of the atmosphere with a given distribution of relative humidity, J. Atmos. Sci., 24, 241– 259.

Morrison, H., Gijs de Boer, Graham Feingold, Jerry Harrington, Matthew D. Shupe and Kara Sulia, 2012: Resilience of persistent Arctic mixed-phase clouds. Nat. Geo., 5, 11-17.

Naud, C. M., J. F. Booth, A. D. Del Genio, 2014: Evaluation of ERA-Interim and MERRA cloudiness in the Southern Ocean. J. Climate, 27, 2109-2115.

Ramnanathan, V., R, Muvva, G, Roberts, Dohyeong Kim, Craig Corrigan, Chul Chung, David Winker, 2007: Warming trends in Asia amplified by brown cloud solar absorption, Nature, 448, 575-578.

Randall, D. A., M. Khairoutdinov, A. Arakawa, and W. Grabowski, 2003: Breaking the cloud-parameterization deadlock, Bull. Amer. Meteor. Soc., 84, 1547-1564.

Rosenfeld, D., Ulrike Lohmann, Graciela B. Raga, Colin D. O'Dowd, Markku Kulmala, Sandro Fuzzi, Anni Reissell, Meinrat O. Andreae, 2008: Flood or Drought: How do Aerosols Effect Precipitation, Science, 321, 1309 -1313.

Satoh, M. T. Matsuno, H. Tomita, H. Miura, T. Nasuno, and S. Iga, 2008: Nonhydrostatic icosahedral atmospheric model (NICAM) for global cloud resoliving simulations. Journal of Computational Physics, 227, 2486-3514.

Soden, B. J., and G. A. Vecchi (2011), The vertical distribution of cloud feedback in coupled ocean-atmosphere models, Geophys. Res. Lett., 38, L12704, doi:10.1029/2011GL047632.

Solomon, S., K. H. Rosenlof, R. W. Portmann, J. S. Daniel, S. M. Davis, T. J. Sanford, G.-K. Plattner, 2010: Contributions of Stratospheric Water Vapor to Decadal Changes in the Rate of Global Warming, Science, 327, 1219-1223.

Stevens, B. and G. Feingold, 2009: Untangling aerosol effects on clouds and precipitation in a buffered system. Nature, 461, 607-613.

Stieglitz, M. S. J. Dery, V. E. Romanovsky, and T. E. Osterkamp, 2003: The role of snow cover in the warming of Arctic permafrost. Geophys. Res. Lett., 30, doi: 10.1029/2003GL017337.

Stroeve, J. C., M. C. Serreze, M. M. Holland, J. E. Kay, J. Malanik, A. P. Barrett, 2012: The Arctic's rapidly shrinking sea ice cover: A research synthesis. Climate Change, DOI: 10.1007s10584-011-0101-1.

Tao, W.-K., J.-P. Chen, Z. Li, C. Wang, and C. Zhang (2012), Impact of aerosols on convective clouds and precipitation, Rev. Geophys., 50, RG2001, doi:10.1029/2011RG000369.

Trenberth, Kevin E., John T. Fasullo, 2010: Simulation of Present-Day and Twenty-First-Century Energy Budgets of the Southern Oceans. J. Climate, 23, 440–454. DOI 10.1175/2009JCLI3152.1

Twomey, S., 1977: The Influence of Pollution on the Shortwave Albedo of Clouds. J. Atmos. Sci., 34, 1149–1152.
doi: http://dx.doi.org/10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2

Wetherald, R. T., S. Manabe, 1988: Cloud Feedback Processes in a General Circulation Model. J. Atmos. Sci., 45, 1397–1416. doi: 10.1175/1520-0469(1988)045<1397:CFPIAG>2.0.CO;2

Williams, K. D., A. Bodas-Salcedo, M. Deque, S. Fermepin, B. Medeiros, M. Watanabe, C. Jakob, S. A. Klein, C. A. Senior and D. L. Willimason, 2013: The Transpose-AMIP II experiment and its application to the understanding of Southern Ocean cloud biases in climate models. J. Climate, 26, 3258-3275.

Wood, R., 2012: Stratocumulus Clouds. Mon. Wea. Rev., 140, 2373-2423, DOI: 10.1175/MWR-D-11-00121.1.

Zelinka, M. D., and D. L. Hartmann (2010), Why is longwave cloud feedback positive?, J. Geophys. Res., 115, D16117, doi:10.1029/2010JD013817.

2.4 Aerosols

Most applications involving aerosol particles require some information about aerosol amount (horizontal distribution), aerosol vertical distribution, and aerosol type. However, the requirements vary immensely with the application, and measurement capabilities vary enormously with observing conditions. For example, the most significant observational challenge for energy budget applications is constraining the global vertical distribution of absorption, scattering and emission, whereas for air quality, near-surface particle concentration, size, and chemical speciation are paramount.

Aerosol science is an inherently interdisciplinary field, crossing the boundaries of most core earth science disciplines. Important feedbacks occur between cloud and aerosol life cycles, between atmospheric temperature and secondary organic aerosol (SOA) formation, and between wind, surface moisture, and soil dust mobilization. Laboratory researchers, field in situ experimenters, remote sensing scientists, and modeling investigators need to interact to address key aerosol-related questions. Support for interdisciplinary research should be strongly encouraged, with small investigator teams from diverse backgrounds being particularly efficacious.

In addition, the sensitivities of aerosol remote-sensing measurement techniques vary considerably with observing conditions, and detailed particle microphysical properties are unobtainable from spacecraft. So specific advances are also needed in both remote-sensing and in situ instrumentation, to improve accuracy and sensitivity with which key aerosol parameters can be measured, and to increase the range of conditions under which they can be derived.

Between the third and fourth assessment reports of the Intergovernmental Panel on Climate Change (IPCC AR3, 2001; IPCC AR4, 2007), NASA resources, primarily the MODIS and MISR space-based instruments and the AERONET surface sun photometer network, provided a climatology of monthly, global aerosol optical depth (AOD), which was used to constrain the AR4 simulations, reducing the estimated uncertainty in Direct Aerosol Radiative Forcing (DARF) from “low” to “med-low.” However, the climate models at present are practically unconstrained with regard to the distribution of aerosol type, and are only loosely constrained with respect to the vertical component of the 3-D spatial distribution. Aerosol type and vertical distribution are also ’tall poles’ regarding all aspects of the interactions between aerosols and clouds. Lacking observational constraints, climate model fidelity estimates rely primarily on model diversity, which at best represents a lower bound on actual uncertainty.

To meet the demands of this interdisciplinary field, aerosol data has to evolve into a more integrated system of satellite data and network observations combined with models, and supplemented with targeted field campaigns. Focused efforts on the “how's” of combining datasets are needed, as well as recognition that integrated monitoring and development of new sensor/retrieval techniques are both a science in themselves.

Overarching Recommended Focus Area Priorities

By and large, the measurement requirements laid forth in some previous mission definition studies (such as the Aerosol-Cloud-Ecosystem (ACE) mission), while technologically ambitious, are conceptually good benchmarks for future aerosol study needs. Particular areas of emphasis include:

Top two measurement needs: 3D absorption and aerosol typing/speciation

- Absorption: Perhaps our biggest challenge, with local $\square_o$ uncertainty <0.02.
- Speciation/ Typing: How do we segregate natural versus anthropogenic aerosol types in the vertical and their ultimate radiation and thermodynamic effects by constituent?

Critical observational environments

- Aerosol system in the surface layer and planetary boundary layer.
- Aerosol observations in partly cloudy to cloudy scenes.

Retrievals

- How do we put two satellite data products (either radiance data or retrieval) together for synergistic benefit? (e.g. Polar + Geostationary observations).
- Clear need for technology investment and numerical testbed development for coupled retrievals.

Network observations and field campaigns

- Let aerosol sensitivity findings be a key defining measurement priorities. (E.g., climate hypothesis driven field campaigns)
- Data reduction/development of predictors of aerosol complexity (typing and speciation)
- Tie network observations and field measurements with the climate data record
- Develop and implement a plan to deploy in situ instruments that can provide detailed microphysical properties for the major aerosol airmass types at sensitivity levels that are unobtainable from remote sensing but required by many applications

Error modeling

- AERONET product and retrieval evaluation and improvement is a very high priority.
- Development of other primary and secondary verification methods.
- How do remote sensing observational errors relate to the model background error and ultimately to any combined products?

Key scientific questions are divided into application areas of (1) Climate, (2) Weather and Processes, and (3) Air Quality. Below is a highlight of the key questions in each area.

2.4.1 Climate

Aerosol particles are ubiquitous in the atmosphere and have strong relationships to such overarching climate (i.e., earth system science) themes as radiation, circulation, thermodynamics, hydrology, cryosphere, biosphere, bio-geochemical cycles, composition and chemical processes**.** To focus effort, often aerosol research is framed within a grand challenge question such as for radiation "How do anthropogenic aerosol particles affect total top of atmosphere radiation via all processes such as the direct, semi-direct and indirect forcing?" Similar questions can be related to all of the above systems (e.g., aerosol relationships to hydrology). Although nominal answers to such questions are outside the

bounds of any individual effort, they nevertheless provide a far reaching focal point for coordinating research. However, in regard to immediate needs of the research community, several crosscutting topical areas receive the most attention. These are listed below.

General Climate- Aerosol Interactions

Aerosol-climate feedbacks will become increasingly important in the 21st century as climate changes become increasingly severe.

2.4.1.1 How are natural and anthropogenic aerosols and their impact on the radiation budget at the top, within, and at the bottom of the atmosphere changing in response to a warming climate and an altered hydrological cycle, and how do these changes feed back to the climate system? **(Critical)**

2.4.1.2 How will projected changes in atmospheric circulation affect aerosols within the climate system through generation, transport, and deposition mechanisms and through their impact on biogeochemical cycles? **(Critical)**

2.4.1.3 What is the baseline (i.e., background) atmospheric aerosol loading and how can observed decadal changes to this loading be used to project future climate change? **(Important)**

Climate sensitivity to aerosol particles

The quantification of the sensitivity of the entire earth system to the aerosol system is one of the greatest overarching science challenges facing the community. Indeed, climate change and its sensitivity to aerosol distributions can be viewed as an ensemble of regional impacts. These are all high priority science topics.

2.4.1.4 What regions are most susceptible to aerosol phenomena, how are different aerosol types affecting regional brightening or dimming and what impacts are regional pollution controls and emissions changes having? **(Critical)**

2.4.1.5 What are the teleconnections between forcing in one region and impact in another and what are the underlying transport mechanisms of different aerosol types? **(Critical)**

2.4.1.6 Does the aerosol system have a role in the temperature hiatus, specifically, do numerous, small-scale volcanic eruptions lead to an increase in the global aerosol loading, especially in the upper troposphere to stratosphere? **(Important)**

2.4.1.7 How is heterogeneous ozone chemistry in the stratosphere, which is dominated by aerosols, changing in a changing stratosphere? **(Important)**

2.4.1.8 What are the long-term trends in absorbing aerosol transport and deposition in the polar regions, and how does this affect atmospheric and surface heating rates? **(Important)**

2.4.2 Weather and Processes.

Understanding aerosol impacts on climate is predicated on the quantification of the physical process and short-term weather relationships within the aerosol system. Aerosol lifecycle (including sources, transformation, transport and scavenging/fate), especially in the troposphere happens fundamentally on short time scales. This category addresses the question as to how to appropriately represent the current aerosol system. Knowledge of the vertical distribution of aerosol type, amount, and optical and microphysical properties is crucial to all aerosol process questions. Although to first order, aerosol particles can be thought of as essentially passive tracers, in reality, aerosols interact in an extremely complex manner with other atmospheric constituents such as clouds and precipitation through radiation, chemistry, and dynamics. Below we consider aerosol-meteorology relationships, aerosol microphysics and ice properties, and atmospheric radiation with key questions emboldened.

Aerosol-Meteorology Relationships- Passive

2.4.2.1 What are the meteorological processes that control the distribution, transport, and deposition of aerosols? **(Critical)**

2.4.2.2 Clouds as a transformative and removal processes. **(Critical)**

2.4.2.3 Vertical redistribution, PBL entrainment/detrainment **(Critical)**

Weather as a predictor of aerosol sources

2.4.2.4 What effects do large-scale meteorological modes (e.g., MJO, NAO, ENSO) have on the aerosol system through impacts on aerosol distribution, transport, and deposition and how can these effects be separated from climate scale impacts? **(Important)**

Aerosol-Meteorology Relationships-Feedbacks

2.4.2.5 What are the important underlying mechanisms through which aerosols impact the meteorological cycle? What are the relative magnitudes of the dynamic, radiative, microphysical, and thermodynamic effects, and when and where are these important? **(Critical)**

2.4.2.6 At what temporal and spatial scales and at what loadings do different aerosol types impact the meteorological cycle especially through their activation as cloud droplets? **(Critical)**

Aerosol Chemistry

2.4.2.7 How do biogenic emissions regulate secondary aerosol formation and how is this process influenced by cloud processing? **(Critical)**

2.4.2.8 What do we mean by "brown carbon" and what are its chemical and optical properties? **(Important)**

2.4.2.9 How do we reconcile chemical transport model descriptions of aerosol species, including mixing state, with the optical properties fundamental to aerosol type retrieval using remote sensing? **(Important)**

Aerosol Microphysics

2.4.2.10 How do we reconcile the needs of the atmospheric chemistry and remote sensing fields with regard to aerosol microphysical descriptions? How much complexity is useful and what tells us what we really need to know about aerosols in the earth system? **(Important)**

2.4.2.11 How do we obtain adequate information on global CCN concentrations? What are the types, sources, and spatial distributions of CCN and Ice Nuclei? **(Important)**

Aerosol Ice Physics

2.4.2.12 There is a significant need for exploratory research on the microphysical properties and lifecycle of Ice Nuclei. **(Critical)**

2.4.2.13 What is the Ice Nuclei lifecycle in the sensitive arctic and southern high mid-latitude regions? **(Important)**

Radiation

2.4.2.14 What is the vertically resolved distribution of atmospheric heating due to the presence of different types of aerosols in clear sky, partly cloudy, and cloudy conditions? **(Critical)**

2.4.2.15 How do aerosol changes affect the radiation budget through the diurnal cycle? **(Critical)**

2.4.3 Air Quality

Although for many years climate impact has been the driving concern motivating aerosol research at NASA, aerosols also have a significant impact on air quality through their effects on human health, biological productivity, visibility, and boundary layer chemistry. Presently NASA satellite data is being used to monitor air quality, particularly in locations without surface-based monitoring stations like much of the developing world. The fundamental challenge is relating remotely sensed information on total aerosol load to pollutant amounts in the lowest 10 m of the atmosphere where people live. Understanding aerosol effects on air quality involves cross-cutting questions with tropospheric and stratospheric chemistry, clouds, convection, and radiation. The primary focus of the aerosol community described below is on boundary constituents and processes.

Boundary layer constituents and processes

2.4.3.1 How do different aerosol types, stratified by size and composition, from natural and anthropogenic sources affect air quality in the PBL and surface layer? **(Critical)**

2.4.3.2 Can we distinguish long-term trends in air quality from extreme events? **(Important)**

2.4.3.3 How do human activities such as desertification, deforestation, and water diversion affect aerosol emission sources and downwind air quality? **(Important)**

2.4.3.4 What are the mechanisms controlling the spatial and temporal diffusion of aerosol plumes and long-range aerosol transport? **(Important)**

2.4.3.5 How do aerosols formed from anthropogenic emissions affect air quality throughout the diurnal cycle? **(Important)**

2.4.3.6 How can we develop a system for the global unified monitoring and evaluation of air quality incorporating the best available data from remote sensors, ground-based monitors, and modeling systems? **(Important)**

2.4.4 Required Aerosol Observations

The interdisciplinary nature of aerosol science defies the concept of a set of required observations with precisions and accuracies. Many of the aerosol science questions posed above will push the limit of what is physically possible to measure in the coming decade (e.g., particle properties in any form near the earth's surface). Conversely, current observations can support numerous aspects of posed research provided the observations are processed differently and incorporated into comprehensive modeling systems (e.g., sensitivity studies). In both cases, requirements for observations vary with the application, and measurement capabilities vary enormously with observing conditions. The one thing that is clear is that there is significant demand for improved and more extensive aerosol-relevant data sets. These datasets are best applied in a multi-senor and or type constellation approach.

Within the context of this decadal review it is impossible to definitively assign specific observations to questions. However, there are areas of development that will provide maximum benefit. For example, whereas the past decade saw significant development with the use of aerosol optical thickness (AOT), the coming decade will no doubt see further emphasis on the three-dimensional aerosol structure. Indeed, lidar, stereo/parallax, and spectral determinations of aerosol height are in ever increasing demand for standalone analyses, fusion with other satellite products, or assimilation into models. Second, aerosol particle light absorption, and its associated atmospheric heating is at the heart of numerous outstanding questions-particularly how direct and semi-direct effects influence clouds and circulation.

In the absence of much needed observing system simulation experiments (OSSEs) and sensitivity studies, it is difficult to credibly assign observations and their associated precisions and uncertainties for specific science questions posed in this report. However, based on the previous decadal survey recommendation for the Aerosol-Cloud-Ecosystem (ACE) mission, scientists did go through the process of developing measurement requirements to support science questions related to those posed here. It is recognized,

however, that some of these measurement requirements are technologically ambitious and successes in sub-orbital demonstrations are still limited. We also expect that many of these observational requirements will necessitate the integration of remote sensing observations into a modeling framework, supported by targeted field campaigns. Nevertheless, we have no significant reason to not endorse ACE satellite observational aerosol requirements as the best benchmark for observing needs currently available. These are:

Column requirements

1. Spectral AOD (UV-VIS-SWIR) st. dev. of AOD within the greater of +/- 0.02 or +/-0.05AOD
2. Column integrated aerosol absorption optical depth (AAOD) within +/-0.02 for AOT>0.1
3. Aerosol morphology, column averaged in cloud-free and above clouds in cloudy (required) and vertically resolved (goal). Identify spherical and non-spherical particles and determine the degree of non-sphericity.
4. Aerosol real part of the refractive index across the solar spectrum (UV–VIS–SWIR) resolved into two particle modes. One standard deviation of retrievals to be within ±0.02 for when total AOD > 0.10 in the midvisible. Column averaged in cloud-free and above clouds in cloudy regions.
5. Aerosol size distribution resolved into two particle modes. Modal effective radius for each mode must be determined to with ±10% and effective variance to within ±50%. One standard deviation of retrievals must fall within these uncertainties for AOD > 0.10. Column averaged in cloud-free and above clouds in cloudy regions.
6. Column particle number concentration (±100%) attributed to two size ranges corresponding to a coarse mode (roughly supermicron) and to an accumulation mode (roughly 0.10 μm to 1.0 μm)

Vertically resolved requirements/goals

1. Aerosol extinction (for 1.5 km aerosol layer in free troposphere, 500 m in boundary layer)
    a. 0.065 km^{-1} ($\Delta\tau_a$=0.098 for Δz_a=1.5 km) when single scattering albedo is 0.95
    b. 0.025 km^{-1} ($\Delta\tau_a$=0.038 for Δz_a=1.5 km) when single scattering albedo is 0.85
2. Vertical distribution of aerosol single scattering albedo to within ±0.02 in 1.5 km aerosol layers in the free troposphere, 500 m resolution in the boundary layer.
3. Aerosol morphology. Identify spherical and non-spherical particles and determine the degree of non-sphericity. (goal)
4. Real part of the refractive index for a 1.5 km aerosol layer in the free troposphere, 500 m in the boundary layer to characterize aerosol type, anthropogenic component and infer water content of the aerosol in vertical layers. The goal is to obtain the vertically resolved real part of the refractive index to within ±0.02 for the dominant aerosol mode in the layer when extinction in the layer exceeds 0.05 km^{-1}. In situations when no aerosol mode dominates, or when extinction in the designated vertical resolutions is insufficient for a robust retrieval, the goal is to provide as much vertical

information about the real part of the refractive index as possible and to define the uncertainties of the retrieval.

5. Vertical distribution of aerosol size distribution for a 1.5 km aerosol layer in the free troposphere, 500 m in the boundary layer to characterize aerosol type and anthropogenic component in vertical layers. The requirement is to identify the vertically resolved effective radius of the dominant mode to within ±20% in the layer when extinction in the layer exceeds 0.05 km^{-1}. In situations when no aerosol mode dominates, or when extinction in the designated vertical resolutions is insufficient for a robust retrieval, the goal is to provide as much vertical information about the size distribution parameters as possible and to define the uncertainties of the retrieval.
6. Vertically resolved particle number concentrations (± 100%); vertical resolutions of 500 m in boundary layer and 1.0 to 1.5 km in the free troposphere.

3.0 Atmospheric Radiation

There are six science questions that were judged to be the highest priority in the area of atmospheric radiation and are given in order of priority with question 3.1.1.1 as the highest priority since it acts as the "volume" dial on climate change response to anthropogenic forcing.

The radiation questions stand on their own, but are also clearly related to the cloud, aerosol, convection, atmospheric circulation, and atmosphere-surface exchange questions in other sections of this document. While the radiation science questions tend to be large time and space scale questions related to climate change, the related cloud and aerosol questions contain an additional focus on smaller time/space scale processes. Solving climate change requires both of these perspectives. This perspective is shown in the Figure 3.0.1 below. Process studies are critical to improving physical processes in climate models, while long time/space scale observations are key to testing the ability of climate models to accurately predict climate change.

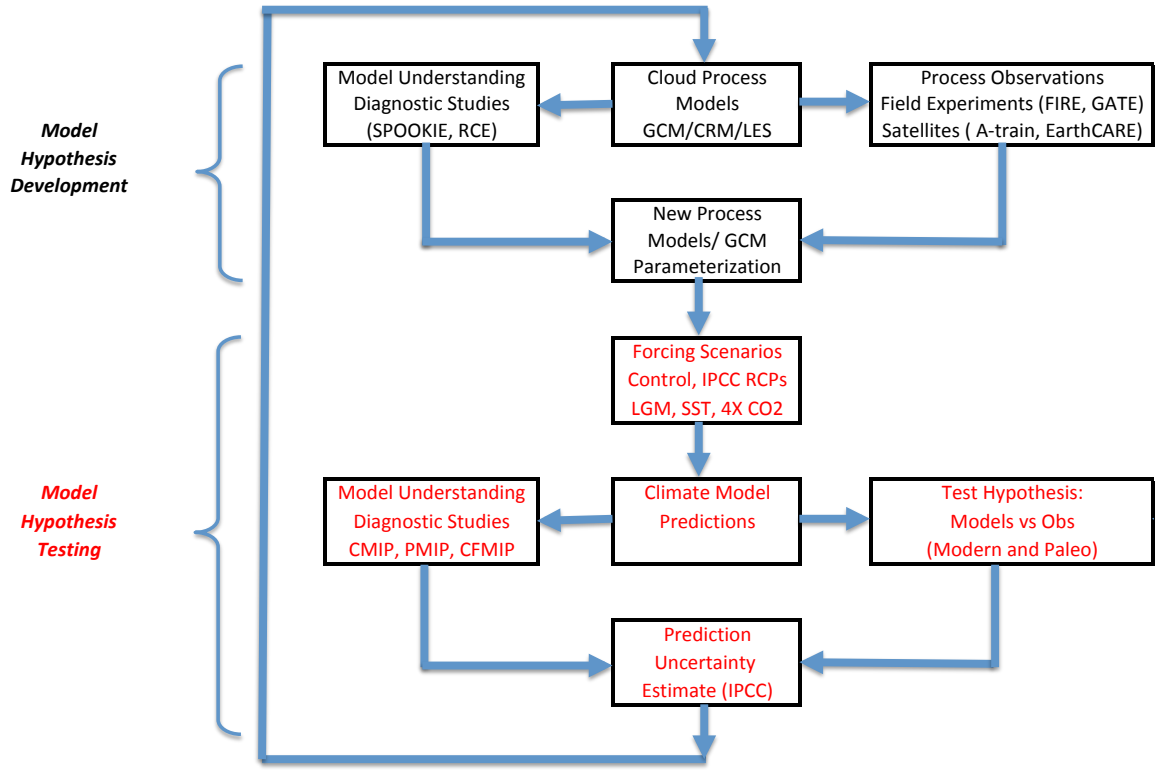

Figure 3.0.1: The interaction of climate model development and testing at short time scales (process in black text) versus long time scales (climate change in red text).

The radiation science questions contain some common themes. The most obvious of these is the need for higher accuracy in a wide range of satellite observations: top of the atmosphere (TOA) and surface radiative fluxes, passive and active cloud properties, passive and active

aerosol properties, Total and Spectral Solar Irradiance, Precipitation, as well as in-situ ocean heat storage observations and boundary layer temperature/humidity profiles. New observations of Far-Infrared spectra also are key to two of the science questions. Finally, the potential use of Ice Bridge type (systematic) aircraft flights to achieve sufficient statistical sampling of cloud and radiation processes is a common theme.
Several crosscutting recommendations also emerged from the discussions and these are listed and discussed after the 6 science questions.

3.1.1 What observations are needed to reduce cloud feedback uncertainty by at least a factor of 2 versus IPCC AR5 results?

Uncertainty in climate sensitivity remains a factor of 4 for 90% confidence bound (*IPCC*, 2007, 2013). Climate sensitivity uncertainty dominates uncertainty in economic impacts of climate change (*SCCM*, 2010). Total feedback uncertainty in turn is dominated by cloud feedback, a factor of 4 larger than water vapor/lapse rate or surface albedo feedback uncertainties (Figure 3.1.1.1 below). Low clouds contribute the majority of the cloud feedback uncertainty; models and observations agree on a positive high cloud feedback, but the model mechanism is unclear.

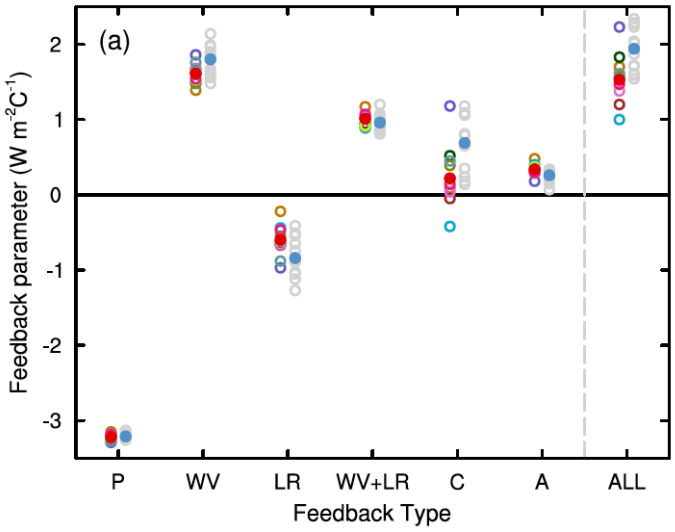

Figure 3.1.1.1: Climate model feedback uncertainties. P = Planck, WV = Water Vapor, LR = Lapse Rate, C = Cloud, A = Surface Albedo feedback. Greyed circles are CMIP3 results, Colored circles are CMIP5.
(figure 9.43 from IPCC, 2013)

3.1.1.1 What are the cloud and radiation properties, with sufficient interannual and decadal climate change accuracy, necessary to reduce cloud feedback uncertainty by at least a factor of 2?
While radiative fluxes can be used to directly estimate decadal changes in cloud radiative effect and SW cloud feedback (*Soden et al.,* 2008), independent verification of the cloud property changes that cause the changes in radiative balance are required to determine if climate models produce cloud feedback by the correct changes in cloud properties. Independent verification of cloud feedback from radiative fluxes and cloud properties will represent a high confidence verification of the cloud feedback physics in climate models. Accuracy requirements are taken from Wielicki et al. 2013 (see their Figure 3b) for SW and LW fluxes for uncertainty factor U_a of 1.2. Requirements for cloud properties require an analogous study to define.

Question	Observation	Resolution time and space	Accuracy (95% conf)	Precision
3.1.1.1	TOA SW TOA LW Net flux Cloud fraction, height, temperature Optical depth Emissivity Particle phase, size Surface air temperature	Monthly, Annual Regional, Zonal, Global	SW:0.3% LW:0.5% T_s: 0.07K for global annual avg. U_a = 1.2 for cloud and T_s as well (Wielicki et al. 2013)	Sufficient to retrieve geophysical variable anomalies at decadal climate change accuracy

3.1.2 What observations and modeling are required to determine the anthropogenic aerosol forcing for direct, indirect and semi-direct effects?

Anthropogenic aerosol radiative forcing uncertainty dominates the uncertainty of anthropogenic radiative forcing over the last several decades. Because aerosols primarily provide a negative climate forcing, aerosols also confuse efforts to use past warming observations to constrain climate sensitivity. Aerosol-cloud interactions force cloud changes with climate forcings that are even more uncertain than those from the aerosols alone. Figure 3.1.2.1 shows recent uncertainty results from the IPCC AR5 (*IPCC*, 2013, Figure 8.13). A factor of 3 total uncertainty in anthropogenic forcing results.

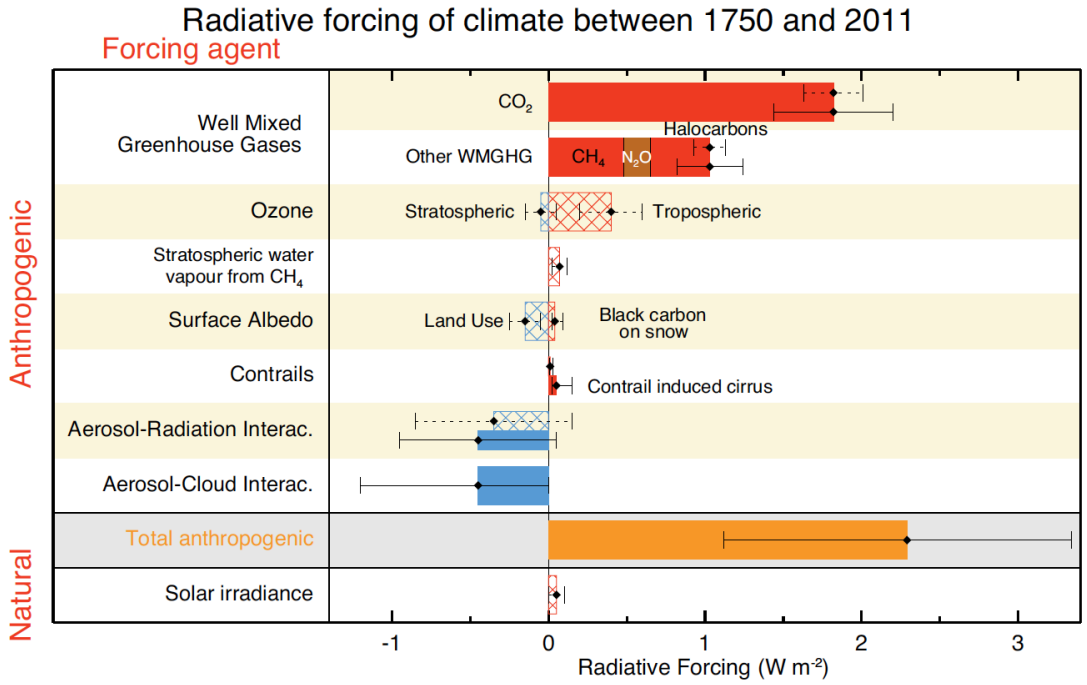

Figure 8.15 | Bar chart for RF (hatched) and ERF (solid) for the period 1750–2011, where the total ERF is derived from Figure 8.16. Uncertainties (5 to 95% confidence range) are given for RF (dotted lines) and ERF (solid lines).

Figure 3.1.2.1 Uncertainties in anthropogenic radiative forcing (IPCC, 2013)

3.1.2.1 What observations and modeling are required to reduce uncertainty in anthropogenic aerosol by at least a factor of 2 relative to the AR5 IPCC estimate?

Question	Observation	Resolution time and space	Accuracy (95% conf)	Precision
3.1.2.1	Anthropogenic aerosol radiative forcing. Aerosol properties (physical, radiative, composition), cloud properties, atmospheric dynamic state, radiative fluxes.	Global, Zonal, Regional; Seasonal and Annual means	0.2 Wm^{-2}	Sufficient for geophysical retrievals at required accuracy

3.1.3 What is the TOA global net radiative balance, variability, and change over time, and their explanation?

The warming hiatus has brought into focus the need for a rigorous understanding of global net radiative balance (90% is ocean heat storage) but challenges remain in both satellite radiation and ocean in-situ observations (*Stephens et al.* 2012; *Loeb et al.,* 2012). Global net radiation is currently constrained for the decade average by ocean in-situ heat storage (ARGO), and for inter-annual variations from the satellite radiation budget (CERES).

3.1.3.1 What is the annual net radiation (1σ) and uncertainty in interannual variations to 0.1 Wm^{-2} (1σ)?

Question	Observation	Resolution time and space	Accuracy (1σ)	Precision
3.1.3.1	TOA SW TOA LW Net flux Ocean temperature profile Surface temperature Total solar irradiance	Global Annual	0.3 Wm^{-2} annual net 0.1 Wm^{-2} interannual	Consistent for geophysical retrievals at desired accuracy

3.1.4 Can we resolve the 15 Wm^{-2} inconsistency in the global net surface/atmosphere energy balance to within less than 5 Wm^{-2} ?

Attempts to reconcile the best estimates of surface radiative, sensible, and latent heat fluxes at the surface, or atmospheric column heating/cooling have failed to achieve consistency within better than 15 Wm^{-2} (*Loeb et al*., 2014; *Stephens et al.,* 2012). This discrepancy represents roughly 20% of the global mean precipitation when expressed as latent heat. Reconciling the inconsistency will help us understand how trends in surface net radiation

(R_a) + sensible heating (S) relate to global trends in latent heating (LP) as shown in Figure 3.1.4.1 below.

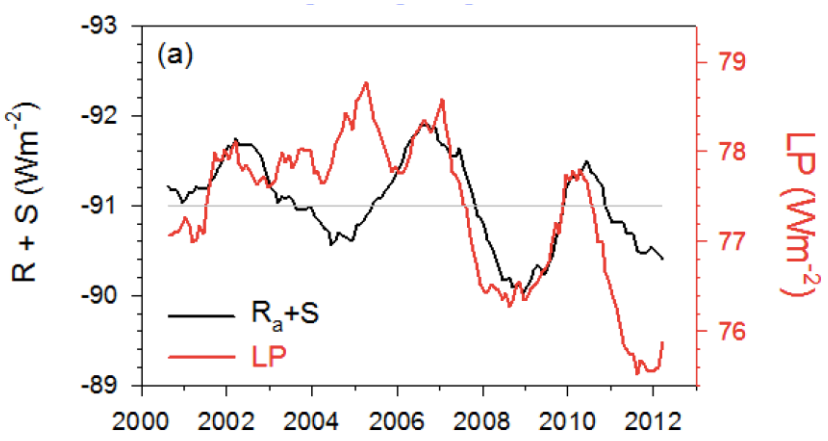

Figure 3.1.4.1: Interannual Variations of global annual (12 month running average) latent heat from precipitation observations (LP in red), net radiation (R_a), and sensible heat flux (S) in black. (Figure 7a from Loeb et al. 2014).

3.1.4.1 What observations are required to close the surface and atmosphere net energy budget to within less than 5 Wm^{-2} ?

Question	Observation	Resolution time and space	Accuracy (1σ)	Precision
3.1.4.1	Precipitation Surface radiative fluxes Surface latent heat flux Surface sensible heat flux TOA fluxes Cloud profile, properties	Zonal and Global annual means	Total budget to less than 5 Wm^{-2}, components to ~ 3 Wm^{-2}	Consistent for geophysical retrievals at desired accuracy

3.1.5 How is the Arctic radiative environment changing as sea ice extent rapidly decreases?

The rapid reduction of sea ice extent in the arctic is changing the radiation environment so rapidly that TOA SW and LW flux trends are statistically significant after only 12 years of observations (*Hartmann and Ceppi,* 2014). It remains uncertain, however, if changing clouds in the arctic are acting to dampen or amplify the surface albedo feedback caused by reducing sea ice cover and thickness in the arctic. Figure 3.1.5.1 shows that statistically significant trends in TOA radiative fluxes for reflected SW and emitted LW are only found in the arctic. As expected for decreasing sea ice extent, reflected SW TOA flux is decreasing while emitted thermal LW TOA flux is increasing. Increased open ocean waters in the spring, summer, and fall in the arctic are expected to change moisture levels and therefore cloud properties. The challenge is to separate the arctic surface and cloud changes into separate feedback components.

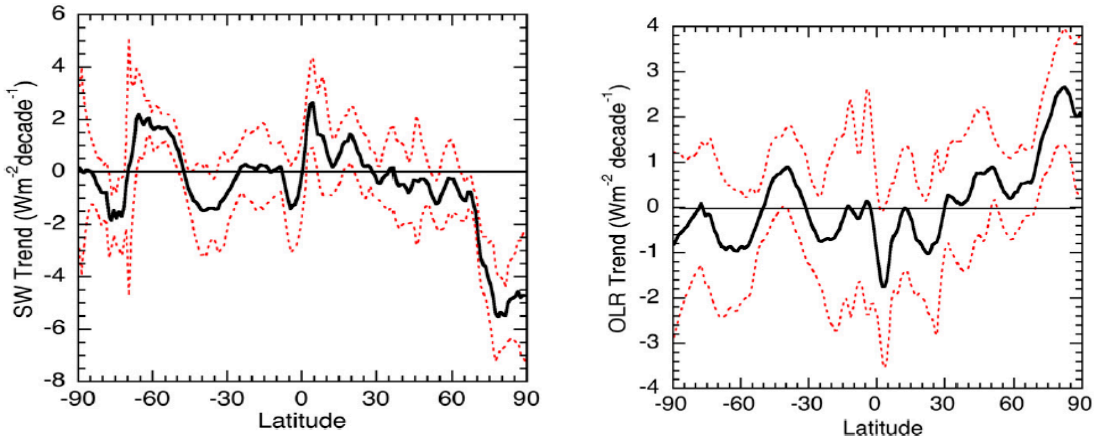

Figure 3.1.5.1: Zonal mean trends in reflected shortwave flux (SW on left) and emitted outgoing longwave flux (OLR on right). Solid black line shows the linear trend over the 2000-2012 period using CERES observations. The red dashed lines show the 5% and 95% confidence bounds on the trends. Taken from Hartmann and Ceppi, 2014.

3.1.5.1 Do clouds in the arctic increase or reduce the rapid warming there? Determine the arctic cloud feedback to within 25% of the sea ice extent driven surface albedo feedback.

Question	Observation	Resolution time and space	Accuracy	Precision
3.1.5.1	Sea ice extent TOA fluxes Surface fluxes Cloud properties Aerosol properties	Arctic Ocean regional and mean. Seasonal and annual means.	Arctic cloud feedback uncertainty to within 25% of sea ice albedo feedback.	Consistent for geophysical retrievals at desired accuracy

3.1.6 Can we observe and confirm the water vapor greenhouse effect and feedback in the far-infrared?

While we have tested the broadband water vapor greenhouse effect using CERES broadband observations, we have yet to verify this effect and the resulting water vapor feedback using spectrally resolved global spaceborne observations of the far-infrared spectral region (15 – 50 □m wavelengths). All of our current satellite infrared spectral observations are limited to mid-infrared from 3.5 to 15 □m wavelength. As a result we have not spectrally observed half of the infrared radiative energy emitted to space. In the polar regions, 60 to 65% of the energy is emitted in the far infrared (see Figure 3.1.6.1 below) (*Mlynczak et al.,* 2001). Most of the water vapor greenhouse effect and water vapor feedback is in the far-infrared. This observation would represent a key closure and verification of our radiative modeling understanding of 1/2 of the Earth's energy emitted to space.

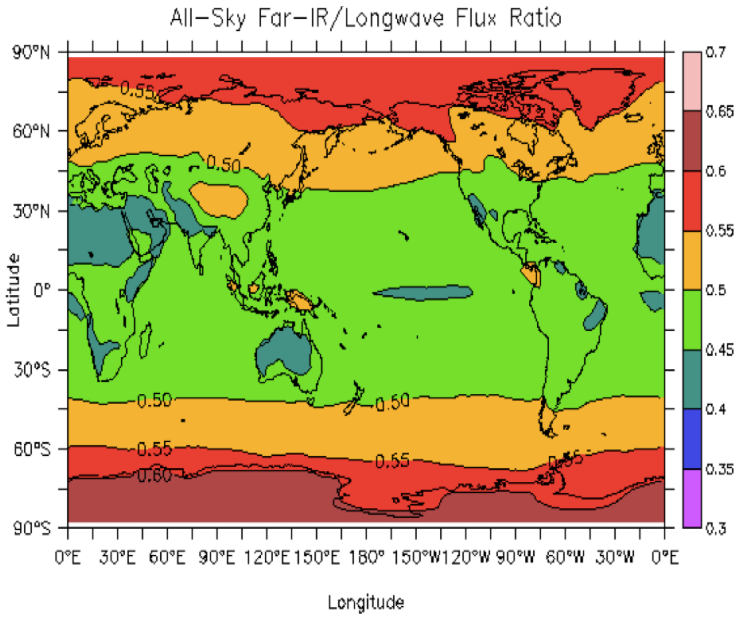

Figure 3.1.6.1: Ratio of the far-infrared radiative energy (>15 μm) to the total infrared energy emitted by the Earth to space. The ratio is given for all-sky conditions including the effects of clouds.

3.1.6.1 What is the far-infrared absorption spectrum at the spectral resolution, coverage, and accuracy sufficient for verification of the water vapor greenhouse effect and water vapor feedback in climate models?

Question	Observation	Resolution time and space	Accuracy	Precision
3.1.6.1	Far IR spectral emission to space (15-50 μm) at 1 cm^{-1} spectral resolution. Cloud property profiles Atmospheric temperature and water vapor profiles	Zonal and Global, Seasonal and Annual averages	TBD from Climate model and observation OSSEs	Consistent for geophysical retrievals at desired accuracy

3.1.7 Radiation Cross-Cutting Recommendations

1. Observing System Simulation Experiments (OSSEs) should be more widely used to improve the rigor of NASA observing system requirements, both for satellites as well as aircraft and field experiments.

While OSSE approaches have been used in weather prediction for some time, very few climate observations have taken advantage of this approach to derive more objective observing requirements (e.g. accuracy, sampling). OSSEs can be applied to decadal climate change observations (using climate models and observation simulators), climate process observations (using GCM/CRM/LES models), as well as field experiments (using GCM/CRM/LES models). OSSEs provide an approach to requirements that is closer to a rigorous hypothesis test. Models represent our physical hypothesis about the climate system. While OSSEs are not justified if models are grossly unrealistic, in most cases they can be a critical tool for improving the rigor of observation requirements. Close coordination is required between modeling and observation scientists.

2. The NASA remote sensing community needs to rigorously determine the level of stability of biases in satellite retrieval algorithms that are used to determine decadal climate change.

Current satellite retrieval algorithms focus entirely on random and bias errors for instantaneous retrievals appropriate to process or weather applications. These algorithms have not been evaluated for their ability to determine decadal climate change. A typical assumption is that they will perform without problem as long as the algorithm code remains constant. But most satellite retrievals have bias errors that greatly exceed the magnitude of decadal climate change. Studies need to be performed that these biases are sufficiently stable during climate change to avoid significant aliasing errors in deriving climate change from remote sensing algorithms.

3. Science questions need to be based on quantitative specific goals, **as opposed to qualitative general research areas of interest. This is critical for defining rigorous observing requirements.**

Science questions which are qualitative in nature are good for exploring totally unknown questions, but Earth science has been attacking such questions for over 30 years. It is time to move from qualitative to quantitative questions: in essence quantitative hypothesis tests, typically with observations testing model hypotheses. This means developing a Traceability Matrix with a quantification of uncertainty at each step. Figure 3.1.7.1 shows a schematic of such a traceability matrix for the challenge of observing decadal climate change and using such changes to test climate model predicted decadal change.

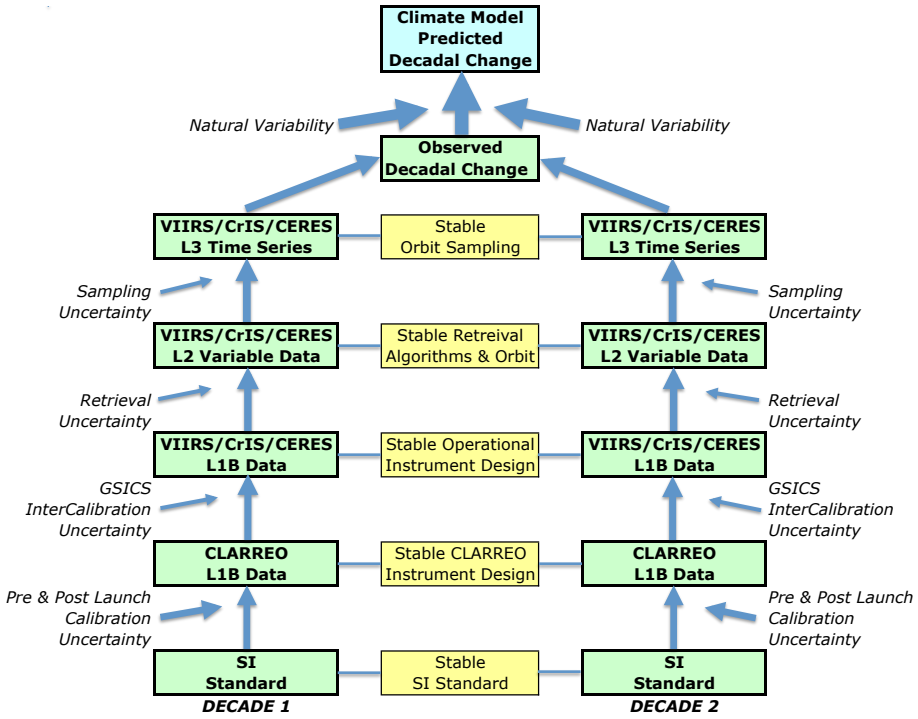

Figure 3.1.7.1 An example of uncertainty traceability for comparison of observed decadal change to climate model predicted decadal change. (From Trenberth et al. 2013)

References (3.0):

Hartmann, D. L. and P. Ceppi, 2014: Trends in the CERES Dataset, 2000–13: The Effects of Sea Ice and Jet Shifts and Comparison to Climate Models. *J. Climate*, **27**, 2444-2456.

IPCC, 2013: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp.

Loeb, N. G., et al., 2012: Observed changes in top-of-the-atmosphere radiation and upper-ocean heating consistent within uncertainty *Nature Geoscience* **5**, 110–113 (2012) doi:10.1038/ngeo1375.

Loeb, N., D. Rutan, S. Kato, and W. Wang, 2014: Observing Interannual Variations in Hadley Circulation Atmospheric Diabatic Heating and Circulation Strength. *J. Climate*. doi:10.1175/JCLI-D-13-00656.1, in press.

Mlynczak, M. G., et al. (2001), The far-infrared: A frontier in remote sensing of Earth's climate and energy balance, in Optical Spectroscopic Techniques, Remote Sensing, and Instrumentation for Atmospheric and Space Research IV, edited by A. M. Larar and M. G. Mlynczak, Proc. SPIE Int. Soc. Opt. Eng., 4485, 150– 158.

Soden, B. J., I. M. Held, R. Colman, K. M. Shell, J. T. Kiehl, and C. A. Shields, 2008: Quantifying climate feedbacks using radiative kernels. *J. Climate*, **21**, 3504–3520.

Stephens, G. L. et al., 2012: An update on Earth's energy balance in light of the latest global observations. *Nature Geoscience,* DOI: 10.1038/NGEO1580.

Trenberth, K.E, et al., 2013: Challenges of a sustained climate observing system. In *Climate science for serving society: Research, modeling, and prediction priorities.* G.R. Asrar and J. W. Hurrell, Eds. Springer Press, 480pp.

Wielicki, B. A., et al., 2013: Climate Absolute Radiance and Refractivity Observatory (CLARREO): Achieving Climate Change Absolute Accuracy in Orbit. *Bull. Amer. Met. Soc.*, **94**, 1519-1539.

4.0 Atmospheric Dynamics

4.1 Circulation

4.1.1 To what degree is the Brewer-Dobson stratospheric circulation accelerating?

4.1.1.1 How will variability and trends of the BDC affect trends and distributions of trace species?

4.1.1.2 How will wave-driven chemical transport, from the mean circulation and mixing, in the stratosphere change in this century?

4.1.1.3 How will intra- and interannual variability of the BDC (e.g., QBO and sudden stratospheric warmings) change in this century?

The Brewer-Dobson circulation (BDC) is critical to understanding the evolution of stratospheric ozone as well as gases relevant to the overall climate, such as water vapor. The BDC governs the lifetime of ozone destroying gases and temperature in the stratosphere (a principal factor in ozone destruction and in the input of water vapor in the tropics). This circulation includes the deep scale circulation (extending all the way into the mesosphere), which is driven primarily by Rossby-wave breaking in the middle to upper stratosphere and a shallower branch driven largely by wave-breaking near the subtropical jet. Since the BDC depends on tropospheric wave forcing and the dynamical (temperature and winds) state of the stratosphere (which affects wave propagation and the locations of the wave breaking that drives the circulation), the BDC can be considered a "proxy" for the overall dynamical state of the stratosphere. The fundamental theoretical framework for understanding the BDC is well-established (*Haynes et al.,* 1991), but important details, including the role of unresolved gravity waves, particularly for the shallow branch, are not well-quantified. Also, the mechanisms of driving the BDC in the tropics, where the classic "downward control" theory breaks down, are an area of active research (*Ortland and Alexander,* 2014). Intraseasonal and interannual variations in the BDC (e.g., sudden stratospheric warmings, ENSO, and QBO variability) have a clear impact on tropical tropopause temperatures and hence the input of water vapor into the stratosphere. The BDC has also been shown to impact tropical convection (*Eguchi and Kodera,* 2007) and tropospheric ozone *(Neu et al.,* 2014).

Question	Observation	Resolution time and space	Accuracy	Precision
4.1.1.1	H_2O N_2O	H_2O: Vertical and temporal to resolve tape recorder N_2O: day and night	2%	H_2O < 0.2 ppm N_2O < 5 ppb
4.1.1.2	O_3, temperature	**Space:** 1 degree, 1 km vertical resolution, **Time:** 3-hourly for T, 6-12 hourly for u and v. u and v direct measurements should be focused on low latitudes.	T < 1K O_3 < 2%	T < 2K O_3 < 5%
4.1.1.3	CH_4, SF_6, CO_2	**Space:** tropics, midlats, polar, 10-20 degrees resolution; Tropopause to 30 km; 1-2 km Vertical Resolution. **Time:** 2 times/month, decadal	SF_6 (2%) CO_2 (.1 ppm) CH_4 (10 ppb)	

4.1.2 How is the mid-to-upper stratosphere temperature responding to GHG increases?

4.1.2.1 How do N_2O, CH_4, CFCs, HCFCs, and HFCs contribute to this change?

4.1.2.2 Are there direct dynamical responses to this temperature change?

4.1.2.3 What is the quantitative variability (both annual and interannual) of temperatures in the mid-to-upper stratosphere?

4.1.2.4 Can we apportion changes amongst various GHGs, and can we detect feedbacks of temperature changes on wave propagation?

Long-term stratospheric temperature trends will occur most prominently in the upper stratosphere as a result of GHG changes. The issue is crucially important because of its direct impact on the photochemistry of ozone in the stratosphere. As CO_2 increases, stratospheric temperature decreases, and ozone increases.

Current observations of stratospheric temperatures are mainly derived from radiosondes and

satellite retrievals. While radiosondes provide reasonable ground-based coverage of the lower stratosphere, the upper stratosphere is primarily observed from satellite systems such as the Stratospheric Sounding Unit (SSU) and the Advanced Microwave Sounding Unit (AMSU). The satellites provide global coverage, while radiosondes provide good coverage over continental regions and sparse coverage over the oceans (particularly in the SH). In the lower stratosphere, temperatures have decreased about 1K since 1979 (*Hartmann et al.,* 2013). The middle-to-upper stratosphere has also cooled, but this change is much less certain because of the quality of the atmospheric observations from the SSU instrument and the merging with the AMSU observations (*WMO,* 2014).

Cross cuts:

- Temperature changes are intimately related to the changes in the Brewer-Dobson circulation.
- Temperature changes drive ozone changes in the mid-to-upper stratophere.
- Related to changes in stratospheric sudden warmings.
- Related to changes in temperatures at the cold-point tropopause and thereby dehydration of the stratosphere.

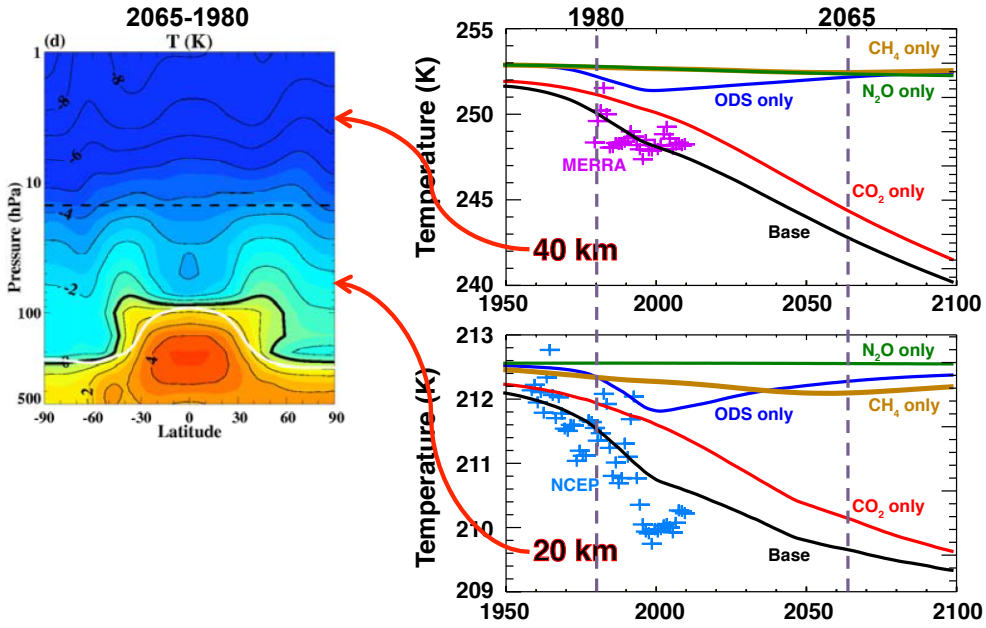

Figure 4.1.1. Model simulated long-term changes in tropospheric and stratospheric temperature based on the IPCC SRES A1B (medium) scenario. The right-hand panels show the global/annual averaged temperature response at 20 km and 40 km to the changes in CO_2 (red line), CH_4 (orange line), N_2O (green line), and all halogenated ozone depleting substances (ODSs - blue line). The response to all source gases combined is shown by the black line. Also shown are global/annual mean temperature observations from the NCEP reanalyses for 1958-2012 at 20 km (blue crosses) and the NASA/MERRA analyses for 1979-2012 at 40 km (red crosses). Stratospheric temperature changes are dominated by CO_2 IR induced cooling. The ODS-induced ozone loss causes reduced solar heating in the

stratosphere and has a maximum effect in 2000. The reduced solar heating caused by N_2O-induced ozone loss is minor throughout 1950-2100. Methane-induced cooling of the stratosphere (mainly due to increased water vapor) is ~0.5K from 1950-2060 at both 20 km and 40 km.

The annual mean difference between 1980 and 2065 (vertical dashed lines) due to the combined response to all source gases is shown in the left hand panel. The impact due to ODS loading is nearly identical in 1980 and 2065, so that the combined effect is due to increases in GHGs, with warming in the troposphere and cooling in the stratosphere. The lower stratospheric response is due to a combination of CO_2 IR cooling and acceleration of the Brewer-Dobson circulation, and is stronger in the tropics than in the extra-tropics. The total cooling increases with height and is due mainly to the CO_2 IR effect in the upper stratosphere.

Question	Observation	Resolution time and space	Accuracy	Precision
4.1.2.1	Upper stratosphere N_2O, CH_4, CFCs, HCFCs, HFCs and temperature	Global, weekly	Trend detection.	
4.1.2.2	Dynamical tracer, temperature	N_2O, ozone, CO, T, winds. resolve responses to wave events. Global, daily		< 2%
4.1.2.3	mid-to-upper stratosphere temperature	Global, monthly	<0.1 K	< 2K
4.1.2.4	CO_2, CH_4, N_2O, temperature, winds	Global, monthly. Winds and T daily.		

4.1.3 What are the transport pathways from the PBL to the stratosphere?

Question	Observation	Resolution time and space	Accuracy	Precision
4.1.3.1	Dynamical tracer, temperature			

4.1.4 How does deep convection contribute to troposphere-stratosphere coupling?

4.1.4.1 How, when, and where does convective transport to the stratosphere occur?

4.1.4.2 What are the characteristic transport behaviors for maritime and deep continental convection?

4.1.4.3 What are the transport pathways and time scales from the planetary and marine boundary layer (PBL; MBL) to the stratosphere?

4.1.4.4 Does summertime injection of H_2O over the US introduce chlorine activation in the lower stratosphere?

4.1.4.5 What is the interplay between emission (sources), chemical lifetime, and transport timescale for short-lived species?

There are significant uncertainties in quantifying convective influences on the lower stratosphere. Although most convective detrainment occurs below ~14 km, extreme overshooting convection can reach up to altitudes ~20 km. No available satellite product provides a complete picture of convective cloud-top heights: geostationary infrared data underestimate cloud tops, TRMM precipitation radar is biased towards continental systems with strong updrafts, and A-TRAIN (CALIPSO and CLOUDSAT) miss the late afternoon and early evening peaks in continental convective activity. Entrainment and detrainment throughout the full vertical profile is likely important for maritime convection but is poorly understood.

Cross cuts: Strong links between convection and composition in the upper troposphere and lower stratosphere. Obvious strong links with clouds and aerosols.

Question	Observation	Resolution time and space	Accuracy	Precision
4.1.4.1	Dynamical tracers in convection			
4.1.4.2	Tracers in maritime and continental convection			
4.1.4.3	Tracers in PBL, MBL, stratosphere			
4.1.4.4	H_2O, chlorine species			
4.1.4.5	Short-lived gases			

4.1.5 How do monsoon circulations contribute to troposphere-stratosphere coupling?

4.1.5.1 How do monsoonal circulations influence tropical-extratropical UTLS coupling?

4.1.5.2 What is the depth of monsoon influence into the lower stratosphere, and how important are monsoons for the stratospheric overworld?

4.1.5.3 How is deep convection coupled to monsoonal circulation and transport in the UTLS?

4.1.5.4 Are there stratospheric influences on monsoon behavior in the troposphere?

Monsoon circulations are a fundamental aspect of the large-scale circulation in the tropics and subtropics, forced as a dynamic response to latent heating within persistent tropical convection. These flows include large-scale persistent anticyclones in the subtropical upper troposphere, with the largest and strongest anticyclone tied to the Asian summer monsoon. Monsoonal circulations enhance transport into and out of the tropics along their eastern and western flanks, although they also tend to isolate air inside the anticyclone. Because this isolated air is often tied to the outflow of deep convection, it is chemically distinct and characteristic of tropospheric composition; the circulation and chemical behavior extend into the lower stratosphere. Monsoons exhibit a high degree of dynamic variability, reflected in constituent behavior that is poorly understood.

Cross cuts: Monsoons play a role in constituent transports and hence tropospheric and stratospheric composition and their coupling. Monsoons are linked to radiation through constituent and aerosol behavior, such as the anomalous ozone, H_2O, and aerosols within monsoon regions. Organized, persistent convection both drives and is influenced by monsoonal circulations.

Question	Observation	Resolution time and space	Accuracy	Precision
4.1.5.1	Tracers in monsoonal convection wrt tropical-extratropical coupling. water vapor and ozone, CO, temperature; trace species including HCN, SO_2, NOx, black and organic carbon	**Space:** Asia and North America, UTLS, fine vertical resolution (~2km) **Time:** NH summer (June to August),		
4.1.5.2	Tracers in lower stratosphere above monsoonal convection: H_2O O_3, CO, T; trace species including HCN, SO_2			
4.1.5.3	Tracers of deep convection. PV (from reanalysis); temperature, vertical velocity; OLR as convective proxy; trace species including CO, ozone, H_20	Satellite measurements of dynamical variables and trace species in UTLS. Space: Asia, minimum 3-5 km vertical resolution Time: June-August		
4.1.5.4	Tracers of stratosphere troposphere exchange			

4.1.6 What are the processes controlling atmospheric transport between the tropics and extra-tropics and between hemispheres, and how are they changing?

4.1.6.1 What are the rates of extra-tropical/tropical transport and NH-SH exchange?

- 4.1.6.1.a What are its mechanisms?
- 4.1.6.1.b Where and when is it occurring (i.e., pathways, spatial/temporal variation)?
- 4.1.6.1.c How could it change with climate?
- 4.1.6.1.d What does it tell us about global tropospheric OH?
- 4.1.6.1.e How do we define the "atmospheric" equator? The ITCZ? The mixing minimum?

4.1.6.2 Are the tropics expanding?

- 4.1.6.2.a Is the Hadley circulation changing?
- 4.1.6.2.b Are the subtropical jets shifting?

Interhemispheric transport refers to the movement of air mass (or tracers) between the NH and SH and essentially describes mixing at the largest meridional scales. It also results in exchanges of energy, momentum and water vapor, thereby potentially impacting large scale circulations. Past studies of interhemispheric transport have largely relied on comparisons of surface measurements of tracer species to derive global mean exchange times (e.g. *Geller et al.,* 1997). These studies have been relied on to validate transport in global circulation models, but provide only limited diagnostics to assess transport mechanisms, since the surface-based tracer measurements do not provide the spatial and temporal resolution necessary to resolve the detailed processes of cross hemisphere exchange, which are largely occurring in the upper troposphere during periods of high convective activity.

Changes in the location of the subtropical jet postulated in response to climate changes will result in shifts in precipitation patterns with concomitant effects on human populations, agriculture, and ecosystems in both the tropics and extratropics. Some recent studies of tropical widening over the past 3-4 decades have claimed poleward expansion of the tropics by as much as 3°/decade, while others have found much smaller and not yet statistically significant trends (*Seidel et al.,* 2008; *Davis and Rosenlof,* 2012) using a number of different metrics for defining the tropics. Attribution for the potential expansion is not clear, with ozone depletion as well as GHG increases considered as potential causes.

Cross-cutting: Questions related to interhemispheric and extra-tropical to tropical transport are directly related to the tropospheric and stratospheric composition topics through atmospheric oxidation chemistry (lifetime) and vertical transport into the stratosphere, both of which are dominated by processes in the tropics.

Question	Observation	Resolution time and space	Accuracy	Precision
4.1.6.1	Latitudinal gradient of one or more tracer (SF_6, HFCs) that has increasing concentration dominated by single hemispheric source; and tropospheric lifetime long relative to mean interhemispheric exchange timescale (~1.3 yr)	**Space:** Latitudinal measurements from NH mid-lat to SH mid-lat from surface to UT, ideally at a variety of longitudes (e.g. W Pacific, E Pacific, Atlantic, etc.) **Time:** Monthly/seasonal		
4.1.6.2	Continuous record of tracer to separate secular trend from interannual variability. CO_2, CH_4, N_2O, HFCs, SF_6			

4.1.7 What role do gravity waves (GWs) play in driving the large-scale circulation?

4.1.7.1 What are the sources of GWs?
4.1.7.2 What are the mechanisms for GW absorption and dissipation?
4.1.7.3 How does the large-scale environment control wave propagation and breaking?
4.1.7.4 To what extent does GW breaking lead to secondary wave generation?
4.1.7.5 What are the impacts GW wave driving on tracer transport?
4.1.7.6 How does GW breaking contribute to mixing and how does such mixing modify advective transport by the large-scale circulation?

GWs are ubiquitous in the atmosphere. These are unbalanced motions - not in geostrophic balance and, at higher frequencies not in hydrostatic balance - with buoyancy as their restoring force. Thus, GWs can propagate in any stably stratified region of the atmosphere, thus almost everywhere. Sources of gravity waves include flow over topography, convective updrafts impinging on stable layers, and *spontaneous* imbalance, in which balance is disrupted by advection. GWs transport horizontal momentum from their source region to where they break or dissipate. This momentum is in the direction of propagation. The propagation of GWs is controlled by the large-scale flow. In particular, GWs break or dissipate when the mean flow wind approaches the wave speed. Thus eastward mean flows support the propagation of westward propagating gravity waves, and *visa versa.*

Because of their small spatial scales (10s to 100s of kilometers) and short periods (minutes to hours) GWs are not observed by conventional meteorological or atmospheric observing systems. Observations depend on high-spatial resolution "snapshots" (e.g. from high vertical resolution GPS or balloon soundings), in which case wave properties must be inferred from

analyzing small-scale "wiggles" in data, or from small numbers of observing systems, such as ground-based lidars, that provide vertical profiles at high temporal frequency.

Therefore, while most atmospheric models, from GCMs to mesoscale models (e.g. WRF) now include parameterized treatments of GWs, these GW parameterizations are not sufficiently constrained by observations, and there is the risk that they can be used to "tune" models and compensate for other model deficiencies. A particular challenge to GW parameterizations is the treatment of non-topographic sources. A newly emerging challenge is the increasing spatial resolution of models, such that a portion of the GW spectrum can be explicitly resolved – i.e. there is a need for *scale-aware* GW parameterization. Improved observations and understanding of the GW spectrum is needed to construct parameterizations that address only a portion of this spectrum.

***Cross cuts**:* While the most prominent effects of GWs are their influences on the mean winds and overturning circulations, especially in the middle atmosphere, GWs also produce rapidly fluctuating vertical velocities and temperatures. These can initiate the formation of cirrus cloud in regions where it otherwise would be absent (e.g. *Haag et al*. 2004), and there is the potential for rectified effects on chemical reactions (e.g. *Hickey et al.* 2003).

Question	Observations (for all questions below)	Resolution time and space	Accuracy	Precision
4.1.7.1	Vertical temperature profiles	High temporal frequency, ~ 1 per minute	0.5 K	0.2 K
4.1.7.2	Temporally continuous observations of temperature	Multiple heights and at horizontal separations smaller than typical GW wavelengths, ~ 10 km	0.5 K	0.2 K
4.1.7.3	Winds	Vertical 100 m, horizontal 10 km	1 m/s	1 m/s
4.1.7.4				
4.1.7.5	Quasi-conserved chemical tracers	High vertical resolution – 100 m	Depends on background vertical gradient of tracer: ~dC/dz *100 m	
4.1.7.6				

References (4.1):

Davis, Sean M., Karen H. Rosenlof, 2012: A Multidiagnostic Intercomparison of Tropical-Width Time Series Using Reanalyses and Satellite Observations. *J. Climate*, **25**, 1061–1078. doi: http://dx.doi.org/10.1175/JCLI-D-11-00127.1

Eguchi, N. and Kodera, K., 2007, Impact of the 2002, Southern Hemisphere, stratospheric warming on the tropical cirrus clouds and convective activity. *Geophysical Research Letters*, 34*: doi: 10.1029/2006GL028744. issn: 0094-8276.*

Geller et al.,Tropospheric SF_6: Observed latitudinal distribution and trends, derived emissions and interhemispheric exchange time, *Geophys. Res. Lett.* 24, 675–678 (1997), doi: 10.1029/97GL00523

Haag, W., and B. Kärcher (2004), The impact of aerosols and gravity waves on cirrus clouds at midlatitudes, *J. Geophys. Res.,* 109, D12202, doi:10.1029/2004JD004579.

Hartmann, D.L., A.M.G. Klein Tank, M. Rusticucci, L.V. Alexander, S. Brönnimann, Y. Charabi, F.J. Dentener, E.J. Dlugokencky, D.R. Easterling, A. Kaplan, B.J. Soden, P.W. Thorne, M. Wild and P.M. Zhai, 2013: Observations: Atmosphere and Surface. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Haynes, P. H., Marks, C. J., McIntyre, M. E., Shepherd, T. G., Shine, K. P., 1991: On the ``downward control'' of extratropical diabatic circulations by eddy-induced mean zonal forces. *J. Atmos. Sci.,* 48, 651--678.

Hickey, M. P., T.-Y. Huang, and R. L. Walterscheid (2003), Gravity wave packet effects on chemical exothermic heating in the mesopause region, *J. Geophys. Res.,* 108, 1448, doi:10.1029/2002JA009363, A12.

Neu, J. L.,T. Flury, G.L. Manney, M. L.Santee, N. J. Livesey, & J. Wroden, 2014. Tropospheric ozone variations governed by changes in stratospheric circulation. *Nature Geosci.*, 7, 340-344.

Ortland, David A., M. Joan Alexander, 2014: The Residual-Mean Circulation in the Tropical Tropopause Layer Driven by Tropical Waves. *J. Atmos. Sci.*, 71, 1305–1322

Seidel, D., Fu, Q., Randel, W.J., Reichler, T.J.,(2008) Widening of the tropical belt in a changing climate, *Nature Geoscience*, 1, 21 - 24 , doi:10.1038/ngeo.2007.38

4.2 Convection

4.2.1 Outstanding problems of atmospheric convection

Atmospheric convection exists in a wide variety of forms and in a broad range of scales, from cumulus clouds, to cumulus congestus, deep cumulonimbus, to mesoscale convective systems. Each form has its unique characteristics and circulation features. It is rarely in a steady state, so that observations are needed during the growing, mature, and dissipating states of the convection, it is highly turbulent so that observations are needed on the sub-convective scale, and both warm and cold-cloud microphysics require data down to the micron scale. A full understanding of convection or any of the roles it plays in the atmosphere requires attention to the specific form taken by the convection as well as all of its internal properties. The interactions of convection with the wide variety of aerosol and important trace constituents of the atmosphere are among the factors that need to be specified in reference to the particular form of convection.

With all of this complexity, atmospheric convection is far from a solved problem. The major challenges in understanding atmospheric convection can be summarized in the following list:

1. Basic in-cloud properties poorly known
To advance knowledge of storm evolution, detailed vertical velocity, microphysical characteristics, and temperature are needed on the sub-kilometer scale and on the scale of minutes.

2. Interaction with humidity environment
The relationship of deep convective clouds and mesoscale convective systems to the humidity field is very poorly understood, especially regarding the way clouds themselves influence the large-scale humidity field. Smaller cumulus and moderate cumulus congestus interact with the humidity of the lower troposphere. Deep cumulonimbus and mesoscale convective systems are major factors in moistening the upper troposphere. Overshooting convection and the anvils of mesoscale convective systems may be affecting the water vapor (and other constituents) of the lower stratosphere. All of these cloud/humidity interactions in the troposphere and stratosphere suffer from a lack of highly accurate humidity data in the high atmosphere.

3. Combined thermal and shear environment
Cumulonimbus clouds occur in various modes that are a joint function of the environmental thermodynamic profile and wind shear. Ordinary cumulonimbus occur in weak shear, and have internal circulations in which the pressure perturbation is determined hydrostatically from the buoyancy. Supercell cumulonimbus occur in strongly sheared environments and develop in-cloud rotation that in turn affects the pressure field in the cloud. In larger mesoscale convective systems, mesoscale layered updraft and downdraft circulations develop as an adjustment to the environment wind shear and thermodynamic profile. The mesoscale systems contain convective-scale centers of action, where water is condensed, ice forms, and numerous chemical transformations and transports occur. Why and how the mesoscale systems evolve is still under study.

4. Surface conditions and diurnal forcing

Convection occurs in a wide range of forms and scales, from cumulus, to cumulonimbus, to mesoscale convective systems. The underlying surface and diurnal forcing determine which types of convection form. The diurnal cycle of convection is a strong function of the scale of the cloud phenomenon, with smaller convection peaking at the time of maximum solar heating. However, larger convection, especially mesoscale convective systems peak long after the peak heating. Often they maximize at night as a result of infrared cooling in the upper-level stratiform cloud layer at night over oceans. The differences between boundary layers over land and ocean lead to vastly different characteristics between maritime and continental mesoscale convective systems. For example, lightning occurs more frequently over land, although the strength of the lightning strikes over oceans is greater. The diurnal behavior on all time and space scales is highly complicated by terrain features such as mountain ranges and coastlines. Holistic understanding of these multiscale diurnal behaviors related to underlying surface conditions has not been achieved.

5. Self aggregation, upscale growth of convection and the nature of mesoscale convective systems

A fundamental question is why convection seeks mesoscale structures and organizations. Recent research is indicating that convection "self aggregates" as a result of seeking convective-radiative equilibrium. Once it reaches mesoscale proportions it adjusts to the thermal and shear environment to form mesoscale convective systems. Exactly how this adjustment is realized remains an important problem. It is known that often this adjustment takes the form of a storm with a leading line of convection and trailing stratiform region. However, when the environmental shear is more complex in speed shear, directional shear, or both, the manner in which the mesoscale motions adjust to the shear is not well understood.

6. Convective populations and interaction with large-scale dynamics

Large-scale circulation driven by tropical convection (especially, monsoons, the Madden-Julian Oscillation, and ENSO) are affected by the whole population of convection. The various forms taken by convective clouds occur in different combinations, comprising a population that is the heating element and vertical momentum exchanger within the larger-scale circulations. As the large-scale circulations develop the relative proportions of shallow, deep, and mesoscale convection vary. Understanding the scale interactions that are critical to large-scale circulations depend on the makeup of the cloud populations. Transports, scavenging, and chemical transformations within the large-scale circulations depend on the nature of the cloud population. The factors controlling the nature of tropical cloud populations are not well understood. Major field programs continue to aim at this understanding.

7. Aerosol environment effects on convection

Rosenfeld et al. (2008) hypothesized that aerosol effects on cloud microphysics can invigorate convection because smaller drops are lofted higher where ice microphysics processes are affected. However, this idea has not been verified, and Fan et al. (2013) have shown that the main effect of increased aerosol in the environment may be to increase the amount of cirriform cloud outflow from the convection. There is some evidence that small-to-moderate cumulus and cumulonimbus with low bases increase in height with increased

near surface aerosol concentration (*Li et al.,* 2011). It is not known whether larger cumulonimbus and/or mesoscale convective systems exhibit similar sensitivity.

8. Downdrafts and cold pools

Convective downdrafts spread out at Earth's surface. The spreading downdraft air is referred to as a cold pool, and cold pools are major factors in triggering new members of the convective populations. Like convection itself, cold pools exist in a spectrum of sizes, from a few km in dimension for small convective showers to up to 1000 km for large mesoscale convective systems (MCSs). The role of cold pools is a primary focus in efforts to understand how convective populations develop. They represent the surface areas affected by convective downward transport.

9. Time resolution

One of the primary inhibitors in understanding how convective processes vary around the globe is the lack of time resolution in observations from space.

4.2.2 Improving understanding of atmospheric composition through studies of convection and its environment

Convective clouds affect atmospheric composition, here defined as both the gaseous and aerosol constituency of the atmosphere. Atmospheric composition may also affect convection to some degree. The manner in which these interactions of convection, gases, and aerosol occur depends strongly on the nature of the convective phenomena. As noted above in Section 4.2.1, convection occurs on a wide range of scales, and the current understanding of convection is far from complete. Much can be gained by a joint effort to simultaneously understand the convection itself along with associated variations in atmospheric composition. Three focus areas for such studies can be identified:

4.2.2.1 Lightning, its production of NOx, the effects on ozone, and connections to storm parameters

Several processes of producing ozone into the troposphere are associated with lightning generation of nitrogen oxides (NO_x). Much is being learned about these processes from the NSF and NASA Deep Convective Clouds and Convection (DC3) field project (*Barth et al.,* 2014). Radar and lightning network measurements verified the close association of the production of graupel and lightning, while aircraft documented the presence of NO_x in outflow layers emanating from the convection. The success of this field program paves the way for using remote measurements to provide a global census of NO_x production by different forms of convection around the globe. In addition, field work and modeling are still needed for attaining the following objectives.

4.2.2.1.1 Quantify how different forms of convection affect lightning-generated NO_x production and subsequent ozone production, namely, ordinary thunderstorms, supercell convection, and mesoscale convective systems.

It is important to learn whether different types of storms produce different amounts of lightning-generated NO_x because of its potential production of O_3 in the upper troposphere

where O_3 is a radiatively active gas. Aircraft measurements suggest that NO_x mixing ratios in the outflow of MCSs are much greater than those in smaller storms, especially ordinary thunderstorms. Much of this difference can be attributed to the number and/or intensity of flashes occurring in the different types of storms, which can be connected to storm characteristics and storm environment and therefore storm types. Lightning characteristics of convection vary between land and ocean and from region to region. Understanding the basis of these differences in convection will be critical to understanding the NO_x production associated with convection. Aircraft field campaigns and subsequent analysis can provide simultaneous detailed information on case studies of cloud electrification, lightning discharges, and lightning-generated NO_x, providing the understanding necessary for the design and interpretation of spaceborne measurements. Spaceborne measurements can show the global census of convective systems with different lightning and NO_x production characteristics, and cloud-resolving models can demonstrate how the differences of dynamics and microphysics among these different types of convection affect ozone production. This research will go hand-in-hand with basic research on convection, which needs better observations of vertical velocity, microphysical properties, and thermodynamic fields to promote better understanding of convective evolution (Convective list item #1). It will also complement efforts to better understand how deep cumulonimbus and mesoscale convective systems adjust to environmental shear and stability (Convective list item #2 and #3). Spaceborne measurements can be used to connect lightning and NO_2 for North America. This topic is ***critical*** to pursue because of the potential for lightning-NO_x to affect O_3 in a climate-relevant region (upper troposphere).

4.2.2.1.2 Develop the capability to predict reliably lightning flash rates in models (from cloud resolving to global models) and to estimate lightning-generated NO_x from other flash characteristics, e.g. flash extent or energy.

To predict the chemical composition of trace gases for field experiment analysis, air quality forecasting, or climate studies, lightning flash rates must be reliably predicted. Recent field experiments can be used to further develop existing and/or develop new methods for predicting flash rates based on various storm parameters (e.g., amount of graupel, vertical velocity, or fluxes). In addition, new parameterizations of lightning-NO_x production based on flash length and/or energy can now be pursued by combining Lightning Mapping Array (LMA) data with aircraft data of NO and NO_2 mixing ratios. Cloud resolving models can test lightning and lightning-NO_x prediction. Models that parameterize convection can be improved and evaluated with CRMs and measurements. Understanding the physics leading to lightning discharges is needed to advance parameterizations of lightning flashes in convective clouds. To predict lightning-NOx in future climate scenarios, it is ***very important*** to have reliable flash rate predictions, which also will be useful for weather forecasts of lightning.

4.2.2.1.3 Measure the spectrum of vertical velocities within both convective-scale and mesoscale systems.

Such measurements are crucial to advancing understanding of convection. All nine items discussed above require better knowledge of in-cloud vertical air motion. These measurements need to be made with better penetrating aircraft platforms. But for development of statistics of vertical air velocities in convective clouds, the measurements

must be made with both cloud radars (e. g., W and Ka band) and precipitation radars (e.g., X, C, and S band). The measurements need to be made with ground-based, ship-based, airborne and spaceborne radars. Obtaining these measurements is ***very important*** since lightning and its associated NO_x production is related to both vertical velocity and ice-phase microphysics produced by strong vertical motions.

4.2.2.1.4 Determine whether downdrafts in deep convection bring lightning-generated NOx to the PBL affecting surface ozone mixing ratios.

Anthropogenic NO emissions continue to decline in the U.S. and Europe. However lightning-produced NOx remains an important source of tropospheric NOx . While boundary layer NOx Convective downdrafts transport the lightning-generated NOx into the boundary layer. Models and future field experiments and modeling studies need to be aimed at determining how much lightning-NOx is transported in downdrafts, and spaceborne measurements with high time resolution are needed to evaluate the global and regional statistics of downdrafts in order to evaluate their overall importance as an NOx sources at low levels of the atmosphere. Determining the contribution of lightning-NO_x to the PBL NO_x and O_3 mixing ratios is deemed to be an ***important*** topic to accomplish.

4.2.2.1.5 Test the hypothesis connecting wind shear to lightning flash length and NOx production from lightning.

It has been hypothesized that lightning flash length is related to wind shear. This hypothesis should be tested by conducting aircraft field experiments in weak and strong shear environments. If validated this hypothesis would suggested the NOx production is more effective in certain wind shear environments. The field studies should measure NO and NO_2. Addressing this topic is an opportunity to take advantage of the availability of LMAs. Learning how much the wind shear affects the lightning-NOx production would be an important factor in future parameterization lightning NOx production. It is ***important*** to evaluate this hypothesis because it will provide answers on how to improve lightning-NO_x parameterizations.

Question	Observation	Resolution time and space	Accuracy	Precision
4.2.2.1.1 4.2.2.1.4 4.2.2.1.5	Aircraft NO, NO_2 Doppler and polarimetric radar data lightning monitoring	1 km, 1 minute	10s pptv 1 dBZ 1 flash/min	10s pptv -20 dBZ < 1 flash/min
4.2.2.1.2	Doppler and polarimetric radar data lightning monitoring	1 km, 1 minute	1 dBZ 1 flash/min	-20 dBZ < 1 flash/min
4.2.2.1.3	vertical velocity from space and from rapid scan ground based radar	minutes, 1 km	10 cm/s	10 cm/s

4.2.2.2 Interaction between convection and environment via transport, scavenging and transformations

Deep convective clouds and mesoscale convective systems affect transport, scavenging and transformations occurring in cloud. The immediate environment of the convection can respond to the disturbance in ways that can further affect atmospheric composition. It is well known that overshooting convective cells locally extend into the stratosphere affecting the radiative balance and chemistry in the stratosphere. Conversely, stratospheric air can be transported into the upper troposphere as seen in DC3 measurements of stratospheric ozone being transported downward in the immediate vicinity of the leading anvil of an intense mesoscale convective system. Isolated convection and mesoscale convective systems contain intense convective cores, or "particle fountains", which are where updrafts, liquid water, and ice particles and likely key chemical transformations are concentrated. The organized layered circulation of mesoscale convective systems advects these fountains into its stratiform region, which is composed of aged and spread-out remnants of the fountains

Measurements in DC3, SEAC4RS, and other field programs successfully show that properties of convective outflow can be measured in such a way as to indicate the transformations and transport that the convection produces. These campaigns sampled the anvils of convective systems detecting a suite of trace gases (e.g., CO, CH_4, O_3, non-methane hydrocarbons, peroxides, HNO_3, SO_2, NO and NO_2) in the inflow and outflow regions. The outflow characteristics compared to inflow properties indicate the processes that have occurred in the convective system. Knowledge of certain key processes in the convective updrafts (fountains), especially scavenging, is as yet incomplete. To understand these processes, aircraft measurements must be combined with radar data that provide information on the storm physics and kinematics. Both DC3 and SEAC4RS have this unique capability, plus SEAC4RS measurements include in situ measurements in small convection providing data on vertical velocities, cloud particles, and trace gases. Further studies of the following types will build on these field studies.

4.2.2.2.1 Processes need to be examined on various timescales, different latitudes, and between land and ocean.

Because convective clouds occur in a wide variety of forms, with different vertical dimensions and vastly different horizontal scales, observational studies and modeling will need to be carefully organized around the these different scales of convective phenomena. The most crucial timescale is the diurnal cycle, and the diurnal variations of cloud related phenomena different for short-lived cumulus and cumulonimbus than for mesoscale convective systems. One of the fundamental problems of convective dynamics is to understand how various scales respond to diurnal radiative processes (Convective list item #4). Understanding how the effects of convection on atmospheric composition vary diurnally will have to take into account the scale of convective phenomena and will go hand-in-hand with basic research on the diurnal variation of the different convective scales. Quantifying the processes occurring in a variety of convection venues is ***critical*** because convective diurnal cycles vary between land, ocean, and mountain environments and differ greatly between shallow and deep convection and between isolated and mesoscale organized convection.

4.2.2.2.2 Collect measurements of chemical species in cloud top regions.

The tops of small convection and anvil regions of deep convection can provide constraints for cloud-resolving modeling studies of convection and its chemical processing of HNO_3, H_2O_2, CH_3OOH, and CH_2O and other soluble trace gases and aerosols. Many more measurements in the vicinity of convective outflows are needed from both field projects and from spaceborne instruments to provide constraints for a wide variety of forms of convection. Applying these atmospheric composition constraints to model simulations will contribute to better basic understanding of the relationship of convection to environmental shear, stability and humidity (Convective list items #2, #3, and #4). High quality measurements of vertical velocity and microphysics made at high time resolution in this effort will in addition be complementary to the need for such measurements to better understand convective evolution (Convective list items #1 and #9). The depth, horizontal extent, and ice water content of convective outflows depends critically on the scale of convective phenomenon (e.g. isolated cumulonimbus vis a vis mesoscale convective system). The nature of the convective phenomena producing significant upper-level outflows depend especially on environment shear (Convective list item #3). Quantifying the degree of scavenging versus transport to the mid and upper troposphere in different types of convection is ***critical*** and determining the net effects of these processes at cloud tops is the net result of many of the in-cloud processes.

4.2.2.2.3 Determine the mechanisms by which overshooting convection and mesoscale convective systems affect stratospheric composition.

Water vapor affects the chemistry and radiation of the stratosphere. Evidence of convective towers penetrating into the lower stratosphere in midlatitudes and the occurrence of large mesoscale tops of extremely low temperature in the tropics continue to raise questions about how convective clouds affect the composition of the stratosphere. These processes must be pursued through high-resolution modeling of the convective systems that are validated with aircraft observations. This work is a key element in understanding how deep convection interacts with the water vapor field (Convective list item # 2). Principal reasons for this importance are that deep stratospheric convection may play a large role in transitioning the current climate state from a low- CO_2/Dry-Stratosphere to a High-CO_2/Moist Stratosphere, and deep stratospheric convection can impact the catalytic conversion of inorganic chlorine to its free radical form (with subsequent loss of ozone).. While there have been some observations over the U.S. in summer that suggest this process is occurring, further studies must be done. The combination of aircraft, NEXRAD radar networks and satellite observations have established the importance of deep stratospheric convection over the U.S. in summer. Because this issue has been raised by measurements over the U.S. it is ***very important and possibly critical*** to verify the importance of this mechanism (or otherwise) to the composition of the stratosphere.

4.2.2.2.4 Identify the mechanism and measure the amount of stratospheric ozone that is transported downward near deep convective mesoscale convective systems.

One of the most understudied aspects of convection is the broad spectrum of small to large cold pools that reach the surface at the bases of downdrafts (Convective list item #8). Much active research is being directed toward understanding cold pools. In addition, atmospheric composition studies have found evidence of stratospheric ozone in convective cold pools. The mechanism of this transport needs to be coupled with research on convective cold pools. Additional field studies and cloud resolving models will be needed for this effort. There is also evidence of stratosphere to troposphere transport via wrapping around leading anvils of mesoscale convective systems. The processes creating this lesser-realized cross-tropopause transport must be determined via cloud-scale modeling followed by additional measurements to determine the frequency of these events as well as identify the cross-tropopause transport mechanisms and contribution to upper troposphere O_3. Because of the importance of O_3 to the troposphere, both radiatively and as a key species promoting oxidation, it is ***very important*** to examine thoroughly these stratosphere-to-troposphere mechanisms.

4.2.2.2.5 Evaluate and improve convective parameterization schemes for global and regional models in representing chemical processing by deep convection including mesoscale systems.

Convective parameterization schemes have been perpetually in a state of development for nearly 50 years. This area of research remains active and evolving, with parameterization schemes endeavoring to account for all the forms of convection, from shallow nonprecipitating cumulus to mesoscale convective systems. Parameterizations of chemical processing by deep convection need to be consistent with state-of-the-art parameterization schemes. Convective parameterizations are informed by the more explicit calculations of cloud resolving models, and the chemical processes to be parameterized need first to be tested within cloud resolving models. These modeling studies need to be constrained by field measurements of chemical species, especially in the vicinity of inflow and outflow regions. These field studies can be conducted along with studies of chemical species in cloud top regions (Section 4.2.2.2.2). Measurements of chemical species in inflow and outflow regions will be powerful constraints on the development of parameterization schemes. These field studies must integrate with better documentation of the microphysical, thermodynamic, and dynamic variables in clouds (Convective list item #1). Pursuing this topic is ***very important*** because the numerical models that are used for policy decisions and assessments rely on the parameterization to represent convection.

4.2.2.2.6 Gather information on the composition, velocity, liquid and ice water contents (LWC and IWC), size distributions in storm cores.

In concert with composition information in the inflow and outflow regions of convection describe in Section 4.2.2.2.5, it is necessary to obtain detailed knowledge of the microphysical, thermodynamic, and dynamic variables along with chemical species in the interior of the active regions of convection. That is measurements and modeling of the internal structure of convective cores (Convective list item #1) need to be contemporaneous and co-located with measurements of chemical species on fine time and space scales. Previous field experiments have not included the information on chemical composition

within the storm cores of hail-producing convection, and even the basic cloud dynamical, thermodynamic, and microphysical information have often been lacking. Having such measurements will provide insight on the processes occurring in these high LWC and mixed phase regions. This activity requires coordinated field measurements with a storm penetrating aircraft. To realize the processes that are occurring in vigorous convection, it is ***very important*** to gather information within the storm cores.

4.2.2.2.7 Understand how the stratiform regions of tropical and midlatitude mesoscale convective systems redistribute trace gases in the atmosphere and contrast this information to the convective region.

The stratiform region of mesoscale convective systems develops their own updraft and can have cloud top temperatures colder than the convective region, yet has not been explored for their contribution to trace gas redistribution. As noted in Convective list item #5 of Section A, mesoscale convective systems take on various forms, depending on the environment shear profile. One implication of this variety of forms is that the redistribution of trace gases associated with the mesoscale systems might vary with environmental shear and stability conditions. The structure and arrangements of convective and stratiform components of these large convective systems will vary with the environment shear. It is therefore important for the examination of trace gas redistribution by mesoscale convective systems to be done in conjunction with modeling and field studies of mesoscale systems. The processes occurring in the stratiform regions of convective storms are ***very important*** to learn about in terms of their contribution to scavenging and redistribution of trace gases, aerosols, and moisture.

Question	Observation	Resolution time and space	Accuracy	Precision
4.2.2.2.1	All	<3 h to get diurnal cycle	1 km, 1 minute	
4.2.2.2.2	All	Minutes, < 1 km	< 1 km, 1 minute	
4.2.2.2.3	spaceborne Doppler radar	<1 km	+ or – 1 m/s	1 m/s
4.2.2.2.4	ground-based Doppler radar and aircraft sampling of composition	~100 m		
4.2.2.2.5	aircraft measurements of chemical species in convection inflow and outflow	~100 m		
4.2.2.2.6	aircraft measurements at many altitudes	~100 m	+ or – .25 g/kg	.1 g/kg
4.2.2.2.7	aircraft measurements of inflow and outflow gases	~100 m		

4.2.2.3 Aerosol interactions with convection

Aerosol effects on convection have been investigated mostly with numerical modeling studies, but to some extent with satellite and/or aircraft observations. Rosenfeld et al. (2008) have hypothesized that aerosol effects on cloud microphysics can invigorate convection because smaller drops are lofted higher where ice microphysics processes are affected. However, this idea has not been verified, and Fan et al. (2013) have shown that the main effect of increased aerosol in the environment may be to increase the amount of cirriform cloud outflow from the convection. In addition, aerosol effects on individual convective clouds may be quite different than what may happen on regional scales and/or short (~2 hours) versus long time scales. Van den Heever and Cotton (2007) demonstrated that aerosol microphysics effects on convection can switch from reducing precipitation during the first couple of hours of a strong convective system to enhancing precipitation after 12 hours or more of convection. There is also some evidence that small-to-moderate cumulus and cumulonimbus with low bases increase in height with increased near surface aerosol concentration (*Li et al.,* 2011). Many studies have been conducted with cloud resolving models and have little to no verification with in situ observations. The cloud seeding community have had limited success in attributing precipitation production to cloud condensation nuclei or ice nuclei number concentrations. Understanding the effect of aerosols on convection is therefore a challenging issue that must be addressed. It is not yet known if large mesoscale convective systems are influenced by the presence of aerosols. However, mesoscale convective systems are fed by deeper layers of the atmosphere than are the boundary-layer based plumes of smaller convective elements, and the aerosol environment of the entire lower troposphere would need to be taken into account. Scavenging of aerosol by convection is very likely, but a coherent global picture of effects of different forms of convection on scavenging has not yet emerged. Some ideas have been suggested for both positive and negative ("buffering") feedbacks between aerosol-cloud microphysics effects and dynamical effects. Satellite studies have not yet provided a global picture of scavenging of aerosol by convection. With all of these unknowns in mind, the following studies appear to be needed.

4.2.2.3.1 Address aerosol effects on convection from space platforms in order to capture the global variability.

The level of detail needs to be determined. However, such knowledge of the global variability will contribute vital information for testing climate model calculations. This effort will be hindered by the presence of cirrus anvils that obscure the spaceborne views of aerosol optical depth. Nevertheless climatological studies of aerosol from existing and future satellites are needed. The A-Train satellite data need to be more fully exploited for this purpose. Ground-based remote sensing of aerosol validated by aircraft will be needed to address aerosol profiles below cirrus decks. Future satellite instrumentation needs to be designed to address this problem. It is ***critical*** to obtain a global picture of aerosol effects on convection.

4.2.2.3.2 Quantify the transformation of all types of aerosols (dust, black carbon, sulfate, and nitrate) in the context of different forms, scales, and strengths of convection.

Wet deposition is the primary removal process of aerosols and therefore it controls their lifetime in the atmosphere. Furthermore, cloud processing of aerosols can alter aerosol optical properties. Reliable aircraft measurements are needed as a first step, with more case studies like those in SEAC4RS and DC3. Specifically, aircraft inlets that remove interference of cloud particles are needed. The topic is critical because it will help answer the question of how all forms of convection from small cumulus to giant mesoscale convective systems affect atmospheric aerosol composition (Convective list item #7). The scavenging and redistribution of aerosols are ***critical*** to know in terms of understanding the potential impact of the cloud-processed aerosol on the radiation budget.

4.2.2.3.3 Enhance ground-based networks to sample the full vertical profile of aerosol through both the PBL and free atmosphere.

We need to understand how aerosol does or does not affect deep and mesoscale convective systems to obtain a complete answer to the general question of how aerosol does or does not affect convection (Convective list item #7). Because mesoscale convective systems transport and process deep layers of environmental air, it is critical to know not only the boundary layer aerosol composition but rather the entire vertical profile of aerosol through the depth of the troposphere. Measurements of aerosol vertical profiles near field mesoscale convective systems should be done by ground-based remote sensing (e.g. lidar data) supplemented by aircraft. Satellite observations in the near field of convection are blocked by upper level cloud so that ground-based observations will be essential. To understand how aerosol does or does not affect deep and mesoscale convective systems is ***critical*** to obtaining a complete answer to the general question of how aerosol does or does not affect convection

4.2.2.3.4 Determine the minimum type of aerosol (chemical composition) required for understanding and modeling aerosol effects on convection including precipitation and transport of water vapor to upper troposphere.

A first order subdivision of aerosol in relation to convection is cloud condensation nuclei (CCN) versus ice nuclei (IN). An important question is how much further detail is needed to answer how all forms of convection interact with the aerosol environment (Convective list item #7). Model experiments with detailed microphysical schemes will be required to answer this question. Determining the minimum aerosol composition for studies of aerosol effects on convection is ***very important*** to representing these processes in numerical models.

4.2.2.3.5 Create a model that treats processes in a unified and consistent manner, e.g. cloud particle microphysics and transformation.

Many models separate the treatment of trace gases and aerosols from the physical processes. For example, wet deposition of aerosols and trace gases is often calculated separately from the determination of precipitation in the cloud physics and convective parameterization routines. It is critical that the same cloud/precipitation particle interactions in the model are

used to compute hydrometeor size distributions and aerosol scavenging. Current state-of-the-art models need to have a consistency among all processes. ***This is an aspirational, but critical goal.***

Question	Observation	Resolution time and space	Accuracy	Precision
4.2.2.3.1				
4.2.2.3.2	Aerosol in situ composition and size distribution Doppler and polarimetric radar	<3 h to get diurnal cycle	1 km, 1 minute 1 dBZ	1 km, 1 minute -20 dBZ
4.2.2.3.3	ground based lidar			
4.2.2.3.4	Radar	Storm cases,	1 dBZ	-20 dBZ
4.2.2.3.5	None			

References (4.2):

Barth, M. C. et al., 2014. The Deep Convective Clouds and Chemistry (DC3) Experiment, 2014. submitted to *Bull. Amer. Meteor. Soc*.

Li Z., F. Niu, J. Fan, Y. Liu, D. Rosenfeld and Y. Ding, 2011: Long-term impacts of aerosols on the vertical development of clouds and precipitation. *Nat Geosci.,* **4**, 888–894.

Fan, J., L. R. Leung, D. Rosenfeld, Q. Chen, Z. Li, J. Zhang, and H. Yan, 2013: Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds. *Proceedings of the National Academy of Sciences*, www.pnas.org/cgidoi/10.1073/pnas.1316830110

Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A., and Andreae, M. O., 2008. Flood or Drought: How Do Aerosols Affect Precipitation?, *Science*, doi:10.1126/science.1160606, **321**, 1309–1313.

Van den Heever, S. and W. R. Cotton, 2007. Urban aerosol impacts on downwind convective storms, *J. Appl. Meteor. and Clim*., **46**, 828-850.

Appendix A

NASA Workshop on Outstanding Questions in Atmospheric Composition: Attendees

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