Megacity Air Pollution Studies–Seoul

(MAPS–Seoul)

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Executive Summary

For the last 10 years, despite comprehensive management efforts, the Seoul Metropolitan Area (SMA) has been confronted with perplexing air quality problems especially in $PM_{2.5}$ and O_3. With this, the second phase of the air quality mitigation plan proposed by the government, which is set to start in 2015 has been set to further improve the SMA's air quality for the next 10 years. Identifying the attainable air quality policy and laying out of the practical emission reduction plan should be based on scientific evidences and an assessment of the current and future status of air quality. However, understanding and predicting the behaviors of air pollutants in the SMA remain very difficult as large discrepancies still exist between the modeled simulation and observed concentrations for these species. These issues are mostly caused by lack of detailed understanding of precursor emissions, transport paths, and chemical mechanisms and by insufficient representation of these processes in current air quality simulations.

Integrated research covering chemical transport model, ground, and satellite monitoring is the primary scientific tool used to assess present chemical characteristics, future trends, emission sources, transport mechanisms, and model uncertainties in air quality. In addition, it provides the key evidences necessary to identify air quality issues, revise policies, and evaluate the effectiveness of the implemented policy. This plan proposes a scientific research program integrating in-situ observations with one preliminary (in 2015) and two intensive (in the summer of 2016 and the winter of 2017) field campaigns in the SMA with the use of remote and satellite sensors, and modeling platforms. This will be partly conducted in collaboration with the National Aeronautics and Space Administration (NASA) and other international scientific communities. This plan utilizes the latest research tools and best available collaboration to achieve the proposed scientific goals.

The anticipated outcomes can be identified as part of the capacity-building process of air quality model simulation and monitoring techniques, the development of improved emission inventories, and the regional assessment of long-range transports of air pollutants, realistic projections for future air quality along diverse scenarios, assessment of air quality impact on ecosystem, health, and climate over the SMA.

1. Introduction

The Seoul Metropolitan Area (SMA) has faced challenging and ongoing air quality issues despite its recent 10-year efforts to improve the SMA's air quality to the level of Tokyo and Paris by 2014. In 2005, the Korean government had legislated a Special Act to implement a series of strict policies of reducing air pollutant emissions to attain the levels to 40 μg/m^3 and 22 ppbv by 2014 from 69 μg/m^3 and 38 ppbv of PM_{10} (particulate matter) and NO_2 in 2003, respectively. Despite the successful performance of comprehensive reduction plans for NO_2, PM, and VOCs (volatile organic compounds) emissions from major primary sources, such as mobile and large-scale industrial sources, the air quality in the SMA has still not reached the levels as planned, especially for NO_2 and O_3 (Figure 1).

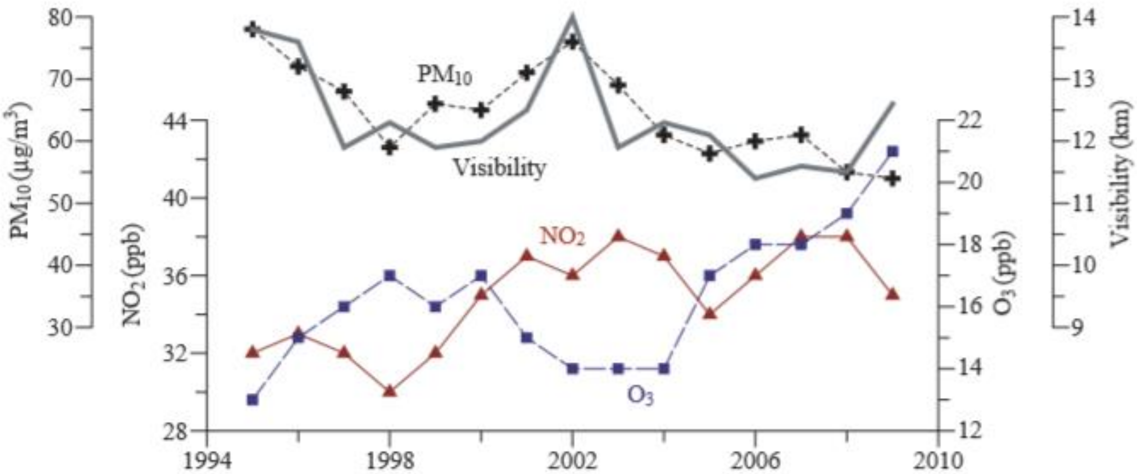

Figure 1. Annual variations of selected air pollutants in Seoul (source: Ghim, 2012).

The annual mean NO_2 and O_3 showed noticeable increases over the measured period. Despite the fact that the PM_{10} level has decreased consistently not only in the SMA but throughout the rest of Korea, it still frequently exceeds the corresponding air quality standards; for example, only 4 (1.7%) out of 239 nationwide monitoring stations satisfied the 24-hr–based PM_{10} standard in 2011 (Kim et al., 2013). In addition, considering the observed O_3 increase for the past 10 years, and the fact that $PM_{2.5}$ mostly originated from secondary sources of photochemistry, it cannot be ruled out that $PM_{2.5}$ may also have been increased in the same time frame. Despite the decrease in annual mean PM_{10} concentrations, there has been an increase in episodic regional pollution events causing significant public health issues and deteriorating visibility in the SMA. The scientific assessments on regional pollution transport and photochemical processes are critical to understand the ineffectiveness of current control strategies and emission reductions in achieving targeted air quality levels in some cases, such as annual NO_2, O_3, visibility, and PM during regional-scale pollution incidents.

Chemical transport models play important roles in the air quality forecasting and control strategies for the public. However, large discrepancies between the simulated and observed air quality concentrations still exist, indicating poor scientific understanding on the processes related to air quality at present. In particular, the air quality model used in the forecast tends to underestimate the observed PM mass concentrations (Figure 2). Uncertainties associated with emissions, meteorology, and physical and chemical processes in the model could cause this discrepancy.

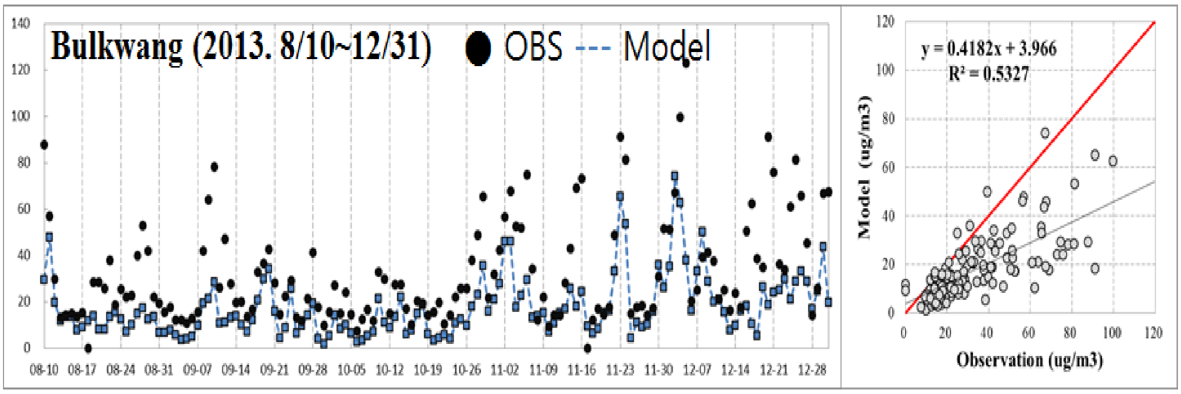

Figure 2. Time series (left) and scatter plot (right) comparisons between the observed and simulated $PM_{2.5}$ mass concentrations at the Bulkwang site in Seoul, Korea for August 10 – December 31, 2013.

Individual processes in the model should be checked given that the model should be tested against observations for better evaluation of their performances as a whole. With this, no rigorous evaluation of the model that focuses on chemical species and individual processes has been attempted in Korea so far. Such evaluation would bring a clearer picture on what causes the bias in the model relative to observations. Therefore, there is a high need to have extensive observations to diagnose issues, including the chemical mass closure of PM_{10} and $PM_{2.5,}$ and the evaluation of each chemical species contributing to PM and O_3 in the model, and to further update scientific understanding regarding the subject.

The air quality goals and effectiveness of emission regulation utilizing air quality models have been heavily based on the Clean Air Policy Support System (CAPSS), which is compiled and maintained by the National Institute of Environmental Research (NIER) and the Korea Ministry of Environment for emission inventory in Korea. However, it is clearly indicated that CAPSS does not have the capacity to resolve the annual trends of these species because of either unidentified emission sources or unreliable emission factors and activities. It is also critical to characterize the impact of natural background VOC emissions (BVOCs; biogenic volatile organic compounds) on production of secondary air pollutants; organic carbon fractions in $PM_{2.5}$ mostly originate from BVOCs controlled by anthropogenic pollution levels (Carlton et al., 2010). However, the emission and photochemical processes of BVOCs have not been clear enough to constrain regional pollution issues (Lee et al., 2013).

The trans-boundary transport from continental outflows is another important factor to the SMA's air quality (Park et al., 2011). Trans-boundary source contributions both to annual mean PM and ozone concentrations have not been well characterized because of uncertainties in local and regional emission sources. With this, the impact of continental outflows from China has been clearly evident over Northeast Asia and has been more frequent in the recent years (Yang et al., 2013). Thus, spatially and temporally extensive studies considering emission source strength, meteorological conditions, and transport pathways are required to accurately assess the contributions of transport.

Capabilities of satellite remote sensing of tropospheric gases and aerosols have been expanded since the launch of Terra into the sun synchronous orbit in 1999. Onboard Terra, the Measurement of Pollution in The Troposphere (MOPITT), and the MODerate resolution Imaging Spectroradiometer (MODIS) have provided valuable information on

CO and aerosols in the troposphere over 10 years. The SCanning Imaging Absorption spectrometer for Atmospheric CHartographY (SCIAMACHY) onboard Envisat provided concentrations of O_3, HCHO, SO_2, NO_2, NO, NO_3, CO_2, CH_4, N_2O, and BrO from 2002 to 2012, and the Ozone Monitoring Instrument (OMI) onboard Aura has provided concentrations of O_3, NO_2, SO_2, HCHO, BrO, OClO, and aerosol from 2004 to the present. The Global Ozone Monitoring Experiment (GOME) and GOME-2 have also provided valuable information on ozone and relevant gases. Ozone Monitoring Profiler Suite (OMPS) aboard NASA's Suomi National Polar-orbiting Partnership (NPP) Satellite is measuring profiles of ozone, together with aerosol and column densities of SO_2 and NO_2 since its launch in 2011. In the future, the TROPOspheric Monitoring Instrument (TROPOMI) is planned to be in low earth orbit (LEO) by 2016 to measure most of the aforementioned gases' concentrations and the Geostationary Environment Monitoring Spectrometer (GEMS) in a geostationary earth orbit (GEO) by the end of 2018.

International campaigns conducted previously provided valuable dataset and improved our understanding of atmospheric chemistry and their role in regional climate change. The Distributed Regional Aerosol Gridded Observation Network (DRAGON)–Asia campaign in spring of 2012 produced sun photometer dataset in mesoscale grid over China, Japan, and Korea to characterize the physical and chemical properties of aerosol, and to evaluate and improve the satellite remote sensing using the Geostationary Ocean Color Imager (GOCI). Although some of these campaigns contributed to the evaluation and improvement of the satellite remote sensing products, such have been limited in terms of facilities and scales. With this, a large demand to carry out the campaign for aerosols and gases together with their precursors three-dimensionally using both ground-based and airborne platform still exists. Furthermore, by the end of 2018, the Asian region will be presented with the first geostationary mission called the Geostationary Environment Monitoring Spectrometer (GEMS) to monitor tropospheric gas concentrations in both high temporal and spatial resolutions, together with the Advanced Meteorological Imager (AMI) and GOCI-2 onboard the Geostationary Korea Multi-Purpose Satellite (GEO-KOMPSAT)-2. Thus, it is crucial to carry out an airborne campaign to measure the concentration of gases and aerosols in order to fully prepare for the success of GEO-KOMPSAT-2 Mission.

The present air quality policy in the SMA focuses on two major issues. The first one is the dense smog occurrences that often occur during the late fall to early spring season despite such events being episodic. This phenomenon is mostly caused by continental outflows driven by the prevailing northwesterly wind over the East Asian region including the Korean Peninsula. In a high-aerosol episode, the stagnant high-pressure system over Korea played a decisive role in the accumulation of air pollutants (Lee et al., 2013).These episodic heavy stagnant events have been worsening in a sense that PM_{10} and SO_2 concentrations have been observed with higher concentrations over the past decades. These observations are consistent to recent emission trends of China (Gu et al., 2013) and long-term satellite studies over this region (Mijling et al., 2013; Zhao et al., 2013).

The second issue is the increasing trend of O_3 and NO_2 for the last decades despite nationwide efforts to reduce its precursors, NO and VOCs. Despite the constant increase of automobiles in the SMA, the nitrogen oxide (NO_X) emission decreased steadily because of the successful NO_X emission control strategy (Figure 3). At the same time with the significant decrease of NO_X, it is unsettling to observe that NO_2 has relatively increased for the last two decades. No significant annual trend in the relative fraction of NO_2 in NO_X

emission ($f(NO_2)$) at roadside measurement sites was found although NO_X emission control strategies, such as diesel particulate filter (DPF) or diesel oxidation catalyst (DOC), should have direct influences on $f(NO_2)$ values (Sohn et al., 2011).If it is to be assumed that the national emission control strategy has been effective as claimed, then the reasonable speculations would be 1) additional NO_X sources either from regional transport or/and 2) longer NO_X photochemical chain length that could extend the chemical lifetime of NO_X. The latter reason may be able to potentially explain the increasing trend of secondary photochemical pollutants, such as O_3, as they are the direct reaction by-product of the NO_X chain cycle. However, it should also be noted that the observed NO_2—which make up most of the NO_X pool—is known for NO_2*, which implies that there is a higher level of NO_2 than the actual NO_2 level in the atmosphere because of artifacts from thermally unstable NO_y species such as PANs or HNO_3.

The emission control strategy during the Special Act Period relied more on NO_X than VOCs; for instance, NO_X emission reduction by 53.0% and VOCs by 38.7% until 2014. However, it turned out that VOC emission control measures were not effective to reduce VOC emissions in recent years. Based on the yearly VOC emission in Seoul, which has been highly uncertain so far, the decreasing rate of VOC emission has been much slower than those of NO_X for the last 10 years (Figures 3 and 4).

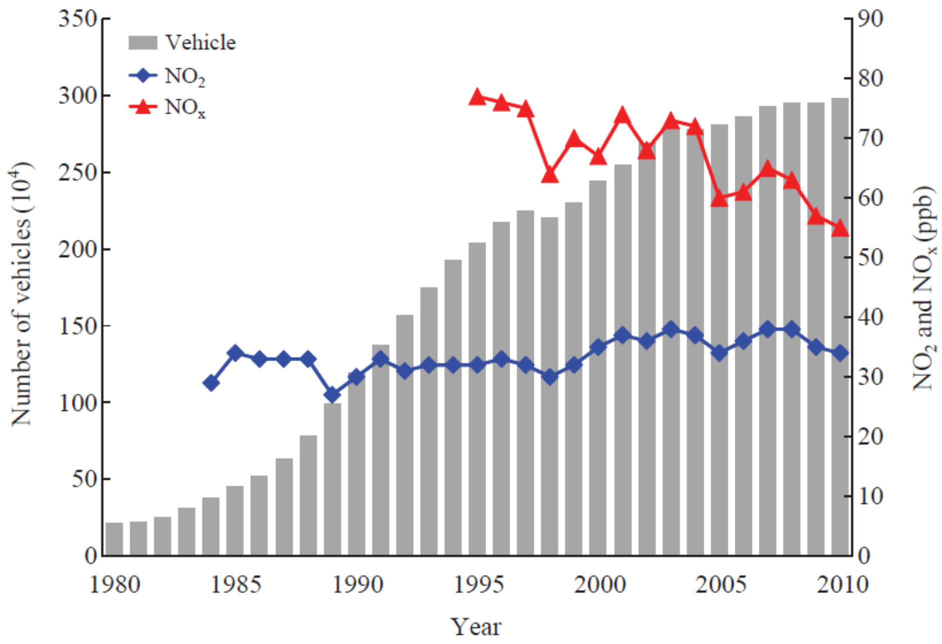

Figure 3. Trend of the yearly average ambient concentrations of nitrogen dioxide, the oxides of nitrogen, and number of vehicles in Seoul between 1980 and 2010 (Kim and Yeo, 2013).

In the absence of recently reported VOC concentrations in the SMA, it can be assumed with caution that an increase ratio on VOCs/ NO_X in the SMA is due to their unbalanced emission changes. It is well known that photochemical air pollution can worsen under the increasing ratio of VOCs/ NO_X with the decreasing concentrations of NO_X in most metropolitan areas (Seinfeld and Pandis, 1998). Tang et al. (2009) investigated average O_3 trends in Beijing over the period from 2001 to 2006 and interpreted that reduced NO_X emissions and elevated VOC emissions contributed to the increased O_3 concentrations. It is very likely that the increasing ozone can be attributed partly to the increasing VOCs/

NO_X ratios in Seoul as well. With this, tropospheric ozone concentrations have been rapidly increasing over Asia. At the remote mountain site of Mt. Happo, which lies directly downwind of the Asian mainland, O_3 has generally increased at a faster rate than that of European and North American sites. This rapid increase has been attributed, at least partially, to the rapid increase of O_3 precursor emissions that has occurred in East Asia (Tanimoto et al., 2009). Ozone pollution was originally identified as a local and urban problem; however, it has been recognized as a regional issue given that ozone and its precursors are chemically stable to be transported in the range of the synoptic scale (i.e., a few days of one's lifetime). Thus, there is a need to identify the degree of contributions from the transported origin or changes in baseline tropospheric ozone concentrations that are tied to the increasing O_3 level in Seoul. A forest study near Seoul suggested that BVOC emissions could enhance regional daytime ozone production from 5 to 20 ppbv. The observed temporal variations evidently reflected the influence of BVOCs on ozone formation and such findings strongly indicated that interactions between anthropogenic pollutants and BVOCs must be understood and quantified in order to assess the photochemical ozone formation in Seoul (Kim et al., 2013a).

The previous air quality policy in the SMA has focused on likely controllable pollutant types and sources (for example, transportation and industrial sectors), and this has made significant progress in terms of mainly primary pollutants such as PM_{10}, NO_X, and CO. However, a number of serious air quality issues remain unresolved; in particular, the contributions of air pollutants transported and their chemical evolutions during the transport; the recent change of oxidizing capacity in the SMA and background region; air quality impacts because of the changing VOCs/ NO_X ratio, and BVOC emission in the SMA and its surrounding region. The aforementioned gas-phase photochemistry can be directly applicable to the understanding of secondary organic aerosol formation processes, especially for $PM_{2.5}$ or lower, which is mostly formed by condensable gases such as sulfuric acid and organic acid from photochemical reactions. In addition, it is also anticipated that the emissions of key pollutants over Northeast Asia will continue to grow.

To further improve the SMA's air quality for the next 10 years with the second government mitigation plan, which is set to start in 2015, a new legislation and regulation should be reinforced or evaluated in conjunction with scientific findings. To be effective for SMA's air quality management, a series of high-priority scientific researches in chemical perspectives will need to address all of these current challenges. In summary, the accumulated experience in air pollution research and policy implementation in the past decades of the field clearly brings up the need to establish an integrated research program to investigate emission–photochemistry–transport phenomenon affecting the SMA's air quality with the most advanced research tools, such as in-situ and remote observations, so that emission inventories and regional air quality models can be thoroughly constrained. These scientific outcomes will eventually serve as solid basis to establish an air quality policy that can be implemented with great confidence.

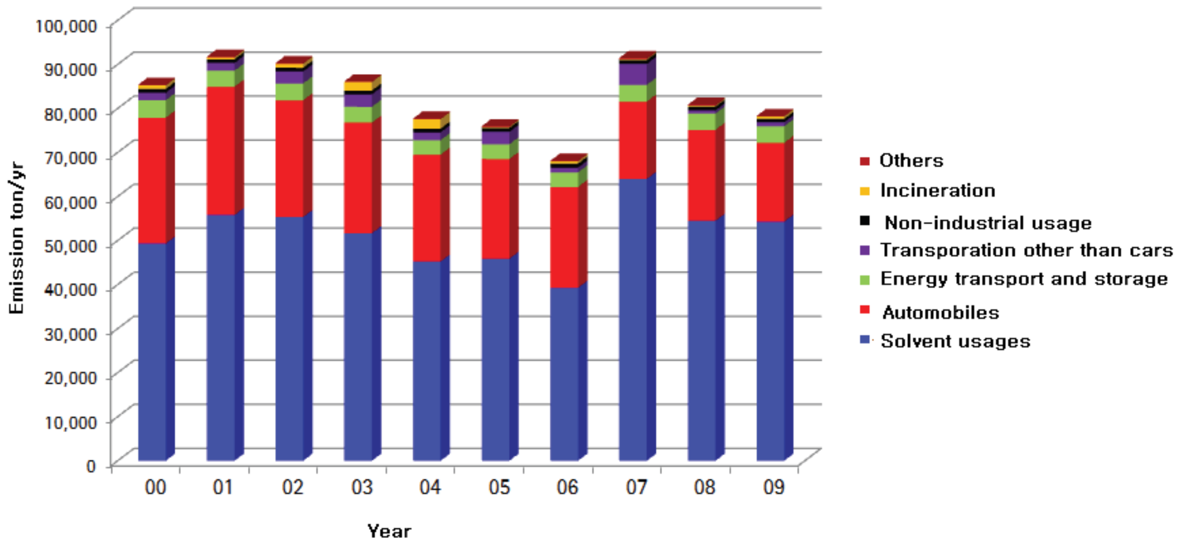

Figure 4. Trend of the yearly VOCs emissions in Seoul between 2000 and 2009 (Choi and Ko, 2013).

2. Overall Goals

The overall goal of MAPS–Seoul is to establish a scientific research program that integrates in-situ observations, remote sensing such as satellite sensors, and modeling platforms to provide snapshots of current air quality status and improve forecasting tools that can be utilized routinely now (daily–seasonal forecasts) and to eventually evaluate the effectiveness of air pollutant reduction policy scenarios. MAPS–Seoul's objectives are as follows:

① Increase the performance of the air quality model prediction by reducing the bias in model simulation, and strengthen management decisions in policy development
- Improve emission inventories, particularly for anthropogenic NO_X, VOCs, NH_3, CO, SO_2, and biogenic VOCs (BVOCs)

② Assess long-range transports of pollutants from Asian megacities, including Beijing, Shanghai, and the SMA
- Characterize the chemical evolutions of oxidants and aerosols in and out of the SMA, including O_3, NO_y, secondary organic aerosol (SOA), and their precursors to precisely constrain critical photochemical processes that should be considered in the air quality forecast modeling framework

③ Validate aerosol and selected gaseous products of GOCI and other satellite sensors to integrate satellite observational products for reliable urban and regional air quality and emission inventory analysis
- Characterize the optical/physical/radiative properties of aerosols in the SMA and surroundings to resolve the effects of air quality on visibility reduction and urban weather forcing

Field studies with one preliminary and the two intensive field campaigns in summer and winter in the SMA are proposed and would be conducted in collaboration with NASA and other international scientific communities. This plan utilizes the latest research tools and best available collaboration to achieve the proposed scientific goals.

3. Scientific Aims

Previous studies illustrate that a comprehensive approach is essential to understand the current status of air quality problems as there are multiple fronts of uncertainties in emission, transport, and chemistry that have been reported in the regional scale. The current method for resolving discrepancies between observed and model-predicted targeted air pollutants (e.g., O_3 and PM) is to apply empirical correction factors in the prediction model system without solid scientific basis. This may work unless there are no significant physical and chemical environmental changes affecting the photochemical processes in the region. However, it is well known that the East Asian region is experiencing rapid environmental changes caused by global-scale climate change and ambitious pollution reduction plans by the Chinese local and federal governments. The top aim for this program is to implement an intensive field observation experiment to study the ozone and $PM_{2.5}$ pollutions in the SMA governed by secondary chemical processes and transport processes. The specific goals targeted within this program will include the following:

① It is crucial to improve the representation of critical processes in models for use of air quality forecasting with spatially comprehensive observations of in-situ grounds, flights, ships, and satellites. The critical processes in the model include natural and anthropogenic emissions, chemical transformation, wet and dry deposition, and atmospheric transport of air pollutants and their precursors.

② The attained research findings must anticipate and provide scientifically reliable information for the future air quality mitigation strategy over the SMA.

③ Within the framework of an internationally collaborative field campaign, it is important for Korean scientists to build scientific capacity in air quality observations, especially in aircraft and radical measurements.

④ It is very important to assess the accuracy of satellite measurements of aerosol optical depth and column density of trace gases through intercomparison with airborne and ground-based measurements. Hourly aerosol products from GOCI can be provided and validated with ground-based and airborne measurements. The first version of the GEMS retrieval algorithm can be tested using the radiance spectrum of either OMI or TROPOMI to be in low earth orbit by 2016. The hourly aerosol products from GOCI in particular are crucial to estimate the effect of long range transport from continents to the Korean peninsula.

The experimental design will be fully coordinated with modeling and remote sensing research communities to validate and improve their capability to be integrated into a comprehensive air quality diagnosis and prediction system.

4. Research Questions and Directions

1) Scientific Questions for Atmospheric Chemistry

① Is it feasible to establish a 'baseline' condition that characterizes air quality relatively free to the onset of most local and anthropogenic activities in the SMA? An analysis of baseline air quality therefore provides an opportunity to identify the contributions from local and regional sources and offers the capacity to trace the long-term air quality changes in regional scale.

② Can we observe or determine the production and loss rates of oxidants (O_x, H_2O_2, RO_2, HO_x) along VOC/ NO_X ratios at different parts of the SMA? What is the most dominant VOC species for oxidant productions in an urban area, and what extends urban NO_X influence to downwind ozone production? The ozone in the urban area of the SMA is VOC-limited and NO_X-inhibited, while the reverse is true in the vegetated outskirts of the SMA. The variation of oxidants at different VOCs and NO_X regime in the SMA is critical to track the chemical evolutions of photochemical O_3 and SOA along the megacity plume.

③ What are the production rates of secondary aerosols during the photochemically active periods or the transported events? In addition, what precursor species are the main drivers for high aerosol loadings?

④ Can we segregate the contributions from anthropogenic and natural origins for photochemical oxidants and aerosol species? Or can we quantify the contributions from different source regions, especially transported vs. local sources?

⑤ How much do the nighttime NO_3 and Cl radicals contribute to the production of aerosol and photochemical species in consecutive daytime periods? Although no measurements for these radical species have been made in the SMA, it is reasonable to consider that these nighttime radicals may play significant roles in the production of oxidized hydrocarbons that are, in turn, essential ingredients for ozone and secondary aerosols.

⑥ What is the extent of the ground ozone and aerosol interaction or exchange with those in elevated levels? The detailed vertical profiles and their temporal and spatial progression of ozone, aerosol, and precursor species will provide the insight about the unresolved sources of ozone and $PM_{2.5}$ in the SMA. To determine the manner in which the horizontal and vertical distributions of air pollutants relate to synoptic meteorological conditions (e.g., stagnation, clouds). It is necessary to identify the meteorological conditions in which in situ and column-integrated optical properties agree. To identify the role of the vertical structure of the boundary layer and the turbulent mixing processes during pollution episodes.

⑦ What are the effects of heterogeneous chemistries on aerosol and oxidant productions in the SMA? While homogeneous reactions are familiar pathways to form the secondary gases and aerosol species, more evidences have converged on the conclusion that heterogeneous reactions over the aerosol and aqueous phase droplets may lead to a large enhancement of aerosol and oxidant production, especially in conditions of heavy aerosol loads such as in the SMA.

⑧ What is the relationship between aerosol properties and their radiative forcing? It is necessary to investigate the effects of locally emitted and long-range transported pollution aerosols on the radiation budget in the SMA.

2) Air Quality Models and Emission Perspectives

① Develop "the best-available" domestic and foreign anthropogenic emissions of O_3 and PM precursors and improve them using the field campaign measurement interactively.
- Develop top-down emissions inventories of aerosols and their precursors over East Asia using adjoint inverse modeling.

② Quantitative evaluation of the systematic model under-prediction in PM_{10} and $PM_{2.5}$ forecasts and mechanistic analysis of model processes.
- Analyze PM_{10} and $PM_{2.5}$ mass closure.
- Diagnose the most important PM species for the bias in the model.
- Quantify source-receptor relationships of O_3, PM, and their precursors between East Asian countries: China and Korea; China and Japan; Korea and Japan.
- Understand simulated transport of O_3, PM, and their precursors in association with the horizontal and the vertical structure of a marine boundary layer.

③ Understand the role of natural sources in O_3 and PM formation and their impact on air quality degradation including biogenic VOC emissions in SMA.

④ Assess the impact of the marine boundary layer on O_3 and PM using vertical profiles of measured air pollutants and meteorological parameters in and around Korea.

3) Satellite Application

① Estimate the effect of long-range transport from continents using satellite remote sensing data in high temporal and spatial resolution

② Test GEMS retrieval algorithms under development using the raw data of OMI(and/or TROPOMI and/or airborne remote sensing)

- Investigate the vertical distribution of trace gases and aerosols and their effects on retrieval accuracy
- Quantify the effect of clouds on the retrieval processes of trace gas concentrations

③ Inter-compared observations from ground-based, airborne and satellite platforms.

- Validate the trace gas products of OMI and OMPS (and/or TROPOMI) to be launched in low earth orbit by 2016 with PANDORA, DOAS and LIDAR
- Validate the aerosol products of GOCI and other relevant satellites
- Understand the effect of aerosol properties on the retrieval accuracy of NO_2

5. Recent Progress and Findings

1) Air Quality Models and Emission Studies

Scientists in Korea have made significant improvements in the regional emission inventory for Korea and East Asia. The National Institute of Environmental Research (NIER) and Konkuk University (KU) jointly developed an Asian emissions inventory, named NIER/KU-CREATE (Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment). For anthropogenic emissions, it has 54 fuel classes, 201 sub-sectors and 13 pollutants, including SO_2, NO_X, CO, NMVOC, NH_3, OC, BC, PM_{10}, $PM_{2.5}$, CO_2, CH_4, N_2O, and mercury. It also includes biogenic and biomass burning emissions inventories using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) and BlueSky–Asia frameworks, respectively. Because the CREATE emission inventory was developed using the international climate and air quality assessment modeling framework (i.e. GAINS) and is fully connected with the most comprehensive emission processing/modeling systems (i.e. CMAS of US EPA), it can be used in support of various climate and air quality modeling researches and field experiments (Woo et al., 2013). However the domestic emissions of trace gases and aerosols are still very uncertain as often indicated by large discrepancies between the simulated and observed CO and PM_{10} concentrations in surface air. CO, which has a relatively long lifetime, can be affected by the trans-boundary transport from countries upwind, but errors with the national emission inventory cannot be eliminated and could imply possible missing sources of domestic emissions such as barbecuing and other minor sources in Korea.

During 2005–2010, the emissions of SO_2 and $PM_{2.5}$ in East Asia have decreased by 15% and 12%, respectively, mainly because of the large-scale deployment of flue gas desulfurization (FGD) of China's power plants and the promotion of highly efficient PM removal technologies in China's power plants and cement industry. During this period, the emissions of NO_X and NMVOC have increased by 25% and 15%, respectively, driven by the rapid increase in emissions from China because of inadequate control strategies. In contrast, the NO_X and NMVOC emissions in East Asia except China have decreased by 13.2% mainly because of the implementation of stringent vehicle emission standards in Japan and Korea. Under current regulations and levels of implementation, NO_X, SO_2, and NMVOC emissions in East Asia are projected to increase by about one-quarter by 2030 relative to the 2010 level, while $PM_{2.5}$ emissions are expected to decrease by 7% as shown in Figure 5 (Wang et al., 2014). South Korea has ambitiously set its second-phase capitol air quality improvement program labeled as the Seoul Metropolitan Area Air Quality Management Plan (SAQMP), targeting the year 2024. The air quality improvement targets for 2024 are 30 $\mu g/m^3$ and 20 $\mu g/m^3$for PM_{10} and $PM_{2.5}$, respectively. Emissions of PM_{10} and $PM_{2.5}$ are required to be decreased by up to 35% and 45%, respectively, from their future baseline level (Figure 6). Various special measures, such as cap-and-trade, LNB, and EURO standards programs, will be implemented to control emissions over the SMA (Kim Y. et al., 2014). Recent high ozone and PM episodes over the SMA and the entirety of Korea, however, would indicate that the recent penetration of aggressive emission control measures over China and Korea would not be that effective as expected. The transport modeling studies with activity/emission factor/control measure–based comprehensive emissions inventory and precise measurement constraints over in-region boundaries should be conducted to understand the present emissions level and the effectiveness of emission control technologies and policy measures over the regions.

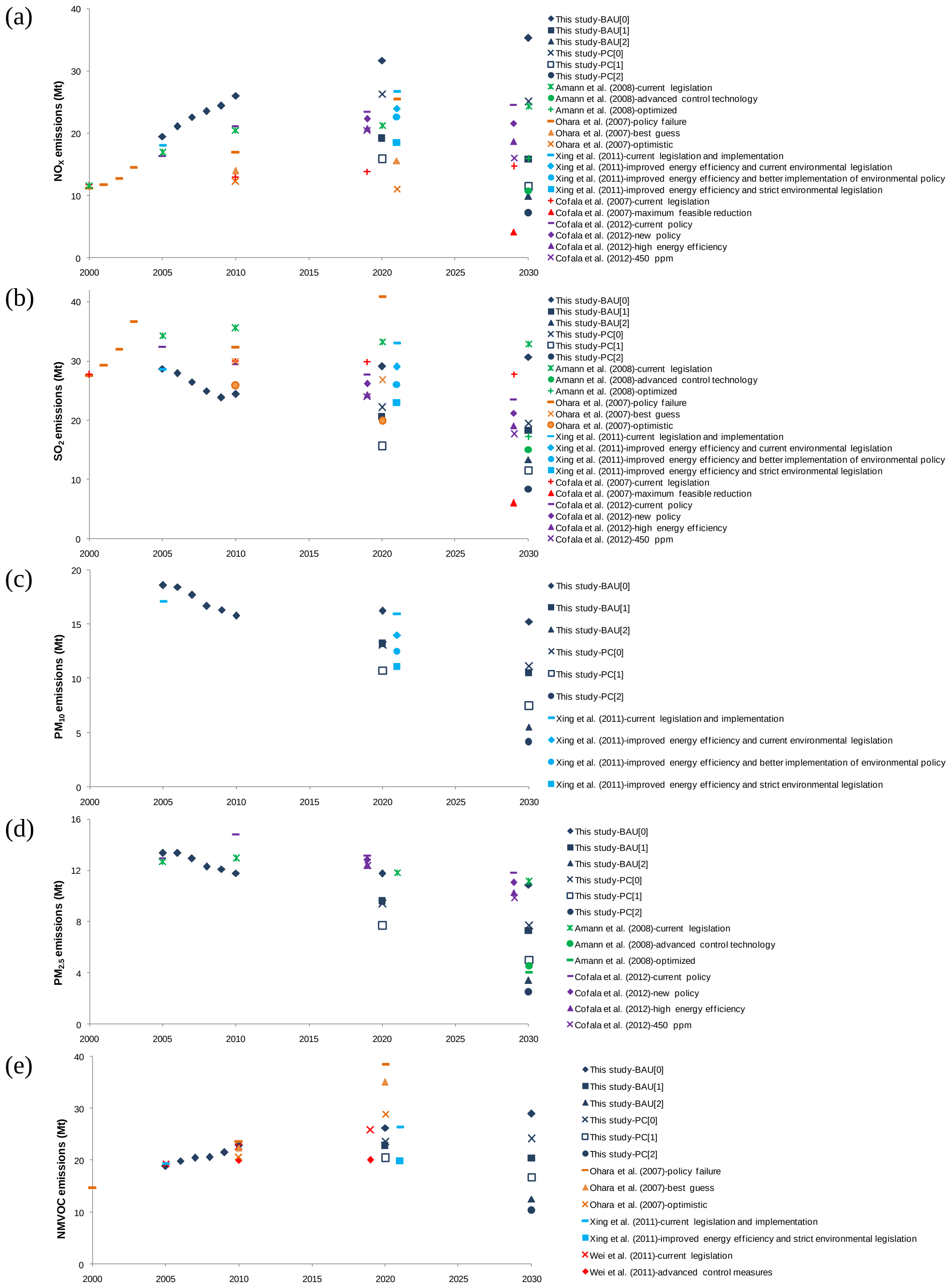

Figure 5. Comparison of emission estimates in this study with other studies: (a) NO_X; (b) SO_2; (c) PM_{10}; (d) $PM_{2.5}$; (e) NMVOC. Scenarios from the same study are shown with symbols of the same color, and only the historical emissions for the first scenario are shown (Wang et al, 2014).

(a) SO_2

(b) PM_{10}

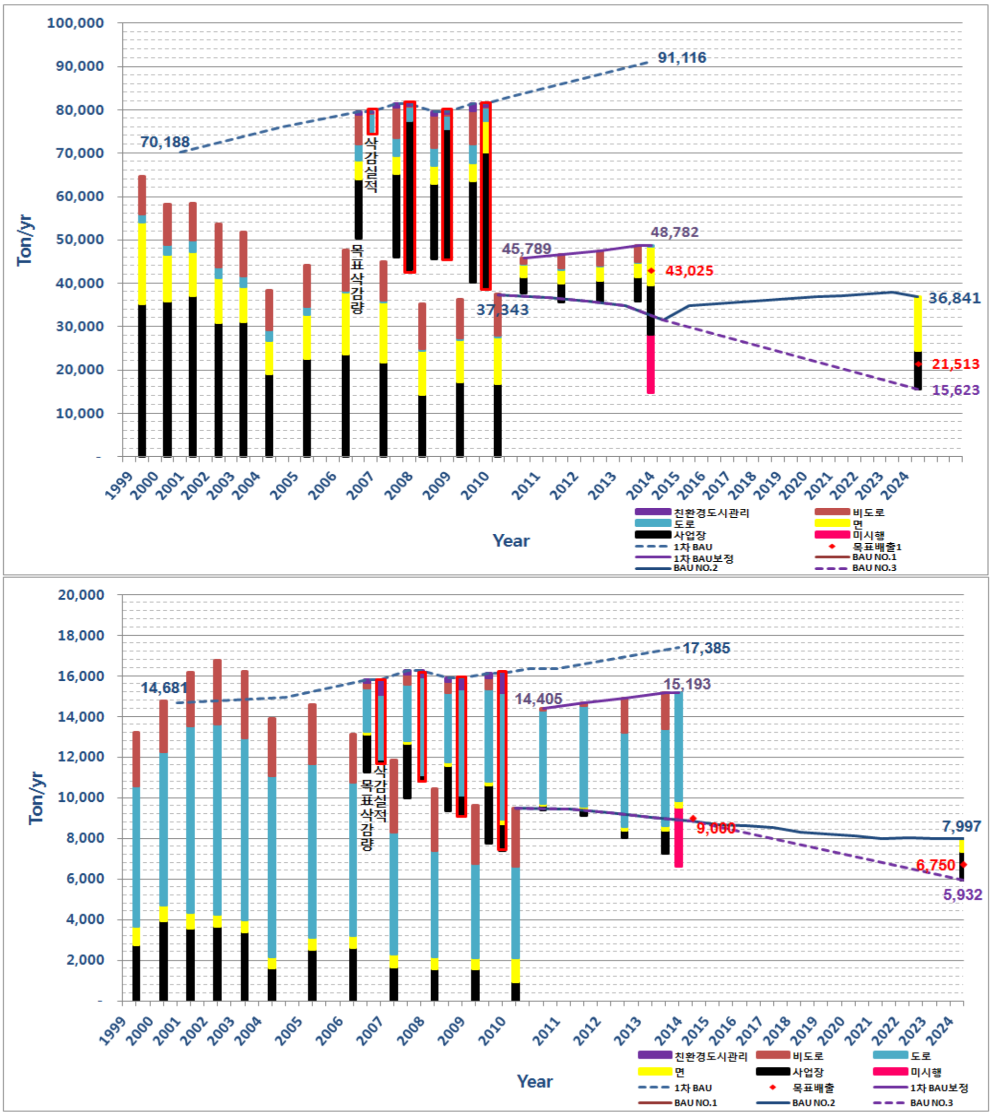

Figure 6. Historical trend and future reduction perspective for the emissions of Seoul Metropolitan Area. The emission reductions are estimated for 1st and 2nd SAQMP periods (Gong et al, 2014).

Air quality models normally underestimate the observed PM_{10} mass concentrations in Seoul and large cities in Korea; however, the reasons for the bias are still subject to in-depth investigation for a clearer understanding. Sulfate aerosol was the dominant chemical species for PM and is relatively well reproduced on a regional scale by a global chemical transport model as shown in Figure 7. However, the underestimate issue still exists on a local scale as shown in Figure 8 in which the simulated value is by a factor of 2 lower than the observation at the Bulkwang site (NIER) during the second half of 2013. Sulfate aerosol has gradually been superseded by organic matter (OM) aerosols because of the deployment of flue gas desulfurization (FGD) and the less use of sulfur containing coals in Korea. The model also significantly underestimates OM concentrations (Figures 8 and 9). As previously mentioned, a clear explanation on the reasons for the bias has not been provided yet and thus would be a critical factor for PM forecast. Observations of PM composition and their precursors would be necessary to diagnose a cause for PM forecast errors, to evaluate the model including its emission, and to improve the model performance. At this moment, the composition data consisting of PM and its precursors are very limited to the modeling community in Korea.

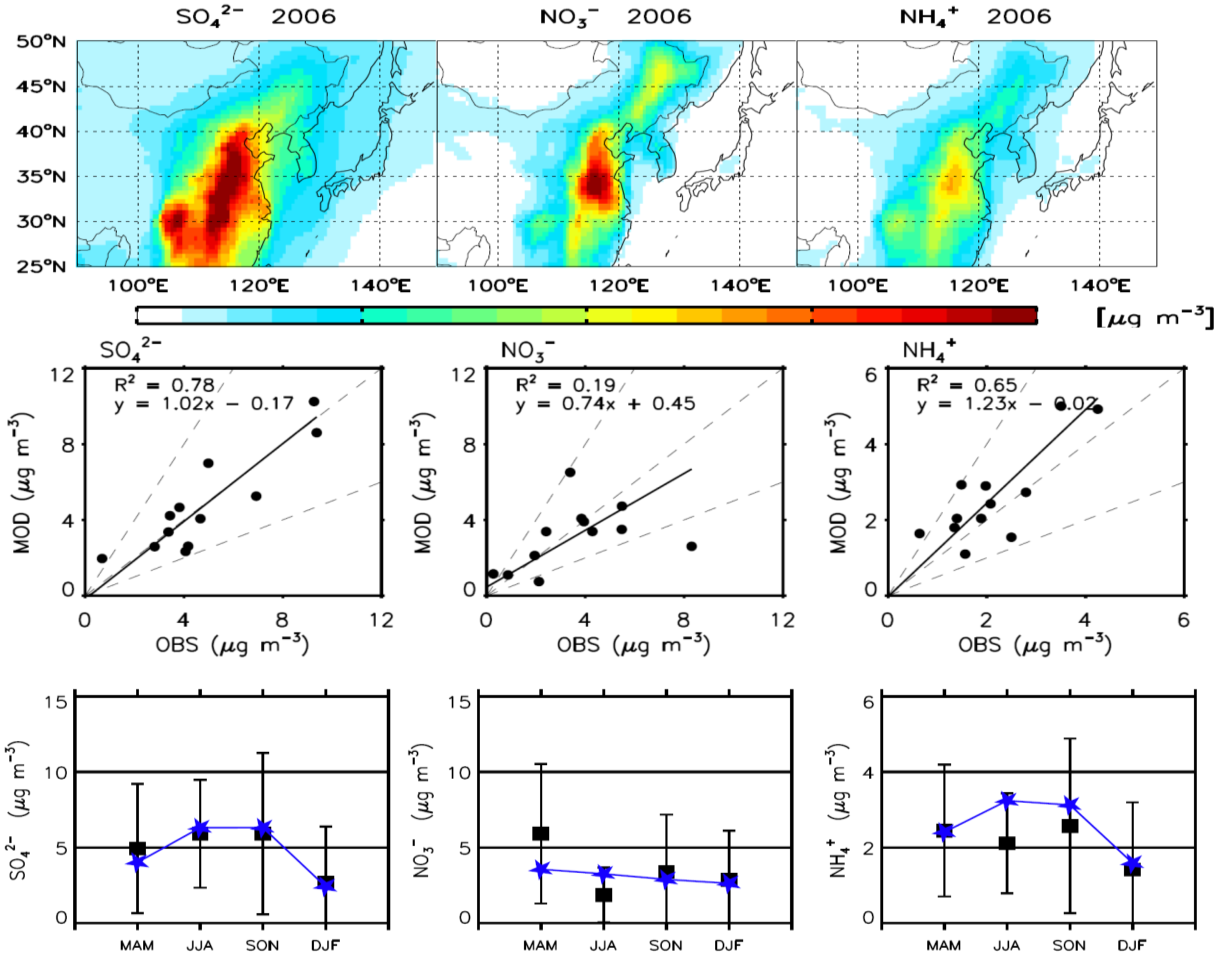

Figure 7. Simulated annual mean concentrations of SO_4^{2-} (left), NO_3^- (middle), and NH_4^+ (right) aerosols in surface air over northeastern Asia for March 2006 - February 2007 (top panel). Scatter plots of the observed and simulated monthly mean SO_4^{2-}, NO_3^-, and NH_4^+ concentrations at the site in Seoul are shown in the middle panel. Comparisons of the observed and simulated seasonal mean concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ aerosols at the site in Seoul are shown in the lower panel; adapted from Jeong et al.(2011).

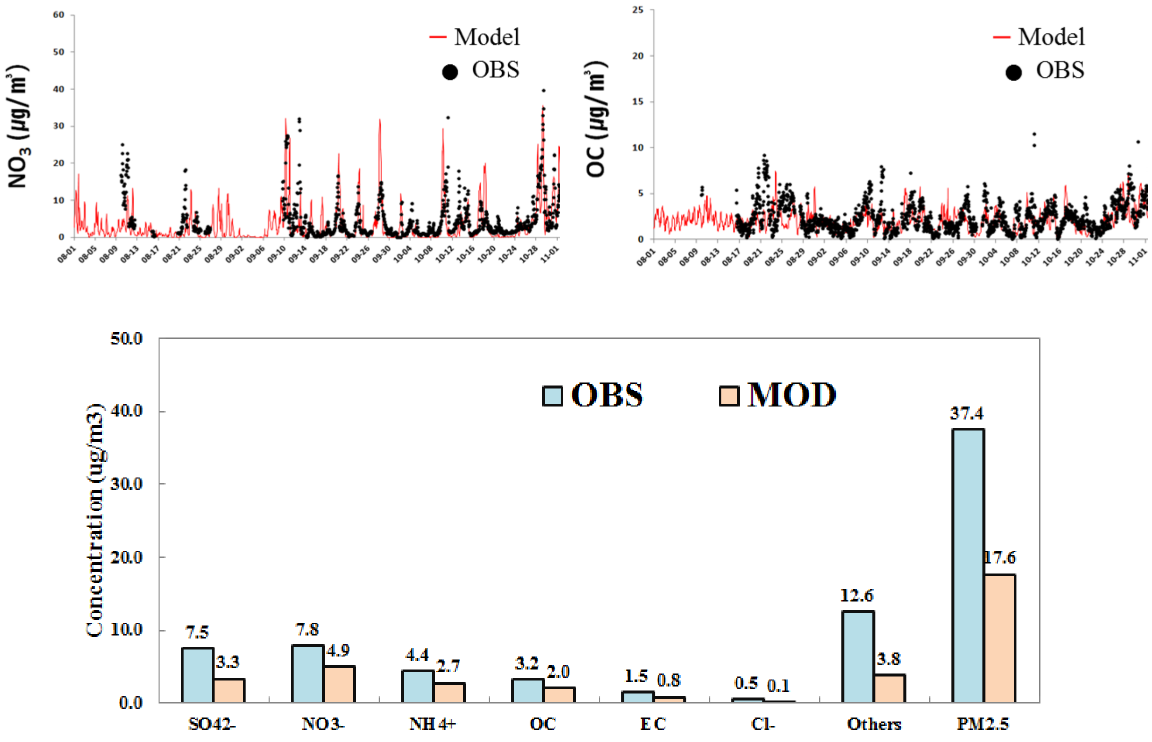

Figure 8. A comparison of the simulated and observed $PM_{2.5}$ composition data at the Bulkwang site in Seoul.

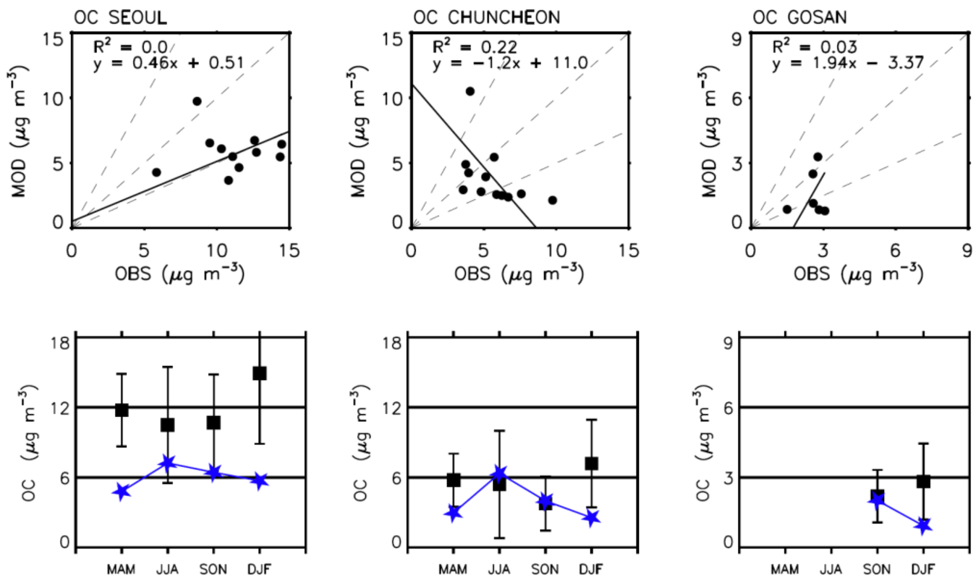

Figure 9. Scatter plots of the observed and simulated monthly mean OC concentrations at sites in Seoul (left), Chuncheon (middle), and Gosan (right) for March 2006 - February 2007. Comparisons of observed (squares) and simulated (stars) seasonal mean concentrations of OC aerosol (lower panel) in surface air at Seoul, Chuncheon, and Gosan sites. One standard deviation of the observed values is plotted with vertical error bars. The observation data at the Gosan site were only available for September 2006-February 2007; adapted from Jeong et al.(2011).

Mountains and other vegetative areas, where BVOC emissions considerably occur, surround Seoul and other large cities in Korea. O_3 concentration in Seoul is normally low because of the titration of high NO_X from the heavy traffic; however, it sometimes increases rapidly when high NO_X in polluted air is mixed with natural VOCs from the surroundings. Thus, O_3 simulations show high sensitivity to emission models of BVOCs (Kim et al., 2014), which is often parameterized using surface types and meteorological variables. Significant errors in surface types specification in Seoul and South Korea are often found in the U.S. database leading to large uncertainties in BVOC emissions (Park et al., 2014), which affect both O_3 and OM aerosol simulations. With this, extensive observations of VOC species would be very helpful to better estimate biogenic emissions and deepen the understanding of their roles in O_3 and PM air quality in Seoul and its surroundings.

Satellite observations of column concentrations of trace gases have been extremely successful in detecting changes in anthropogenic emissions and even useful as top-down constraints to update a bottom-up emission inventory with the inverse modeling technique (Han et al., 2009; Han et al., 2011). The retrieved vertical column concentrations, however, are very sensitive to air mass factor (AMF), which is used to convert slant column into vertical column. With this, the specification of AMF depends on the assumed vertical profile of retrieved species, which is obtained from CTMs. Thus, the validation of modeled vertical profiles of species is critical for the success of satellite observations and has been very lacking in East Asia. The profile validation is particularly important for the Korean GEMS whose missions are to measure the diurnal variation of short-lived chemicals. Low-orbiting satellites conduct measurements for the same local overpass time over particular regions and do not need a priori information with high temporal variation. However, the GEMS requires at least hourly a priori information of species profiles and the validation of those a priori information should be a high priority during the campaign.

For top-down emission estimates, the analytical inverse modeling has been widely used; however, it is limited in giving full constraint to the emissions of ozone precursors over East Asia because of the great length of the lifetime of aerosols and the precursors that allows enough time for the tracers to be transported beyond the range of the observable time span (e.g., hours). Adjoint (4-D var) inverse modeling can utilize each model grid as a state vector and consider the chemical and physical feedback of pollutants at the receptor site (e.g., transport; Figure 10: the case of O_3). Using the adjoint sensitivity of precursor emissions (e.g., SO_X, NO_X, and NH_3 emissions) with respect to the aerosol concentration at the receptor site, the ambient or trans-boundary impact of aerosol precursors on the aerosol concentration over Korea can be identified. The chemical data assimilation can be applied to species including NO_X, SO_2, NH_3, and BC or OC (to be confirmed). A procedure of the chemical data assimilation using the 4-D variational method is as follows: 1) build the adjoint modeling framework of GEOS-Chem to use satellite observations (e.g., OMI, SCIAMACHY, TES), 2) constrain emissions of aerosol precursors (NO_2, SO_2, NH_3) from East Asia through satellite observations, 3) constrain emissions of aerosol precursors from East Asia with the combination of satellite observations and aircraft measurements (from MAPS). This mission will deepen the understanding on significant aerosol precursors over East Asia along with the characteristics and main contributors to the trans-boundary pollution event.

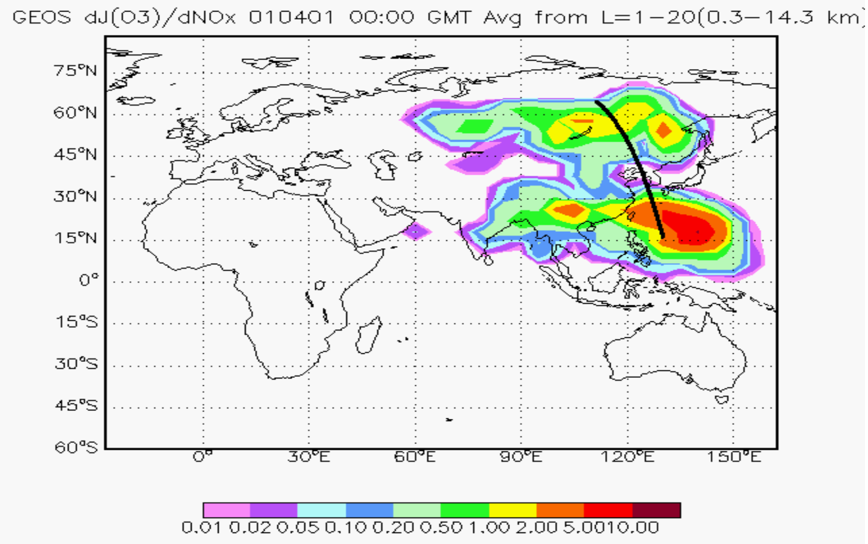

Figure 10. The adjoint sensitivity ambient NO_X emissions with respect to TES O_3 along the AURA satellite track (black line) in April 11, 2007 (Sandu, 2007).

Vertical profiles of recent in-situ measurement of O_3, NO_X, CO and meteorological variables show a distinct effect of advection, and the transport process of air pollutants especially over the western marine area of Korea and multiple elevated mixed or boundary layers are often found. These are closely related to the transport process of air pollutants. Case-by-case–based analysis showed that measurements have a vertical distribution of the air pollutant properties. For example, a general dust case in Asia for May 29, 2008, showed only single peak of PM_{10} at around 1,200 m, based on the diagnosis of the measurement of vertical profiles of meteorological parameters. However, as illustrated in Figure 11, the Asian dust case observed on May 22, 2008, showed two peaks of vertical profiles: the first peak was shown about 800 m and the second appeared about 2,500 m (Figure 11). This suggests the complexity of the transport process associated with boundary layer dynamics over Northeast Asia. A modeling study requires detailed information of boundary layer structure in order to interpret the transport process of measured species during the campaign.

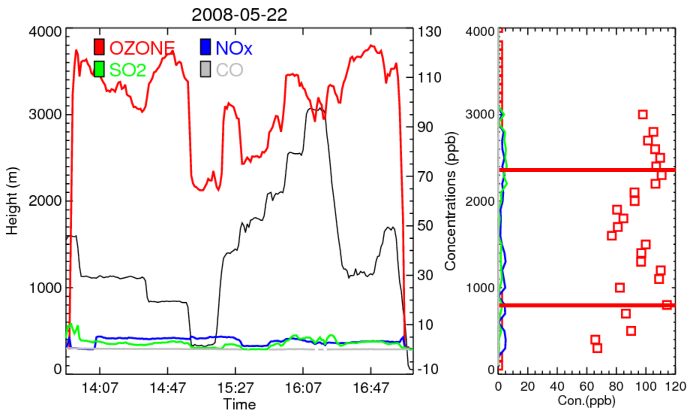

Figure 11. Recent measurement of vertical distribution of air pollutants and meteorological variables. Red square, blue, green, and gray indicate O_3, $\mathbf{NO_X}$, SO_2, and CO, respectively (left panel), and blue and black indicate vapor mixing ratio and potential temperature (right panel), respectively, observed at May 22, 2008.

2) Atmospheric Chemistry Studies

● Ozone and Its precursor Chemistry

It is well known that background ozone levels have steadily increased, especially over Northeast Asia with its levels at the highest rate for the last 20 years. However, there are recent indications that the increase in baseline O_3 concentrations has slowed and the accelerations are also generally, statistically, and significantly negative in the Japanese background station of Mt. Happo (Figure 12)–these findings, however, are very prone to change with the data produced in recent years and so should be considered with caution (Parrish et al., 2012). Xu et al. (2008) also reported a decrease in the average ozone concentration at a regional background station in eastern China from 1991 to 2006. However, they also pointed out that all other long-term trends, such as increase in the daily amplitude of the relative diurnal variations, increase in the monthly highest 5% of the ozone concentration, decrease in the monthly lowest 5% of the ozone concentration, and increase in the frequencies at the high and low ends of the ozone distribution, indicated that the variability of surface ozone has been enhanced because of increased NO_X concentrations.

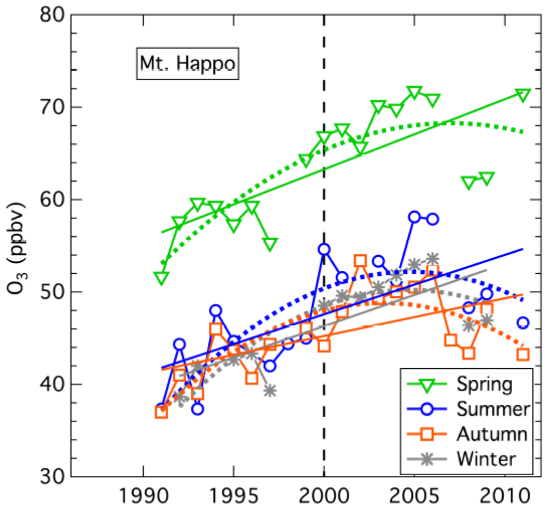

Figure 12. Seasonal O_3 averages measured at high elevation (1.9 km) sites in Japan at Mt.Happo. Lines indicate linear and quadratic regressions for the complete data sets (Parrish et al., 2012).

In contrast to the recent slowdown in increase rates of ozone in background atmosphere, the long-term trends of tropospheric O_3 in the seven major cities in Korea in Figure 13 showed consistent increases in tropospheric O_3 levels of 118 ± 69% in the seven cities over a 22-year period from 1989 to 2010 (Susaya et al., 2013). This study has confirmed that increases in VOC/ NO_X emission ratio would affect increases in O_3 concentrations, but no significant evidence was found that ozone concentrations were affected by changes in emissions of HCl or Cl. They concluded that emission patterns of precursor compounds and regional/long-range transport combined with meteorological parameters are likely to play a significant role in increasing O_3 levels over this region. Although local emissions were still major contributors for O_3 in South Korea, Chinese emissions were the dominant contributor in a highly episodic case with NO_X and VOC emissions from China, which contributed approximately 82% and 91%, respectively, to a maximum 8-hr ozone over this region (Choi et al., 2014).

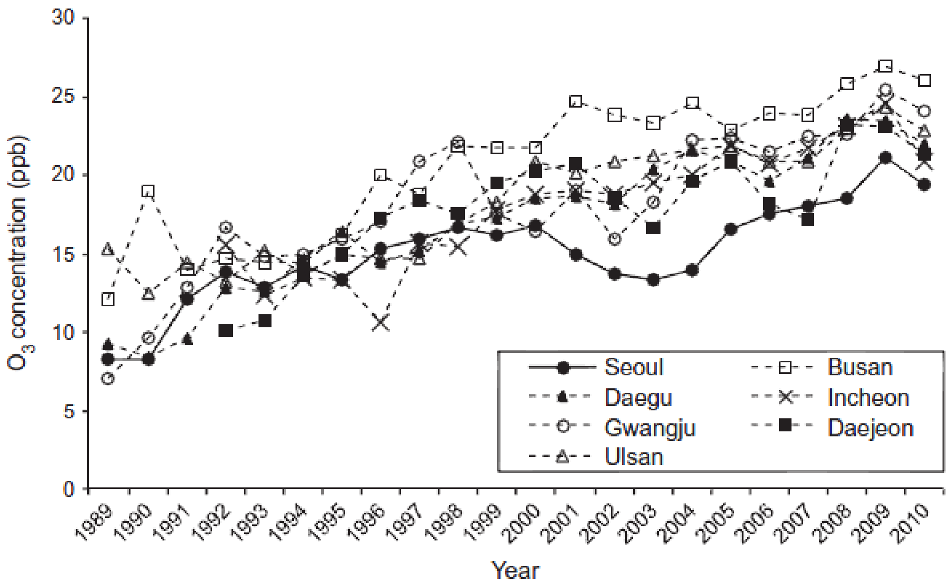

Figure13. Plot of annual mean concentrations of tropospheric O_3 measured from seven cities in Korea for a 22-year period (1989–2010) (Susaya et al., 2013).

Peroxy acetyl nitrate (PAN) is regarded as a more effective indicator of photochemical air pollution because it originates solely from tropospheric photochemical reactions unlike O_3, which has stratospheric origin in part. In recent measurements, PAN concentrations in the marine boundary layer between China and Korea were about two times higher than those of previous studies during the late 1990s over a similar area (Lee et al., 2012a). The observed increase rate of PAN is much steeper than that of O_3 in the regional background tropospheric airs. Its fast increase is also coherent to the fact that the average PAN/O_3 ratios in the SMA and large cities in China was much higher than those observed in cities of Europe and North America where control strategies have been enforced effectively to reduce hydrocarbon emissions (Lee et al., 2008; Zhang et al., 2011). These studies showed that levels and compositions of VOCs in the SMA and Chinese megacities are particularly favorable to fast production of photochemical species. Despite the importance of VOCs onto ozone and $PM_{2.5}$ chemistry, studies on the subject with regard to the SMA have been very sparse except for a selected few (Na et al., 2003; Na et al.,2004; Shin et al.,2013). In addition to the anthropogenic VOCs, it was found that BVOC photochemistry could play a significant role in ozone formation in the suburban region of the SMA (Kim et al., 2013). This study showed the importance of natural VOC emissions in regional photochemistry near megacities.

● Aerosol and Its Precursor Chemistry

Aerosol concentration mostly depends on its local sources in the SMA, such as mobile and industrial emissions, similar to other typical megacities. However, evidences are accumulated that prove that transported origins from the outside can comprise the significant proportion of aerosols in the SMA. A positive matrix factorization (PMF) study predicted that the major contributors of $PM_{2.5}$ in the SMA were secondary nitrate (20.9%), secondary sulfate (20.5%), gasoline-fueled vehicles (17.2%), and biomass burning (12.1%) followed by lesser contributions from diesel emissions (8.1%), soil (7.4%), industry (6.7%), road salt and two-stroke engine (5.1%), and aged sea salt (2.2%; Heo et al., 2009). The same study identified that both secondary sulfate and secondary nitrate factors were regional sources from the eastern coastal industrial area of China, especially for secondary sulfate. It also suggested that the high contribution of secondary nitrate observed in Seoul was likely influenced by ammonium emissions from large agricultural regions in China.

Jeong et al. (2013) applied a new index to estimate the change in mean mixing ratio of a target species ata receptor site because of the contribution of the long-range transport in the SMA. The inter-annual variations in the change caused by the long-range–transported SO_2 to its mean mixing ratio in Seoul was estimated to range from 0.40 to 1.03 ppb, which account for 8–21% of the ambient mean SO_2 mixing ratio in Seoul. A study at a costal SMA (Incheon) successfully identified major local sources of $PM_{2.5}$, including motor vehicles, sea salt, combustion processes, and soil. The statistical analysis with potential source contribution function (PSCF) also indicated that likely regional pollution sources included the southwest coast of industrialized China for secondary aerosol, northern Asia for forest fire combustion particles, and Mongolia for yellow sand particles from both desertification and industrial emissions (Choi et al., 2013). Jeong et al. (2011) demonstrated that the domestic source contributions were the most significant, with values of around 74% for OC (9% from fossil fuels and 65%from biofuels) and 78% for BC (42% from fossil fuels and 36% from biofuels) aerosol concentrations on an annual mean basis while the trans-boundary transport of Chinese sources was also found to be an important factor in Korea, with contributions of 13% and 20% to the OC and BC concentrations, respectively. The OC emission from under fired charbroiling meat cooking was assessed to contribute to about 2.5% of the ambient PM_{10} levels in Seoul, although it was projected that the contribution of $PM_{2.5}$ from meat cooking restaurants should be much greater to atmospheric PM composition than that of PM_{10} (Lee et al., 2011).

Cayetano et al. (2011) observed that the chemical characteristics of aerosol particles evolved as they undergo long-range transport over the Yellow Sea from continent source region. Although the chemical components of aerosol were strongly related to distinct source regions, enrichment of acidic secondary aerosols on mineral dust particles and even of sea-salt components during transport was observed. In relatively clean air mass with limited preexisting aerosol loadings but with sufficient precursors available, new particle formation events were often witnessed. Strong new particle formation events were frequently observed over the Yellow Sea. With this, Kim et al. (2013a) found that strong new particle formation events were observed on 7.5% of all days (60 days of a total of 800 days) at Gosan station in Jeju Island and 14.6% of all days (140 days of a total of 958 days) in Anmyun Island. The maximum occurrence season of strong new particle formation events was spring at Gosan, but winter in Anmyun Island with a fast-moving cold and dry air mass from the Asian continent after the passage of a frontal system.

3) Satellite studies

There have been significant efforts to improve the accuracy of aerosol products for air quality studies (e.g., Levy et al., 2013) and to meet the air quality community's need for high spatial resolution products (e.g., C. Li et al., 2005). Most aerosol properties retrieved from satellite remote sensing have been from low earth orbit (LEO) satellite, including the Sea-Viewing Wide Field-of-View Sensor (SeaWiFS), MODIS, and the Visible Infrared Imager Radiometer Suite (VIIRS; Figure 14), and increasing products have been from geostationary orbit(GEO) including the conventional meteorological imager(MI), including GOES and the Communication Ocean and Meteorological Satellite(COMS; Figure 15). Recently, as the first ocean color imager in GEO, GOCI started to provide valuable information on aerosol properties in high temporal and spatial resolution in East Asia since 2010.

In order to understand the mesoscale distribution and processes of aerosol properties, the Distributed Regional Aerosol Gridded Observation Network(DRAGON)–NE Asia Campaign was held in Korea (http://aeronet.gsfc.nasa.gov/new_web/dragon.html) in collaboration with NASA of U.S. and NIER of Korea. With the dataset obtained during the campaign, aerosol properties retrieved from GOCI and MI were compared, and a more accurate aerosol model was established in this region to improve the accuracy of satellite remote sensing products. With the PM samplers operated together with AERONET sun photometers during the campaign, surface PM was estimated from satellite and ground-based observation of aerosol optical depth (AOD), which showed a reasonably good correlation. This would extend the spatial coverage of PM estimation to the satellite's field of view.

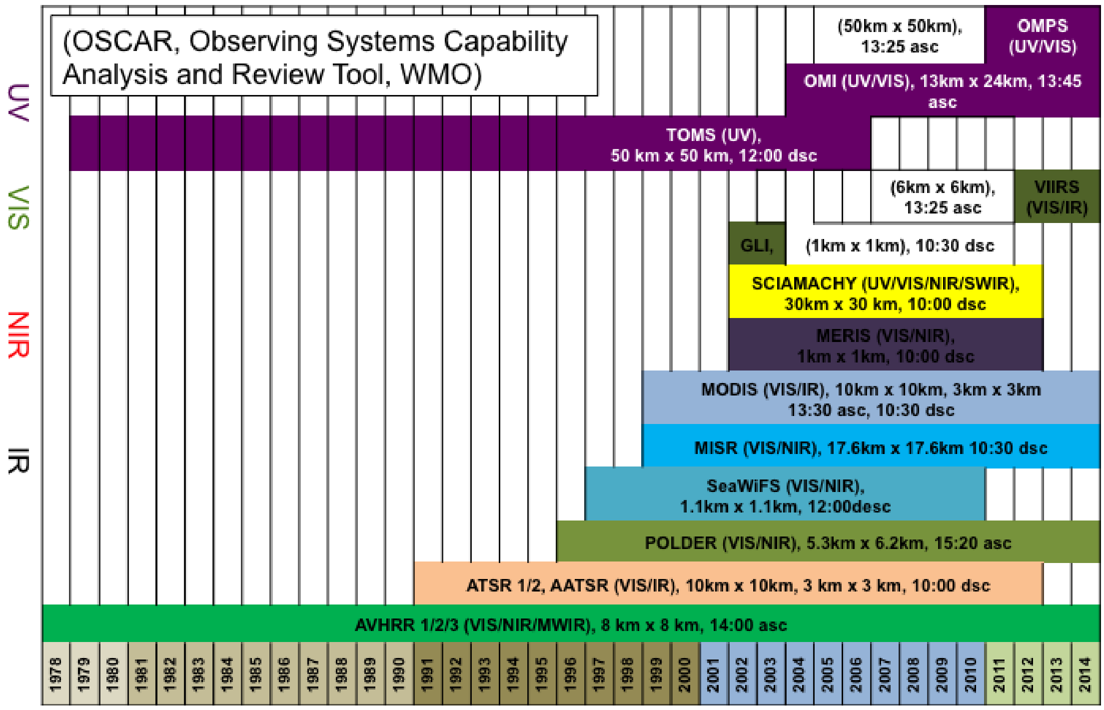

Figure 14. Aerosol retrieval using satellite (LEO) measurement: past and present (sensor list, AOD spatial resolution)

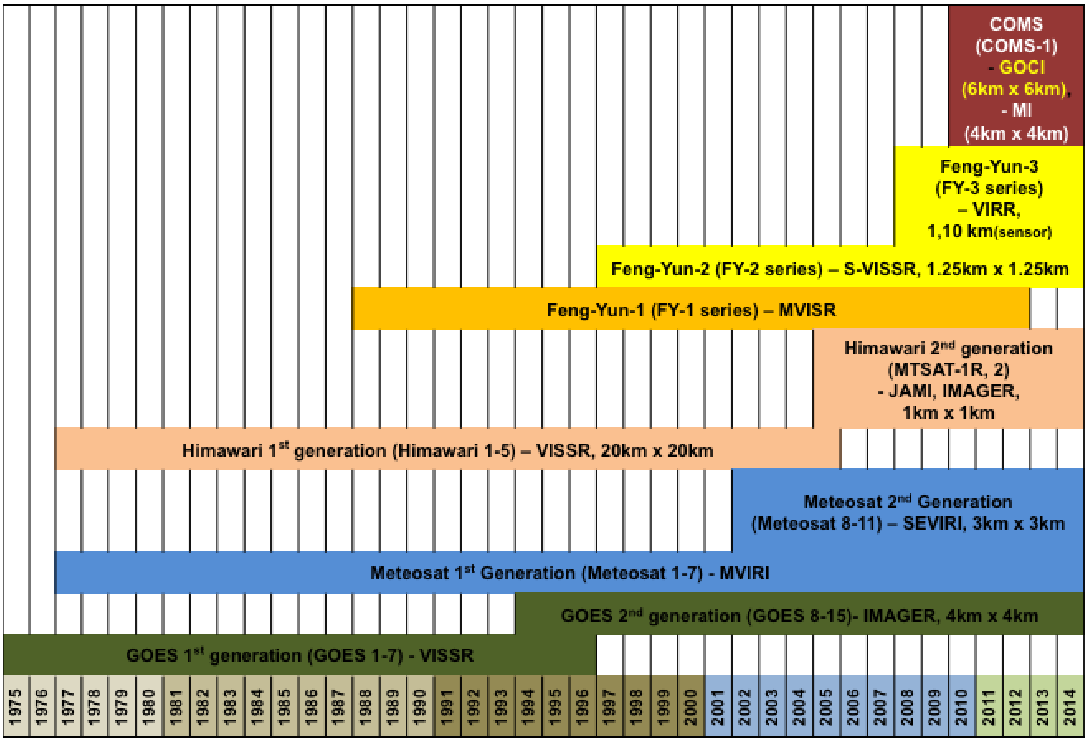

Figure 15. Aerosol retrieval using satellite (GEO) measurement: past and present (project, satellite, and sensors)

The accuracy of aerosol products from MODIS has been better over the ocean because of less noise from the surface, while that over land has been poor. However, the recent progress in MODIS Collection 6 showed improved performance over land as well as seen in Figure 16. The aerosol products from MODIS onboard Terra and Aqua have been available for the past decade (http://modis.gsfc.nasa.gov/data/).

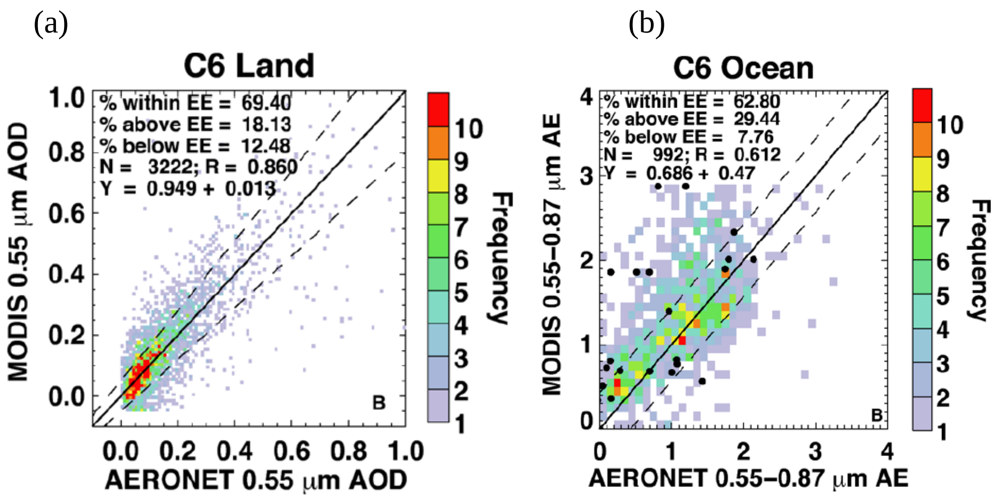

Figure 16. Comparison of MODIS Collection 6 against AERONET for (a) AOD and (b) Angstrom exponent (Levy et al., 2013).

Remer et al. (2012) investigated the data availability of aerosol products for different spatial resolutions using MODIS. Retrieval of aerosol properties in finer resolution provides much more frequent data than that in coarser resolution; that is, retrievals with a nominal 4-km footprint will lose 60–70% of those that would have made with a nominal

0.5-km pixel instrument. However, further investigations are required to assess the accuracy changes with respect to the resolution. Lee et al. (2012b) took the non-sphericity and dynamic behavior of aerosol into consideration to improve the accuracy of aerosol products from MODIS. VIIRS, which was launched in 2013, continues the mission of MODIS with improved instrumentation (Cao et al., 2013).

For the aerosol remote sensing from geostationary orbit, there have been extensive efforts to retrieve information on aerosol with its advantage of high temporal resolution coupled with its disadvantage of having a limited number of visible channels. To overcome such limitation, Kim et al. (2014) improved the accuracy of aerosol products in urban areas by considering background aerosol correction and critical reflectance. However, with the launch of the first ocean color imager, Geostationary Ocean Color Imager(GOCI)-1 onboard the GEO-KOMPSAT 1(also known as “Chollian”), it is now possible to retrieve aerosol optical properties using multiple channels in visible and near infrared (NIR; Lee et al., 2012b).

Because of its strength of global coverage, there have been many attempts to evaluate PM from the satellite observation of AOD either by empirical techniques (e.g., Zhang et al., 2009; Kloog et al., 2011) or by means of a chemical transport model (CTM; Choi et al., 2009; van Donkelaar et al., 2011; van Donkelaar et al., 2012). Song et al.(2009) analyzed the spatial and seasonal variations of surface PM_{10} and MODIS AOD over China, where they found strong spatial correlation between the two but variable temporal correlation depending on region. Seo et al. (2014) estimated PM concentration from the AOD measurements of AERONET and MODIS during the DRAGON–NE Asia campaign, identified important predictors such as boundary layer height and effective radius, and found variable correlation depending on the type of location.

The capability to measure tropospheric concentration of trace gases from space has evolved since the regular monitoring of the stratospheric ozone layer with the Solar Backscattered Ultraviolet (SBUV) and the Total Ozone Monitoring Spectrometer (TOMS) on Nimbus-7 in late 1978. These measurements have continued since 2004 to date with the Ozone Monitoring Instrument (OMI) on EOS Aura. With the recent development of spectrometers in UV-visible with sub-nm spectral resolution and development of retrieval algorithms, the generation of estimates of the column amounts of atmospheric O_3, NO_2, SO_2, HCHO, CHOCHO and other constituents in the troposphere and stratosphere is now possible. The evaluated accuracy in trace gas concentration from OMI can be summarized as in Table 1.

To date, all the UV-visible satellite missions to monitor trace gas concentrations in the atmosphere have been in low earth orbits (LEOs), usually allowing one observation per day. With the advent of new UV-visible instruments on geostationary (GEO) platforms, the diurnal variation of these components will be captured. The GEMS is to be launched into orbit at the end of 2018, positioned over Asia. The instrument is a step-and-stare scanning UV-visible imaging spectrometer, with a scanning Schmidt telescope and an Offner spectrometer. A UV-enhanced 2-D CCD takes images, with one axis spectral and the other north–south spatial, with east–west scanning over time. By 2020, the geostationary orbits are expected to be filled with three UV visible spectrometers—NASA Tropospheric Emissions: Monitoring of Pollution (TEMPO) over North America, the ESA Sentinel-4 Ultraviolet Visible Near infrared (UVN) spectrometer over Europe, the KARI GEMS over Asia, with the TROPOspheric Monitoring Instrument(TROPOMI) flying underneath in LEO.

Table 1. Accuracy of OMI measurements

	Correlation coefficient(R)	a, Slope	b, Intercept	RMSE	Error (%)
O_3 (Total)	0.82 – 0.97	0.83–0.97	35.5 DU	*7%*	
O_3 (Trop)	0.5 – 0.8	0.5 – 0.9	0–15 DU	*5–10 DU (10–20%)*	3–6 DU *(10–20%) [depending on SZA]*
HCHO	0.81 (0.57 – 0.77)	0.80 (0.75–0.88)	0.01×10^{16} (-2.3–$1.8. \times 10^{15}$)		
NO_2	0.8	0.5	3.0 [$10^{15}cm^{-2}$]	*N/A*	20%
SO_2	0.7	1.0	0.5 DU	*N/A*	50 –100%
Cloud Fraction	0.90	0.9 – 1.1	*N/A*	*N/A*	2 – 5%
Cloud Pressure	0.80	0.9 – 1.1	*N/A*	*N/A*	5 – 20%
Surface Reflectance	0.70 – 0.91	*N/A*	*N/A*	*0.03*	5 – 40%
AOD	0.7	*N/A*	*N/A*	*T/V*	30% at AOD > 0.1

Recognized by the Committee on Earth Observing Satellite (CEOS) Atmospheric Composition Constellation (ACC), the geostationary constellation of UV-visible spectrometers will enlighten everyone on the global distribution of ozone, aerosol, and their precursors. To integrate the dataset for global measurements, harmonized data quality is very important. Thus, the inter-calibration among the three different UV-visible satellite instruments is very important, in addition to the quality of the data processing. Therefore, the standardization of data products and pre-calibration/post-calibration/validation methods are being discussed.

6. Research Strategies

1) Air Quality Models and Emission Strategies

Korean modeling experts will be involved in all stages of preflight planning, flight execution, and post-flight data interpretation. They will use fine-resolution regional CTMs (CMAQ and WRF-Chem) and a global CTM (GEOS-Chem and its adjoint). A global model will provide boundary conditions for regional CTMs to account for intercontinental transport of pollutions.

i) At the Pre-campaign stage

Air quality models will be used in discussion of what scientific issues can be explored. After the collection of the ideas, pre-runs of CTM models will be made for feasibility tests of the collected ideas until the campaign actually begins.

- Air quality forecasting(AQF)

Several numerical experiments will be conducted in order to investigate (i) major source regions in detail for each $PM_{2.5}$ component and reasons causing the model bias, (ii) "best-performing" options for high PM_{10}/$PM_{2.5}$ episodes, (iii) configuration for frequent trans-boundary high PM_{10}/$PM_{2.5}$ plume pathways, and (iv) comparisons to outputs from the NIER AQF system.

➢ Ensemble forecast (2015-2016)

a. Continuous/routine daily Air Quality Forecast: 2015-2017

b. Model: WRF/CAMx, WRF/CMAQ, WRF-CHEM, GEOS-CHEM

c. Emission: NIER/KU-CREATE emissions inventory by incorporating up-to-date control measures and growth factors

d. Routine evaluation: surface + satellite

e. Development and testing of "campaign-oriented" analysis tool (e.g. pseudo- flight tracking)

ii) During the Campaign

AQF will support plan of a flight track ahead in order to better capture the trans-boundary air pollutants by allowing occasional targeted measurement of pollution plumes from the upwind region. This targeted measurement helps in understanding on the chemical evolution of anthropogenic plumes during transit from the upwind source to Korea.

Multi-model ensembles will be used to reduce modeling errors associated with meteorology, chemical schemes, and physical processes. The participating models including global CTMs (GEOS-Chem, GRIMs-Chem) and regional CTMs (CMAQ, WRF-Chem) can be categorized as online versus off-line models based on the simulation ability for the two-way interaction between the chemical and the meteorological processes. The off-line models are normally driven by the assimilated meteorology and thus are less subject to errors in meteorological fields. On the other hand, the effect of the chemistry–meteorology interaction would be investigated using the online models.

The trans-boundary effect of anthropogenic emissions from China is a convoluting factor to add the simulated uncertainty and is a particular interest for the Korean community. Even though China is a standing source of air pollutants, its effect on air quality in Korea is determined by synoptic-scale meteorological transport, of which the accurate simulation is somewhat challenging for a regional meteorological model owing to the highly unpredictable Asian monsoon system. Therefore, modeling system will be set up by combining preexisting CTMs with regional or global weather forecasting models that are best suited for the East Asian climate. This system is expected to be useful for preflight planning to detect pollution pathways as well as for post-mission data interpretation to understand the source–receptor relationship.

Finally, the same bottom-up emission inventory will be used for all participating models to understand the characteristics of different parameterizations and schemes. The intercomparison of models with observational constraints helps in the understanding of the determining factors and in identifying the optimized scheme for air quality models in Korea.

➢ On-campaign Support (2015 and 2016)

a. Ensemble forecast

b. Real-time evaluation: surface observation + satellite + aircraft

c. Emission : nowcasted modeling emission fields using SMOKE, model–based biogenic emissions.

d. Flight planning support

i. Backward/forward HYSPLIT trajectory
ii. Vertical structure analysis using pseudo- flight track
iii. On-demand super-fine nesting (Flexi-nesting)

iii) At the Post-campaign Stage

Post-campaign observation analysis will be conducted by models used in the campaign. A few detailed plans are discussed below and will be updated in the near future.

- CTMs have been modified to more comprehensively consider, for example, hydroxyl radical reactivation in isoprene chemistry (Butler et al., 2008; Han et al., 2013), complicated parameterizations for reaction probability of N_2O_5 during night (e.g., Riemer et al., 2003; Evans and Jacob, 2005; Davis et al., 2008), etc. CTMs will be validated three-dimensionally using the data that will be obtained from the MAPS—Seoul campaign. In particular, the vertical distributions of NO_2, SO_2, and HCHO are of primary importance because the model-calculated vertical profiles have been used in the satellite retrievals of such gas species. The possible unrealistic vertical profiles have resulted in systematic biases in the satellite-retrieved gas products in connection with air mass factors (AMFs) and averaging kernels (AKs; Eskes and Boersma, 2003; Lamsal et al., 2014). This issue will be intensively investigated in close collaboration with the algorithm development teams of Korean GEMS sensor. The aforementioned issue is also closely related to investigation on the accuracy of bottom-up pollutant emissions and CTM model parameterizations (Han et al., 2014). These investigations will also be carried out comprehensively at the post-campaign stage using the observation data available after the campaign.

● MAPS-Seoul will perform in-depth analyses of results from CTM runs during the campaign with high-resolution and high-quality campaign observational data as well as satellite data available. These analyses include: (i) cross-comparison of model performance with various CTM runtime options, (ii) model intercomparisons among CAMx, CMAQ, WRF-CHEM, GEOS-CHEM and other photochemical models, iii) source contribution assessment, (iv) process contribution (including chemistry) assessment. This study will then compile the results of all analyses to make recommendations for emission inventory improvement.

iv) A Period for the MAPS Campaign

The chemical environment in Korea and a broad part of East Asia is strongly tied to natural climate variability. In particular, the East Asian summer monsoon normally accompanies a large amount of precipitation that efficiently washes out air pollutants. Therefore, the planned aircraft campaign should avoid the normal rainy monsoon season (late June – mid-August). One of the campaign's objectives is to validate air quality models, which will be advanced based on the new understanding from the planned observations. In this context, a season for the worst air quality would be the best target period for the mission, which would be either from December to January or from May to early June. The first period is a season with large primary pollutant emissions from household heating and other miscellaneous sources, which were not yet accurately accounted for the present inventory. The latter period experiences higher secondary pollutant concentrations than the first such that ozone and fine PM concentrations reach their peak values owing to the rapid photochemical production. In addition, the export of continental pollution plumes is very efficient for the latter period.

The seasonal air mass transport varies significantly over East Asia. In winter, the northwesterly wind from Northern Asia is dominant and the pollutant emissions over northeast Chinese urban/industrial area usually significantly contribute to air pollution over East China, Korea, and Japan (Figure 17). In spring, the westerly wind is generally dominant and the main Chinese emission strongly impact on the air quality of Korea, Japan, and even North America by trans-boundary transport of pollutants, which are more activated by warmer temperature and enhanced soil moisture (Figure 17). However, the air transport in summer is very different; Asian monsoon drives southwesterly wind from west-Pacific, which has relatively cleaner, humid, and warmer air mass. Although the southwesterly wind can drive the pollutants over southern China toward Korea and Japan, the characteristics of the impact are different from the patterns in other seasons (Figure 17).

Given the distinctly different transport pathways over East Asia, the campaign should be ideal if the observation is conducted when the different patterns of air transport is clear. Then it can be an opportunity to understand the impact of air mass from (1) northwest (northeast Chinese emissions), (2) west (central Chinese emissions), and (3) southwest (western Pacific and south Chinese emissions) on downwind areas (coastal China, Korea, and Japan).

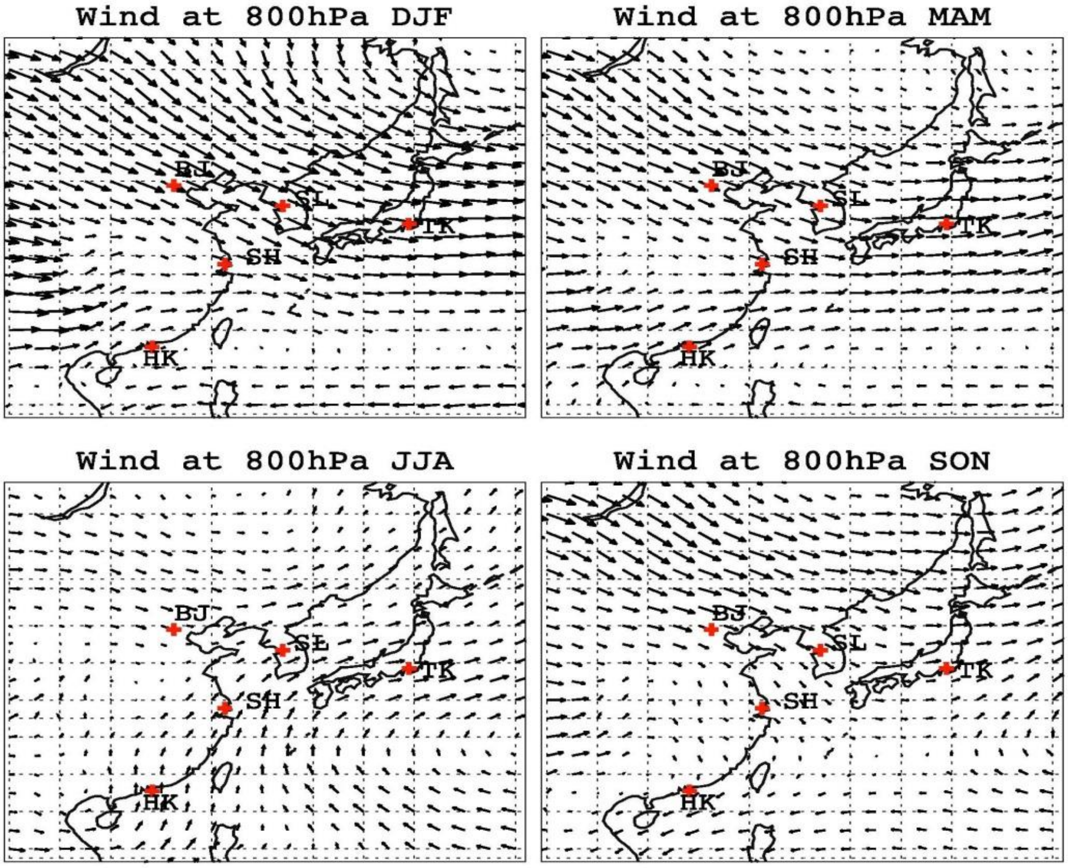

Figure 17. Distinctly different seasonal air mass transport over East Asia. The horizontal wind directions near 800 hPa shown here have been generated by GEOS-4 in 2007 (Shim et al., 2008).

2) Field observations

i) Proposed Campaign Periods

MAPS–Seoul program has been planned for five years and includes three intensive field campaigns and data analysis. The first field campaign will be held in 2015, and the research will focus as a preparatory field campaign to build up the measurement capacity and preliminary data production for the following two intensive campaigns in 2016. The second field campaign to be held in May to June 2016 will be the first collaborative research (named KORUS–AQ) with U.S. scientists mainly associated in NASA DC-8. The research focuses are the photochemical formation of ozone and $PM_{2.5}$ in the SMA and their interactions between chemistries, transport, and various sources.

Korean participants will deploy another field campaign in winter 2016 or 2017 to study the regional haze events in the cold season. The winter campaign will take place in the period when the continental outflows are prevailing and transported contributions in air quality are dominant over the SMA, utilizing full research resources and capacities developed in the preceding two field campaigns.

ii) Proposed Measurements and Platforms

MAPS–Seoul will execute a series of ground, ship, and aircraft experiments in order to examine the chemistry, transport and emission characteristics during the distinct pollution episode periods in the SMA. A variety of chemical and physical properties should be measured in detailed temporal and spatial resolutions. The priorities of proposed measurements with related scientific questions are listed in Table 2. A number of well-equipped ground sites in the region of the SMA with full set of data during the research periods will be accessible to this study. However, the measurement capabilities on radical species, such as, OH, NO_3, and Chlorine, should be further developed soon. The proposed platforms employed during the program are shown in Table 3 and Figure 18. It includes three NIER super sites (Bangyeong Island, Seoul, Jeju super site) and one forest site in outskirts of Seoul.

A total of two King Air units and NASA DC-8 will be deployed during the KORUS–AQ campaign period. All aircrafts are going to be coordinated to accomplish their own scientific goals and full research capacities. Although the detailed collaboration will be designed under KORUS–AQ science plan, the King Air units will cover the major trace gases and aerosol distributions in the SMA region (Table 3) while DC-8 will take part in the measurements of comprehensive chemical species in the primarily regional scale covering the SMA in part. Flight scenarios should be designed to collect sufficient information on diverse research targets related to chemistry in and out of the megacity plume, continental outflows, biogenic and biomass burning emission, plumes of large industrial sources, nighttime and early morning chemistry, and intercomparison purposes. Plausible flight scenarios not yet proven to be practical are depicted in Figure 19. The specific flight plans should be further developed under flight opportunities, such meteorological conditions, and flight restricted zones. During the KORUS–AQ period, onboard measurements in DC-8 by a number of Korean scientists would surely be taken into consideration of mutual agreement between NIER and NASA. The research vessel (R/V) provides unique opportunities to collect the information about the regional background conditions, the oceanic emissions, and continental outflows. Any participation of R/V during the campaign is highly uncertain because of its own schedule and unsecured budget allocation at this stage of preparation.

● **Canopy or building scale fluxes: ground measurements on a tower or building**

Ground-based observations on an above-canopy tower or building platform will provide i) a comprehensive characterization of the physical, chemical, and biological processes controlling atmosphere-surface exchange and the atmospheric processing of air pollutants, ii) insights into the interactions between air pollutants and natural emission within/above the canopy in terms of O_3 production and SOA formation, and iii) better estimate or improvement of current emission inventory from the SMA. In order to estimate the atmosphere-surface fluxes of trace gases, the eddy covariance (EC) method will be applied. This method is well-known as the most reliable and direct flux measurement method, and requires a fast response sensor (e.g. 5-20 Hz data) with high sensitivity (Baldocchi, 2003). For example, PTR-TOF-MS (Proton Transfer Reaction – Time Of Flight – Mass Spectrometer) system is suitable for this application. In particular, the PTR-TOF-MS is a unique instrument that can measure an unprecedented number of VOCs simultaneously (including OVOCs missed in previous studies) with very high time resolution, breaking through instrumental and technical limitations and allowing a new approach to measuring VOC fluxes between the atmosphere and biosphere while also observing their oxidation products fluxes (Park et al., 2013). In addition to VOC measurement, O_3 and NO_X (NO & NO_2) flux will be also measured for investigating their vertical movement. Along with flux measurements, vertical gradient measurements are important for characterizing source strengths and the importance of oxidation processes along vertical transects for species with differing reactivity.

- **Regional scale fluxes: airborne and shipborne measurement**

In the urban and sub-urban area including the SMA, quantifying inflow, outflow, emission, and deposition of air pollutants and their oxidation products is crucial in identifying the origin (e.g. foreign or domestic) of air pollution. To do so, airborne and shipborne EC flux measurement is novel approaches to quantify regional scale fluxes for chemical species (e.g. VOCs, NO_X, O_3, and etc.), and these observations have been successfully conducted so far (Karl et al, 2009; Bariteau et al, 2010). In MAPS-Seoul, using a research vessel and a small air plane (e.g. Hanseo King Air, KMA Kisang I) it is possible to measure 3-D fluxes of air pollutants and their fate over inland and surrounding oceans of Korea.

The combined flux observations at the canopy/building scale, and regional scale will allow comprehensive investigation of mechanistic processes controlling sources and sinks of air pollutants and their Lagrangian evolution in both vertical and horizontal directions as they are transported away from the source regions and react chemically in the atmosphere. Therefore, these flux observations will provide critical data for not only precisely establishing emission and deposition inventories that will aid air pollution control policy in Korea, but also will provide a more comprehensive understanding of atmosphere-surface interaction processes in the Earth system. Specifically, the improvement in our understanding on O_3 and SOA formation by emissions from anthropogenic/biogenic source and depositions into the surface/ecosystem can be achieved by integrating the results from the observations into regional CTM models (e.g. WRF-CHEM, CMAQ, and etc.).

Table2. Measurements needed to answer the scientific questions with a chemistry perspective.

Parameters to Be Measured	Q 1	Q 2	Q 3	Q 4	Q 5	Q6	Q7	Q8	Comments
O_3	√√√	√√√		√√	√√√	√√√	√√	√√	
$PM_{2.5}$ mass and chemical compositions	√√√		√√√	√√	√√√	√√√	√√√	√√√	
Aerosol number and size distribution	√√√		√√√	√√	√√√	√√√	√√√	√√√	
EC/OC	√√√		√√√	√√	√√√	√	√√√	√√√	
VOCs speciation	√√√	√√√	√√√	√√√	√√√	√√	√√		
NO, NO_2, NO_X	√√√	√√√	√√√	√√	√√√	√√	√√	√	
CO, SO_2	√√√		√√√			√√	√√	√	
HCHO and aldehydes	√√	√√√		√√	√√√		√√		
Alkyl nitrate, PANs, HNO_3	√√	√√√	√√√	√√	√√√	√	√√		
BVOCs and OH reactivity	√√√	√√√	√√√	√√√	√√√		√√√		
H_2O_2, CH_3OOH	√√	√√√	√√√	√√	√√√	√√	√√		
Aerosol scattering and absorption	√√√		√√√				√√	√√√	
Black carbon	√√√						√	√√√	
Aerosol profile						√√√	√	√√√	
High resolution aerosol speciation	√√		√√√	√√√	√√√	√√	√√√	√√	
Aerosol trace metals	√			√√√			√√	√	
HONO, NH_3			√√√	√√		√	√√√		
Oxidized hydrocarbons		√√√	√√√	√√	√√√		√		
OA speciation			√√√	√√√			√√√		
OH radical		√√√	√√√		√√√				
Carbon and Nitrogen isotopes	√√	√√	√√	√√√			√√		
NO_3 radical, N_2O_5		√√	√√√	√	√√√		√√		Highly uncertain
Cl and $ClNO_2$		√√	√√√	√	√√√		√√√		Highly uncertain
Meteorology including sondes						√√√			

Q1–7: scientific questions listed in section 4. 1), √√√=essential, √√=important, √=useful, none=not relevant

Table 3. Proposed instruments and platforms

Parameters to Be Measured	G1	G2	G3	G4	A1	A2	RV1	Instruments
O_3	√	√	√	√	√		√	UV absorption
$PM_{2.5}$ mass and chemical compositions	√	√	√	√	√		√	Filter pack
Aerosol number and size distribution	√	√	√	√		√	√	CPC and SMPS
EC/OC	√	√	√	√				Sunset
VOCs speciation	√	√	√		√			Canister,GC/MS
NO, NO_2, NO_X	√	√	√		√		√	Chemiluminescence
CO, SO_2	√	√	√		√		√	NDIR, Fluorescence
HCHO and aldehydes	√	√						DNPH/HPLC
Alkyl nitrate, PANs, HNO_3	√							CIMS, DC-8
BVOCs and OH reactivity	√						√	PTR-MS
H_2O_2, CH_3OOH	√							HPLC (Derivatization)
Aerosol scattering and absorption	√	√	√			√	√	SMPS
Black carbon	√	√	√					SP2, DC-8
Aerosol profile	√	√	√					LIDAR
High resolution aerosol speciation	√	√	√					AMS, DC8
Aerosol trace metals	√	√	√					Realtime XRF
HONO, NH_3	√							AIM or DOAS
Oxidized hydrocarbons		√						Tbb, CEAS, DC-8
OA speciation	√	√						High volume filter /GC-MS
OH radical	√							CIMS
Carbon and Nitrogen isotopes	√	√	√	√				Accelerated MS
NO_3 radical, N_2O_5		√						Tbb, CRDS, DC-8
Cl and $ClNO_2$	√							Tbb, CIMS, DC-8

G1: Upwind ground site of SMA
G2: Downtown site in SMA
G3: Downwind forest site from SMA
G4: Regional background site in Jeju Island
A1: Hanseo Univ. King Air
A2: KMA King Air, to be built by 2016
RV1: R/V KMA Kisang I
DC8: possibly loaded in NASA DC-8; Tbb : need to be built before the experiment

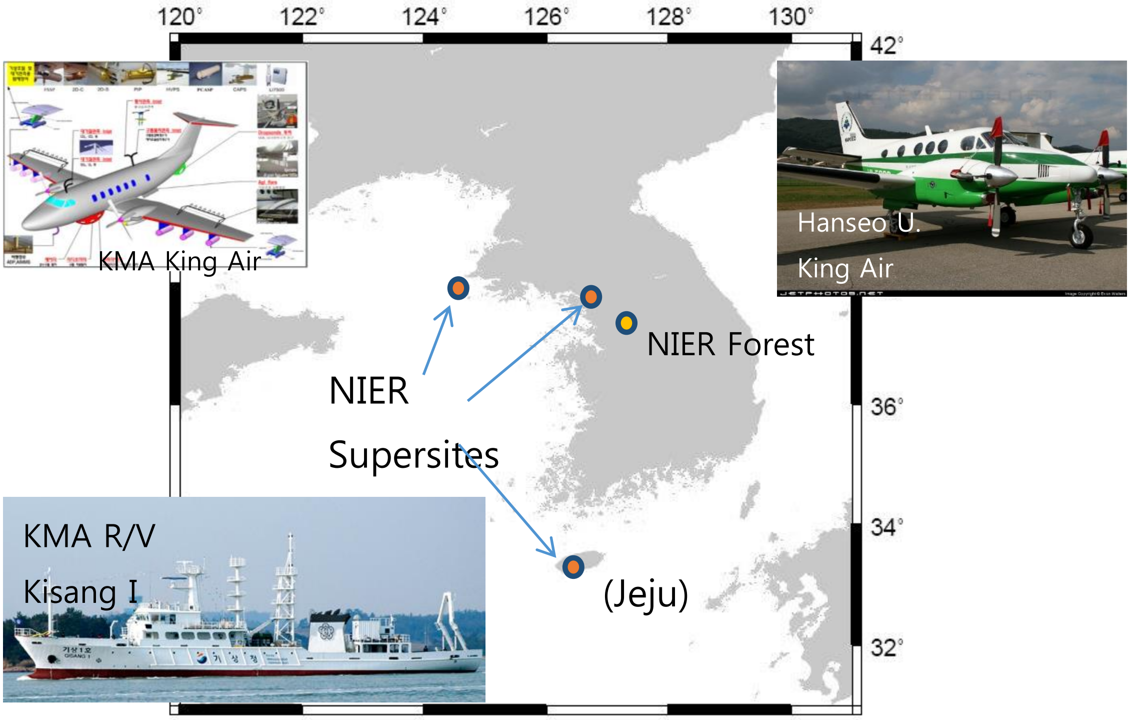

Figure 18. The proposed platforms in the MAPS–Seoul field campaign.

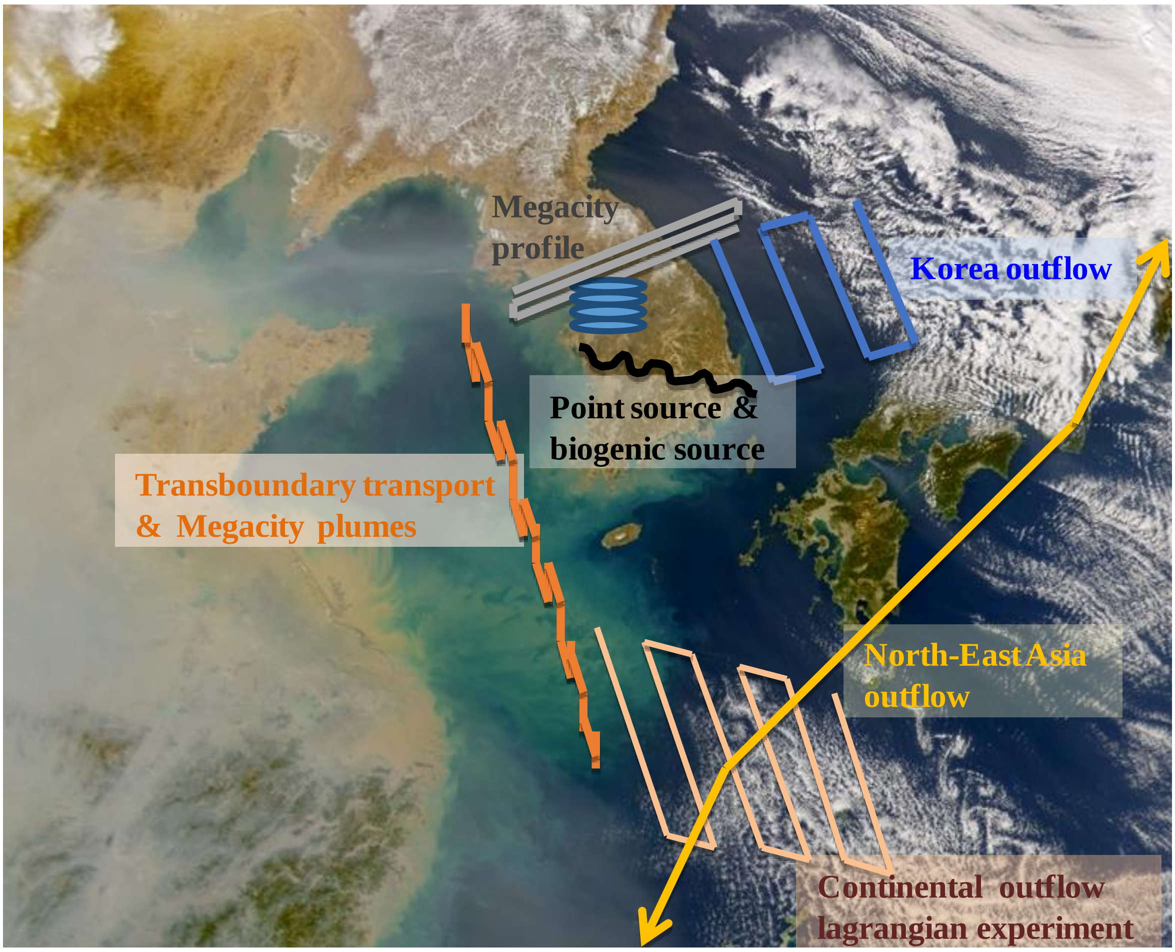

Figure 19. Proposed flight plans for two units of King-Air and DC-8

3) Satellite and other remote sensing measurements

The satellite team will be in charge of providing timely dataset during the campaign. Hourly information on aerosol properties including AOD, fine mode fraction (FMF), and single-scattering albedo (SSA) can be provided to intercompare with airborne and ground-based observations. The dataset can also be provided to the modeling team for data assimilation and evaluation. With the planned launch of TROPOMI in 2016, daily information on trace gas concentration can also be provided for intercomparison.

If the funding is available for the airborne version of GEMS based on its engineering model (EM), the developed GEMS data processing algorithm can be tested and the processed data can be supplied to the modeling and ground-based observation team. Otherwise, GEOTASO from the U.S. Science Team will be a very valuable contribution to substitute the airborne version of GEMS.

The capabilities from the ground-based optical and satellite platform during the campaign can be summarized as in Table 4.

Table 4. Capabilities from the ground-based optical and satellite platform

Parameters to be measured	S1	S2	S3	S4	G1	G2	G3	G4
O_3	√		√	√	√			√
NO_2	√		√	√	√		√	
SO_2	√		√	√				
HCHO	√			√				
AI	√		√	√				
AOD	√	√	√	√		√		√
SSA	√	√		√		√		
FMF		√				√		
CO				√				

S1 : OMI
S2 : GOCI
S3 : OMPS
S4 : TROPOMI
G1 : PANDORA
G2 : AERONET
G3 : DOAS
G4 : LIDAR

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