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Review Paper

Visibility Impairment by Atmospheric Fine Particles in an Urban Area

Young J. Kim* and Kyung W. Kim

Advanced Environmental Monitoring Research Center (ADEMRC) Department of Environmental Science and Engineering Kwangju Institute of Science and Technology (K-JIST), Kwangju, Korea

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Abstract

Visibility impairment in an urban area is mainly caused by airborne fine particulate matters. Visibility in a clean air environment is more sensitive to the change of $PM_{2.5}$ particle concentrations. However, a proportionally larger reduction in fine particle concentration is needed to achieve a small increment of visibility improvement in polluted areas. Continuous optical monitoring of atmospheric visibility and extensive aerosol measurements have been made in the urban atmosphere of Kwangju, Korea. The mean for fine particulate mass from 1999 to 2002 at Kwangju was measured to be $23.6 \pm 20.3\ \mu g/m^3$. The daily average seasonal visual range was measured to be 13.1, 9.2, 11.0, and 13.9 km in spring, summer, fall, and winter, respectively. The mean light extinction budgets by sulfate, nitrate, organic carbon, and elemental carbon aerosol were observed to be 27, 14, 22, and 12%, respectively. It is highly recommended that a new visibility standard and/or a fine particle standard be established in order to protect the health and welfare of general public. Much more work needs to be done in visibility studies, including long-term monitoring of visibility, improvement of visibility models, and formulating integrated strategies for managing fine particles to mitigate the visibility impairment and climate change.

Key words : Visibility, Fine particulate matter, Light extinction, Human health, Climate

1. INTRODUCTION

Generally, "visibility" has been defined as "the greatest distance at which an observer can just see a black object viewed against the horizon sky". An object is usually referred to as the threshold contrast when the difference between the brightness of the sky and the brightness of the object is reduced to such a degree that an observer can just barely see the object (Malm, 2000). Whether we define visibility in terms of visual range or in terms of some parameter more closely related to how visitors perceive a visual resource, the preservation or improvement of visibility requires an understanding of what constituents in the atmosphere impair visibility as well as the origins of those constituents. Figure 1. illustrates the three processes affecting visibility.

In addition to their effects on human health, fine particulate matter in the atmosphere contributes to visibility impairment. With Ambient $PM_{2.5}$ the major source of visibility impairment results from direct particle

* Corresponding author.
Tel : +82-(0)62-970-3401, E-mail : yjkim@kjist.ac.kr

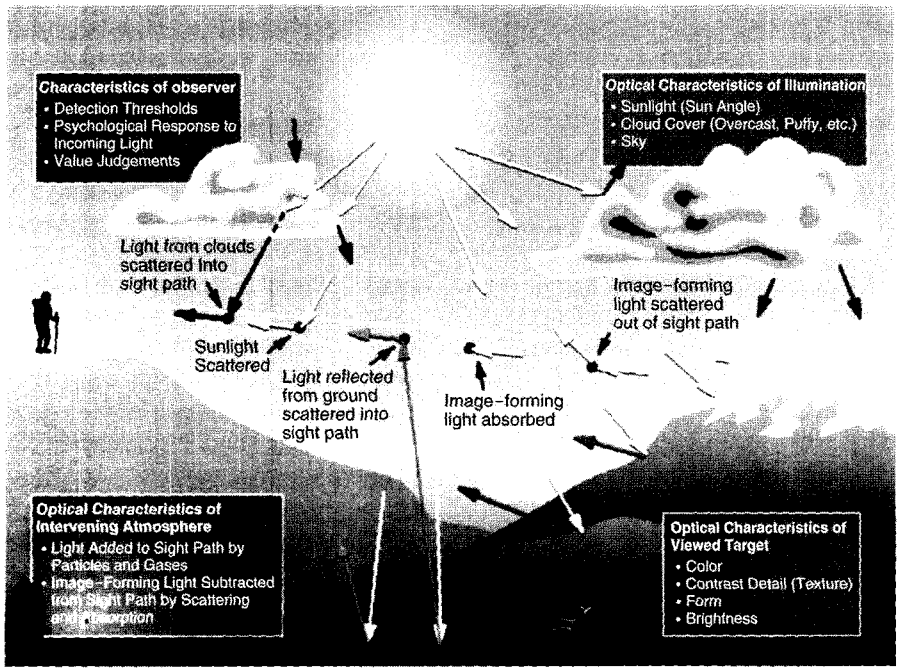

Fig. 1. Illustration of the three processes by which particles and gases in the atmosphere affect visibility-the scattering of image-forming light, the absorption of image-forming light, and the scattering of light into the sight path (Malm, 2000).

emissions (carbon and soil dust) and secondary particles (generated by atmospheric reactions of precursor gas emissions). Ambient PM concentration is also influenced by meteorology. The major precursor gases are sulfur dioxide, nitrogen oxides, ammonia, and volatile organic compounds (VOCs). Ambient PM mass is thus a mixture composed mostly of sulfate, nitrate, ammonium, organic carbon, black carbon and soil dust (Malm *et al.*, 1996). The degree of visibility impairment is closely linked to PM concentration, chemical composition, size distribution and ambient relative humidity.

The impact of air pollution by anthropogenic sources on human health and visibility has been studied for decades. Many analyses have been conducted worldwide not only to estimate the health benefits from air pollution abatement but also to identify scientific and technical understandings of how the air pollutants impair visibility (Clancy *et al.*, 2002; Kim *et al.*, 2001a; Wilson and Suh, 1997; Johnson *et al.*, 1990; Larson *et al.*, 1988; Dzubay *et al.*, 1982). It is known that visibility impairment can result directly or indirectly from particle emission and is dominated by fine particulate matter. Recently, the North American Research Strategy for Tropospheric Ozone (NARSTO) published a PM Science Assessment Report (NARSTO, 2003), which summarized the state of scientific understanding of atmospheric PM as it relates to policy questions and managing the implementation of new PM standards and goals. The Ministry of Environment in Korea recently funded a multi-year study, Fine Particles and Visibility Impairment in the Seoul Metropolitan Area (KOSAE, 2003), which consists of intensive measurements, modeling studies, and analysis work to formulate policy recommendations. One major goal of this study is to build the capacity for forecasting visibility impairment.

2. RULES AND REGULATIONS

Visibility has long played a role in air quality regulation and measurement. The first recorded law to control air pollution was issued in 14th century London and prohibited the use of coal based on a high correlation between black chimney plumes and reduced visibility, soot deposits, and respiratory distress (Brimblecombe, 1987). The U.S. Congress passed the first Air Pollution Control Act in 1955 identifying air quality as a national problem and initiating research for understanding it. This 1955 law included no measures for reducing emissions, quantifying human exposure, or examining excessive haze (Fleming and Knorr, 2002). The 1970 CAA (Clean Air Act) initiated emission reductions in areas with excessive concentrations of sulfur dioxide (SO_2), nitrogen dioxide (NO_2), CO, O_3, lead (Pb), and suspended particulate matter (PM). The U.S. EPA set a National Ambient Air Quality Standards (NAAQS) for airborne particles in 1971. Over time, the standards have evolved with an increasing focus on size. Current $PM_{2.5}$ standard requires that the 3-yr average of the 98th percentile of 24-hr average concentration must not exceed 65 $\mu g/m^3$, and the 3-yr average of annual arithmetic mean concentration must not exceed 15 $\mu g/m^3$. California's new standard has more stringent than national $PM_{2.5}$ standards (less than 12 $\mu g/m^3$) and visibility standard (less than 17 km). Korea's current PM standard sets ambient PM_{10} concentration not to be exceeded 150 $\mu g/m^3$veraged over 24 hours and 70 $\mu g/m^3$ averaged over a year. The Canada Wide Standards (CWS) for $PM_{2.5}$ is 30 $\mu g/m^3$ averaged over 24 hours to be achieved by 2010 (NARSTO, 2003). The Clean Air for Europe (CAFE) set a limit value for PM_{10} of 40 $\mu g/m^3$ (24-hr average), not to be exceeded more than 35 times a calendar year as of 2005, and a goal of 20 $\mu g/m^3$ (24-hr average) by 2010. The current PM standard for the United Kingdom sets a limit value for PM_{10} of 40 $\mu g/m^3$ (24-hr average). Japan has a suspended particulate matter (SPM) standard, which is the daily average of hourly values, shall not exceed 100 $\mu g/m^3$, and hourly values shall not exceed 200 $\mu g/m^3$. The existing PM standards of several nations are summarized in Table 1.

Table 1. Existing PM and visibility standards

Year Adopted	PM_{10}		$PM_{2.5}$		Visual Range
	24-hr	Annual	24-hr	Annual	
	($\mu g/m^3$)				(km)
1997 US EPA	150	50	65	15	
1999 CAFE (2005)[a]		40			
1999 CAFE (2010)[a]		20			
2000 CWS			30		
2000 UK		40			
1973 Japan	100, 200[b]				
2001 Korea	150	70			
2001 Arizona					VR = 40 km 250 days/year b_{ext} = 76 Mm^{-1} when RH < 70%
1995 Colorado					51.2 km b_{ext} = 230 Mm^{-1} when RH < 70%
2002 California	50	20		12	17 km

[a]: Attainment target date: 2010 goal subject to review
[b]: Daily average of hourly values and hourly average values

The 1977 CAA (Clean Air Act) amendments addressed visibility in non-urban areas under the Prevention of Significant Deterioration of Air Quality (PSD). The PSD program intends to protect so-called Class I areas which includes national parks, national wilderness areas, national monuments, national seashores, and other areas of special national or regional natural recreational, scenic, or historic value that remain relatively pollution-free from becoming more polluted. The 1990 CAA amendments added a section entitled "Visibility". This section requires an expansion of visibility monitoring networks, assessment of source contributions to visibility, adaptation of regional air quality models for visibility source attribution, and studies of atmospheric chemistry and physics related to haze formation and transport (Mathai, 1995). The 1999 U.S. EPA's Regional Haze Rule is possibly the most ambitious and the most stringent air quality goal ever promulgated. It intends that U.S. anthropogenic emission be reduced to

a visibility that is not noticeably worse than it would be under 'natural conditions' in the Class I areas by year 2065 (Watson, 2002).

3. PM's IMPACT ON HUMAN HEALTH, VISIBILITY, AND CLIMATE

In principle, particles with an aerodynamic diameter of less than 5 μm follow the motion of the air and may remain suspended for days in ambient air. Larger particles, with an aerodynamic diameter of greater than 10 μm, are removed within a matter of hours either by settling or by rain. There is increasing scientific and medical evidence that exposure to fine ($PM_{2.5}$) and ultra fine (smaller than 0.1 μm) particulate matter could have relatively more significant health implications than exposure to larger particles.

The particle deposition efficiencies shown in Figure 2 have been corrected to account for inhalability. Inhalability is the sampling efficiency of the nose and mouth for particulate matters (Phalen *et al.*, 1986). For 10 μm particles, inhalability is about 77 percent and for $PM_{2.5}$ inhalability is greater than 90 percent (Vincent, 1999). Here it is important to note that different, size-dependent ranges of aerodynamic behavior exist. The deposition of larger particles is influenced mainly by inertial forces and gravitational settling, while deposition of particles less than about 0.5 μm diameter are influenced by Brownian diffusion. Consideration of deposition data of this kind as well as the size distribution of ambient PM was a major factor leading to the establishment of $PM_{2.5}$ as indicator metrics for human health impacts.

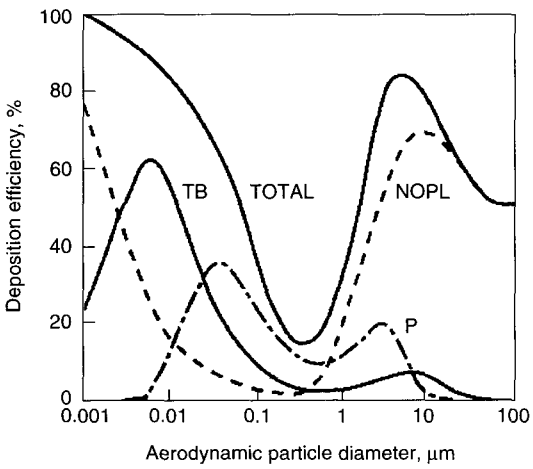

Fig. 2. Particle deposition in the major regions of the human respiratory tract during normal respiration corrected for size dependent inhalability (NOPL, naso-oro-pharyngo-laryngea). region; TB, tracheo-bronchial region; and P, pulmonary region). Developed from the National Council on Radiation Protection Model (NCRP, 1997) by Phalen (2002).

Acute exposure studies linked to indices of acute morbidity or mortality provide little information about how air pollution affects longer-term morbidity or mortality rates or the potential role of air pollution in the process of inducing chronic disease. Chronic exposure studies evaluate the effects of ambient exposure that persists for long periods of time as well as the cumulative effects of repeated acute exposure episodes. Chronic exposure has been reviewed by Pope and Dockery (1999), CEPA/FPAC (1999), California EPA (2002) and the U.S. EPA (2002). Key results from the studies are shown in Table 2. In the Six Cities Study, excess mortality associated with $PM_{2.5}$ was 26 percent greater for the most polluted city compared to the least polluted city. In the American Cancer Society Study (ACS) study, the excess mortality was 15 percent and 17 percent greater for the most polluted compared to the least polluted area based on sulfate and $PM_{2.5}$, res-

Table 2. Comparison of mortality risk ratios for air pollution from the six cities and ACS prospective cohort studies (Pope and Dockery, 1999).

Cause of death	Particulate air pollution (Most vs least polluted city)		
	Six cities ($PM_{2.5}$)	ACS ($PM_{2.5}$)	ACS (SO_4^{2-})
All	1.26 (1.08~1.47)	1.17 (10.9~1.26)	1.15 (1.09~1.22)
Cardiopulmonary	1.37 (1.11~1.68)	1.31 (1.17~1.46)	1.26 (1.16~1.37)
Lung cancer	1.37 (0.81~2.31)	1.03 (0.80~1.33)	1.36 (1.11~1.66)
All others	1.01 (0.79~1.30)	1.07 (0.92~1.24)	1.01 (0.92~1.11)

pectively, as indicators. The WHO Guidelines for Air Quality report presents the current knowledge on exposure-response relationships with respect to particles smaller than 10 μm (WHO, 2000). The most important finding from recent health effect studies is that no thresholds level exists between ambient PM concentrations and health effect as shown in Figure 3 (Schewela, 2003).

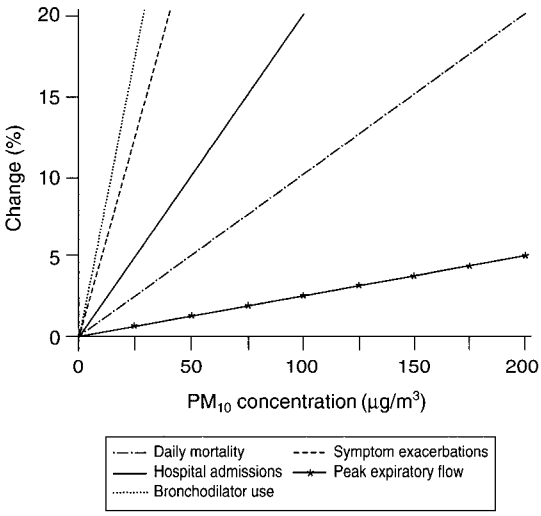

Fig. 3. Relationship of PM_{10} with different health effect indicators (Schewela, 2003).

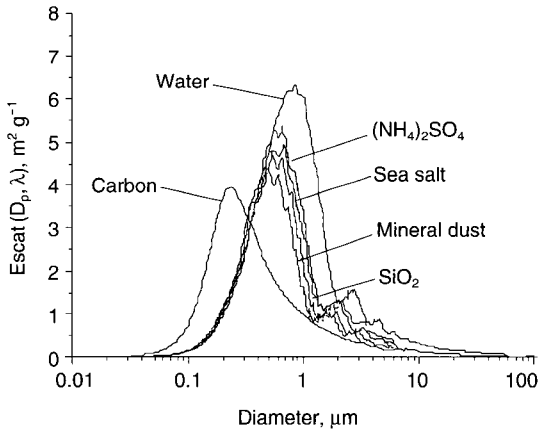

Fig. 4. Mass scattering efficiencies of homogeneous spheres of $(NH_4)_2SO_4$, NaCl, mineral dust, carbon, silica, and water droplet at λ= 550 nm (Kim *et al.*, 2001a).

In addition to their effects on human health, fine particulate matter in the atmosphere contributes to impairment of visibility more than larger particles and also affects the climate change. Optically, particulate matter interferes with visibility by absorbing or scattering visible light as shown in Figure 1. For example, BC absorbs light and sulfate is highly reflective. Light scattering is roughly proportional to the mass concentration of fine particles, while light absorption is roughly proportional to the mass concentration of the light-absorbing species such as BC. The relationship between the light extinction coefficient and the PM mass concentration depends on the size and chemical composition of the particles, and the ambient relative humidity. The effectiveness of particles at scattering light depends on their sizes. Particles whose diameters are approximately the same as the wavelength of light are the most efficient, in terms of light scattering per unit mass (the scattering efficiency; E_{scat}). As shown in Figure 4, particles whose diameters are between 0.3 and 1.0 μm tend to scatter the most light per unit mass (Kim *et al.*, 2001a). Since the human eye is most sensitive to light with wavelengths in the vicinity of 0.5 μm, these effects are very evident to human observers. Larger particles, such as those in the coarse mode with diameters between 2.5 and 10 μm, are not very efficient at scattering light. Particles with diameters in the general vicinity of 0.5 μm also account for most of the mass in the fine particle fraction ($PM_{2.5}$). As a result, the $PM_{2.5}$ mass concentration corresponds to those particles that are most efficient at scattering light.

Extensive measurements of the fine particulate matter species that cause light extinction have been carried out in rural areas of the United States for the past decades. Figure 5 shows the distribution of average light extinction over the United States, calculated from particle measurements made by the IMPROVE (Interagency Monitoring of Protected Visual Environments) network at national parks and wilderness areas (Malm, 2000; Sisler, 1996). This figure shows the dramatic difference between the relatively impaired visibility in the

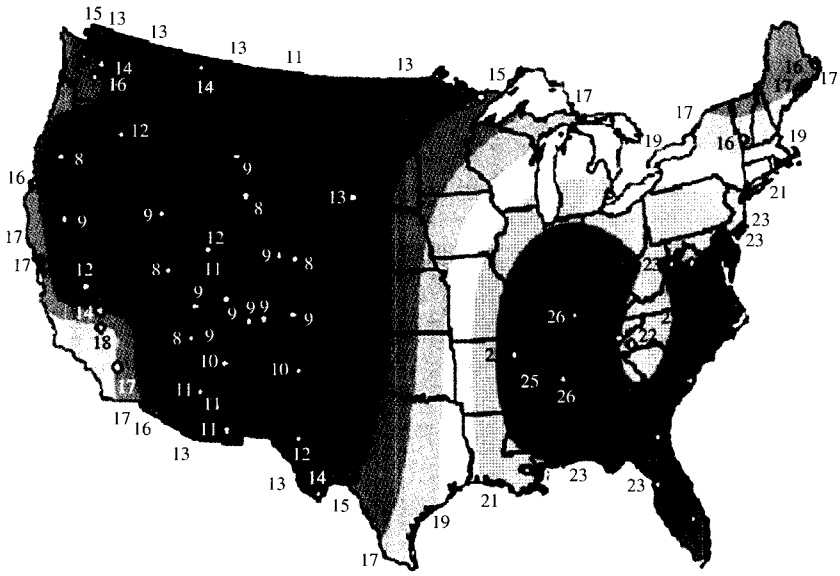

Fig. 5. Distribution of average light extinction in national parks and wilderness areas of the United States, in units of deciviews, as calculated from particulate matter measurements in the IMPROVE network (Malm, 2000).

more humid and more populated East area and the much clearer visibility in the arid and relatively unpopulated intermountain West. Visibility along the Pacific Coast is again poorer because of increased population density and, in the Northwest, higher humidity. Urban areas normally have greater particulate matter concentrations and thus the average visibility there can be expected to be worse than that shown in Figure 5.

In Korea, extensive optical visibility monitoring has been conducted in the urban area of Kwangju from 1999 to 2000 (Kim *et al.*, 2001b) and during ACE-Asia intensive observation period 2001 (Kim *et al.*, 2002b). Additional measurements have been made in the rural area of Anmyon in 2000 (Kim *et al.*, 2003) and in the urban area of Seoul since July 2002 (KOSAE, 2003).

The major components of fine particulate matter affecting visibility impairment are sulfate and nitrate compounds, organics, elemental carbon (EC), and soil dust. It is found that characteristics of light extinct budget (% contribution of each species to the total light extinction) for fine particulate matter are quite different among regions and nations as shown in Table 3. Sulfate and organics species are the single largest contributors to visibility reduction at all monitoring sites. In the United States, sulfate species make up over 70% of the measured visibility impacts, they are responsible for between 20 and 30% in other regions. In the eastern United States, organics contribute between 10 and 15% of estimated extinction. In other regions, organics contribute about 20 to 40% of the extinction. EC contributes about 4 to 6% to extinction in the eastern United

Table 3. Comparison of light extinction budgets of major components for fine particulate matter affecting visibility impairment among nations and regions.

Chemical component	U.S.A[a]		Korea		
	West	East	Kwangju[b]	Anmyon[c]	Seoul[d]
	(%)				
Sulfate	30~40	60~75	34	39	24
Nitrate	30~40	3~10	17	12	19
Organics	20~40	10~15	29	41	30
EC	7~15	4~6	15	1	23

[a]: (Malm, 2000), [b]: (Kim *et al.*, 2001b)
[c]: (Kim *et al.*, 2003), [d]: (KOSAE, 2003)

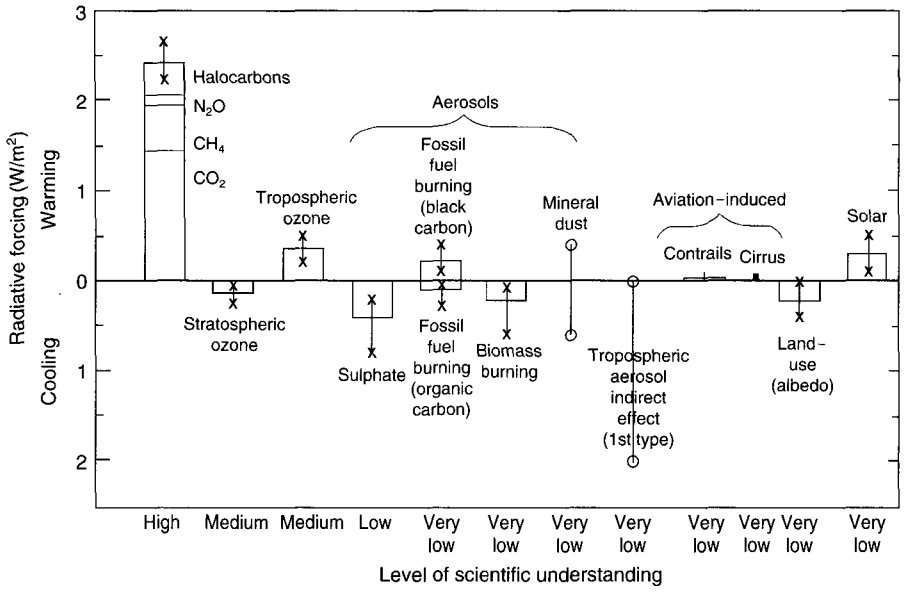

Fig. 6. Global, annual mean radiative forcings (W/m²) for the period from pre-industrial (1750) to the present. The height of each rectangular bar denotes a best estimate value while its absence denotes no best estimate is possible. The vertical line about the rectangular bar with "x" delimiters indicates an estimate of the uncertainty range. A vertical line without a rectangular bar and with "o" delimiters denotes a forcing for which no central estimate can be given owing to large uncertainties. A level of scientific understanding is accorded to each forcing, with denoting high, medium, low and very low levels, respectively. The figure shows that the dominant effect of atmospheric particles is a negative forcing (IPCC, 2002).

States, about 7 to 15% in the western United States, and about 12 to 23% in the urban areas of Korea, respectively.

Particles in the atmosphere influence the balance between the solar radiation that reaches the earth's surface. This process occurs both directly (direct aerosol forcing), as the particles scatter and absorb incoming solar and outgoing infrared radiation, and indirectly (indirect aerosol forcing), as the particles influence cloud formation and precipitation, which in turn affect the radiation balance. Changes in the radiation balance due to changes in the amount and type of aerosol, natural or anthropogenic, are believed to contribute to climate change through a net cooling of the atmosphere (IPCC, 2002).

Figure 6 summarizes the current understanding and level of uncertainty attached to the influences of greenhouse gases and atmospheric particles over the last 250 years. Carbon dioxide and the other well-mixed greenhouse gases represent the largest forcing at $+2.4\ W/m^2$ with a high level of scientific understanding. Net increases in particulate matter concentrations are estimated to counter this forcing (i.e., they contribute to cooling) by anywhere from a negligible amount to more than about $-2.5\ W/m^2$. As this large range suggests, the uncertainties relating to aerosol radiative forcing remain very large, mainly because the values rely to a great extent on estimates from global modeling studies that are difficult to verify (NARSTO, 2003).

4. VISIBILITY THEORY

Visibility impairment is primarily due to the interaction of light with particles in the atmosphere; gaseous pollutants usually play a small role. Particles interact

with light by two important mechanisms: they can absorb the light, and scatter the light in a direction different from that of the incident light (Friedlander, 1977). Despite the diversity of these physical phenomena, a variety of available techniques make it possible to characterize the optical properties of the atmosphere and to identify and quantify the pollutants that can affect visibility degradation.

The extinction coefficient, b_{ext}, is an important measure of atmospheric transparency and is a measure most directly related to the composition of the atmosphere. It is the measure of the fraction of light energy, dE, lost from a collimated beam of energy, E, in traversing a unit thickness of atmosphere dx: $dE = -b_{ext}\,Edx$. The extinction coefficient has dimensions of inverse length (e.g., Mm^{-1}). The extinction coefficient comprises four additive components:

$$b_{ext} = b_{scat} + b_{abs} = b_{Ray} + b_{sp} + b_{ag} + b_{ap} \quad (1)$$

where b_{Ray} is light scattering by gas molecules. Gas scattering is almost entirely attributable to oxygen and nitrogen molecules in the air and often creates natural "blue-sky". It is obviously unaffected by pollutant gases and is 12 Mm^{-1} at the wavelength of 550 nm at sea level. b_{sp} is light scattering by particles. This scattering usually is dominated by fine particles, because particles at the size range of 0.1 ~ 1.0 μm have the greatest scattering efficiency as shown Figure 4. Many airborne particulate pollutants are in this size range. b_{ag} is light absorption by gases. Nitrogen dioxide (NO_2) is the only common atmospheric gaseous species that significantly absorbs light in the visible spectral region. b_{ap} is light absorption by particles. Absorption arises entirely from elemental carbon particles.

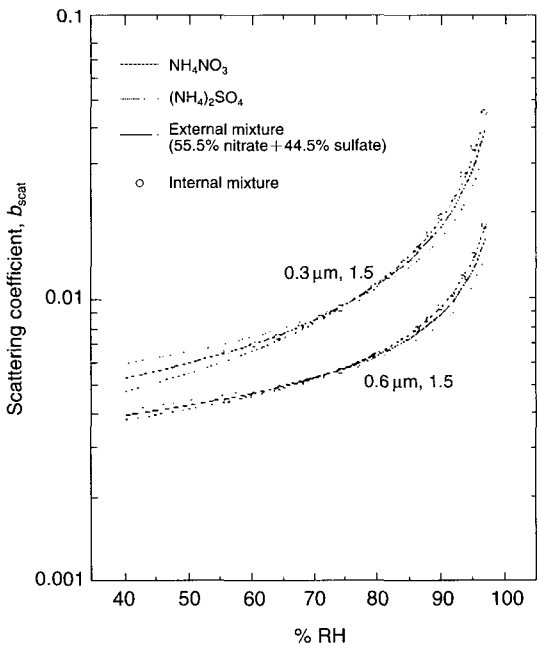

Fig. 7. Effect of relative humidity on light scattering by mixtures of ammonium nitrate and ammonium sulfate (Tang, 1996).

The relationship between the light extinction coefficient and the PM mass concentration depends on the size and chemical composition of the particles, and the ambient relative humidity. Particles that contain SO_4^{2-} and NO_3^-, along with other soluble salts (NaCl), have long been known to absorb liquid water, thereby growing into size ranges that scatter more incident light (Tang, 1996). The growth pattern varies for different compounds: fairly smooth and continuous for sulfuric acid (H_2SO_4) but with abrupt, step-function increases at ~80% RH for $(NH_4)_2SO_4$, ~62% RH for NH_4NO_3, and ~75% RH for NaCl a phenomenon known as deliquescence. Some organics also may absorb water, but few experiments have shown this to be very substantial. Saxena and Hildemann (1996) identified many water-soluble organic compounds that have the potential to be in the particle phase and absorb water.

5. SIGNIFICANCE OF SECONDARY ORGANIC AEROSOL (SOA)

Most of the ambient $PM_{2.5}$ mass usually originates as gases (SO_2, NH_3, NO_x, VOCs) and, through physicochemical processes, is transferred to the condensed phase. Emitted aerosol particles (black carbon, organics, dust, sea salt, fly ash) have both a direct and an indirect role in the formation of secondary $PM_{2.5}$. Pri-

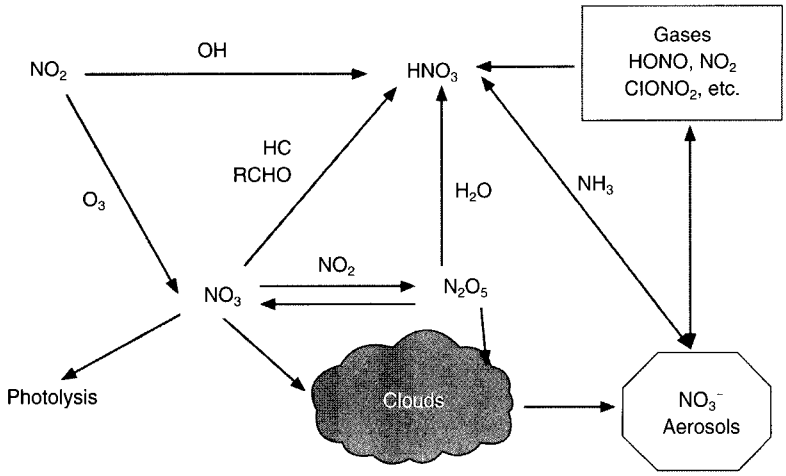

Fig. 8. Schematic of the formation of gas phase nitric acid and particulate nitrate in the atmosphere. Formation of particulate nitrate from nitric acid requires either reaction with gas-phase ammonia or sea salt or alkaline dust (NARSTO, 2003).

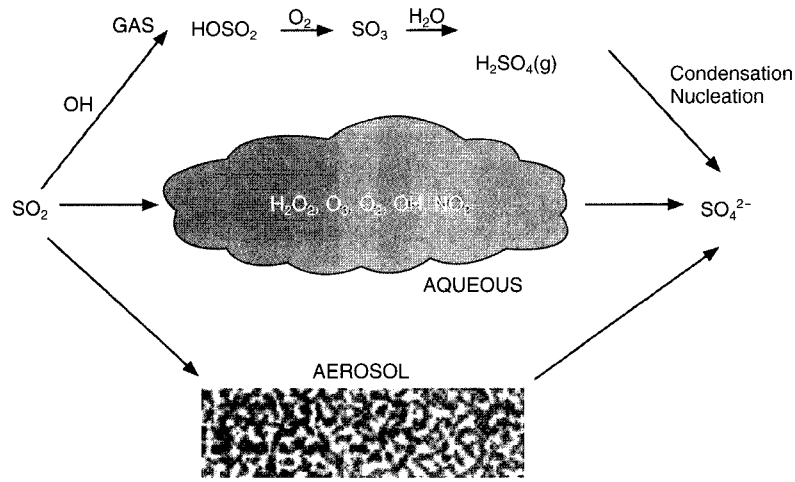

Fig. 9. Schematic of the three pathways (reaction in the gas, cloud, and condensed phases) for the formation of sulfate in the atmosphere. Some of the reactions in the aqueous and condensed phases are catalyzed by trace amounts of metals (NARSTO, 2003).

mary particles can provide the reaction sites for the formation of new aerosol material. The formation of nitrates by the reaction of sea salt or alkaline dust with HNO_3 vapor is an example of such reactions (Seinfeld and Pandis, 1998). Other important reactions include the formation of SO_4^{2-} on alkaline particles and the oxidation of primary organic aerosol compounds to more hydrophilic ones (Zhang *et al.*, 1993). This fraction of the $PM_{2.5}$ mass is called secondary organic aerosol (SOA).

Nitrates are formed from the oxidation of NO and NO_2 either during the daytime (reaction with OH) or during the night (reactions with ozone and water) (Wayne, 1991). Nitric acid is continuously transferred

between the gas and the condensed phases (condensation and evaporation) in the atmosphere. It naturally prefers the gas phase when left alone, but reactions with gas-phase NH_3, sea salt, and dust result in its transfer to the condensed phase (Seinfeld and Pandis, 1998). The formation of aerosol NH_4NO_3 is favored by the availability of NH_3, low temperatures, and high relative humidity as shown in Figure 8 (NARSTO, 2003). The formation of SO_4^{2-} from the oxidation of SO_2 is an important process in most areas of the world. The H_2SO_4 formed by three different pathways; the oxidation of SO_2 in the gas phase by the hydroxyl radical, OH, the dissolution of SO_2 in cloud, fog, or rain-water, and the oxidation of SO_2 in reactions in the water of the aerosol particles as shown in Figure 9 (NARSTO, 2003). The sulfates formed from those pathways reacts readily with NH_3 to form ammonium sulfate, $(NH_4)_2SO_4$. If there is insufficient NH_3 present to fully neutralize the available H_2SO_4, part of the particles exist as ammonium bisulfate, NH_4HSO_4 and the associated particles are acidic. In extreme cases, sulfates can exist in particles as H_2SO_4. During the past twenty years, much progress has been made in understanding the first two pathways, but some important questions still remain about the smaller third pathway.

The organic component of ambient particles is a complex mixture of hundreds or even thousands of organic compounds. These organic compounds are either emitted directly from sources (primary organic aerosol) or can be formed in-situ by condensation of low-volatility hydrocarbon-oxidation products (secondary organic aerosol). As organic gases are oxidized with species such as OH, ozone, and NO_3, their oxidation products accumulate. Some of these products have low volatilities and condense on available particles. The ability of a given volatile organic compound (VOC) to produce SOA during its atmospheric oxidation depends on four factors as shown in Figure 10: its atmospheric abundance, its chemical reactivity, the availability of oxidants, and the volatility of its products (NARSTO, 2003). Despite significant progress that has been made in understanding the origins and properties of organic particles, it remains the least understood chemical component of $PM_{2.5}$. Its chemical complexity and the difficulty in unraveling its chemical composition, even by state-of-the-art methods, are limiting progress. Our understanding of the contribution of primary, secondary, biogenic and anthropogenic organic

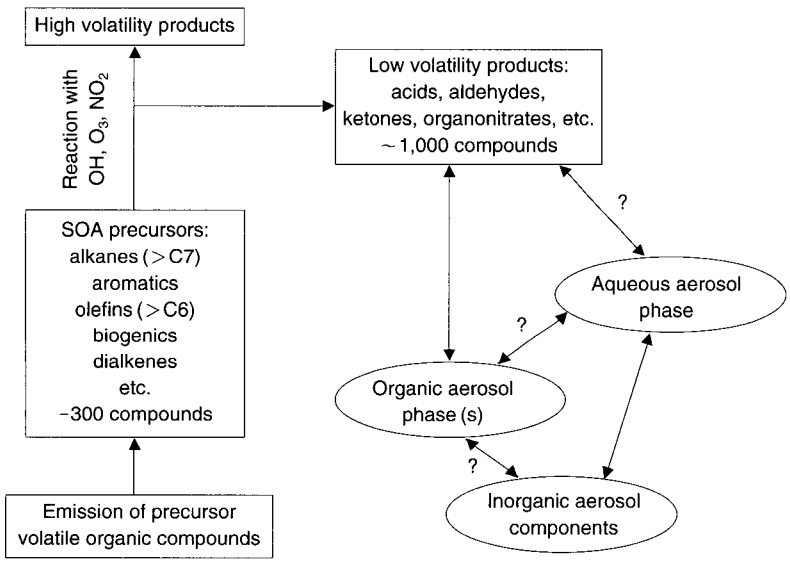

Fig. 10. Schematic process of the formation of secondary organic aerosols in the atmosphere (NARSTO, 2003).

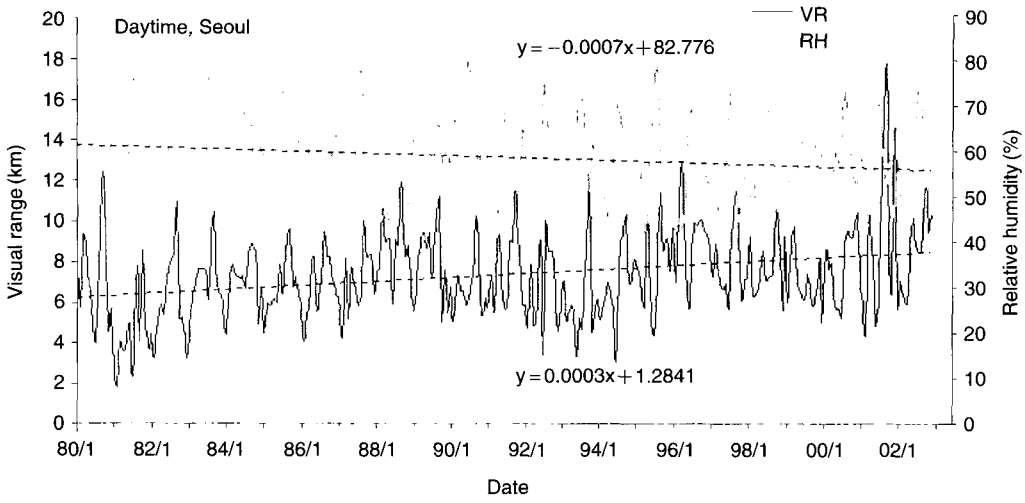

Fig. 11. Visibility trend in Seoul in the 1980~2002 (KOSAE, 2003).

$PM_{2.5}$ compounds should be enhanced. The development of methods to speciate particle-phase organic compounds in atmospheric and source samples over the past decade has led to the development of molecular marker source apportionment models (Schauer and Cass, 2000) that use unique organic compounds emitted from sources as tracers to quantify the contribution of air pollution to atmospheric concentrations of carbonaceous particulate matters and the mass of particulates.

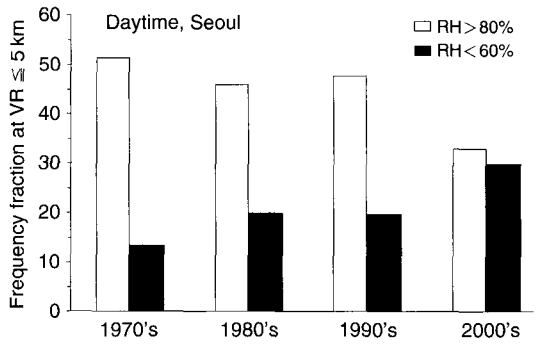

Fig. 12. Variation of frequency fraction of visual range observed below 5km under both RH>80% and RH<60% conditions in Seoul in the 1970s~the 2000s (KOSAE, 2003).

6. CHARACTERISTICS OF VISIBILITY IMPAIRMENT IN KOREA

6. 1 Visibility Trends in Korea

The spatial and temporal trends as well as the man-made cause of atmospheric haze have been paid considerable attention from Korean researchers (KOSAE, 2003; Ghim *et al.*, 2002; Lee *et al.*, 1996; Park *et al.*, 1994). Visibility trends in Seoul have been reviewed in this study for the period of the 1970~2002. Visibility measurements were based on human eye observations. Visual range observations (prevailing visibility) have operated at meteorological stations by the Korea Meteorological Administration (KMA) since 1940. Visual range has increased gradually from 6.2 km to 7.9 km in Seoul over past 20 years as in Figure 11. Relative humidity has, however, decreased during the same period mainly due to temperature increase. Figure 12 shows decadal variations in the frequency fractions of the visual range observed below 5 km under both RH> 80% and RH<60% conditions in Seoul in the 1970s~ the 2000s. This result reveals that visibility impairment occurred mainly under high relative humidity conditions in the 1970s but more frequently under low relative humidity condition in the 2000s. Under dry condi-

tion, the frequency of visual range observed below 5 km increased approximately double of the 2000's from the 1970's. The visibility impairment of the 1970's in Seoul might be mainly affected by fog, mist, or rain in which condition relative humidity was higher than 80%. However, the serious haze of the 2000's was not only caused by the effect of relative humidity, but also resulted from the particulate matter under dry conditions, which might have increased over the decades.

6. 2 Monitoring Methodology

Visibility is a parameter closely related to air quality, which represents the status of the surrounding air in a close relation with human health and welfare. Although there have been much improvements in the past decades to reduce the emission of primary air pollutants in Korea, the degree of visibility degradation in urban areas show little signs of waning. Visibility impairment, which results from the scattering and absorption of light by particles and gases in the atmosphere, is considered a direct result of air pollution (Kim *et al.*, 2001b). Continuous visibility monitoring was performed in the city of Kwangju for three years beginning 23 May 1999. For continuous monitoring of atmospheric visibility in the city of Kwangju, Korea, a transmissometer system consisting of a transmitter and a receiver was installed at a distance of 1.91 km across the downtown of Kwangju, Korea. At the transmitter site a nephelometer and an aethalometer were also installed to measure the scattering and absorption coefficients of the atmosphere, respectively. In this study, both aerosol sampling and visibility monitoring was seasonally and regularly conducted in order to investigate the extent of visibility reduction caused by each of the atmospheric constituents such as sulfate, nitrate, organic, carbon, and soil particles as well as to identify optical, physical, and chemical properties of aerosol observed in Korea.

Optical monitoring provides a continuous quantitative measure of ambient light extinction coefficient representing visibility conditions. Optical monitoring at Kwangju included continuous measurements of b_{ext} (light extinction coefficient), b_{scat} (light scattering coefficient) and b_{abs} (light absorption coefficient) using a LPV-2 long-path transmissometer, a Belfort model 1597 integrating nephelometer and a MageeScience model AE-14U aethalometer. Water vapor in the air can affect the growth of hygroscopic aerosols and thus affect the visibility. Meteorological data collected at the Kwangju Meteorological Office at the receiver site was used for data analysis. The amount of soot in the air affecting the light absorption was determined from the black carbon concentration measurement by the aethalometer. The monitoring methods and measurement parameters of the optical monitoring instruments are summarized in Table 4.

Besides continuous optical monitoring, intensive aerosol monitoring was also carried out seasonally, 12 ~17 and 17~22 July 1999, 14~18 Sep. and 23~26 Nov. 1999, 17~22 Jan. and 21~26 Feb. 2000, and 22 ~29 May 2000, respectively. Additional sampling was conducted during the Asian Dust storm periods of 23 ~30 March and 7~9 April 2000. There were simultaneous intensive sampling periods carried out over the subject areas in both Kwangju and Anmyon during the summer, 1~10 August 2000. During the ACE-Asia IOP (Intensive Observation Period), 22 March~4 May 2001 aerosol measurements were taken in Kwangju. 12 -hour diurnal routine aerosol sampling was also conducted every sixth day for one year from 7 May 2001 to 29 February 2002. Two 12-hour aerosol samples were collected 6 AM and 6 PM each day to investigate

Table 4. Measurement parameters of optical monitoring at Kwangju, Korea.

Monitoring instrument	Measurement parameter	Sampling frequency	Averaging interval
Transmissometer	Total light extinction coefficient	Continuous (16-min integration & 20-min cycle)	1 minute
Nephelometer	Light scattering coefficient	Continuous	1 minute
Aethalometer	Light absorption coefficient	Continuous	1 minute

Table 5. Aerosol sampling methods and analytical methods.

Sampler		Particle size	Filter type	Analytical method
URG VAPS	Right arm	$<2.5\,\mu m$	Quartz (Whatman, 47mm)	Thermal Manganese Oxidation Analysis (EC & OC)
	Center arm	$2.5\sim10\,\mu m$	Polycarbonate (Nuclepore) filter (Costa, $0.4\,\mu m$, 47 mm)	Gravimetric Analysis (mass)
	Left arm	$<2.5\,\mu m$	Teflon (Zeflour) filter (Gelman, $2\,\mu m$, 47 mm)	Gravimetric Analysis (mass) Ion Chromatography Analysis (sulfate, nitrate, etc)
			Nylon (Nylasorb) filter (Gelman, $1\,\mu m$, 47 mm)	
			Denuder (URG, 150 & 242 mm)	Ion Chromatography Analysis (SO_2, HNO_3, NH_3)
URG $PM_{2.5}$ cyclone / WINS sampler		$<2.5\,\mu m$	Teflon (Teflo) filter (Gelman, $2\,\mu m$, 47 mm)	Gravimetric Analysis (mass) Induced Couple Plasma Analysis & Atomic Absorption Analysis (Na-Pb, 20 elements)
MOUDI sampler			Teflon filter (Sartorius,$0.4\,\mu m$, 47 mm)	Gravimetric Analysis (mass) Ion Chromatography Analysis (sulfate, nitrate, etc)
			Teflon (Zeflour) filter (Gelman, $2\,\mu m$, 47 mm)	

the diurnal variation in the aerosol composition and its impacts on visibility impairment. Table 5 summarizes aerosol sampling and analytical methods used in those studies.

The URG-VAPS (Versatile Air Pollutant Sampler), R & P Sequential WINS, MOUDI (Micro Orifice Uniform Deposit Impactor) sampler were used to determine the gravimetric mass, chemical composition of size-differentiated particles, ionic concentration of acidic aerosol (sulfate and nitrate), and artifacts caused by gas phase. Two arms of the URG-VAPS collect both fine particles with an aerodynamic diameter ≤ 2.5 μm on a nylon (Nylasorb) and a quartz filter and coarse particles with aerodynamic diameters < 10 μm on a polycarbonate (Nuclepore) filter. WINS sampler employs a teflon (Teflo) filter to collect aerosol samples with a 2.5 μm cut size inlet. Teflo and polycarbonate Nuclepore filters were used to measure gravimetric mass and concentrations of trace elements using ICP/MS & /AES and AAS methods. Nylon filter samples were used for IC analysis of sulfate, nitrate ions, after being extracted by 10 ml of distilled water. Exposed quartz filters were analyzed by Thermal Manganeseioxide Oxidation (TMO) method for the quantification of elemental and organic carbon.

Documenting visibility events is an important aspect for evaluating visibility impairment. A Pentax PZ-20 self-contained automatic camera for the visibility monitoring was installed and operated at the top of Kwangju Meteorological Office for scene monitoring by taking 6 cuts a day every 2 hour from 08:00 to 18:00.

6. 3 Chemical Analysis

In a typical urban atmosphere, the fine aerosol species that contribute to visibility reduction are classified into five major types: sulfates, nitrates, organic mass by carbon, elemental carbon, and soil dust. Mass concentration of each component was calculated from the masses of the measured elements and ions according to modified IMPROVE (Interagency Monitoring of Protected Visual Environments) programs (Kim *et al.*, 2001a) as summarized in Table 6. Organic mass concentration by carbon [OMC] was calculated by (Malm *et al.*, 1996).

$$[OMC] = 1.4[OC] \quad (2)$$

Organic mass concentration is calculated by multiplying the total carbon by 1.4, which assumes that organic carbon mass concentration accounts for 71% of the organic mass. Since there is little information on the elemental composition of local soil, a fine soil equation from US IMPROVE study was used in this study. Fine soil (FS) mass concentration was calculated by summing the elements associated with soil, such as Al_2O_3, SiO_2, CaO, K_2O, FeO, Fe_2O_3, TiO_2. Each molar correlation factor is considered with a correlation for other compounds such as MgO, NaO, H_2O, and carbonate. A final equation for fine soil after dividing by 0.86 is (IMPROVE, 2002):

$$[FS] = 2.20[Al] + 2.49[Si] + 1.63[Ca] + 2.42[Fe] + 1.94[Ti] \quad (3)$$

In particular, coarse mode particles increased greatly in mass and dominated the visibility impairment during Asian Dust storm period. Mass concentrations of ammonium sulfate, mineral dust, and sea-salt of the coarse fraction ($2.5\ \mu m < D_p < 10\ \mu m$) in the samples were analyzed as shown in Table 6 to assess the scattering efficiency of coarse particles under unusual haze conditions by Asian Dust particles. The optical absorption coefficient of dust particles was determined by an empirical relation with mass concentration of the TSP (total suspended particle) (Parungo *et al.*, 1997, 1994).

6. 4 Results and Discussion

In the atmosphere, the fine aerosols that predominantly affect visibility impairment are classified into five major types: ammonium sulfates (NHSO), ammonium nitrates (NHNO), organic mass by carbon (OMC), elemental carbon (EC), and fine soil (FS). Multiple linear regression analysis has been used extensively to determine the contributions of various particulate species to extinction efficiency of b_{sp} (IMPROVE, 2002). The following modified IMPROVE protocol was used to reconstruct aerosol extinction coefficient (Kim *et al.*, 2001a) in Kwangju;

$$b_{ext}(Mm^{-1}) = 3f(RH)[NHSO] + 3f(RH)[NHNO] + 4[OMC] + 1[FS] + b_{scat,coarse} + b_{abs} + b_{NO_2} + b_{Ray}$$

$$b_{scat,coarse}(Mm^{-1}) = 0.4[SS] + 1.0[NHSOc] + 0.77[MD], (RH \leq 40\%) \quad (4)$$

Where f(RH) is the relative humidity dependent scaling factor which illustrates the relationship between RH and the scattering efficiency for ammonium sulfate aerosol with a mass mean diameter of 0.3 μm and a geometric size distribution of 1.5 μm. where the brackets mean the mass concentration of the component in

Table 6. Composite variables for particulate matters modified IMPROVE program proposed by Kim (2001a).

Particle size range	Component	Specifications	Composite equation
Fine particle ($D_p < 2.5\ \mu m$)	NHSO	$(NH_4)_2SO_4$	4.125[S]
	NHNO	NH_4NO_3	1.29[NO_3^-]
	OMC	Organic mass by carbon	1.4[OC]
	Soil	Fine soil	2.20[Al]+2.49[Si]+1.63[Ca] +2.42[Fe]+1.94[Ti]
	b_{abs}	Absorption coefficient by elemental carbon	10[EC]
Coarse particle ($2.5 < D_p < 10\ \mu m$)	$NHSO_c$	$(NH_4)_2SO_4$ [nss-SO_4^{2-}]	1.375{[SO_4^{2-}]−0.25[Na]}
	SS	Sea-Salt	2.5[Na]
	MD	Mineral Dust	[CM]−{[NHSOc]+[SS]}
	CM	Coarse Mass	[PM_{10}]−[$PM_{2.5}$]
TSP	b_{Dust}	Absorption coefficient by dust particles	TSP concentration dependent
Gas phase	b_{NO_2}	Absorption Coefficient by NO_2 gas	0.33[NO_2] $_{ppb}$
	b_{Ray}	Rayleigh scattering coefficient	Altitude dependent

the unit of μg/m^3. Growth function f(RH) of hygroscopic aerosol as a function of RH proposed by Malm [1996] was used in this analysis. In this equation, it was assumed that coarse particles and fine soil particles are from a single natural source, windblown dust.

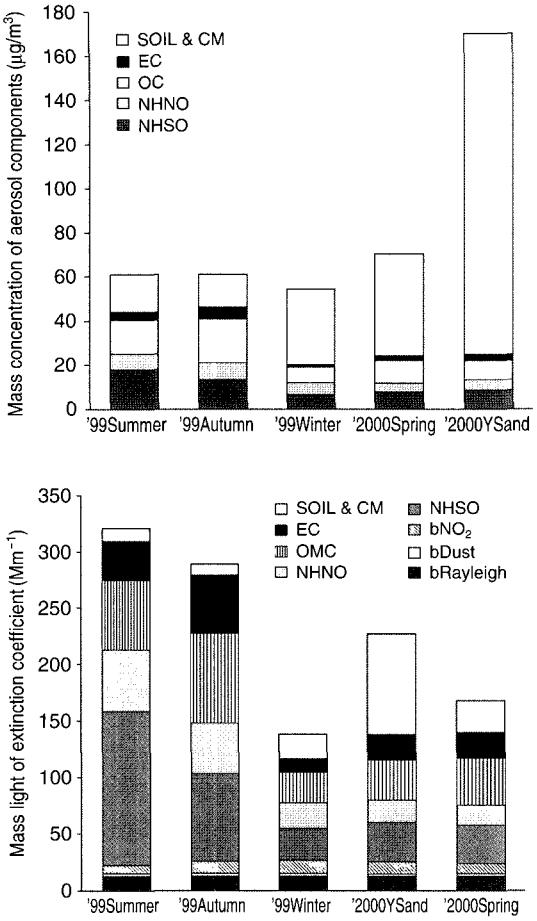

Fig. 13. Seasonal variations of mass concentrations and mean light extinction budgets of aerosol components during the entire intensive monitoring periods (Kim *et al.*, 2001b).

Figure 13 illustrates the average seasonal visibility impairment in terms of light extinction budget-a percent ratio of light extinction coefficient of an aerosol component to reconstructed total light extinction coefficient-during the entire monitoring periods between 1999 and 2000 at Kwangju, Korea. Seasonal average total light extinction measured with a transmissometer is summarized in Table 7. It shows that sulfates and nitrates account for about 45 and 18 % of total average light extinction in the summer, respectively. Sulfate and nitrates plays a particularly significant role in the humid summer months. Organic carbon accounts for about 21 percent of total average light extinction. Elemental carbon is responsible for between 10~20 percent of total light extinction in most seasons. Sulfate, OMC, nitrate, and EC components of fine aerosol were the major contributors to visibility impairment. However, sulfates and nitrates play less of a role in the spring, accounting for about 10 to 20 percent of total average light extinction. In addition, diurnal variation of visibility for Best-case ($b_{ext} \leq 109$ Mm^{-1}) days showed rapid improvement in the morning hours, while it was delayed until afternoon for the Worst-case ($b_{ext} \leq 652$ Mm^{-1}) days during summer 1999 as shown in Figure 14. The aerosol light extinction budget of each aerosol component for the Worst-case was calculated to be 11.7 times larger than the Best-case for NHSO, 20.4 times for NHNO, 2.2 times for OMC, respectively. Results shows that EC and FS were 3.7 and 2.2 times

Table 7. Reconstructed & measured light extinction coefficients of aerosol components and visibility conditions during the entire intensive sampling periods (Kim *et al.*, 2001b).

Intensive period	Reconstructed fine particle					Measured parameter				
	NHSO	NHNO	OMC	EC	Soil & CM	$PM_{2.5}$	PM_{10}	b_{ext}	vr	RH
	Mm^{-1}							Mm^{-1}	km	%
'99 Summer	123	50	62	35	22	45.4	58.7	347	11.3	72.2
'99 Fall	72	41	79	51	20	41.1	53.0	279	14.0	63.3
'99 Winter	24	19	28	11	45	15.3	41.1	141	27.8	48.0
'2000 Spring	34	18	42	23	60	15.3	59.8	189	20.7	56.6
'2000 Yellow sand	32	18	36	22	189	20.9	159.2	444	8.8	49.0

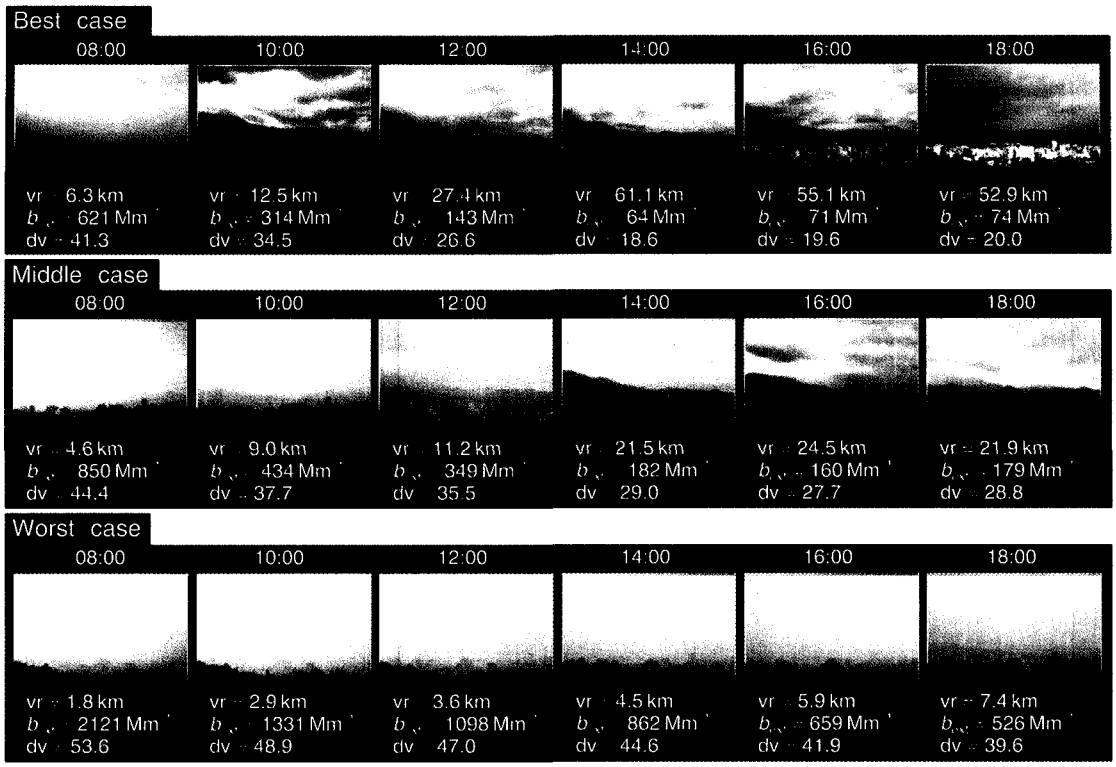

Fig. 14. Diurnal variations of scenic images by haze level during the summer intensive monitoring period, 1999 (Kim and Kim, 2003).

more than those of Best-case, respectively. The sum of total contributions of wet NHSO and NHNO to light extinction was calculated to be 301 Mm^{-1} in the worst -case. However, the sum of contributions by dry NHSO and NHNO was calculated to be 123 Mm^{-1}. Mass extinction efficiencies of fine and coarse particle were calculated to be 5.8 ± 0.3 m^2/g and 1.8 ± 0.1 m^2/g, respectively (Kim and Kim, 2003).

Scatter plots of the light extinction coefficient (b_{ext}) and the visual range (*VR*) averaged for aerosol sampling periods against particle mass concentration under lower relative humidity conditions (less than 60 percent) are shown in Figure 15. From linear regression functions between the light extinction coefficients and the particle mass concentrations of $PM_{2.5}$ and PM_{10}, are obtained as (Kim *et al.*, 2001b):

$$b_{ext}(Mm^{-1}) = 8.28 \times [PM_{2.5}]\ (\mu g/m^3)\ (R^2 = 0.851),$$
$$b_{ext}(Mm^{-1}) = 5.31 \times [PM_{10}]\ (\mu g/m^3)\ (R^2 = 0.556) \quad (5)$$

It was found that the $PM_{2.5}$ particle concentration has a better correlation with visibility than PM_{10} and coarse particle concentrations. As shown in Figure 15, visibility in clean environments is more sensitive to changes in $PM_{2.5}$ particle concentrations than in polluted areas. It may be also concluded that a proportionally larger reduction in fine particle concentrations is needed to achieve a small increment of visibility improvement in polluted areas.

During the ACE-Asia IOP (Intensive Observation Period), aerosol measurements were made 22 March~ 4 May 2001 in the urban atmosphere of the city of Kwangju, Korea. During the Asian Dust storm periods, severe visibility impairment resulted from the enor-

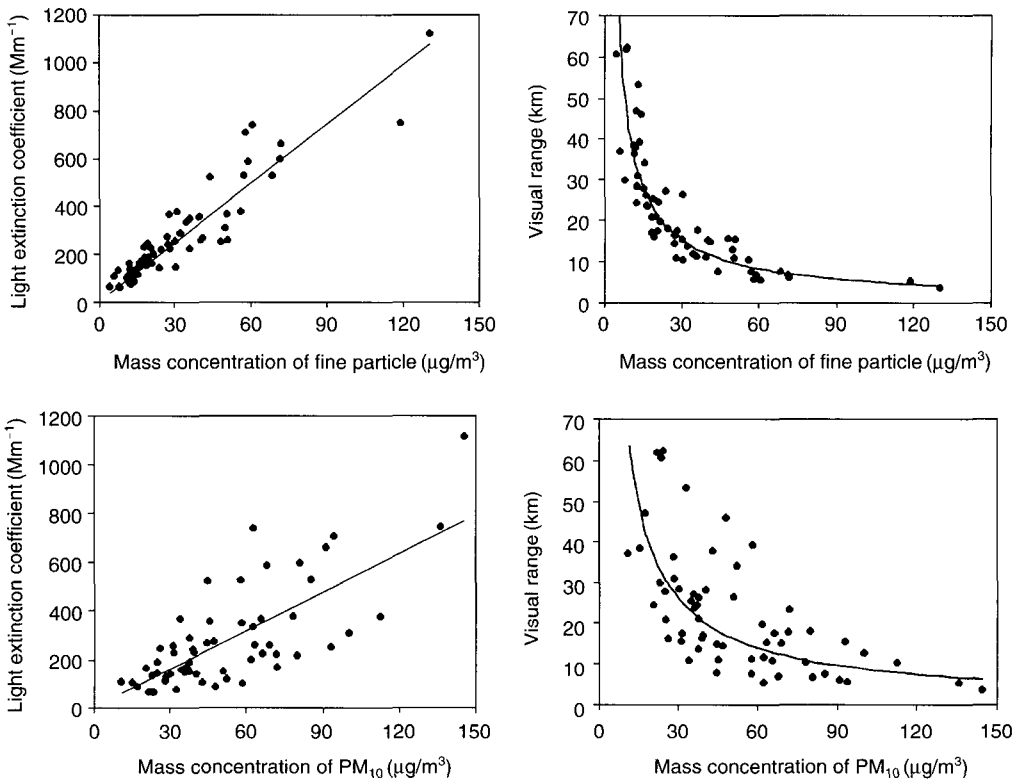

Fig. 15. Scatter plots between light extinction coefficient and particle mass concentrations and between visual range and particle mass concentrations (Kim *et al.*, 2001b).

Table 8. Chemical composition and optical properties of aerosols observed at the Kwangju site during the ACE-Asia IOP (Kim *et al.*, 2002b).

	Fine	Coarse	PM_{10}	Fine particle					Coarse particle					b_{ext}	b_{scat}	b_{abs}	$\sigma_{ext,PM10}$	ω
				EC	OC	nss-SO_4^{2-}	NO_3^-	FS	nss-SO_4^{2-}	Sea-salt	MD	BC_C	RH					
				μg m^{-3}									%	M m^{-1}			m^2 g^{-1}	
CC[a]	11.9	35.6	47.6	1.4	3.0	4.9	1.5	6.0	1.9	1.2	26.3	N/A	34.0	112	89	17	2.4	0.84
NK[b]	23.6	30.3	53.9	3.1	8.7	4.4	2.4	4.8	2.0	0.3	26.1	N/A	36.4	217	168	31	4.0	0.85
SM[c]	22.0	15.0	37.0	3.1	9.1	4.8	2.2	3.1	2.3	0.5	10.6	N/A	56.5	388	263	42	11.6	0.86
SL[d]	26.0	9.0	35.0	3.2	9.8	9.3	3.3	3.1	3.3	0.3	7.2	N/A	51.8	305	235	37	9.5	0.86
1st CD[e]	38.7	246.6	285.3	1.7	4.3	4.7	5.4	37.7	5.8	7.3	213.1	2.3	42.4	1082	952	48	4.1	0.93
2nd CD[e]	37.4	93.0	130.4	2.3	7.3	11.2	5.6	15.4	4.3	2.3	72.3	1.5	57.1	648	571	47	5.0	0.90
NKD[f]	45.4	82.2	127.6	2.5	13.6	5.1	2.4	24.2	1.9	1.6	70.3	2.0	35.5	515	444	63	4.0	0.84

[a]: CC (Clean Continental): Clean condition perturbed by continental aerosol, two events (3/26-D, 3/26-N).
[b]: NK (Northwestern China/Korea): Mixture of continental and regional aerosols, two events (4/4-D, 4/22-D).
[c]: SM (Southern Marine): Mixture of local and marine aerosols , four events (4/7-N, 4/8-D, 4/28-D, 5/4-N).
[d]: SL (Stagnant Local): Hazy condition impacted by local pollution, two events (4/8-N, 4/10-D).
[e]: CD (Continental with Asian Dust): **Asian Dust** particles originated from northwestern Chinese desert regions.
1st CD: two events (3/22-N, 3/23-D).
2nd CD: four events (4/10-N, 4/11-D. 4/12-N, 4/13-D).
[f]: NKD (Northeastern China/Korea with Asian Dust): Mixture of **Asian Dust** particles originated from northeastern Chinese sandy areas and regional aerosol , one event (4/25-D).

mous amount of coarse particles as well as fine particles. To characterize the long-range transport of aerosol (mineral dust, organic, ionic, and element) in the region of interest, air mass pathways were investigated. Major air mass pathway of Asian Dust storm was from either the northwestern Chinese desert regions or the northeastern Chinese sandy areas during the ACE-Asia IOP. Chemical composition and optical properties of six types of air mass pathway are summarized in Table 8. It was revealed that chemical and optical properties of particulate matter affecting visibility impairment varied by the air mass pathway (Kim *et al.*, 2002b). In addition, as large dust particles abundant during the Asian Dust storm period collided and agglomerated with fine BC particles, those BC particles are found in the coarse mode. Single particle analysis was carried out on some samples of fine and coarse particles collected during the Asian Dust storm event (Figure 16) shows that mineral dust particles were black due to the adsorption by carbonaceous aerosol, which also emitted X-rays of majority of C using SEM/EDX (scanning electron microscope/energy dispersive x-ray analysis).

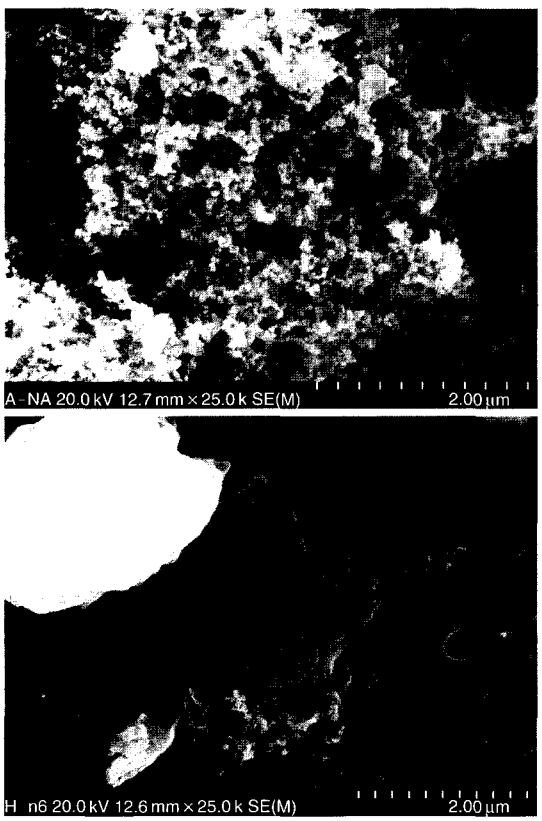

Fig. 16. Scanning electron microscope image (SEM) of agglomerated elemental carbon onto a mineral particle and dust particle adsorbed by carbonaceous particles (Kim *et al.*, 2002b).

Mean measurements for aerosol chemical and optical components are summarized in Table 8. This result shows that average visual range and fine mass concentration in Kwangju was measured to be approximately 9 km and $23.6 \pm 20.3\ \mu g/m^3$ which was a higher value than that of the NAAQS (National Ambient Air Quality Standard, $15\ \mu g/m^3$ of $PM_{2.5}$) and the CAAQS (California Ambient Air Quality Standard, 17 km and $12\ \mu g/m^3$ of $PM_{2.5}$) of the United States.

7. VISIBILITY MODELING

Visibility modeling commonly starts with the application of a chemical transport model (CTM) such as Models-3 (U.S. EPA, 2000), MATCH (Collins *et al.*, 2001), and GOCART (Chin *et al.*, 2000). A plume model is the starting point for a plume blight situation, while a three-dimensional CTM would be used for regional haze. The minimum outputs needed from the CTM for visibility modeling are spatial distributions of

Table 9. Optical and Chemical properties of aerosol observed at Kwangju between 1999 and 2002.

Measurement parameter	Specification	Mean value except AD*	Mean value of AD
FM ($\mu g/m^3$)	Fine mass	23.6 ± 20.3	37.6 ± 3.5
CM ($\mu g/m^3$)	Coarse mass	31.8 ± 40.4	126.1 ± 104.5
b_{ext} (Mm^{-1})	Light extinction coefficient	432 ± 269	680 ± 354
b_{scat} (Mm^{-1})	Light scattering coefficient	342 ± 161	587 ± 286
b_{abs} (Mm^{-1})	Light absorption coefficient	48 ± 25	52 ± 23
ω	Single scattering albedo	0.88 ± 0.6	0.92 ± 0.04

*AD: Asian Dust particle

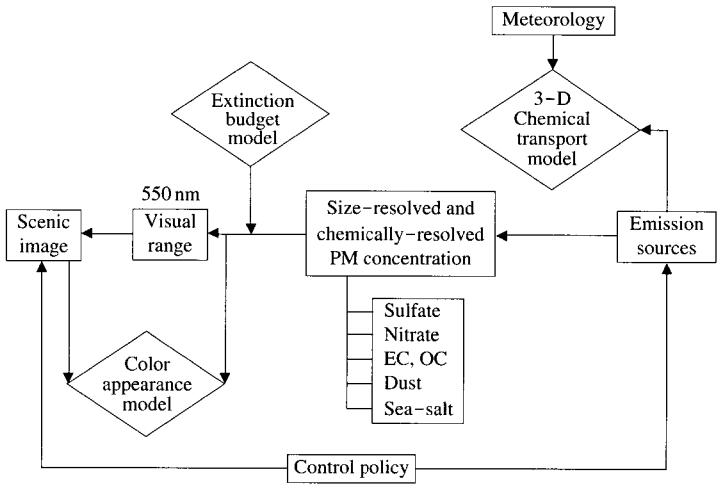

Fig. 17. Schematic diagram of procedure for visibility prediction and control policy.

particulate matter species concentrations (both fine and coarse), NO_2 gas concentrations, and relative humidity. The most simple extinction budget models use the species' concentrations, together with assumed scattering and absorption efficiencies, to arrive at local descriptions of extinction through the empirical formulas. Color appearance models play a role in further understanding the visibility change (KOSAE, 2003). More advanced models calculate particle scattering based on the physics of the interaction of light with particles. Such aerosol optics models also require information concerning the particle size distribution of the aerosol, which generally has to be estimated based on specialized measurements as shown in Figure 17. Since not all of the above information is produced routinely by CTMs, the intended use of the CTM outputs for visibility modeling must be taken into account when setting up the modeling exercise (NARSTO, 2003). Thus the quality of the visibility prediction depends greatly on the skill of the PM model in the CTMs. Consequently, the quality of modeled representations of light extinction based on these PM calculations is currently somewhat uncertain. Much remains to be learned about the important aerosol processes in the upper atmosphere and extinction by secondary organic aerosols. Extensive work is underway to learn more about the particles and their optical properties to improve models.

8. CONCLUSION

Fine particles are responsible for human health, regional haze, and urban visibility impairment. For the public, visibility can be perceived as an indicator of air quality variation. A common symptom of air pollution recognizable to the public is obscured visibility, usually referred to as haze. Nitrogen dioxide is the only common atmospheric gaseous species that significantly absorbs light in the visible spectral region. Absorption arises nearly entirely from elemental carbon particles. Ammonium sulfates, ammonium nitrates, organics are major contributors of light scattering. Thus particulate matter optically interferes with the vision by absorbing or scattering visible light. Scattering usually is dominated by fine particles, because particles at the size range of $0.1 \sim 1.0\,\mu m$ have the greatest scattering efficiency. From linear regression functions between light extinction coefficients and particle mass concentrations of $PM_{2.5}$ and PM_{10}, light scattering is roughly proportional to the mass concentration of submicron fine par-

ticles.

Owing to past efforts to reduce ambient particulate emissions TSP and PM_{10} concentrations have decreased in urban areas in Korea. However, visibility impairment by regional haze is still persistent in most urban areas in Korea since concentration of fine particles has not decreased over the decades. Visibility is relatively insensitive to small changes in PM concentrations in an urban area. Rather a substantial decrease in fine particle concentration is required to achieve perceptible improvement in the visibility. It is highly recommended that the new visibility standard and fine particle standard of $PM_{1.0}$ and/or $PM_{2.5}$ be established for protection of the health and welfare of the general public. Much work need to be done in visibility studies including long-term monitoring, improvement of visibility models, and formulating integrated strategies for managing fine particles to mitigate the visibility impairment and climate change.

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