White Paper for IMPAACT:
Investigation of Multiscale Processes Affecting Atmospheric Chemical Transport

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IMPAACT: Investigation of Multiscale Processes Affecting Atmospheric Chemical Transport

**IMPAACT aims to improve understanding of the transport, removal, and processing of gas- and aerosol-phase emissions of air pollutants and reactive greenhouse gases from East Asia as they are carried across the Pacific to North America and the Arctic.**

1. Introduction

Over the last two decades, the economic output of China has increased more than tenfold (Lin et al., 2014). Pollutant emissions from manufacturing, power generation, transportation, household heating, and incineration in East Asia have increased along with this economic explosion (Lu et al., 2011). Numerous studies have demonstrated that emissions of pollutants from East Asia strongly affect atmospheric chemistry regionally (e.g., Wang et al., 2017; Fang et al., 2009), throughout the Northern Hemisphere (e.g., Verstraeten et al., 2015), and globally (e.g., Li et al., 2016). These pollutants alter many atmospheric chemical processes, including acid deposition, ozone (O$_3$) production and the oxidizing capacity of the troposphere. They also affect climate through O$_3$, methane, halocarbon and N$_2$O radiative forcing, aerosol-radiation interactions, aerosol-cloud interactions, and snow and ice albedo changes, and degrade air quality regionally and across the Pacific in North America (Li et al., 2016; Sand et al., 2013; Verstraeten et al., 2015). Pollution from East Asia can be lifted into the free troposphere (FT) by convection and within the warm conveyor belts (WCBs) of cyclonic systems and transported across the Pacific Ocean above the marine boundary layer (Andreae, et al., 1988; Arimoto et al., 1996; 1997; Jaffe et al., 1999, 2003; Clarke et al., 2001; Van Curen, 2003; Bertschi et al., 2004; Heald et al., 2003, 2006; Zhang et al., 2008; Verstraeten et al., 2015). Some of the pollution can enter the Arctic (e.g., Bourgeois and Bey, 2011) while more reaches the western coasts of Canada and the United States (e.g., Luan and Jaeglé, 2013). Over continental North America the pollutants can be mixed to the surface, affecting the concentration of regulated pollutants, contributing to violations of air quality standards, and potentially affecting human health (Van Curen, 2003; Li et al., 2014; Verstraeten et al., 2015). Increases in O$_3$ observed in North America at surface sites (Parrish et al., 2012, Jaffe et al., 2003a), aloft (Cooper et al., 2011) and through the tropospheric column (Verstraeten et al., 2015) are attributed to increasing emissions from East Asia, although analysis must account for natural variability and stratospheric influences (e.g., Lin et al., 2014; Lin et al., 2017).

Arctic ecosystems and climate are also affected by air pollution transported from East Asian sources (Qi et al., 2017a,b). Aerosol particles and tropospheric O$_3$ contribute to Arctic warming by altering the atmospheric radiation balance both within and outside of the Arctic (Shindell and Faluvegi, 2009; Natarajan et al, 2012, Sand et al., 2013; AMAP, 2015). Deposition of black carbon (BC) to the Arctic surface can reduce snow and ice albedo, accelerating springtime snowmelt and increasing Arctic warming (e.g., Flanner et al., 2007).

Numerical models that accurately simulate the production, transport, transformation, deposition,
and environmental effects of pollutants are essential to understand existing atmospheric conditions, to predict future changes in atmospheric chemistry and climate forcing, and to develop sound mitigation strategies. **Atmospheric observations of key species provide important tests of the accuracy of the emissions and the chemical and physical processes simulated in these models.** Recent comparisons of trace gases and aerosol species in remote areas of the troposphere with global chemical transport and chemistry-climate models show a range of model skill for key species, including aerosol black carbon (BC), organic carbon (OC), and O$_3$—all short-lived climate forcing agents—and for reactive species that participate in aerosol, HO$_x$, NO$_x$, and SO$_x$ chemistry. Most models tend to underestimate CO in northern high latitudes, for reasons that may be related to OH abundance or emissions or transport errors (Shindell et al., 2006; Stein et al., 2014; Monks et al., 2015). Models often struggle to replicate observed vertical distributions of many species (Emmons et al., 2015; Eckhardt et al., 2015; Koch et al. 2009, Schwarz et al. 2017; Tsigaridis et al., 2014; Sand et al., 2017), leading to significant errors in estimates of radiative effects, the lifetime of anthropogenic and natural species in the atmosphere, and deposition to the surface.

**The variable quality of these comparisons and the diversity of model outputs reduce confidence in our ability to estimate current climate forcing, climate response to emission changes, and pollutant processing and deposition** (Fig. 1; Shindell et al., 2008; Koch et al., 2009; Shindell and Faluvegi, 2009; Jiao et al., 2014; Tsigaridis et al., 2014; Wang et al., 2014a; Eckhardt et al., 2015; Sand et al., 2017). Deficiencies in model capability and a lack of observations to constrain the models present major challenges to advancing this understanding and to making credible near- and long-term projections of atmospheric composition and climate forcing (Arnold et al., 2014; AMAP, 2015).

![Figure 1](image-url)
made in an airmass-relative, or Lagrangian, framework. Only a few observation-modeling studies, notably the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) program in 2004 (e.g., Methven et al., 2006) and the Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, of Climate, Chemistry, Aerosols, and Transport (POLARCAT) in 2008 (Law et al., 2014), have conducted long-range transport studies that focus on measuring temporal changes in atmospheric composition during transport over intercontinental scales. Recognition of this issue is a powerful motivation for the modern experimental design presented below incorporating state-of-the-art instrumentation and techniques.

We propose a new study, IMPAACT, that combines in situ and remote sensing observations with modeling on a range of scales to improve understanding and reduce model uncertainty and diversity regarding pollutant transport, removal, and chemical processing on synoptic and intercontinental scales. IMPAACT will focus on the characteristics, transport, and evolution of emissions from East Asia as they are lifted from the boundary layer, carried across the Pacific in the (FT), and enter the Arctic and western U.S. and Canada. IMPAACT will target transport in cyclonic systems in which wet processing and deposition play important and understudied roles in controlling pollutant abundance and speciation. These observations will be closely coordinated with emissions, Lagrangian, and chemical transport models, which will be used for flight and ship track planning, sensitivity tests, and diagnostic studies aimed at identifying model deficiencies and improving representation of key processes. The NASA DC-8 aircraft and two lower tropospheric aircraft (P-3 or C-130-class) are needed to meet the sampling needs of the IMPAACT study.

The purpose of this document is to lay the foundation for more detailed proposals and planning documents for the U.S. NASA, NOAA, and NSF agencies, and for developing partnerships with university and international collaborators that will lead to additional measurements, simulations, and analyses to expand and enhance the research effort. In Section 2 we summarize the primary scientific issues that will be addressed by IMPAACT, and develop science questions that will lead to testable hypotheses. Section 3 describes the observational and modeling approaches that will be used to answer these questions.

2. Scientific background

2.1 Transport from the polluted boundary layer to the free troposphere.

Export of mid-latitude air pollution from the boundary layer to the FT primarily occurs via deep convection or the WCBs of mid-latitude cyclones (e.g., Monks et al., 2009; Dentener et al., 2011). Because WCBs usually generate precipitation, which can efficiently remove some pollutants from the atmosphere. IMPAACT will focus primarily on springtime export of pollution in WCB systems (Fig. 2). An important region for WCB formation is the warm Kuroshio Current (Hirata et al., 2015; Gyakum et al., 1989), which flows northeastward along the east coast of Japan. During winter the main WCB formation region is along the southern coast of Japan with polluted boundary layer air masses in the formation region arriving from the northwest (Stohl, 2001; Eckhardt et al., 2004; Madonna et al., 2014). In summer the WCB formation region is further south and west including Taiwan and subtropical regions of mainland China, with air mass origins in the vicinity of SE Asia (Madonna et al. 2014). Springtime WCB inflow regions and air mass source regions have characteristics that fall between the winter and summer extremes.
Transport of Asian air pollution via mid-latitude cyclones was examined during the PEM West B (Crawford et al., 1997), TRACE-P (Jacob et al., 2003; Miyazaki et al., 2003; Mari et al., 2004), PEACE-B (Oshima et al., 2004), ACE-Asia (Huebert et al., 2003) and A-FORCE (Oshima et al., 2012, 2013) missions. These springtime experiments found that both deep convection and WCBs were important mechanisms for exporting pollution from the boundary layer. More recent research has focused on transport from China (Ding et al., 2007; Li et al., 2012) and South Korea (Lee et al., 2011). Black carbon export from northeastern China during spring is expected to occur by eastward transport within the planetary boundary layer and by lofting in mid-latitude cyclones in the 35°-45° latitude band, while convective and orographic lifting is expected to be important further south in the 25°-35° latitude band (Oshima et al., 2013).

Long-range transport of air pollution from Asia via WCBs has been observed at mid-latitudes where plumes routinely reach western North America during spring (e.g., Heald et al., 2003; Cooper et al., 2004a,b; Jaffe et al., XXXXX, Lin et al., 2012). A satellite-based composite study of aerosol export from East Asia shows a maximum in spring with a branch that crosses the North Pacific Ocean in 6-8 days and a branch that reaches the Arctic in 3-4 days (Fig. 3; Luan and Jaeglé, 2013). Several cases of the transport of anthropogenic and biomass burning emissions from Asia into the Arctic were observed during the spring 2008 ARCPAC and ARCTAS experiments (Brock et al., 2011; Fisher et al., 2010; Jacob et al., 2010).

While the research summarized above has advanced our knowledge of pollution export from the polluted boundary layer to the FT in the region of East Asia, there are many outstanding science questions requiring further evaluation, four of which IMPAACT can address.
Question 1: During transport events, what fraction of boundary layer aerosols and trace gases is transported vertically by quasi-isentropic transport within the warm conveyor belt (WCB) and what fraction is transported by deep convection within or near the WCB?

Although embedded deep convection can constitute an important component of the vertical transport within a WCB (Kiley et al., 2006), the relative contribution of deep convection vs. quasi-isentropic uplift within WCBs has not been quantified by in situ observations. Measurements are needed of the aerosol and trace gas composition outflow from deep convective clouds embedded within WCBs, as well within regions of stratiform lift in WCBs. Repeated sampling of the outflow region of these two types of cloud, coupled with observations of the composition of the boundary layer inflow region and vertical flux estimates from regional scale models, can provide an estimate of the relative contribution of each transport pathway to boundary layer export to the FT. Carbon monoxide and various alkanes, alkenes, and halocarbons are examples of appropriate tracers with a range of lifetimes against oxidation that can be used to evaluate this transport. These species also serve as chemical tracers against species with shorter lifetimes. By measuring the ratio of the species of interest to the tracer species before and after transport from the boundary layer to the FT, as well as during further transport across the Pacific, the net impacts of removal processes on these species can be separated from dilution and mixing.

Question 2: What is the extent of mixing between stratospheric filaments and WCB outflow during transport across the Pacific?

Once boundary layer pollution is lofted, long range transport occurs within upper tropospheric jet streams, which separate the troposphere from the lowermost stratosphere (Shapiro, 1980).
Tropopause folding associated with Rossby wave breaking can lead to interweaving of stratospheric and tropospheric air masses bringing boundary layer pollutants into close proximity to stratospherically influenced air (Fig. 4; Cooper et al., 2004b). Turbulence within tropopause folds can lead to irreversible mixing of polluted and stratospheric air masses (e.g., Fairlie et al., 2007) which can have significant impacts on the chemistry of these air masses.

In situ measurements of tracer-tracer relationships in these intertwined air masses at different stages of mixing can be used to diagnose the degree of incorporation of stratospheric air and its influence on upper and middle tropospheric chemistry. Large scale mixing efficiency can be diagnosed using Lagrangian approaches (Fairlie et al, 2007).

**Question 3: How does variability in transport and emissions (including wildfire emissions) impact \( O_3 \) in the Western Pacific, Arctic and North America?**

Several studies have pointed out the importance of the retention of soluble trace gases in freezing cloud drops in determining the removal of that trace gas by precipitation (Barth et al., 2001; 2007; Stuart and Jacobson, 2003; 2004; Michael and Stuart, 2009; Bela et al., 2016). Analysis of aircraft measurements and cloud-resolving model simulations suggest a strong link between cloud physics processes and convective transport of formaldehyde (CH\(_2\)O), hydrogen peroxide (H\(_2\)O\(_2\)), and other gases, with perhaps varying scavenging efficiencies depending on the dominant cloud physics processes. Sampling of soluble trace gases that are ozone and aerosol precursors (e.g., CH\(_2\)O, H\(_2\)O\(_2\), CH\(_3\)OOH, SO\(_2\), HNO\(_3\)) and insoluble trace gases with longer chemical lifetimes in the storm inflow and outflow regions can provide information on the removal efficiencies of the more soluble species. Determining the trace gas composition in snow and graupel particles can provide new information on these scavenging processes. Measurements of cloud residue particles (i.e., with a counterflow virtual impactor) and of trace gases devolved from the evaporating cloud particles would provide valuable information on the chemical and physical processes involved in the transport of soluble species.

**Question 4: How important are ice-phase cloud physics processes for determining the fraction of boundary layer soluble trace gases and aerosols that are transported to the free troposphere?**

Several studies have pointed out the importance of the retention of soluble trace gases in freezing cloud drops in determining the removal of that trace gas by precipitation (Barth et al., 2001; 2007; Stuart and Jacobson, 2003; 2004; Michael and Stuart, 2009; Bela et al., 2016). Analysis of aircraft measurements and cloud-resolving model simulations suggest a strong link between cloud physics processes and convective transport of formaldehyde (CH\(_2\)O), hydrogen peroxide (H\(_2\)O\(_2\)), and other gases, with perhaps varying scavenging efficiencies depending on the dominant cloud physics processes. Sampling of soluble trace gases that are ozone and aerosol precursors (e.g., CH\(_2\)O, H\(_2\)O\(_2\), CH\(_3\)OOH, SO\(_2\), HNO\(_3\)) and insoluble trace gases with longer chemical lifetimes in the storm inflow and outflow regions can provide information on the removal efficiencies of the more soluble species. Determining the trace gas composition in snow and graupel particles can provide new information on these scavenging processes. Measurements of cloud residue particles (i.e., with a counterflow virtual impactor) and of trace gases devolved from the evaporating cloud particles would provide valuable information on the chemical and physical processes involved in the transport of soluble species.

**Question 5: Does the concentration of aerosols in the boundary layer affect the vertical and horizontal transport of air masses within a WCB or convective system?**

Recent modeling studies have found that pollution can affect precipitation formation in WCB originating in the western North Pacific [Wang et al., 2014a; Wang et al., 2014b; Joos et al., 2016]. These global chemistry-climate simulations of present day and preindustrial scenarios produce varying results. Wang et al. (2014a) report that anthropogenic aerosols invigorate WCBs producing more precipitation, while Joos et al., (2016) report comparable precipitation amounts in present day and preindustrial simulation results.
These effects cannot be directly measured, but documenting aerosol concentrations and hygroscopic and optical properties in the inflow and outflow regions, along with meteorological parameters such as precipitation amounts and FT water vapor concentrations, can provide important constraints for simulations of these effects.

2.2 Ozone and its precursors

Ozone is a significant short-lived greenhouse forcing agent and a regulated secondary criteria pollutant in many countries. Mixing ratios of O\textsubscript{3} in the troposphere are substantial and variable, and are influenced by pollution, biomass burning, natural emissions, and transport from the stratosphere. Existing deficiencies in global CTM and CCM representations of the abundance, distribution, and long-term changes in O\textsubscript{3} (Logan et al., 2012; Parrish et al., 2013; 2014; Derwent et al., 2016) may be caused by errors in emissions, transport, removal, and chemistry.

Tropospheric O\textsubscript{3} is produced by a catalytic oxidation cycle involving NO\textsubscript{x} (NO+NO\textsubscript{2}) and hydrocarbons. The abundance and partitioning of reactive nitrogen (NO\textsubscript{y} = NO\textsubscript{x} + HNO\textsubscript{3} + PNs + ANs + N\textsubscript{2}O\textsubscript{5} + NO\textsubscript{3}· + . . . , where PNs are peroxy nitrates and ANs are alkyl and multifunctional nitrates) affects the abundance of NO\textsubscript{x} and thus the temporal and spatial scale of this production (e.g., Bertram et al., 2013). Accurately simulating tropospheric O\textsubscript{3} production requires that numerical models represent this partitioning with fidelity, as well as the emissions, sinks, lifetime and transport pathways of O\textsubscript{3} and its other precursors. This remains a challenge for many reasons including rapidly changing precursor emissions over time, the large complexity and uncertainty in VOC emissions and oxidation schemes, the multiphase chemistry associated with NO\textsubscript{y} and organic species, and sub-grid-scale nonlinear chemical and physical processes (Yan et al., 2016).

East Asia is a region with very large and rapidly changing anthropogenic emissions of O\textsubscript{3} precursors (Liu et al., 2016), and this is coupled with large interannual variability in emissions from natural and biomass burning sources (Tanimoto et al., 2015). Past in situ and satellite observations show that plumes of reactive nitrogen from East Asia reach the eastern North Pacific frequently. A number of studies have attempted to model O\textsubscript{3} production and sources in the North Pacific with varying success. Several of these, as well as observations (e.g., Bertram et al., 2013) have pointed to PNs, especially peroxyacetyl nitrate (PAN), as a key carrier of reactive nitrogen and therefore greater O\textsubscript{3} production in downwind regions. The processing pathways of both PNs and ANs, and the fraction resulting in recycling of NO\textsubscript{x} from its reservoirs, the importance of nighttime chemistry, the role of halogens, etc., are all under-constrained.

Question 6: What are the abundances and partitioning of reactive nitrogen species and VOCs in the lower troposphere inflow region to WCBs, and in the high-altitude outflow region of WCBs?

There are few recent in situ observations of O\textsubscript{3} precursors over the North Pacific that can be used to constrain high spatial and chemical resolution, newer-generation CTMs. Specifically, measurements are needed of precursor species in the boundary layer prior to lifting and transport (Wang et al., 2017). It is particularly important to comprehensively measure the abundance and partitioning of reactive nitrogen species prior to, during, and after lifting, wet removal in WCBs, and transport to the FT (Fig. 5; Fischer et al., 2011; Bertram et al., 2013; Zhang et al 2008; Neuman et al 2006). The removal of reactive nitrogen in WCBs depends critically on the amount of water-soluble HNO\textsubscript{3} relative to sparingly-soluble or insoluble PNs that is formed prior to lifting and wet removal. If HNO\textsubscript{3} is a large fraction of NO\textsubscript{y}, effective removal of HNO\textsubscript{3} in precipitating WCB systems will substantially reduce the efficiency of NO\textsubscript{x} transport from the boundary layer to the FT and the resulting amount...
of O$_3$ produced in the FT downwind of the cyclonic system. To accurately simulate downstream O$_3$ production, models must capture the abundance and partitioning of NO$_x$ species in the BL over the western Pacific Ocean (Kotchenruther et al 2001; Hudman et al 2004; Systematic in situ measurements that provide statistically-sound information on nitrogen partitioning and VOC abundance and speciation in the region of WCB inflow and in the upper-level outflow are essential to constrain such simulations. The efficiency of transport through WCBs (and deep convection) will likely depend on the amount of liquid-phase vs. ice-phase precipitation formation, since adsorption and reaction of gas-phase species onto ice crystals formed by the Wegener-Bergeron-Findeisen process is typically much lower than is reactivity of the same species in liquid clouds (e.g., Clegg and Abbatt, 2001). Thus it is also important to understand the meteorological details of the cyclonic systems and the cloud physics within the WCB that are involved in precipitation formation and to compare such observations with (necessarily parameterized) representations of these processes in global-scale models.

Question 7: What is the evolution in partitioning, speciation, and reactivity of nitrogen and VOC compounds in the middle and upper troposphere during multi-day transport across the Pacific?

After lifting from the lower troposphere by WCBs or convection, O$_3$ production is limited by the amount of NO$_x$ present and the gradual decrease in total VOC reactivity as more reactive species are lost to oxidation. Recycling of NO$_x$ via thermal dissociation of PNs can occur if the air mass descends and warms (Hudman et al., 2004), leading to O$_3$ formation. The ultimate amount of additional O$_3$ production (or destruction) following uplift will depend on the details of these chemical transformations, which are mediated by transport pathways that are statistically poorly constrained by observations. Additional removal or vertical redistribution of HNO$_3$ by stratiform precipitation during transport may also occur and can affect the ultimate O$_3$ production rates and yields during transport.

Question 8: What is the importance of stratospheric air on the O$_3$ budget of the upper and middle troposphere over the Pacific?
This topic is closely related to Question 2, which focuses on the mixing of stratospheric and tropospheric air. Previous studies indicate that the stratosphere contributes substantially to the North Pacific tropospheric O$_3$ budget (Jaeglé et al. 2003). More recent work has documented the influence of stratospheric O$_3$ on surface mixing ratios in the western U.S. (Langford et al., 2015). Given significant changes in human emissions and new knowledge of significant interannual variability, additional observations and modeling efforts constraining the importance of stratospheric air on middle- and lower-tropospheric O$_3$ mixing ratios are needed.

2.3 Secondary aerosol species

Secondary aerosol particles are those produced from the gas phase, usually after oxidation via gas-phase or aqueous-phase reactions. Sulfur, nitrogen, and organic compounds are the dominant secondary aerosol components, and are mostly found in particles with diameters from 0.1-1.0 µm that have long atmospheric lifetimes in the FT, serve as cloud condensation nuclei (CCN), and scatter and absorb light efficiently. They contribute to acid rain, can contain toxic species such as PAHs and mercury compounds, and affect climate through aerosol-radiation and aerosol-cloud interactions. Inorganic and organic aerosol species can partition between the gas and condensed phases, altering the chemistry of both. Ammonium nitrate is a semi-volatile compound that can play an important role in reactive nitrogen partitioning and the ammonia budget. Wet removal is efficient for most inorganic aerosol species, but is poorly known for aged organic particles in the atmosphere (e.g., Hodzic et al., 2015).

Many global models reasonably replicate observed acid deposition and surface aerosol mass concentrations, demonstrating a broad understanding of emissions and wet scavenging at northern midlatitudes (Vet et al., 2014). However, the formation of CCN and subsequent aerosol-cloud interactions over large regions of the globe is highly sensitive to the amount of anthropogenic SO$_2$ emitted and transported to the FT (e.g., Merikanto et al., 2009), suggesting that a higher level of understanding is needed to constrain the climate impacts of East Asian sulfur emissions. Sulfate aerosol from East Asia has been observed to reach the West Coast of the U.S. (Jaffe et al., 2003; Brock et al., 2004; Dunlea et al., 2009), and model simulations suggest that East Asian sulfate is a substantial fraction of the background sulfate concentration in the western U.S. (Vet et al., 2015). In the Arctic, models have difficulty replicating the observed seasonality at surface sites, as well as the SO$_4^{2-}$/BC ratio, indicating that wet removal in mixed-phase clouds is not being simulated adequately (Fig. 1; Eckhardt et al., 2015; Qi et al., 2017c). Chemical transport models can successfully match ground-based and airborne observations in the Arctic when wet scavenging parameterizations are optimized (Fisher et al., 2010, Bourgeois and Bey, 2011; Qi et al. 2017b,c), further indicating the importance of mixed phase wet removal process. In addition to sulfate and nitrate, soil dust particles produced from the extensive deserts of central and eastern Asia are efficient nuclei for cloud ice formation, play an important role in aerosol-cloud interactions, and can react heterogeneously with gas-phase species (Cwiertny et al., 2008; Tang et al., 2016). Better quantification of the source strength and chemical and ice nucleating characteristics of these particles is needed. The radiative importance of soil dust is discussed in 2.4.

The situation for OA is more complex than that for inorganic aerosols, given the large number of primary and VOC sources, species, and gas-phase and aqueous reactions involved. Global model intercomparisons show wide disparities in the seasonality, vertical distribution, and the magnitude of OA concentrations (Tsigaridis et al., 2014) but are poorly constrained by observations. Most models underestimate OA compared with ground-based measurements in rural areas by a
factor of 2 or more, at least in part because of lack of knowledge of the volatility of VOC oxidation products (Tsiganidis et al., 2014). This disparity is important; OA to make up ~30% of fine aerosol mass in the Arctic during the springtime (e.g., Brock et al., 2010) and ~40% at coastal sites across the East China Sea downwind of China (Jimenez et al., 2009).

The aerosol-radiation interactions associated with East Asian OA are dependent upon particle hygroscopicity and the amount of light absorption by brown carbon (BrC). Although both of these parameters may change during transport due to oxidation and photobleaching in biomass burning plumes (Forrister et al., 2015), this evolution has not been directly studied for anthropogenic plumes. Most SOA, which forms within ~1 day after emission of VOCs, is believed to be lost by wet removal during initial lifting to the FT. Because SO$_2$ is not removed as efficiently by wet deposition, sulfate/SOA ratios in pollution transported from East Asia across the Pacific are believed to be higher than in the polluted boundary layer over East Asia (Brock et al., 2004; Dunlea et al., 2009; van Donkelaar et al., 2008;). This differential removal of SOA relative to SO$_2$ has been inferred rather than directly measured.

**Question 9: What is the fraction of gas-phase secondary precursors, especially SO$_2$ and VOC, that is transported into the FT? How does this transport efficiency compare to that of particulate SO$_4^{2-}$ and OA and to conserved trace gases? Is transport of precursor species efficiency higher when ice-phase precipitation processes dominate?**

Global models (e.g., Merikanto et al. 2009) suggest that CCN abundance in the remote troposphere is strongly affected by the transport of anthropogenic SO$_2$ to FT. Similarly, SO$_4^{2-}$ aerosol production during trans-Pacific transport may contribute to background sulfate concentrations in the western U.S. and Canada. The efficiency of transport of SO$_2$ to the FT (Fig. 5) is poorly constrained, and is probably quite sensitive to the microphysics of precipitation formation.

**Question 10: How much nitrogen is partitioned into the aerosol phase as NO$_3^-$ and NH$_4^+$ during transport across the Pacific in the FT?**

Because ammonium nitrate is semi-volatile, it can serve as a temporary reservoir of significant amounts of reactive nitrogen which can produce O$_3$ if released to the gas-phase. This depends on whether or not the volatilized aerosol NO$_3^-$ photolyzes to produce HONO (Ye et al 2016). The partitioning of the NO$_3^-$ budget prior to lifting, the wet scavenging efficiency, and the partitioning in the FT all help control the reactive nitrogen available in the FT for this chemistry. Observations suggest that little inorganic nitrate is found in particles in the FT because partitioning favors evaporation (e.g., Dunlea et al., 2009), but further measurements are needed to confirm this expectation over a range of thermodynamic conditions and concentrations and speciation of gas-phase reactive nitrogen compounds. Small amounts of nitrate may be found in organic species, but these are unlikely to repartition to the gas phase.

**Question 11: What is the hygroscopicity of SOA at low altitudes before transport, what fraction is being transported into the FT, and what is the hygroscopicity in the FT on both sides of the Pacific and in the Arctic? Is the aerosol internally mixed (sulfate, nitrate, and organics), or do we need to consider the hygroscopic and optical characteristics of external mixtures, as is done in most global models?**

Organic aerosol abundance and hygroscopic and optical properties (and their evolution with time) are very poorly characterized over the scales of
synoptic transport. The removal efficiency of the OA will depend in part on how mixed it is with hygroscopic inorganic species, its own intrinsic hygroscopicity (which may vary with degree of oxidation), and the mechanisms of precipitation formation. Most observations (e.g., Wang et al., 2010) suggest that, even in source regions, internal aerosol mixtures evolve quickly by condensation. How models represent the mixing state and hygroscopicity of the aerosol may lead to large variability in removal efficiency (e.g., Tsigaridis et al., 2014). Observations downwind of East Asia, which provides strong sources of BC, soil dust, and inorganic and organic species, will constrain model representations of aerosol mixing state and hygroscopicity.

**Question 12: Is there aqueous production of SOA, especially during lifting to the FT? Is there further chemical production or loss of SOA as it is transported across the Pacific after lifting?**

There is some evidence for aqueous-phase production of SOA through many proposed mechanisms and processes (Ervens et al., 2011; McNeill, 2015). In situ measurements have generally been limited in scope and scale, focusing on small convective clouds or fog (e.g., Sorooshian et al., 2006; Gilardoni et al., 2016). Aqueous pathways are incompletely represented in global atmospheric chemistry models. Measurements are needed to provide constraints on the magnitude of SOA aqueous production at the intercontinental and global scale (e.g., Waxman et al., 2013). Increases in oxalate and other organic acids and of the oxidation products of glyoxal and methylglyoxal relative to tracer species following lifting in WCBs would provide evidence of aqueous production of organic compounds (McNeill, 2015). However, without comprehensive speciated measurements of compounds produced by aqueous processes, observations during IMPAACT are unlikely to fully quantify SOA formation from this channel, and will provide only limited constraints on models.

### 2.4 Absorbing aerosol

Some light-absorbing aerosol species, such as black carbon, also result in negative health effects (Jannson et al., 2012; Mauderly and Chow, 2008) and contribute, with much uncertainty, to global climate forcing (Bond et al., 2013, Choobari et al, 2014). Uncertainty in their climate effects is due to multiple factors, including 1) the difficulty in experimentally determining the very small fraction of aerosol extinction caused by absorption (rather than light scattering), 2) the large spatial and temporal variability of these species (driven by their varied sources and short but strongly altitude dependent lifetime), and 3) aging processes such as coatings of organic and inorganic species,

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**Figure 5.** Schematic showing how wet scavenging in WCB belts removes soluble and hygroscopic species, while permitting less soluble and hygroscopic compounds to reach the free troposphere, affecting the downstream composition and abundance of both gas-phase and particulate species.
which can change their optical properties and lifetime. Global models struggle to capture these characteristics and the complex dynamical feedbacks associated with diabatic heating due to in situ absorption. Of the light-absorbing species, black carbon (BC), produced both from wildfires and anthropogenic combustion (crop clearing, diesel engines, coal burning, etc.), is primarily responsible for most absorption at visible wavelengths and has specifically been linked to poor health (Geng et al., 2013). Brown carbon (BrC), a class of materials that tend to have highly wavelength-dependent absorption cross-sections, are a more recent focus of attention, and likely contribute significantly to net aerosol absorption at short wavelength while also strongly affecting photochemistry (e.g., Liu et al., 2017). Mineral dust, which globally averaged is predominantly naturally emitted, is the largest contributor to absorbing aerosol mass, yet does not dominate short wave absorption on a global scale. Because mineral dust particles are typically much larger in size than BC and BrC, their lifetimes against transport and wet and dry removal are shorter. Hence mineral dust has different spatial distributions, and interacts with atmospheric processes and affects air quality differently than do the other absorbing components.

In spring, substantial amounts of pollution from anthropogenic activities, agricultural burning in SE Asia, and Siberian wildfires are transported out of East Asia and towards the Arctic and North America (Qi et al., 2017c). The radiative forcing attributable to these species occurs both inside and outside of the Arctic (Natarajan et al., 2012, Sand et al., 2013), while air quality impacts from this pollution can extend to the West Coast of the U.S. and beyond (Jaffe et al 2004; 2005; Laing et al., 2016). Observations during IMPAACT will improve understanding of basic processes that determine the effect of primary absorbing aerosol on global and regional climate and on air quality in the continental US by quantifying the amount exported out of Asia, determining the mixing state of absorbing and non-absorbing components, identifying the mechanisms of long range transport across the Pacific, and determining the fraction surviving in the atmosphere over long distances and its characteristics.

**Question 13: How efficient is the transport of BC/BrC/Dust aerosols during initial lifting and long-range transport over the Pacific relative to each other and gas-phase tracers? What processes control these differential efficiencies? What is the importance of small-scale features not resolved in models or satellite observations?**

A key and poorly constrained parameter in global models is the time-dependent wet scavenging efficiency of primary absorbing aerosol species. Emissions of BC, BrC, and dust from Asia can be linked to the airmasses that carry them via gas-phase tracer species that are not removed over the relatively short timescales over which aerosol losses occur. Note that dust is not co-emitted with any distinguishable gas-phase tracers, but can still be correlated with anthropogenic emissions because transport processes in the East Asian region often mix air masses from arid desert and urbanized regions (Fairlie et al, 2010). In IMPAACT, this work will be strongly supported with satellite and modeling analysis. In situ observations will be combined with satellite measurements of aerosol optical depth (AOD) in visible (Remer et al., 2005) and ultra-violet (Torres et al., 2007) wavelengths, infrared retrievals of carbon monoxide and dust (DeSouza-Machado et al, 2010), and global air quality forecasting and assimilation systems (MOZART, AM3, GEOS-Chem, RAQMS). In situ measurements can be used to assess quantities derived from the satellite products, and to counter possible biases from the lack of satellite retrievals in cloudy conditions.

**Question 14: What are the light absorbing and scattering properties of BC/BrC/Dust from East Asia, and how do they evolve throughout long range transport and removal? How do
they contribute to direct radiative forcing? To indirect and semi-direct forcing?

The optical properties of absorbing aerosol change with aging, due to changes in morphology and coating and internal mixing of BC and dust with other materials (Liu et al., 2017), as well as photobleaching and secondary production of BrC (Forrister et al., 2015). These changes can also affect the way these aerosols take up water and their efficacy as cloud nucleating/ice nucleating particles. Hence, the evolution of these properties are fundamental to interpreting how primary absorbing aerosol species affect climate and air quality, and how these effects vary during transport and atmospheric processing.

**Question 15: What are the sources, emissions, chemical processing and deposition for Hg in the Western Pacific and Arctic regions?**

Given that Asian emissions represent the largest source of Hg on the planet (Jaffe et al 2005; Selin et al 2007; Strode et al 2008) IMPAACT presents an opportunity to improve knowledge regarding this important neurotoxin. There are large uncertainties in chemical processing and deposition (e.g. Bieser et al 2017). Observations of the vertical and horizontal distributions and gradients during IMPAACT would provide key constraints to global models and improve our understanding of Hg transport into the Arctic.

**3. Observational and modeling approach**

IMPAACT is designed to address the above science questions by systematically sampling the abundance and distributions of key species during export from Asia, both in the boundary layer and after processing during long-range transport. These measurements must be coupled with modeling on a range of scales to help develop the process-level understanding that is necessary for global-scale model improvement. Sampling by either the chemically-instrumented NOAA P-3 or NSF C-130 aircraft in the boundary layer just downwind of East Asia prior to vertical transport will be coupled with free tropospheric sampling in the outflow region immediately downwind of the Asian continent by the NASA DC-8 to determine the export efficiency of pollutants relative to tracers, and to quantify the speciation of reactive nitrogen, sulfur, and organic species. Sampling by the DC-8 further downwind as a function of transport time/distance, and following cloud processing and removal of soluble species, will provide data-driven constraints for improved parameterizations in modern CTMs. Either the NOAA P-3 or the NSF C-130 can provide measurements within the middle regions of WCB outflow, and may be deployed to measure the composition of air entering the Arctic and continental U.S. A global class U.S. ship will sample measurements of aerosols and trace gases downwind of the Asian continent in the Sea of Japan and/or along the southern and eastern coasts of Japan. The 24/7 sampling onboard the ship will provide continuous measurements in the boundary layer for comparison to the aircraft snapshots. A suite of additional ground sites along the Pacific Cordillera will provide continuous observations of selected species for spatial and temporal context. Geostationary satellite observations from the new GEMS instrument over East Asia and the new TEMPO instrument over North America will provide continuous coverage of AOD over the region of WCB formation and over the populated regions of the West Coast. Instruments on polar-orbiting platforms, such as the A-Train constellation, will be provide information on the column abundance of several key gas-phase species, including CO, as well as AOD. The data obtained during IMPAACT, coupled with focused modeling efforts, will systematically quantify the processes driving the chemistry of background O₃ production and loss, the abundance and characteristics of aerosol species, and the episodic but significant role that wet removal plays in modulating the long-range transport of the pollutants across the northern Pacific Ocean.

**3.1 Why East Asia?**
Frequent transport of pollutants from the boundary layer to the free troposphere in WCBs occurs primarily in two areas of the northern hemisphere: near the East Asian coastline in the western Pacific Ocean, and near the east coast of North America in the western Atlantic Ocean (Fig. 6). In principle the processes of pollutant transport, removal, and processing could be studied in either location. However, East Asia is favored as a research venue for the following reasons:

- The levels of pollution in the eastern U.S. and Canada are modest and continue to decline (e.g., Hidy et al., 2014). The relatively low pollutant abundance in North America makes it more challenging to unambiguously apportion measurements between anthropogenic, stratospheric, and biogenic sources (e.g., Methven et al., 2006).
- While China has begun to reduce the emissions of some pollutants, notably SO$_2$, East Asia remains heavily polluted and will continue as the largest regional source of pollution in the middle and high latitudes. Transport and processing of this pollution can be readily observed with both in situ and satellite measurements (Luan and Jaeglé, 2013; Oshima et al., 2012, 2013).

An observational project in East Asia presents logistical challenges and costs. It is unlikely that foreign research aircraft will be permitted in Chinese airspace. Although Chinese airborne measurement capabilities are growing, Chinese researchers will probably not be able to provide

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**Figure 6.** Top panel: Regions of initial uplift into WCBs determined from back-trajectory calculations averaged into 5° x 3° cells (Stohl et al., 2001). Bottom panel: Average April precipitation amounts (mm/day) from 1998-2011 from high-resolution Tropical Rainfall Measuring Mission (TRMM) satellite sensor measurements. A region of precipitation associated with WCBs and cyclone development is evident in the vicinity of Okinawa and Japan extending northeastward over the Pacific Ocean. Image from [http://pmm.nasa.gov/trmm/trmm-based-climatology](http://pmm.nasa.gov/trmm/trmm-based-climatology).
the comprehensive airborne scientific payloads necessary to answer the scientific questions posed above. Thus the primary platforms for airborne measurement will likely be large, U.S. agency-operated aircraft, which will limit operations to international, Japanese, and South Korean airspace over the East China Sea and the Pacific Ocean proper. This does not present a critical problem, since the inflow zones for Asian WCBs are located in these regions, and the FT transport will occur between Japan and North America. However, it will not be possible to characterize the pollution and emissions directly over the source regions of China. Instead, aged pollution in the marine boundary layer and lower free troposphere over the East China Sea will be characterized prior to vertical transport. Pollution that has already been lifted to the free troposphere by convection or synoptic lifting over the Asian continent will also be sampled before transport across the Pacific. It is hoped that scientific colleagues in China will be able to make limited airborne measurements as well extensive ground-based observations that can be used to help evaluate emissions, transport, and near-source chemical processing.

3.2 Airborne observations

The scientific questions detailed above require aircraft payloads capable of sensitive and accurate measurements of reactive gas species, tracers compounds, photolytic fluxes, meteorological parameters, and aerosol microphysical, chemical, and optical properties. A list of measurement requirements is provided in Table 1, separated into essential measurements and those that provide complementary information that would aid in analysis and interpretation. Specific instruments and techniques are not identified.

The primary goal of airborne measurements during the IMPACT project is to quantify the changes in pollutant abundance and characteristics during transport from the boundary layer to the free troposphere and across the Pacific Ocean, and the continued transformation of these pollutants as they are carried into the Arctic or western North America. Once lifted, FT transport typically

Figure 7. Schematic of airborne sampling strategy in the WCB. Large turboprop aircraft sample the WCB inflow, cloudy region, and mid-level outflow, while the DC-8 aircraft samples the high-altitude outflow and trans-Pacific regions. Remote sensing aircraft probe cloud and precipitation structure while satellites measure cloud properties and pollution abundances and spatial distributions in cloud-free air.
occurs in narrow, laminar sheets and filaments, which can be mixed with adjacent air by intermittent turbulence. These small-scale features are not captured by the synoptic weather observation network (particularly sparse over the North Pacific) nor by global-scale models. In the face of these uncertainties in transport, IMPAACT will not try to repeatedly sample the same exact air parcel at multiple times and locations as it is carried across the North Pacific. Instead, IMPAACT will use a statistically based, quasi-Lagrangian approach. Probability distribution functions (PDFs) of pollutant characteristics in the inflow region to a WCB system will be developed by making repeated transects at different altitudes and locations in the airmass. Similar observations will then be made in the outflow region of the synoptic systems, and at various intervals downwind during trans-Pacific transport in the FT (Fig. 7). If one of the aircraft is equipped with a counterflow virtual impactor for sampling cloud residue particles and/or a cloud water collector, sampling will be done in the cloudy region of the WCB below freezing level where airframe icing risk is low. Finally, measurements may be made over Alaska and the western regions of North America in airmasses that have passed through WCBs and across the Pacific. The PDFs of airmass chemical characteristics in all of these environments will be directly compared to global model output, which is typically at a resolution of 10s to 100s of km. Perhaps the most useful comparisons between the measurements and model output will be of species-species relationships, which will show discrepancies that will be attributable to initial conditions (diagnosed in the inflow region) or to chemical or mixing processes (in the outflow/FT region). For example, a PDF of the relationship of BC to CO (which are often co-emitted) in the WCB inflow region in the boundary layer will be compared to model values to assess the accuracy of the emissions inventories and initial transport and aging. After vertical transport in the WCB system, similar observations will be made in the outflow region in the FT, likely indicating that BC has been efficiently scavenged compared to CO (e.g., Oshima et al., 2012). Mixing of stratospheric air and air from other airmasses will be assessed and accounted for using independent tracer species and their relationships to one another (e.g., Xiao et al., 2007; Parish et al. 1996; Methven et al., 2006). Values of BC/CO and of the tracer relationships from the simulations will then be compared to the observed values and used to identify limitations in model representations of transport, wet removal, and chemistry. Continued comparisons at further distances downwind will allow diagnosis of chemistry and mixing during FT transport across the Pacific (e.g., Bertram et al., 2013; Xiao et al., 2007).

This statistically based, quasi-Lagrangian sampling strategy means that sampling must occur at several points during transport across the Pacific. The sampling aircraft must be able to fly from an appropriate airport to a remote location over the Pacific, perform statistically representative sampling, and then return to the departure airport or a new destination while carrying a comprehensive payload. Only a large, long-range aircraft capable of reaching the outflow altitudes of cyclonic systems (~300 hPa) can be used to examine transformations in airmasses as they are carried across the Pacific (Fig. 8). There is only one research aircraft capable of meeting all of these needs: the DC-8 operated by NASA’s Armstrong Flight Research Facility. The NSF G-V and DLR HALO aircraft have the altitude and range capability, but are not large enough to carry the payload needed to address all the scientific questions (Table 1).

A case can be made that the DC-8 alone could meet all of the observational requirements, including sampling in the inflow regions, in the FT across the Pacific, and over the Arctic and western North America. However, cross-Pacific transport in the FT occurs over periods of 4-7 days (Luan and
Jaeglé, 2013) in the springtime. To fly every day while following a pollution transport event from the East China Sea to North America in a single aircraft would create unacceptable crew fatigue and instrument readiness issues. Instead, a more tractable approach would be to use one or more aircraft with the payload, range, and altitude capabilities to sample in the low-altitude WCB inflow region, within the WCB, and the lower portions of the WCB outflow, while the DC-8 samples the high-altitude WCB outflow and subsequently downwind to North America. There are three research aircraft capable of these low-to-mid-altitude measurements with the desired payload: either of two WP-3Ds operated by NOAA (Warneke et al., 2016), a P-3B operated by NASA, and a C-130Q operated by NSF (http://data.eol.ucar.edu/master_list/?project=WIN TER). High-quality scientific payloads capable of addressing most, if not all, of the IMPAACT science questions have been developed for these aircraft. Shared calibration standards and protocols and in-flight comparisons must be an integral part of IMPAACT if changes in species abundances and relationships measured on different platforms are to be attributed with confidence to atmospheric processes.

There are several additional aircraft with more limited range or payload that could be operated in the vicinity of Japan, over Alaska and Arctic Canada, and over the western coast of North America to address specific portions of the larger science questions. These include the U.K. FAAM's BAE-146, the German DLR's Falcon and G-V HALO, the French Falcon and ATR-42, the Alfred Wegener Institute's BT-67s (DC-3s), and the U.S. NSF's Wyoming KingAir and G-V. Additional measurements may be made by colleagues from the Republic of Korea and Japan using chartered aircraft and by French and Russian collaborators on a YAK-42D aircraft over eastern Siberia (Paris et al., 2010; Berchet et al., 2013; Kuz'michev et al., 2016), where East Asian pollution is frequently transported. These aircraft will have more limited instrumentation but could contribute substantially to specific subsets of science questions if payloads can be crafted appropriately and calibrated well.

### 3.3 Ship-borne and surface observations

A global class U.S. ship will make measurements of aerosols and trace gases downwind of the Asian continent in the Sea of Japan and/or along the southern coast of Japan and the East China Sea (Fig. 9). The actual cruise track will depend on the synoptic meteorological conditions driving pollution export and removal and the status and planned flight tracks of the aircraft platforms. The global class vessel will allow for the placement of five 20-ft lab containers housing in situ instrumentation high on the foredeck to minimize contamination from the ship’s stack. Remaining
deck space will be available for remote sensing instrumentation and lab containers dedicated to sampling surface seawater properties. This complement of in situ and remote observations will allow for a full characterization of clouds and precipitation, aerosol properties, atmospheric composition, and surface seawater in the sampling region. Ship positioning and sampling will be made in coordination with aircraft, satellites, and nearby ground stations. The ship will complement measurements made by the other platforms by providing continuous near-source data up- and downwind of Japan. These data will help assess mechanisms of pollution transport and removal and provide constraints for model parameterizations.

The Japanese R/V Mirai will frequently be operated at the fixed "K2" location for oceanographic research. Japanese researchers are expected to operate instruments to measure BC and CO, and possibly other species, for extended periods of time in 2021. These observations will help constrain the mixing of polluted FT airmasses into the boundary layer over the central Pacific Ocean.

Surface station observations will focus on augmenting or resurrecting a number of existing and past sites located on both sides of the Pacific. Figure 8 shows the location of the proposed sites; many of them are at high altitude with a history of observations and are thus well-suited for the scientific aims of IMPAACT. The goal will be to have as many consistent high-quality observations at as many sites as possible. We anticipate that observations of CO, black carbon, ozone and other species will be made at many of these sites. There is currently no universal standard for black carbon, so consistency and collaboration will be critical for this species. Other target species that could be measured at some sites include key reservoir species (e.g. PAN) that are thought to be important for long-range transport. Observations of aerosol scattering and size distribution will also be implemented at a subset of sites.

3.4 Satellite remote sensing observations

Satellite remote sensing observations relevant to IMPAACT include both polar orbiting and geostationary satellite platforms. Trace gas measurements from the polar orbiting Cross-track Infrared Sounder (CrIS) and Advanced Technology Microwave Sounder (ATMS) on the Joint Polar Satellite System (JPSS-1 and JPSS-2), the Infrared Atmospheric Sounding
Interferometer (IASI) onboard the EUMETSAT MetOp platforms, and the TROPOspheric Monitoring Instrument (TROPOMI) on board of the Copernicus Sentinel-5 Precursor satellite will provide measurements of tropospheric O3, CO, CO2, CH4 (CrIS/ATMS, IASI, TROPOMI), and high spatial resolution NO2, HCHO (TropOMI) for tracking intercontinental trace gas transport. The Visible Infrared Imaging Radiometer Suite (VIIRS) instrument on the JPSS series will provide aerosol optical depth retrievals for tracking intercontinental aerosol transport.

In addition to these polar orbiting satellites, we will have a suite of geostationary satellites for observing global air quality including three ultra-violet (UV) and UV plus visible (UV-Vis) spectrometers in geostationary orbit over Asia, North America, and Europe. These geostationary UV and UV-Vis spectrometers will provide hourly retrievals of ozone (O3), nitrogen dioxide (NO2), sulfur dioxide (SO2), and formaldehyde (HCHO) columns as well as aerosol optical depth (AOD) over the major industrialized regions of the northern hemisphere.

The Geostationary Environment Spectrometer (GEMS) instrument is planned to be launched by the Korea Aerospace Research Institute in 2019 onboard the Geostationary Korea Multi-Purpose Satellite (GeoKOMPSAT). The Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument is part of the NASA Earth System Science Pathfinder (ESSP) Earth Venture (EV) program and is planned to be launched no later than 2021 as a hosted payload instrument onboard a commercial satellite. The TEMPO viewing footprint will cover the continental U.S., including the West Coast, and extend north to the Canadian oil sands, and south to Mexico City and the Yucatan Peninsula. The European Sentinel-4 UV-Vis and Near-infrared sounder (UVN) is planned to be launched onboard the Meteosat Third Generation Imaging (MTG-I) satellite in 2022.

3.5 Modeling

Numerical modeling of pollutant emission, transport, evolution, removal, and effects on air quality and climate must be an integral part of the IMPAACT project. They are the tools with which we understand the consequences of observed characteristics and processes, analyze future scenarios, and develop policies that can alter future atmospheric composition and climate. Models on a variety of scales, from box models to global CCMs will be used.

Diverse comparisons between models and observations and between different models is a primary motivation for IMPAACT. In particular, relatively poor comparisons between observations and modeling of tropospheric ozone distribution and trends (Derwent et al., 2016; Emmons et al., 2015; Parrish et al., 2014), of CO (Shindell et al., 2006) and of aerosol BC and SO4 (especially in the Arctic) (Koch et al., 2009; Schwarz et al., 2010) have pointed to the importance of parameterizations of wet removal on these species (Q. Wang et al., 2014; Fisher et al., 2011). Ongoing sensitivity studies support the assertion that transport and removal of precursor species in WCB systems is a key determinant in model performance. Prior to the IMPAACT observation period, planning would benefit by using existing model output to identify key species subject to model diversity. If possible, additional analyses that would investigate how transport through mixed-phase vs. liquid-only clouds affects this diversity, and others focused identifying regions of initial uplift in WCB systems, would be helpful for mission planning.

High-resolution models ranging from regional to global in scale have been developed and used as forecasting tools for observational field programs (e.g., Barth et al., 2015). Some of these models use forecast weather with offline chemistry, while others are fully coupled CTMs that include feedbacks between atmospheric
composition and meteorology. A third class of models assimilates satellite observations of key gas phase species (such as CO) and aerosol optical depth (AOD) to improve prediction of atmospheric chemistry (e.g., RAQMS; Fig. 4). Finally, Lagrangian forward trajectory models (with or without parameterized or explicit chemical schemes) have been used to identify transport from specific sources or regions.

All the above model types have been used to guide observational platforms during field projects. Each has strengths and weaknesses and provides useful information that can be used to advise decision-making in the field. Even over the North Pacific, where meteorological observations are sparse, chemical transport models can provide significant skill that can be very useful in flight planning. As an example of this capability, in Fig. 10 we show output from the GEOS-5 CTM used during the recent ATom flight campaign. This model predicted a narrow band of highly polluted air from East Asia over the central North Pacific associated with a wave cyclone, by chance lying along the predetermined flight path of the NASA DC-8 aircraft. The aircraft did indeed penetrate the narrow band of pollution. The exact altitude of the polluted layers, their vertical and horizontal scale, and the mixing ratio of CO within the layers did not align perfectly with the CTM output. This example shows that modern CTMs have sufficient accuracy to track Asian pollution events across the Pacific Ocean and can provide useful flight planning guidance even in this data-poor region. It also shows that there are limitations to the accuracy of the simulations for flight planning and for data analysis, and emphasizes that comparisons of species-species correlations within the polluted regions in the measurements and models may provide more useful insights into model performance instead of directly comparing values at specific locations and times.

Figure 10. 96-hr forecast CO mixing ratio from fossil fuel combustion only at 850 hPa (top panel) and cross section along planned aircraft flight track (middle panel) from the GEOS-5 model. Lines with waypoint labels show the planned aircraft flight path. Bottom panel: altitude-time plot of NASA DC-8 flight track during ATom-2 flight on 1 February 2017 from Anchorage, Alaska to Kona, Hawai‘i, color-coded by measured CO mixing ratio. Note that the aircraft encountered high mixing ratios of CO over the central N. Pacific, but not at the precise locations predicted by the model. Model plots courtesy of H. Bian and. Aircraft position data courtesy of P. Bui, and CO mixing ratios courtesy of R. Commane, B. Daube, and S. Wofsy.

The successful exploration of the IMPAACT science questions will require tightly coupled post-campaign observational analysis and modeling activities. This analysis will rely on a cascade of models that can be applied to the
range of attribution studies, process-based explorations, and global impact assessments envisioned as part of IMPAACT. To that end, the analysis will engage modelers specializing in Lagrangian transport modeling, large-eddy simulation, regional and global chemical transport modeling, and chemistry-climate modeling.

Table 1. Airborne measurement needs, payload priority, and science questions addressed.

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<tr>
<th>Parameter</th>
<th>Priority</th>
<th>Science questions</th>
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<td>NO$_y$, NO, NO$_2$, HNO$_3$, PAN, PPN, ANs. N$_2$O$_5$, NO$_3$ if nighttime measurements made.</td>
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### Aerosol lidar: vertical structure and optical properties

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<td>Cloud radar: cloud structure and phase</td>
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### Table 2. Shipboard measurement needs, payload priority, and science questions addressed

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<td>Aerosol thermal volatility at 230C</td>
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<td>Cloud liquid water path</td>
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