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No. 145

March 2017

JICA Research Institute



JICA Research Institute

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A Comparative Study of Urban Air Quality in Megacities in Mexico and Japan: Based on Japan-Mexico Joint Research Project on Formation Mechanism of Ozone, VOCs and PM_{2.5}, and Proposal of Countermeasure Scenario*

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Mitsuhiro Yamamoto^{*}, Takuro Watanabe^{**}, Tsuneaki Maeda^{**}, Akira Mizohata^{***}

Abstract

Photochemical ozone and black carbon are key substances both for regional air pollution and global climate change. These two pollutants are so-called SLCPs (Short-Lived Climate Pollutants). International comparison studies among megacities with widely different conditions are effective in clarifying the formation mechanisms of SLCPs. A comparison study in megacity areas of Japan and Mexico mainly focusing on ozone, VOCs (volatile organic compounds) and PM_{2.5} was conducted based on air pollution trend analysis and field measurements including vertical soundings of ozone and meteorological parameters. In this study, co-beneficial countermeasure scenarios based upon the obtained scientific data has been proposed. Photochemical ozone, EC (elemental carbon; a major SLCP), and NO_x (nitrogen oxides) and VOCs (NO_x and VOCs are implicit SLCPs) need to be controlled to improve the regional and global atmospheric environment. In Japan, countermeasures including the whole Asian area will be necessary because there is considerable contribution from trans-boundary air pollution. In Mexico, regulation of VOCs including energy shift and diesel exhaust gas control will be effective. These findings will be utilized to formulate and/or evaluate ProAire (Program for Air Quality Improvement) for the three studied megacity areas of Mexico.

Keywords: Mexico, SATREPS (Science and Technology Research Partnership for Sustainable Development), Ozone, VOCs (Volatile Organic Compounds), PM_{2.5}, Countermeasures

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This paper is part of a number of country cases within the JICA Research Institute's research project, "A Study on Urban Air Quality Improvement in Asia," which focuses on PM_{2.5} and other air pollution problems and analyzes relevant policies in primarily Asian countries.

This work was supported by the "Joint Research Project on Formation Mechanism of Ozone, VOCs, and PM_{2.5} and the Proposal of Countermeasure Scenarios" supported by the Japanese agencies JST and JICA under SATREPS scheme.

1. Introduction

Air pollution in megacities of developing countries needs quick countermeasures. Just as the scale and speed of economic growth are greater than those experienced by developed countries, the increase in the severity of air pollution is more rapid. The rise of such megacities worldwide has consequences not only in regional health problems but also in global climate change. Specifically, ozone and particulate matter (PM) are known to affect the global climate. Unlike classical air pollutants such as carbon monoxide (CO) and sulfur dioxide (SO₂), the complex chemistry involving ozone and PM is not fully understood even in developed countries.

Mexico City is representative of such megacities in developing countries. After its rapid industrialization in the late 1980s, Mexico City was named the “most polluted city on the planet” by the United Nations (Mage et al. 1996). Around this time, air pollution mitigation measures from developed countries including Japan were introduced, and the current air pollution level is equivalent to those in megacities in developed countries (e.g., the ozone concentration level in Mexico City is approximately the same as in Los Angeles (Parrish et al. 2012).

International research on air pollution in Mexico has been conducted mainly by US researchers (e.g., Molina et al. 2010). The target of this research was Mexico City or US-border cities such as Tijuana. This was especially true of the MILAGRO (Megacity Initiative: Local and Global Research Observations) campaign conducted in March 2006, which aimed to understand the mechanism of air pollution in Mexico City and the effect of megacity air pollution on regional and global scales, by investigating the characteristics of Mexico City atmosphere at an unprecedented scale in terms of human resources, scientific equipment, and spatial coverage.

While the US contribution has tended to focus on the scientific aspects of air pollution, the Japanese contribution has emphasized technology transfer and the development of human resources. JICA (Japan International Cooperation Agency) initiated the CENICA (The National Center for Environmental Research and Training) project in 1995 in collaboration with INE (National Institute of Ecology) of Mexico to confront the problems of air pollution and waste. Since 2012, with the promulgation of the General Law on Climate Change, INE was reorganized into INECC (National Institute of Ecology and Climate Change). Air pollution-abatement measures in Mexico and supporting projects from Japan to Mexico starting in 1980 are shown in Figure 1.

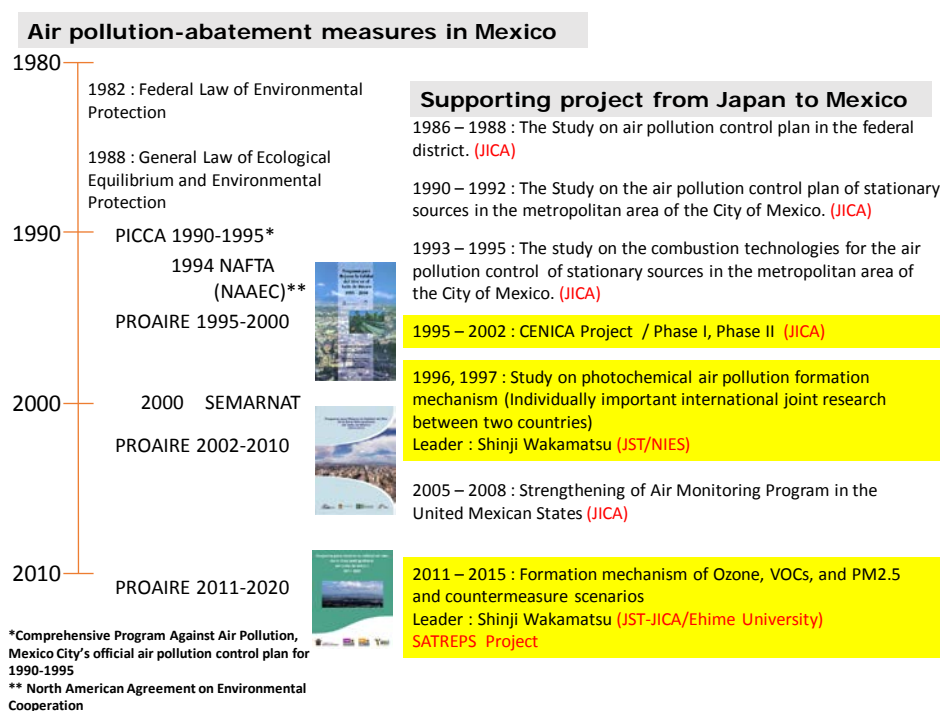


Figure 1. Air pollution-abatement measures in Mexico and supporting projects from Japan to Mexico starting in 1980.

Source: Author

Based on the CENICA project from 1995, an international research study entitled “Joint Research Project on Formation Mechanism of Ozone, VOCs, and PM_{2.5} and Proposal of Countermeasure Scenario” was conducted between Ehime University and INECC from 2011 to 2015 under the support of SATREPS (Science and Technology Research Partnership for Sustainable Development), which was jointly supported by JST (Japan Science and Technology Agency) and JICA (Japan International Cooperation Agency). The SATREPS project, which focused more on scientific research, conducted comparison studies of megacity areas of Japan (TMA: Tokyo Metropolitan Area) and Mexico (MCMA: Mexico City Metropolitan Area, GMA: Guadalajara Metropolitan Area, and MMA: Monterrey Metropolitan Area). It should be noted that Guadalajara and Monterrey, the second and the third largest cities in Mexico respectively, have received little attention from previous international research projects. Characteristics of the three Mexican metropolitan areas are shown in Figure 2.

The urban core of MCMA lies on a plateau surrounded by mountain ranges on the west, east, and southwest sides, with a prevailing wind from the northern gap. Many of the heavy-industry factories have been relocated out of the Mexico City basin since the worst air pollution periods around 1990. Today, the main economic activity is commerce and the largest source of air pollutants is the automobile. Although the number of registered vehicles in MCMA is considerably more than in GMA or MMA, the strictly enforced exhaust-gas regulations suppress the emission per vehicle (Kanda et al. 2016).

GMA is located on a lower plateau than MCMA, and consequently experiences higher daytime temperatures. The prevailing wind is the westerly sea breeze coming from the Pacific Ocean, about 230 km to the west. As is true in MCMA, there are not many factories of heavy industries in GMA, the primary economic activity is commerce, and most air pollutants are emitted from automobiles. However, because there is no exhaust-gas inspection scheme in GMA, the proportion of old cars is much higher than in MCMA.

MMA is located on a still lower plane than GMA or MCMA. The prevailing wind comes from the Gulf of Mexico on the east. The climate is arid-steppe with high daytime temperatures (typically beyond 30 degrees Celsius) but with a large day-night difference. The main economic activity in MMA is manufacturing. Near the city center, there are large cement and steel plants together with smaller related industries. Quarry mining in the west and an oil refinery in the east are also significant sources of emission , as are automobiles. Emission intensity per vehicle is estimated to be the highest among the three cities (Benítez-Garcia et al. 2014).

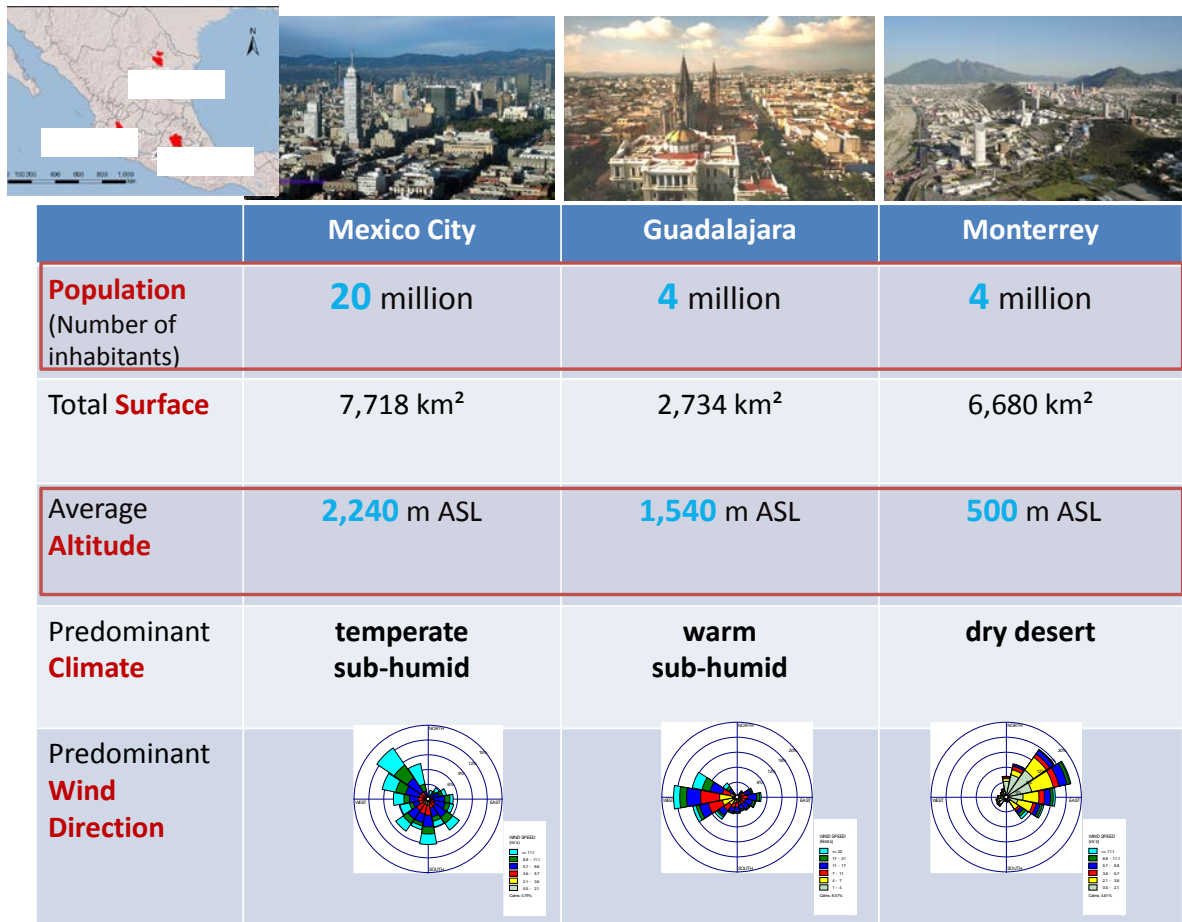


Figure 2. Characteristics of Mexican three Metropolitan Area.

Source: Benítez-Garcia et al. (2014)

As shown in Figure 3., the formation of urban ozone and $PM_{2.5}$ are related to each other. In particular, the involvement of VOCs makes the relationship complicated because VOCs are composed of several hundreds of chemical compounds and the behaviors of individual compounds are not understood well. To understand this complex relationship, the project consisted of six working groups (WGs) (WG1: ozone, WG2: VOCs, WG3: $PM_{2.5}$, WG4: personal exposure and roadside air pollution, WG5: monitoring, emission, and modeling, WG6: co-beneficial countermeasure scenarios), which focused on individual topics while communicating closely with each other (Figure 4.).

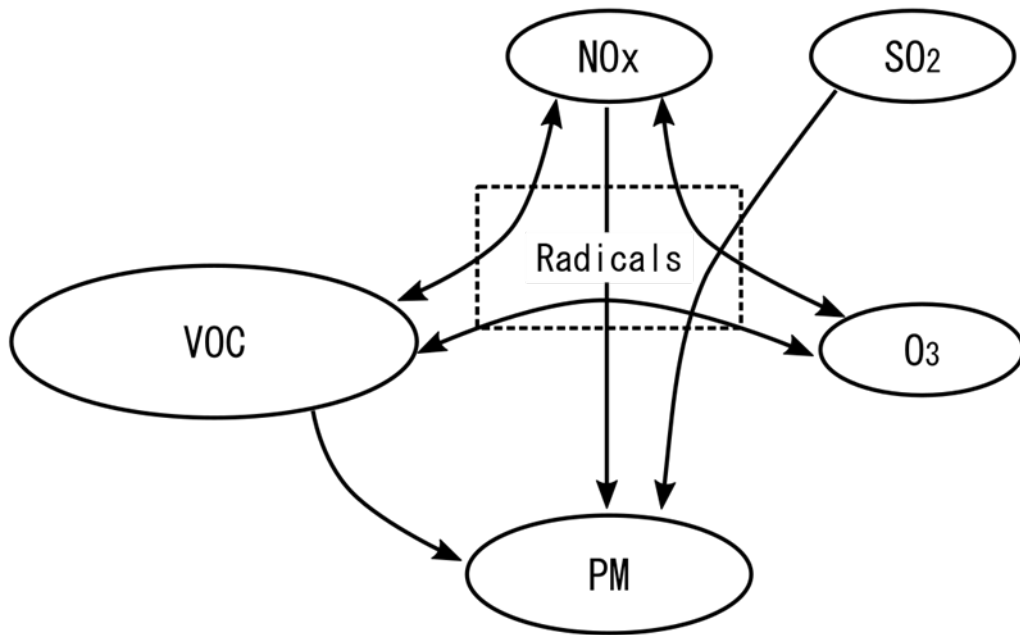


Figure 3. Relationship between VOCs, NO_x, SO₂, Ozone and PM in urban atmosphere.

Source: Author

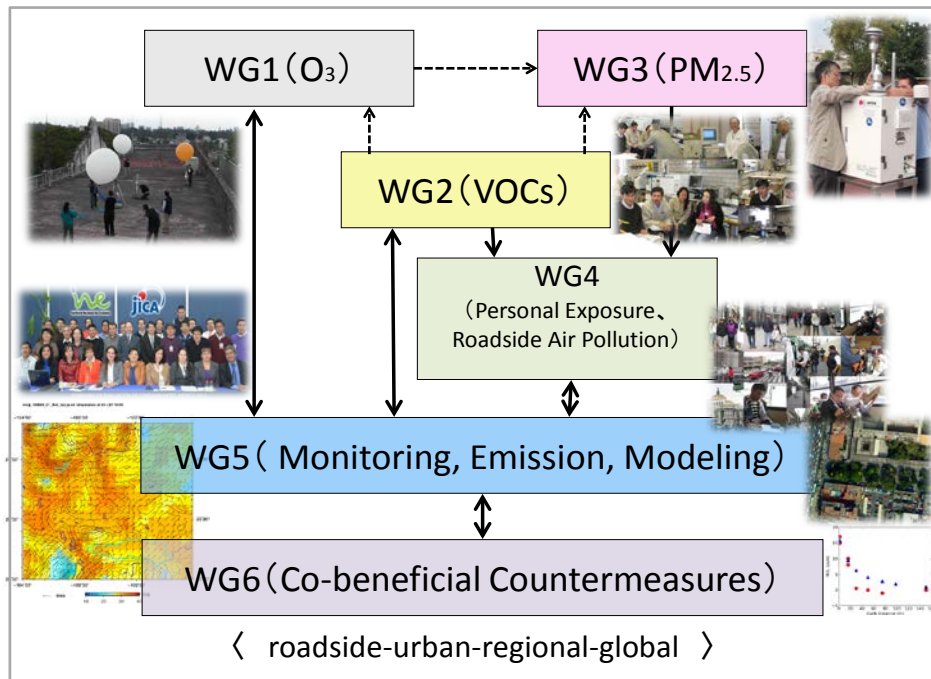


Figure 4. Relationship between the six working groups (WG)

Source: Author

The purpose of this paper is to review the activities of the WGs. Firstly, an analysis of air-quality monitoring data by WG5 is presented; secondly, field observations and related technology transfers by WG1, WG2, and WG3 are explained, and finally, proposals for air-pollution mitigation by WG6 are described. The originality of the project, the work presented in this paper and those already published elsewhere, lies in its comparison of different urban areas: MCMA, GMA, MMA, and cities in Japan. The distinctly different air pollution characteristics in these areas were investigated concurrently to reveal the pollution formation mechanism more clearly than when single areas were studied in separate projects.

2. Air pollution trends in Japan and Mexico (WG5)

To provide an idea of the state of air pollution in the two countries, we present statistics of ozone concentrations in the target areas. We choose ozone here because it plays a central role in the atmospheric chemistry (Seinfeld and Pandis 2006). For other air pollutants, the reader is referred to Benítez-García et al. (2014). A key point is that ozone is generated and destroyed through complex photochemical reactions involving NO_x and VOCs. Emphasis is placed on the comparison between Mexico and Japan.

Figure 5. shows the trends of annual means in TMA of the daily maximum Ox (photochemical oxidants consisting mostly of ozone) concentration, the daily mean of NO_x concentration, and the early-morning mean of NMHC (Non-Methane-Hydro-Carbons: a subset of VOCs) concentration. Despite the clear decreases in the precursor substances NO_x and VOCs, the concentration of Ox has not changed much over the past decade.

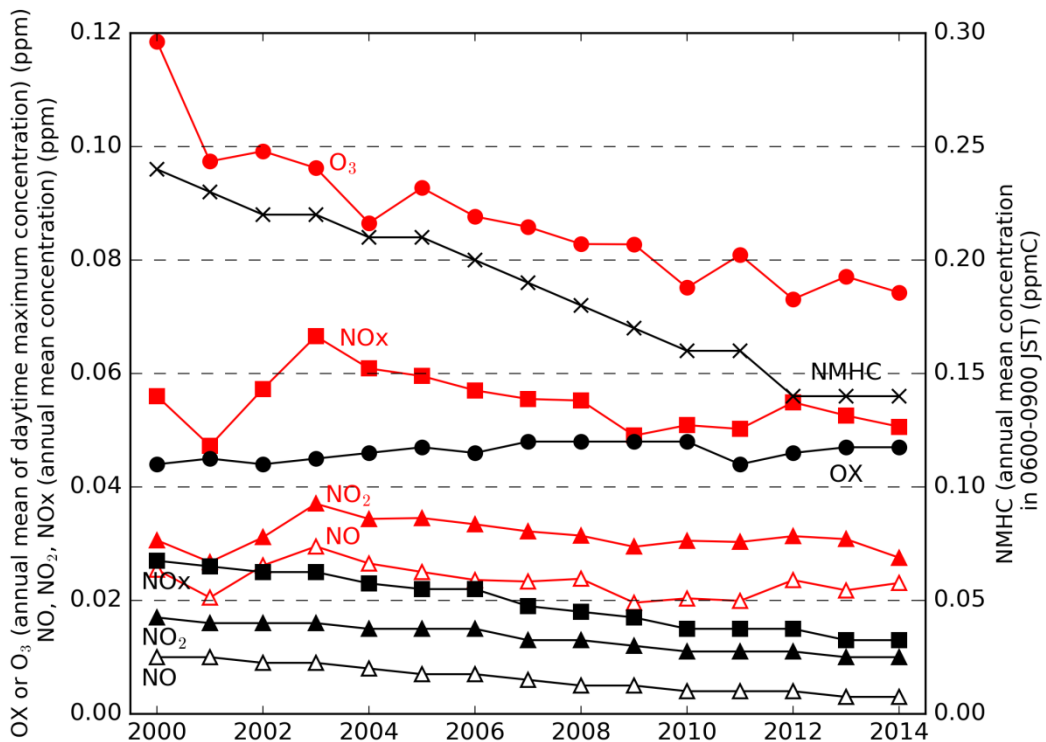


Figure 5. Annual Means of O_x, NO, NO₂, NO_x, and NMHC in Mexico (MCMA) and Japan (TMA)

Sources: MOEJ, Mexico City government

Notes: Annual means of the daily maximum O_x, daily average NO, NO₂ and NO_x, and early-morning (6:00 – 9:00) average in TMA (black), and corresponding O₃, NO, NO₂ and NO_x concentrations at four representative stations (MER, PED, UIZ and XAL) average in MCMA (red)

MER:Merced, PED:Pedregal, UIZ:UAM Iztapalapa, XAL:Xalostoc

Annual statistics for ozone concentration in megacity areas of Mexico exhibit different trends in different areas (Figure 6.). Note that the O₃ concentration in Figure 6. is the annual mean of daily averages whereas that in Figure 5. is the annual mean of the daily maximum. In MCMA, the annual mean (based on the daily and spatial averages) has been

decreasing until recently when it seems to revert upward, but in GMA and MMA, the annual means have generally been increasing. The increase in GMA has been particularly large.

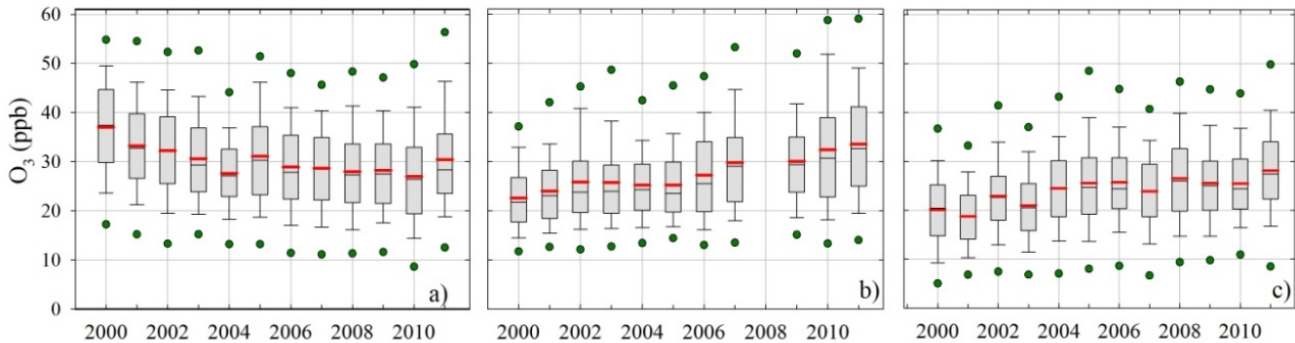


Figure 6. Trends of annual statistics of O_3 concentration in a) MCMA, b) GMA, and c) MMA.

Source: Benítez-García et al. (2014)

Notes: The panels show the annual mean (thick line), median (thin line), 2nd and 98th percentiles (green circles), 10th and 90th percentiles (whiskers), and 25th and 75th percentiles (gray boxes) of the daily average concentrations.

Diurnal variation of the ozone concentrations in Japan and Mexico are shown in Figure 7. The difference between the daily maximum and minimum is larger in Mexico than in Japan, with the largest difference occurring in Mexico City. This is because the daytime production of ozone and the nighttime titration of ozone by nitric oxide (NO) are more intense in Mexico than in Japan (Benítez-García et al. 2015).

The reasons for these differences between Japan and Mexico can be understood in terms of the effects of global-warming, long-range transport of ozone, and non-linear relationship between NO_x, VOCs, and ozone. However, because the weights of these parameters are different depending on the emission intensity, meteorology, and geographical

characteristics, extensive investigation including emissions estimation, field observations, and numerical modeling is necessary to understand the reasons.



Surface monitoring data of O₃

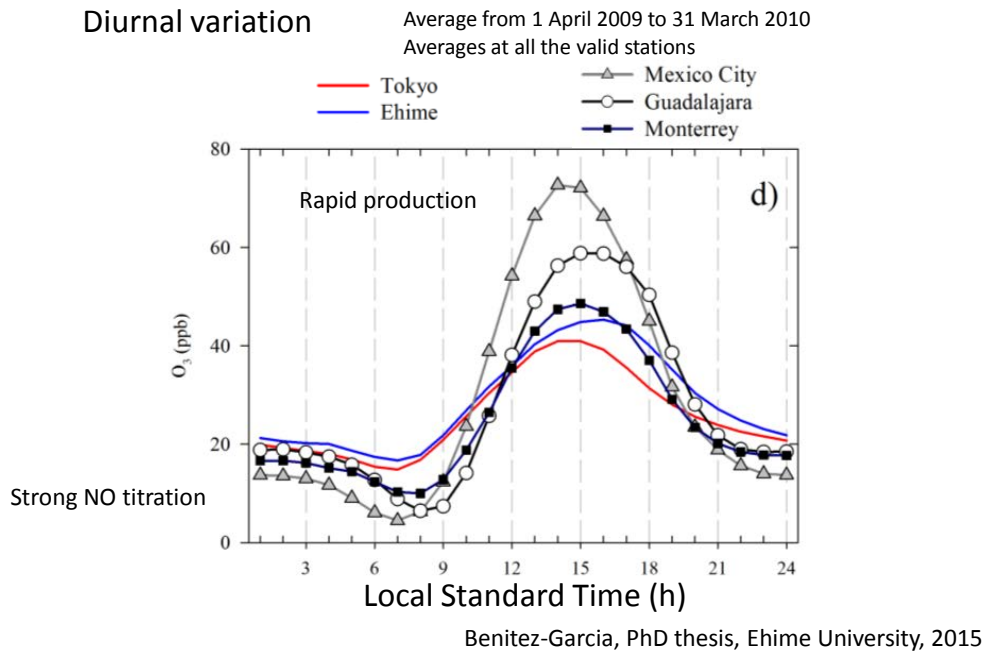


Figure 7. Diurnal variation of ozone concentration in Japan and Mexico

Source: Benítez-Garcia et al. (2015)

3. Vertical ozone distribution in Japan and Mexico (WG1)

In urban areas, the spread of ozone in the atmosphere can become fairly wide depending on meteorological conditions, because chemical reactions and meteorological conditions contribute equally to the production and transport of photochemical ozone. Driven by meteorological motions such as synoptic-scale horizontal wind, local circulations like land-sea breezes and mountain-valley winds, and mixing in the convective boundary layer, photochemical ozone can spread over 100 km horizontally and 3-4 km vertically. Such factors can cause high ozone concentrations in regions far away from the source of the ozone

precursors NO_x and VOCs (Seinfeld and Pandis 2006). The distribution of photochemical ozone in three dimensions is thought to be influenced considerably by the size, emission characteristics, orography, elevation, latitude, and climate of the individual city. The vertical distribution of ozone is strongly affected by the removal processes in the surface boundary layer where ozone reacts with NO generated mainly by human activities and is also decomposed on solid surfaces. The residual high-zone air in the upper free troposphere has been known to travel long distances (Neu et al. 1994; Kleinman et al. 1994). Due to the above characteristics, three-dimensional observation is required to understand the behavior of photochemical ozone in an identified region. In general, airplane observation is often employed to take three-dimensional measurements of meteorology and ozone (e.g., Wakamatsu et al. 1981 and references therein; Molina et al. 2010), but ozonesonde observation is a less costly alternative to take vertical profiles. Vertical profiles of ozone concentration provide valuable information not only for understanding the formation mechanism of ozone but also for planning countermeasures to reduce photochemical ozone. Because three-dimensional observations bring into scope the whole airshed of the target city, efficient and rational countermeasures can be devised. In this project, the first of its kind in Mexican cities, we estimated overall ozone production based on the measurements of ozone and meteorological elements in the convective mixing layer.

For vertical distributions, ozonesonde observations were conducted in Mexico City and Guadalajara. In both cities, the observation balloons were launched from the SMN (Servicio Meteorológico Nacional), which are routine observation sites for vertical sounding. The instruments used were GPS radiosonde for obtaining meteorological parameters (temperature, relative humidity, wind speed, and wind direction) and ozonesonde (GPS radiosonde attached to an electrochemical concentration cell (ECC) ozone sensor) (Kanda et al. 2014). The instruments ascended at a rate of about 5 ms⁻¹ up to about 16 km ASL (above sea level) for radiosonde and to about 30 km ASL for ozonesonde (Figure 8).

The following characteristics were noted. In MCMA and GMA, ozone was uniformly distributed with a high concentration in the relatively thick mixing layer where the potential temperature was approximately uniform. In TMA and GMA observations, long-range transport of ozone was considered to be the cause of these concentration peaks in the mid-troposphere (Figure 9).

Vertical ozone profiles

Mexico City 2012 Mar 8

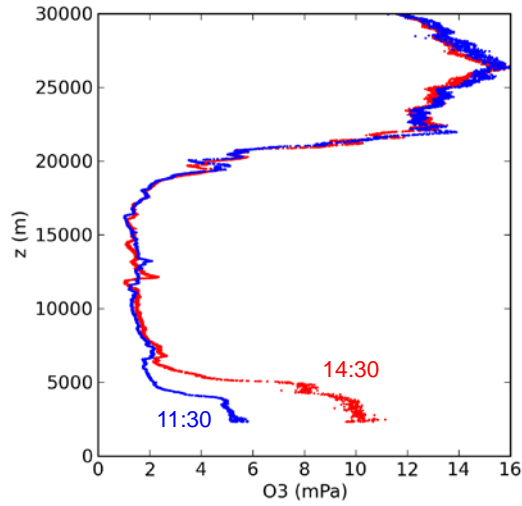


Figure 8. Vertical ozone profiles in Mexico City

Source: Author

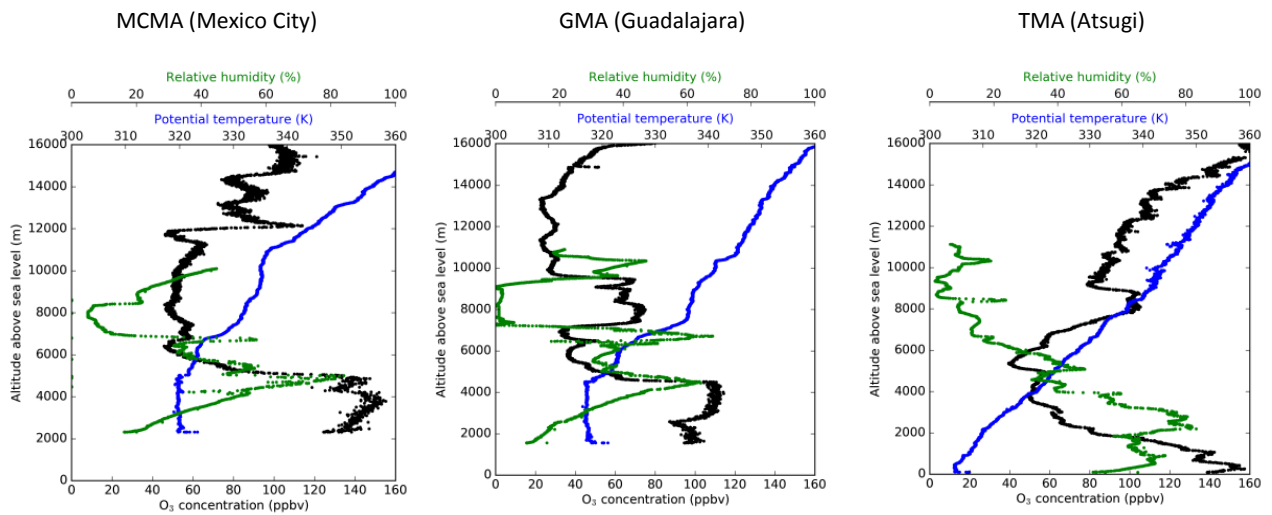


Figure 9. Vertical ozone distribution in Mexico (MCMA, GMA) and Japan (TMA).

Source: Author

Note: Black dots: O₃ concentration; blue dots: potential temperature; green dots: relative humidity.

4. VOCs in Japan and Mexico (WG2)

There are more than 100 major VOCs; some are photochemically reactive, others are toxic, and still others are both photochemically reactive and toxic. Benzene, for example, has high toxicity, but low photochemical reactivity. VOCs with both high toxicity and photochemical reactivity that have to be controlled in terms of ozone production and toxic effect include aldehydes, 1-3 butadiene, and toluene. In the photochemical production of ozone, both reactivity and the amount of VOCs are relevant. Low-reactivity VOCs can increase cumulative ozone production if present in large amounts and reacting steadily (e.g., Stevenson et al. 2000 for methane). Therefore, knowing the atmospheric composition of individual VOC species is important to countermeasures against both photochemical air pollution and toxic hazards. In the measurement of VOC species, it is necessary to develop and design methods of preparing standard gases, calibrating analytical instruments, sampling ambient air, and monitoring routinely. In this project, we conducted field measurements of VOC species and technology transfer on the methods of standard-gas production.

4.1 Characteristics of VOC concentrations

In a previous study of a JICA project (see Figure 1), the concentration of VOCs in MCMA in the year 2000 was found to be more than 10 times higher than in TMA, especially for low carbon number compounds such as propane and butane (Figure 10).

Figure 10. depicts VOC data from Teotihuacan (rural) at the top of a pyramid 65 m above ground level, is located 50 km north-east of Mexico City; Urawa (urban), located about 20 km north of Tokyo; and Niijima Island (remote), located 160 km south of Tokyo.

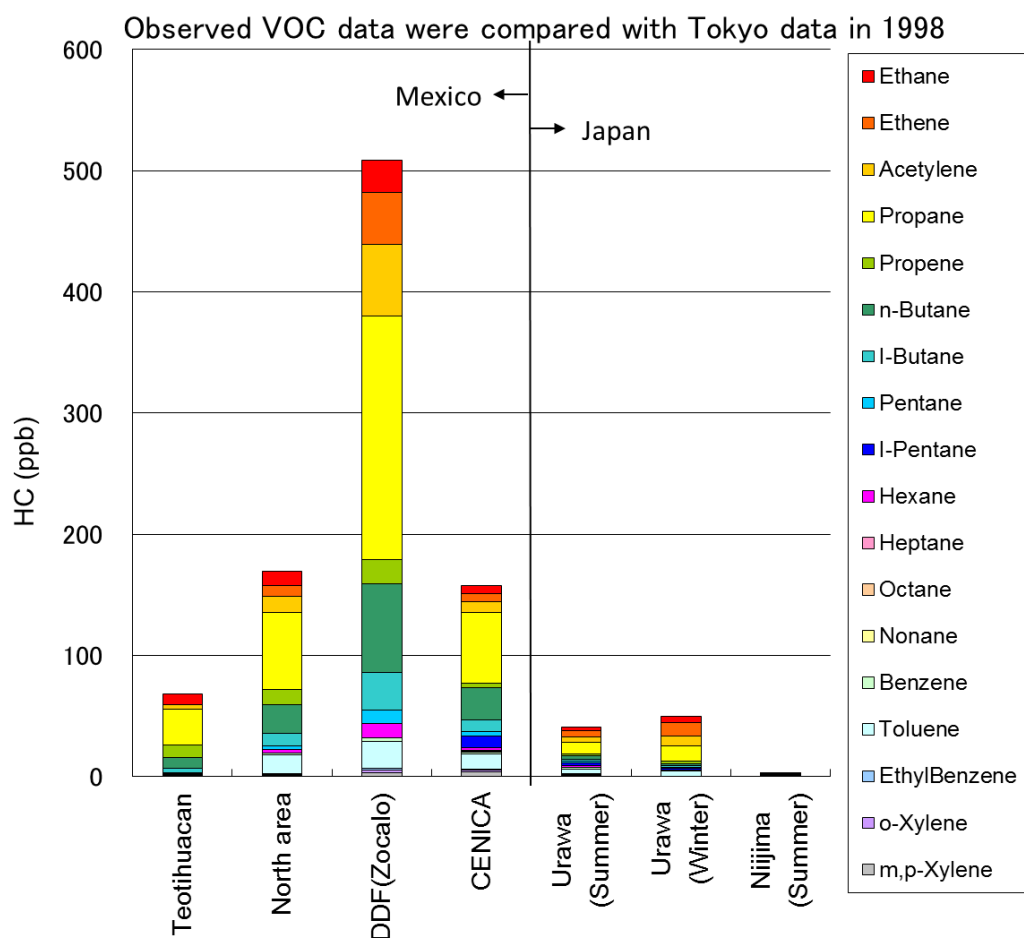


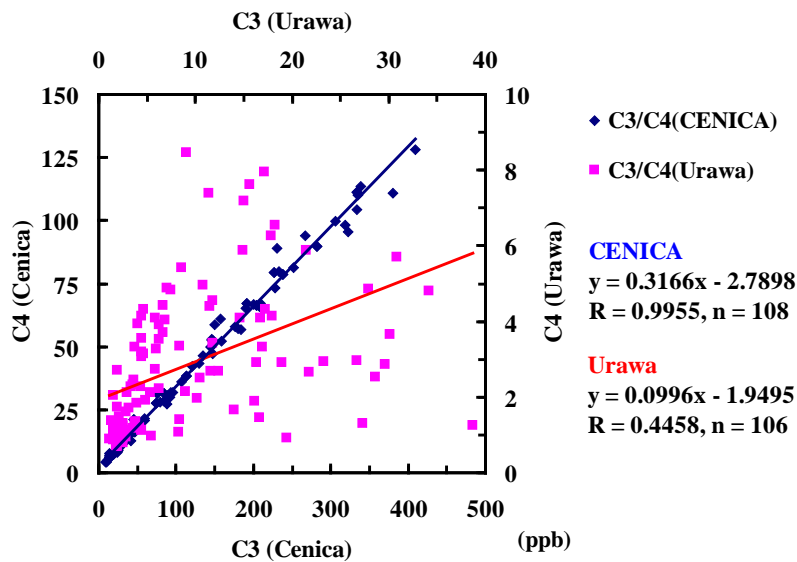
Figure 10. VOC species in Mexico City and Japan (Urawa and Niijima).

Source: Prepared by Dr. Tazuko Morikawa

Note: Teotihuacan (rural), northern area of Mexico City (urban), DDF Zocalo (urban center of Mexico City), CENICA (urban), Urawa (urban), Niijima (remote).

Scatter plots of propane (C3) and butane (C4) at CENICA and Urawa observed in 1999 are shown in Figure 11. Note that the full scales of C3 and C4 in CENICA are 500 ppb and 150 ppb, respectively, whereas those in Urawa are 40 ppb and 10 ppb, respectively. Hence, the approximate magnitudes in CENICA were 10 times higher than in Urawa. The C4/C3 ratio in CENICA was 0.3 and the correlation coefficient was approximately 1.0. This indicates the

emission sources of C3 and C4 were identical. On the other hand, in Urawa, the correlation between C3 and C4 was not significant and a non-negligible intercept is observed in C4. This indicates the emission sources of C3 and C4 were different in Urawa.



Correlation between propane and butane at CENICA from March 24 to March 29 and Urawa from August 1 to August 9, 1999.

Figure 11. Correlations between C3 and C4 concentrations in Mexico City (CENICA) and Japan (Urawa)

Source: Author

In Figure 12, correlation plots are shown for propane, butane, and toluene observed in CENICA in 2000 and 2012. The results in 2012 were obtained in the SATREPS project. Note that the full scales in 2000 of propane, butane and toluene are 450 ppb, 150 ppb and 100 ppb, respectively, and 140 ppb, 50 ppb and 100 ppb, respectively, in 2012. Propane and butane decreased by approximately one-third over the 12 years, but their concentrations were still three times higher than those observed in Urawa in 1999 (Figure 11). The concentration of toluene was almost the same in 2000 and 2012. The average concentrations obtained in March

2006 between 9:00 and 18:00 during the MILAGRO campaign (Apel et al., 2010) are comparable—38 ppb for propane, 20 ppb for n-butane, and 11 ppb for toluene—although the sampling location (Mexican Institute of Petroleum) was different,

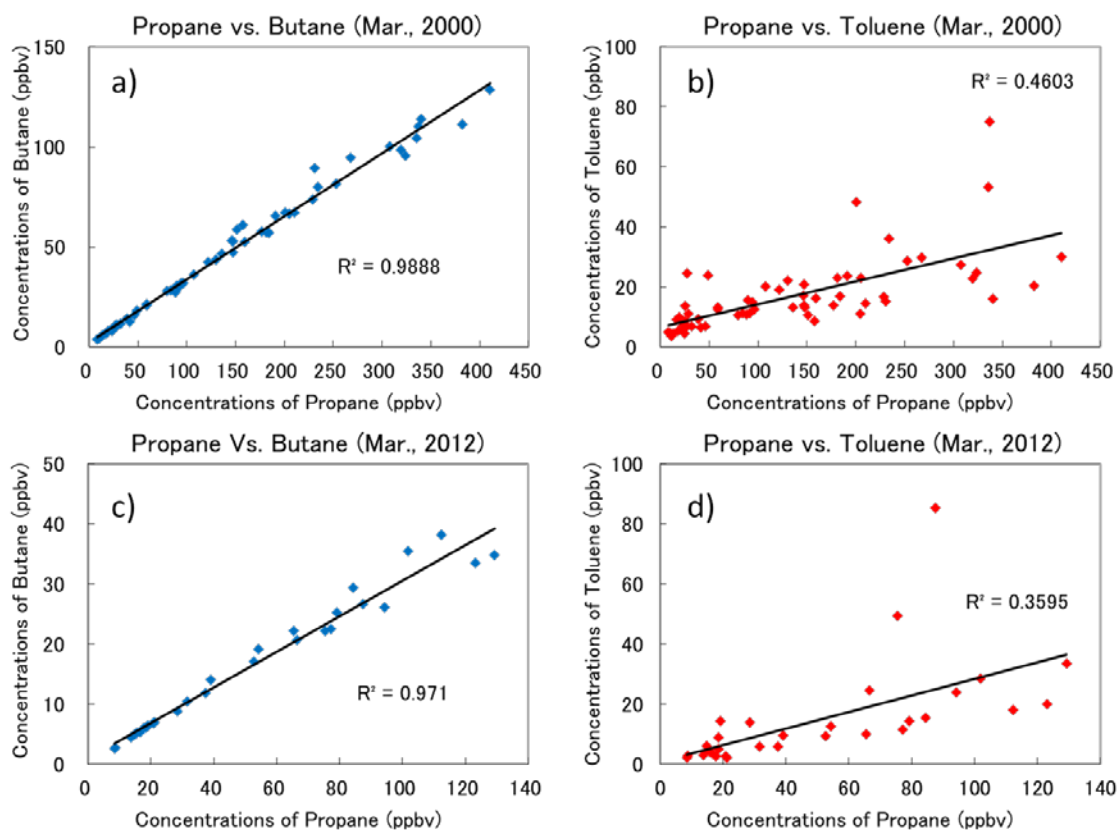


Figure 12. Correlations between concentrations of propane vs. butane and propane vs. toluene

Source: Author

Note: a) propane vs. butane (Mar. 2000); b) propane vs. toluene (Mar. 2000); c) propane vs. butane (Mar. 2012); d) propane vs. toluene (Mar. 2012). The observation site was CENICA.

A major source of these Alkane species (propane and butane) is LPG (Liquefied Petroleum Gas) (Velasco et al. 2008, Garzon et al. 2015). In Japan, the regulation of VOCs from stationary sources started in 2006 and since then the high percentile values of the daily maximum ozone concentrations have decreased.

4.2 Technology transfer of standard gases to CENAM

The supply system of standard gases in Mexico and quality control of measurement results was examined through VOC monitoring in Mexico, leading to the conclusion that a unified quality control system including environmental criteria needs to be established. The scheme of technical support and technology transfer in the monitoring field of VOCs in Mexico are summarized in Figure 13 (Watanabe and Maeda 2016).

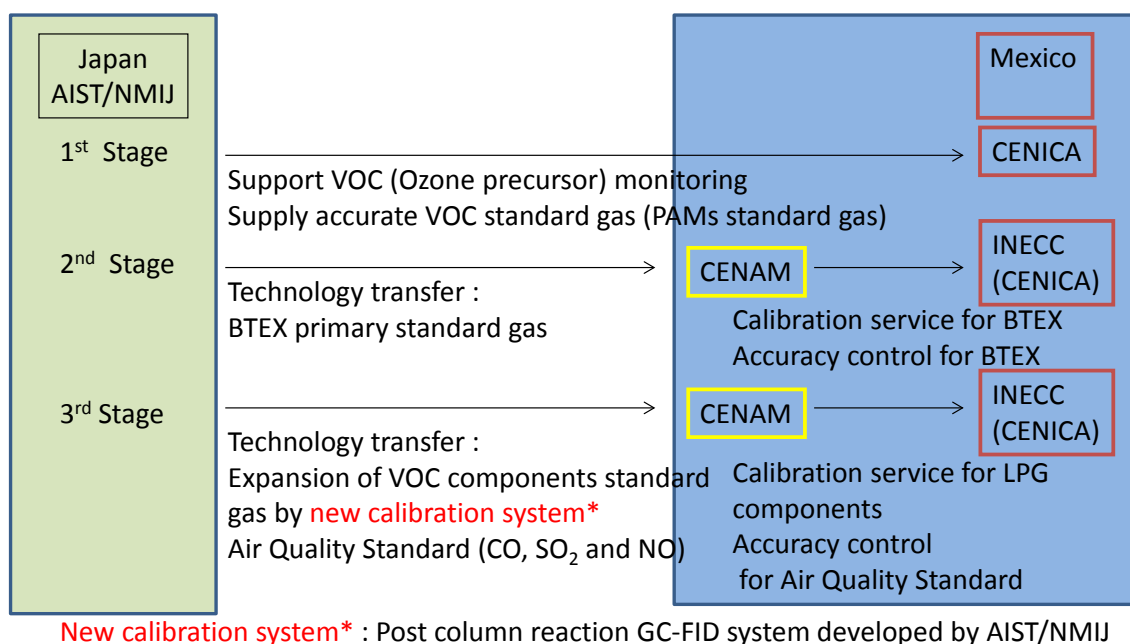


Figure 13. Scheme of technical support and technology transfer in the air monitoring field in Mexico.

Source: Author

Notes: BTEX (Benzene, Toluene, Ethyl benzene, Xylene),

CENAM (Centro Nacional de Metrologia: National Metrology Center),

AIST (The National Institute of Advanced Industrial Science and Technology), NMIJ (National Metrology Institute of Japan),

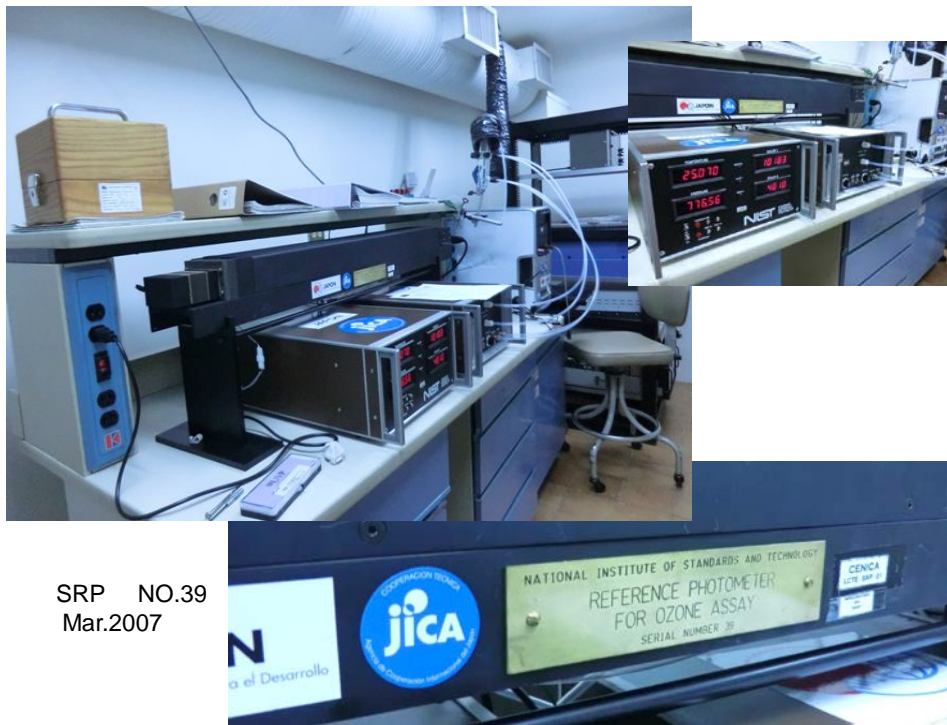
INECC (Instituto Nacional de Ecología y Cambio Climático: National Institute of Ecology),

PAMS (Photochemical Assessment Monitoring Stations)

The transfer of technology related to BTEX standard gases preparation/calibration (2nd Stage in Figure 13) was carried out in the following manner. Engineers for CENAM were invited to AIST/NMIJ and were trained to prepare and validate BTEX standard gases (concentration level: 1 $\mu\text{mol/mol}$). After the training, CENAM established its own calibration service for BTEX standard gases in Mexico. In July 2014, CENAM began providing the calibration service under MoU (Memorandum of Understanding) between CENAM and INECC. The second stage was thus successfully completed.

The third and final stage is a future challenge following the decommission of the project. CENAM will supply SI-traceable (traceable to International System of units) standard gas to INECC, and INECC will be responsible for maintaining the quality of the air monitoring data in Mexico.

CENAM and INECC renewed the MoU in 2015 and will continue their strong collaboration in this field. Under the renewed MoU, it is expected that they will establish a similar collaboration system to that between the NIST (National Institute of Standards and Technology) and the EPA (Environmental Protection Agency) in the USA. Technology transfer on standard gases from AIST/NMIJ to CENAM will contribute to building such a system of collaboration. Such activities are effective for continuous and self-sustained improvement in the ambient monitoring in Mexico not only of VOCs but also of criteria pollutants. All these will contribute to the air pollution prevention program in Mexico. AIST/NMIJ will continue to cooperate with CENAM in the area of standard gases. Such collaboration is expected to expand to other Latin American countries through Mexico.



SRP NO.39
Mar.2007

Figure 14. SRP (Standard Reference Photometer) introduced to CENICA

Source: Author

The air quality monitoring system was also improved by the introduction of SRP (Standard Reference Photometer: The NIST SRP developed jointly with the US-EPA to provide the traceability for ozone measurement.) (Figure 14.) for ozone, and SI traceable standard gases for VOCs and criteria pollutants (CO, SO₂, NO₂). A reliable air pollution monitoring network system, using SRP and SI traceable standard gases established in this project, should be disseminated in the future not only in Mexico but also in Latin American countries.

5. PM species in Mexico (WG3)

Environmental standards for PM_{2.5} are defined in terms of mass concentration. However, the substance composition varies widely by region and season (Tai et al. 2010), and PM_{2.5} originates both from natural and anthropogenic sources. Therefore, it is necessary to know the composition of PM_{2.5} to devise effective countermeasures. The predominant components of PM_{2.5} are inorganic substances such as sulfate and nitrate, and carbonaceous substances such as elemental carbon (black carbon) and organic matter. Though metal components comprise only a small portion of the total mass, they provide valuable information for identifying emission sources (Querol et al. 2001). In this project, we conducted field measurements and analysis of PM_{2.5} components.

5.1 Field observation

Characterization studies of PM_{2.5} species in the Mexican major metropolitan areas of Mexico City, Monterrey, and Guadalajara were conducted to clarify the PM_{2.5} formation mechanism and propose countermeasure scenarios for improving the atmospheric environment in these regions.

PM_{2.5} was collected for 24 hours using two kinds of filters: a stretched Teflon filter with a support ring which meets the EPA standards and quartz fiber filter. The quartz fiber filter was heated in advance to remove carbonaceous compounds. The mass concentration of PM_{2.5} was measured using the gravimetric method on samples collected on the Teflon filter.

Concentrations of various elements in PM_{2.5} were also measured in the Teflon filter samples using the energy dispersive X-ray fluorescence (EDXRF) method. Concentrations of carbon components, organic carbon (OC) and elemental carbon (EC) in PM_{2.5} were measured from the quartz filter samples using the thermal optical reflectance carbon analysis method

with the IMPROVE-A protocol. Water soluble ionic species were also measured using the quartz filter samples using the ion chromatography method.

PM_{2.5} mass concentration can be explained by adding up the major individual constituents. This sum is called the reconstructed fine mass concentration (RCFM). The major PM mass constituents are (NH₄)₂SO₄, particulate organic matter (POM), EC, crustal particles (soil), biomass aerosol, sea salt, NH₄NO₃, and NH₄Cl. RCFM should provide a reasonable estimate of the dry PM_{2.5} mass concentration in the atmosphere. Inconsistency between RCFM and the weighed PM_{2.5} mass concentration indicates errors in one or more of the processes.

In this study, RCFM was defined by the following equation.

$$\text{RCFM} = 4.125[\text{S}] + [\text{POM}] + [\text{EC}] + [\text{Crustal}] + 1.4[\text{KNON}] + 2.5[\text{Na}^+] + 1.29[\text{NO}_3^-] + [\text{NH}_4\text{Cl}]$$

POM was defined by $[\text{POM}] = \text{C1}[\text{OC}]$, where C1 = 1.6 was selected from the typical range from 1.4 to 2.2 accounts for the non-carbon elements in POM.

Crustal was defined by $[\text{Crustal}] = 2.32[\text{Al}] + 2.63[\text{Si}] + 1.72[\text{Ca}] + 1.67[\text{Fe}] + 2.05[\text{Ti}]$, where 10% of the mass was assumed to be water.

KNON (non-soil potassium) was defined by $[\text{KNON}] = [\text{K}] - \text{R0} [\text{Fe}]$, where R0 is the ratio of [K] to [Fe] in coarse particles.

The results of the RCFM analysis are listed in Table 1.

Table 1. Major PM_{2.5} mass constituent contribution (µg/m³) in the atmosphere of the metropolises in Mexico.

	Mexico City		Monterrey		Guadalajara	
PM _{2.5}	41.2		29.5		17.7	
POM	13.4	33%	8.5	29%	5.5	31%
EC	5.2	13%	2.0	7%	1.7	10%
(NH ₄) ₂ S	6.9	17%	5.0	17%	3.4	19%
O ₄						
NH ₄ NO ₃	3.0	7%	3.2	11%	0.7	4%
Crustal	11.5	28%	6.6	22%	4.3	24%
Biomass	0.5	1%	0.3	1%	0.2	1%
NH ₄ Cl	0.3	1%	0.6	2%	0.2	1%
Sea salt	0.4	1%	0.6	2%	0.1	1%
RCFM	41.2	100%	26.8	91%	16.1	91%

Source: Author

Numbers of valid observation dates were approximately 80, 30, and 25 days in Mexico City, Monterrey, and Guadalajara and the average values of PM_{2.5} concentrations were 41.2, 29.5, and 17.7 µg/m³, respectively. In all the cities, PM_{2.5} concentrations exceeded the Mexican environmental standard of 15 µg/m³ for the annual average, although the sampling season could have affected the result considerably.

In Mexico City, RCFM was almost the same as PM_{2.5} mass concentration, whereas in Monterrey and Guadalajara, RCFM was about 9% smaller than PM_{2.5} mass concentration. The compositions in the three metropolitan areas resembled each other, though in Mexico City, the EC concentration was more than two times higher than in other cities.

Figure 15. shows the scatter diagram between the EC concentration and the concentration ratio of OC and EC (OC/EC). A high EC concentration exceeding 10 µg/m³ was observed only in Mexico City. The ratio of OC/EC decreases as the EC concentration

increases. Since a high EC and low OC is indicative of diesel exhaust particles (DEP), the contribution of DEP is considered to be large in Mexico City.

In Monterrey, a higher Ca percentage than in natural soil was observed. At the Obispado station in Monterrey, the ratio of Ca and Si in the PM_{2.5} was 4.03 (in general, the average of Earth's crust is Ca / Si = 0.24). Sources of this enriched Ca are considered to be limestone mining processes and eroded soil related to these facilities. In Monterrey, a high iron concentration was also noted, which indicates some industrial emission sources.

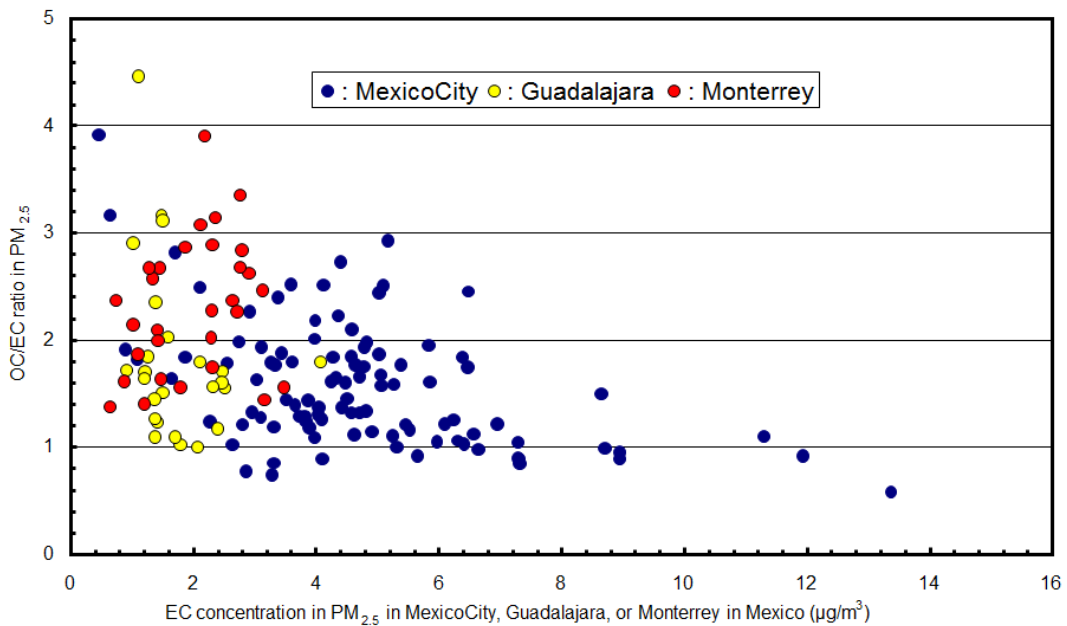


Figure 15. Scatter plot between EC concentration and concentration ratio of OC to EC.

Source: Author

5.2 PM_{2.5} data in the megacities of the world

The annual mean of diurnal PM_{2.5} variations at selected monitoring stations in Mexico (Mexico City, Monterrey), USA (Houston, Los Angeles, New York, and Philadelphia), China (Beijing), and Japan (Tokyo, Osaka, Okayama, Hiroshima, Kagawa, and Ehime) are shown in Figure 16. The concentration level in Beijing is 2-5 times higher than in other cities. Diurnal peak occurs

at noon in Mexico City, but at midnight in Beijing. In Mexico City, photo-chemically formed $PM_{2.5}$ is considered dominant. On the other hand, in Beijing, a different formation mechanism is considered prevalent. The formation mechanism of this night-time $PM_{2.5}$ have not yet been clarified in detail, but it is likely to involve heterogeneous processes and to be dependent on meteorological factors such as relative humidity, temperature, wind speed, and inversion height.

As mentioned previously, the most abundant component in $PM_{2.5}$ in Japan is sulfate, but in Mexico is POM. In MCMA, the contribution of EC is significant.

In MCMA, measured data showed considerably higher concentrations of PM_{10} , $PM_{2.5}$ and VOCs than in other regions except for Beijing, likely due to the local anthropogenic and natural emissions.

A comparison of $PM_{1.0}$ between Japan and Mexico should be conducted to clarify the contributions of individual anthropogenic sources. A speciation study of $PM_{1.0}$ will be important in future research.

PM2.5 : Diurnal Variation (Annual Mean)

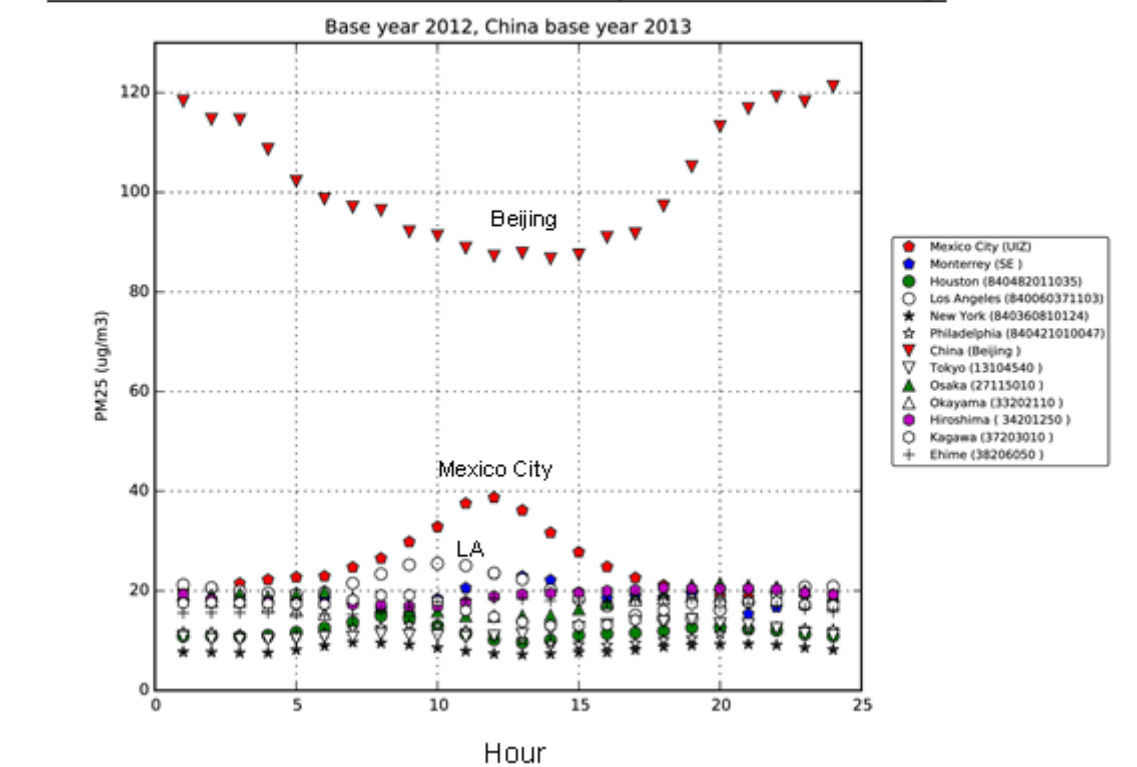


Figure 16. Diurnal variations of PM_{2.5}

Source: Author

Notes: Letters in parentheses indicate identification codes of the monitoring stations.

6. Countermeasures for PM_{2.5} and O₃ in Mexico and Japan (WG6)

Based on the collaborative field measurements in MCMA, GMA, and MMA, and the analysis of other air pollution and socio-economical issues, the Japanese and Mexican researchers sought solutions to mitigate the air pollution in the Mexican urban areas. In this project, we proposed countermeasure scenarios based on the outputs of the six WGs and on discussions between the Japanese and Mexican researchers. The proposed scenarios will be utilized to formulate and/or evaluate ProAire (Program for Air Quality Improvement) for three modeled megacity areas of Mexico.

ProAire is a set of action plans for air-quality improvement composed by the relevant state or municipal government under the supervision of the federal government. Mexico City and the Jalisco state where GMA is located have regularly published ProAires, with the current ones running from 2011 to 2020. The Nuevo Leon state where MMA is located is preparing to build one as well. The federal government plans to complete ProAires in all the major airsheds by 2018. Scientific evidence and comprehensive socio-economical analysis are indispensable to these programs.

The following 4 items summarize the proposals based on the results from co-operative investigation conducted during the SATREPS project.

(1) Measures for VOCs emission control related to photochemical Ozone

As discussed in Kanda et al. (2016), ozone production in MCMA and GMA is considered to be in the VOC-limited regime. In the VOC-limited regime, ozone concentration increases when NO_x emissions are reduced, but decreases when VOCs emissions are reduced. Therefore, measures focused on VOC reduction are considered important in the Mexico City metropolitan area.

Probable reduction targets include paint solvents that occupy a large proportion of stationary emissions. Propane and butane that were identified to derive from LPG leakage are also of concern because of their high concentrations despite their low photochemical reactivity.

In Japan, VOC emission control measures for stationary sources have been promoted by amending the Air pollution Control Law in 2004 to reduce high-ozone events. Emission control was implemented in a combined manner by imposing regulations on large-scale sources as well as promoting voluntary efforts by small-scale sources. The scheme was called the “best mix” approach. Legal regulation started in 2006, and the goal of 30% reduction compared to the emissions in 2000 was achieved in 2010. (Wakamatsu et al. 2013)

Japan addressed LPG leakage by allowing the supply of LPG to households only in the form of factory-prepared cylinders. When the installation of large LPG storage tanks was allowed in 1997, it was permitted only on the ground or underground, but prohibited on rooftops because no appropriate methods have been established that prevent the escape of LPG liquid remaining in the pipe during the supply process to rooftop tanks.

(2) Measures for black carbon and PM₁₀

Suspended particulate matter, especially the small-sized PM_{2.5}, has been associated with various health problems. Therefore, monitoring systems for PM_{2.5} have been developed in recent years. Among various components of PM_{2.5}, black carbon (BC) is attracting attention today because it is not only a health hazard but also an SLCP related to global warming. Therefore, developing countermeasures against BC has become a pressing issue.

According to the results of WG3, the mean EC (Elemental Carbon; equivalent to BC) concentration at a monitoring station (Merced) in Mexico City was 5.1 µg / m³ (12% of PM_{2.5}). On the other hand, the EC concentration in Guadalajara was 1.3 µg / m³ (6%), and that in Osaka, Japan, was 1.0 µg / m³ (6%). Therefore, the concentration and proportion in PM_{2.5} of EC are relatively high in Mexico City.

In Mexico, the primary sources of BC are domestic cooking and heating using bio-fuel (about 50%), and the secondary source is vehicles (about 25%). In urban areas where heavy-duty diesel vehicles are considered to make a large contribution, the installation of PM removal devices (DPF: Diesel Particulate Filter) on diesel vehicles is considered an effective measure.

(i) BC measures from heavy duty diesel vehicles in Japan

In Japan, since a majority of BC is discharged from heavy-duty diesel vehicles, such as trucks and buses in urban areas, exhaust gas regulations for PM from these vehicles have been frequently upgraded, resulting in an emission limit in 2009 of 1/70 of the value in 1994 (1994: 0.7 g/kWh, 1999: 0.25, 2003: 0.18, 2005: 0.027, 2009: 0.01). In addition, in three major metropolitan areas (Tokyo, Osaka, and Nagoya), the NOx / PM Law was enacted as a special measure to reduce both nitrogen oxides and PM in automobile exhaust gas.

As of 2002, new emission standards were applied to vehicles already in use, and exhaust-gas testing was implemented in vehicle inspection (annually for large vehicles). For example, the emission standard of PM for large buses and trucks that are larger than 3.5 tons is 0.49 g/kWh; the diesel vehicles manufactured before 1998 do not meet this criterion. If vehicles do not pass the inspection, they can no longer be used on public roads. As this regulation resulted in a large financial impact on the transportation industry, a national subsidy (subsidy rate: 1/4) was provided for switching to new vehicles and installing DPF within a grace period in order to enforce the law effectively.

(ii) Measures for suspended particulate matter in Monterrey

In Monterrey, PM₁₀ has exceeded the environmental standard significantly and PM_{2.5} has also been high. Therefore, measures for PM are urgently required. From analysis of the monitoring data, PM₁₀ had a tendency to be high in the morning, suggesting that the concentration of PM₁₀ becomes higher right after the human activity begins. It was also high in the winter, as it is thought that the northwesterly wind and less rain characteristic of December and January is related to this high concentration of PM₁₀. As mentioned previously, the concentration of PM₁₀ is considered to be greatly affected by limestone quarries, dust from limestone that piles up on the road, dust from the cement factory, and so on. In addition, because the road conditions are

not very good and drainage is not smooth after the rain, there appears to be room for improvement related to deposited dust on the road.

(3) Measures for evaporative gasoline vapor emission related to photochemical ozone and personal exposure

At gas stations, worker exposure to gasoline vapor as they refuel vehicles is a health concern. Also because VOCs in gasoline vapor are precursors to photochemical air pollution, gasoline vapor recovery is a viable measure in terms of both workplace safety and emission control.

In May 2012, WG4 selected a large gas station in Guadalajara, and using passive samplers, measured exposure to employees who regularly refuel vehicles. At the same time, a reference measurement was conducted at the rooftop of a shopping mall 250 m away from the gas station.

It was found that the concentrations of BTEX (benzene, toluene, ethylbenzene, and xylene) deriving from gasoline were much higher at gas station than at the shopping mall. On the other hand, no notable differences in the concentration for CO or carbonyls due to vehicle exhaust gas were observed. Therefore, employees at the gas station were obviously exposed to gasoline vapor at a high concentration level. In Guadalajara where vapor return system at refueling has not been established, some measure of vapor emission control is desired.

(4) Measures to ensure the accuracy of air monitoring data

(i) The need to ensure the accuracy of monitoring data

In order to devise proper measures based on the correct picture of environmental air pollution, accurate ambient monitoring is essential. Also, the accuracy of monitoring data greatly affects the quality of estimations of the effect of reducing emissions by model simulations and implementing future scenarios.

As there are many factors that cause errors in the data, it is important to properly utilize the equipment for calibrating the sample analysis processes by using standard gases whose quality is of prime importance.

It is especially important to accurately identify the environmental concentrations of VOCs, which are important precursors of photochemical pollution.

(ii) Establishment of standard gas supply system

WG2 measured the environmental VOCs components in Mexico using the calibration system of the Advanced Industrial Science and Technology (AIST), the certification body of the standard gas in Japan. In addition, in collaboration with the Mexico National Metrology Center (CENAM), the production technology has been transferred. As a result, elucidation of the dynamics of VOCs in the ambient air in Mexico has become possible with a high degree of accuracy.

Furthermore, SI traceable standard gases used in Japan for all ambient criteria gases in Mexico have been introduced to CENAM, and traceability and comparability for air monitoring of criteria pollutants have become possible in Mexico. The MoU between INECC and CENAM was updated in February 2015, and supply and use of the standard gas in Mexico has become possible.

Monitoring data will be more reliable, and is expected to contribute to the improvement of air quality in Latin American countries through the transfer of the acquired technology of CENAM, making use of international schemes such as JICA's South-South cooperation.

7. Summary and future direction

The collaborative research project “Joint Research Project on Formation Mechanism of Ozone, VOCs, and PM_{2.5} and Proposal of Countermeasure Scenario” was conducted from 2011 to 2015. Air quality in three urban areas in Mexico—MCMA, GMA, and MMA—was studied intensively using the same experimental and analytical methods in Mexico and Japan. This article described part of the results of the project.

- Trends in the state of air quality over a recent decade were compared among Japanese and Mexican urban areas. In Japan, the ozone concentration has been increasing gradually despite significant decreases in the NO_x and NMHC (non-methane hydrocarbon) concentrations. In Mexico, the ozone concentrations have been increasing in GMA and MMA, but decreasing in MCMA. There are various factors that are considered to be responsible for these different trends, but no definitive explanations exist yet.
- Vertical structures of ozone concentration were investigated using ozonesonde. In MCMA and GMA, the daytime ozone was found to be distributed vertically uniformly in a convective mixing layer that was considerably thicker than found in the neighborhood of Tokyo, Japan. Both in GMA and near Tokyo, ozone concentration maxima were often observed in the mid-troposphere, indicating long-range transport. The ozonesonde results provided valuable validation data for CTM simulations for estimating the effects of modified emissions.
- In MCMA, a considerable reduction in the concentration of VOCs from 2000 to 2012 was confirmed based on the same sampling and analysis methods. However,

the concentration levels of propane and butane in MCMA in 2012 were still much higher than those measured near Tokyo in 1998. A high correlation between concentrations of propane and butane in MCMA indicated that the dominant source of these species was LPG facilities. To establish supply and quality control systems of standard gases in Mexico, Japanese technologies were transferred for CO, SO₂, NO₂, BTEX, and PAMs gases.

- In MCMA, GMA, and MMA, PM_{2.5} was sampled and the components (carbon, inorganic ions, and metals) were analyzed. The proportion of EC was distinctly higher in MCMA, indicating a large contribution of diesel vehicles. In MMA, high proportions of Ca and Fe attributable to the local industries were noted. Otherwise, similar compositions were found in MCMA, GMA, and MMA. Diurnal behaviors of PM_{2.5} concentrations were compared among major cities of the world. In MCMA, PM_{2.5} peaked at mid-day, indicative of photochemical smog, whereas in Beijing, PM_{2.5} peaked at night, the reasons for which are not well understood.
- The project proposed countermeasures against air pollution, which are to be incorporated in the ProAires in Mexico. The proposals include VOCs emission control to mitigate photochemical ozone, reduction of black carbon from diesel vehicles, reduction of primary coarse particles in MMA, gasoline vapor control at gas stations for both reduction of photochemical ozone and toxic exposure to workers, and improvement of air-quality monitoring.

In closing this article, a future direction is discussed briefly in relation to global climate change. Photochemical ozone and black carbon (or elemental carbon: EC) are

pollutants that cause both global warming and human health problems (Akimoto et.al. 2015). These pollutants are called SLCP because of their relatively short lifetimes of less than several months in the atmosphere. Global warming has positive feedback on the production of photochemical ozone. This is especially important in megacity areas of the world. Because VOCs and NO_x are major precursors of photochemical ozone in the troposphere and the NO_x concentration is highly correlated with the EC concentration, they are regarded as implicit SLCPs.

Photochemical ozone, EC (major SLCP), and NO_x and VOCs (implicit SLCP) need to be controlled to improve the regional and global atmospheric environmental quality. The relationship between regional and global atmospheric environments is shown in Figure 17.

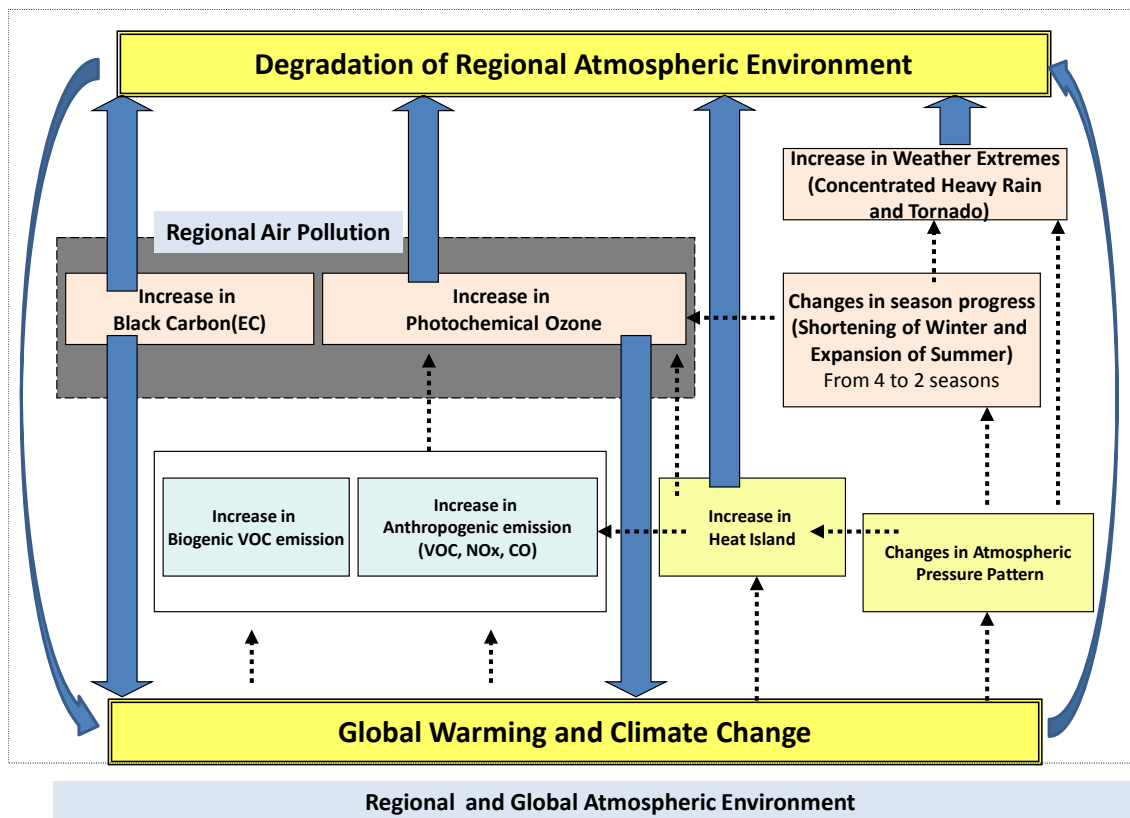


Figure 17. Relationship between regional and global atmospheric environments

Source: Author

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Abstract (in Japanese)

要約

光化学オキシダントとブラックカーボン、大気汚染と地球規模の気候変動の両方にとって重要な物質である。これらの2つの汚染物質はSLCP (Short-Lived Climate Pollutants) と呼ばれているものである。条件の異なる大都市間の国際比較研究は、SLCPの形成メカニズムを明らかにする上で有効な方策といえる。

光化学オキシダント、揮発性有機化合物 (Volatile Organic Compounds (VOC))、PM2.5を中心とした日本とメキシコの大都市圏における比較研究は、大気汚染の傾向分析と、光化学オキシダントや気象パラメータの鉛直分布測定などのフィールド測定に基づいて実施された。本研究では、得られた科学的データに基づく対策シナリオを提案している。

地域および地球規模の大気環境を改善するためには、光化学オキシダント及びEC (Elemental Carbon) といった主要なSLCP、また、NO_x (窒素酸化物) およびVOCといった潜在的なSLCPを管理する必要がある。

本研究の成果として、日本では、国境を越えた大気汚染の影響が大きいため、アジア全域を含む対策が必要であり、メキシコでは、エネルギーシフトやディーゼル排ガス規制などのVOC規制が効果的であることが示されている。本研究により得られた知見は、Mexico City、Guadalajara、Monterreyのメキシコの3大都市圏のProAire (Program for Air Quality Improvement) の策定や評価への活用が期待される。