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Evolution of $PM_{2.5}$ Measurements and Standards in the U.S. and Future Perspectives for China

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ABSTRACT

National Ambient Air Quality Standards (NAAQS) were first established in the United States to protect public health and welfare, and the concept has been adopted in China and many other countries. For particulate matter (PM), the NAAQS indicator evolved from total particle mass concentration, to PM_{10} and $PM_{2.5}$ mass concentrations as defined by the PM size-selective properties of the monitoring instrument and human inhalation characteristics. Particle measurements started with optical microscopy in the early 18th century, and scientific research over the past 300 years has related particles to adverse environmental and health effects. Several options for $PM_{2.5}$ measurement and assessment are available to China and other developing countries as they implement new $PM_{2.5}$ ambient air quality standards. Although much can be learned from the experience of North America and Europe, China can leapfrog ahead in terms of $PM_{2.5}$ monitoring and emission reduction technology. China-specific guidance documents should be created for network design, equipment selection and operation, quality control and quality assurance, database management, and interpretation. Future air quality management and standards will need to consider multiple pollutants and their effects on visibility, climate, materials, and ecosystems in addition to the primary concerns about public health.

Keywords: $PM_{2.5}$; PM_{10}; Aerosol sampling; Size-selective inlets; Multipollutant; NAAQS.

INTRODUCTION

The Chinese and U.S. National Academies (NRC, 2008), after examining parallels between the evolution of air quality management and energy issues in both countries, recommended China's adoption of $PM_{2.5}$ (particles with aerodynamic diameters [d_p] < 2.5 micrometers [μm]) mass concentration as an indicator of adverse health effects. Since then, China's State Council has approved $PM_{2.5}$ national ambient air quality standards (NAAQS) requiring cities to attain concentrations below 35 μg/m^3 annual arithmetic average and < 75 μg/m^3 for 24-hr periods beginning in 2016 (MEP and AQISQ, 2012). Questions have arisen in the country about how $PM_{2.5}$ came to be measured and regulated as an indicator of adverse effects, first in the United States, and later in other parts of the world. The objectives of this paper are to: 1) provide a brief summary of the evolution of size-specific fractions for suspended particulate matter (PM); 2) review relationships between inhalation properties and their health effects; 3) illustrate changes in PM measurement methods and air quality standards; and 4) look to the future of multipollutant air quality management.

AMBIENT PM SIZE FRACTIONS

The basic form of atmospheric PM size distributions was elucidated by Whitby *et al.* (1972) in Los Angeles, California, using newly developed instrumentation (Liu *et al.*, 1974). These researchers originally concluded that typical particle size distributions could be explained by two log-normal curves superimposed on each other, with a minimum in the 1 to 3 μm region. Fig. 1 illustrates current understanding of ambient particle size distributions with multiple modes, elaborating on the original Whitby bimodal distribution. $PM_{2.5}$ and PM_{10} (d_p < 10 μm), as indicated by the arrows at the top of Fig. 1, are the regulated size fractions in the U.S. and many other countries. PM_{10} includes $PM_{2.5}$, and a "coarse" fraction is defined as the difference between the two mass measurements ($PM_{10-2.5}$). The $PM_{2.5}$ fraction was often termed the "fine" fraction, but was later given the $PM_{2.5}$ designation to specify the 2.5 μm upper size limit.

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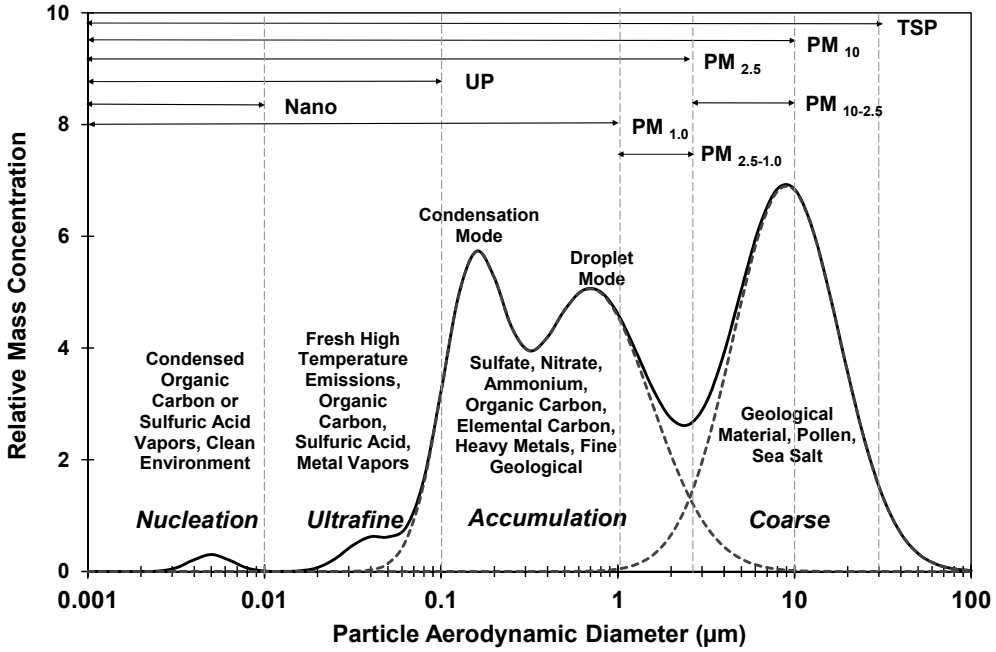

Fig. 1. Idealized example of an ambient particle size distribution, patterned after Chow (1995) and Watson (2002). TSP = Total Suspended as measured by a high-volume (hivol) sampler in the particle size range of 0 to ~30–50 μm. Nucleation and ultrafine modes denote particles less than 0.01 and 0.1 μm, respectively. The accumulation mode contains of most of the fine particles from ~0.1 to ~2 μm. The ~0.2 μm condensation mode results from gas phase reaction products while the ~0.7 μm droplet mode results from gas absorption and reactions in water droplets. The coarse mode extends from ~2 or 3 μm to 100 μm. UP = Ultrafine Particles. Nano = Nanoparticles.

Relative magnitudes of the nucleation and ultrafine modes are exaggerated in Fig. 1, as ultrafine particles (UP; d_p < 0.1 μm or 100 nm) dominate particle number counts but constitute a small (< 5%) fraction of $PM_{2.5}$ mass.

Using low-pressure impactors to obtain better chemical resolution within the size distribution, Hering and Friedlander (1982) and John *et al.* (1990) noticed shifts in the maxima of the accumulation mode (~0.1–2 μm) under different meteorological conditions. They identified a larger droplet mode, peaking near 0.7 μm, formed from growth of particle nuclei and by aqueous-phase reactions in fogs and clouds (Penkett *et al.*, 1979). Sulfate, in particular, forms from gaseous sulfur dioxide in water droplets, resulting in larger particles when the droplet evaporates. Under dry atmospheric conditions, gas-phase conversion of sulfur dioxide and oxides of nitrogen creates smaller particles in the condensation mode, which peaks at ~0.2 μm. Another interpretation of these modes for relative humidity (RH) > 80% is that the water-absorbing sulfate and nitrate compounds grow into the droplet mode while the water-repellent soot and some organic carbon retain their original sizes (Zhang *et al.*, 1994).

Limited chemical speciation studies in China (e.g., Zhang and Friedlander, 2000; He *et al.*, 2001; Wu *et al.*, 2003; Ye *et al.*, 2003; Sun *et al.*, 2004; Xu *et al.*, 2004; Cao *et al.*, 2005; Louie *et al.*, 2005a, b; Chow *et al.*, 2006a; Duan *et al.*, 2006; Ho *et al.*, 2006; Wang *et al.*, 2006; Song *et al.*, 2007; Shen *et al.*, 2007, 2010; Zhang *et al.*, 2010; Cao *et al.*, 2011; Deng *et al.*, 2011; Shen *et al.*, 2011; Yang *et al.*, 2011; Cao *et al.*, 2012a; Huang *et al.*, 2012; Li *et al.*, 2012; Tao *et al.*, 2012; Wang *et al.*, 2012; Xu *et al.*, 2012; Zhang *et al.*, 2012) show that the $PM_{2.5}$ size fraction is dominated by sulfate, nitrate, ammonium, organic carbon and elemental carbon, with small (< 10%) portions from fugitive dust except during Asian dust episodes (Zhao *et al.*, 2010). The $PM_{10–2.5}$ fraction is dominated by geological material, usually in the form of fugitive dust. $PM_{10–2.5}$ also contains sea salt (near coastal areas), bioaerosols (e.g., pollens, spores, and plant parts), and uncontrolled industrial emissions. Newer industries in China are equipped with pollution control devices (e.g., baghouses and precipitators; Hu *et al.*, 2010) that more efficiently remove $PM_{10–2.5}$ and larger particles than the smaller accumulation mode particles.

At the lower end of the size spectrum in Fig. 1, UP are both directly emitted by combustion sources and form from condensed gases as secondary aerosols (Watson *et al.*, 2002; Chang *et al.*, 2004; Chow and Watson, 2006; Watson *et al.*, 2006a, b; Chow and Watson, 2007). Coal and biomass are the most commonly used solid fuels in combustion. Combusted liquid fuels from petroleum refining include residual oil, diesel, kerosene, aircraft, and gasoline. The cleanest burning fuels are gases such as methane, propane, and butane. Fossil fuels, especially coal, include trace amounts of sulfur that can be oxidized to sulfuric acid. As organic gases and sulfuric acid cool upon dilution with ambient air, they may condense onto larger particles or nucleate into UP. Black carbon soot is produced by incomplete combustion in oxygen-starved conditions. Some of these may be UP, but they often grow due to condensation

and adsorption of vapors or coagulation with other UP. UP are also produced in clean remote environments. Volatile organic compounds emitted in the gas-phase by natural (e.g., plant life and wildfire) and anthropogenic (e.g., fossil fuel and biofuel combustion) sources transform into semi-volatile organic compounds with lower vapor pressures in the presence of sunlight. These gases can nucleate when their vapor pressure exceeds the saturation vapor pressure at a given temperature. This nucleation usually occurs when PM mass concentrations are low, as the vapors preferentially condense on larger particles when they are available in sufficient quantities.

Both UP and $PM_{10-2.5}$ have relatively short lifetimes of minutes to a few hours after emission or formation. UP diffuse rapidly to surfaces or grow into the accumulation mode (McMurry, 2000). Coarse particles, including those > 10 μm, gravitationally deposit to surfaces (Hinds, 2001). $PM_{2.5}$ in the accumulation mode follows updrafts and can remain suspended for several days, up to a week (Watson *et al.*, 2000). As a result, $PM_{2.5}$ mass tends to be more homogeneously distributed with distance from the emission source and as secondary sulfates and nitrates form over regional scales (Chow *et al.*, 2002).

PM INHALATION PROPERTIES AND HUMAN HEALTH EFFECTS

People have associated visible air pollution and odors with displeasure and adverse health effects since the 12th century (Halliday, 1961; Brimblecombe, 1976, 1978; Boubel *et al.*, 1994). Over 300 years ago, Rammazini (1703) related particle number counts measured by an early optical microscope with different respiratory ailments contracted by workers in 52 different occupations. More detailed studies on human inhalation and lung deposition properties were motivated by increased understanding of the hazards posed by nuclear testing (Chamberlain and Dyson, 1956). Fig. 2 summarizes some of the early inhalation and deposition efficiencies, determined by measuring inhaled and exhaled inert particles. Inhalation and lung deposition estimates served as part of the basis toward selecting the $PM_{2.5}$ and PM_{10} size fractions for regulation. Inhalation properties have also been measured experimentally using casts of human airways (Gurman *et al.*, 1984a, b; Zhou and Cheng, 2005) and simulated by computational fluid dynamics (CFD) models (Rostami, 2009), showing the same general patterns as those of Fig. 2.

Fig. 2 demonstrates that the human nose and mouth do not exhibit step-function cut-offs at 2.5 and 10 μm; approximately 50% of the particles at these sizes penetrate the nose and mouth, respectively. The volume and rapidity of breathing also affect the inhaled size range, with the cut-point shifting to lower sizes during exercise than when at rest. The upper cut-point for coarse particles was originally believed to be 15 μm (Miller *et al.*, 1979), and in the late 1970s the U.S. Inhalable Particulate Network (IPN) characterized the PM_{15} fraction (Watson *et al.*, 1981; Rogers and Watson, 1984). The 15 μm upper limit for inhalable particles was criticized because a mouthpiece was used to deliver the aerosol to human subjects that might have minimized the influence of normal airway obstructions (Swift and Proctor, 1982; Lippmann, 1984; Lodge, 1984; Swift and Proctor, 1984), and U.S. EPA (1982, 1986) settled on the PM_{10} indicator for regulation.

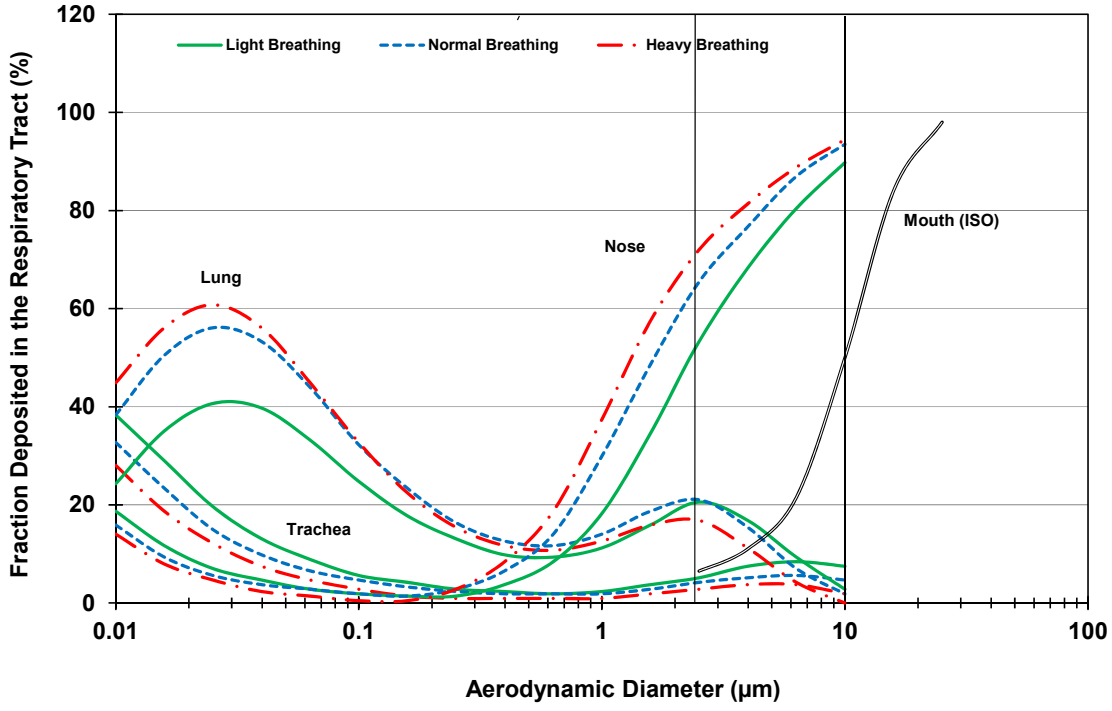

Fig. 2. Inhalation and deposition properties for the human respiratory system. Adapted from Chow (1995) using data from Phalen *et al.* (1991). The International Standards Organization (ISO) for mouth breathing is also shown (ACGIH, 1994).

The "fine" fraction had been recognized as another potential indicator of adverse PM health effects (Friedlander, 1973; Friedlander and Lippmann, 1994), but ambient data were lacking to establish appropriate correlations. The Harvard Six Cities Study (Spengler and Thurston, 1983; Chow and Spengler, 1986a, b; Dockery *et al.*, 1993) established this relationship using a dichotomous sampler (Evans and Ryan, 1983) with 15 and 2.5 μm 50% cut-points on its inlets for total (PM_{15}) and fine ($PM_{2.5}$) particle size fractions, respectively. Subsequent epidemiological studies (Chow *et al.*, 2006b; Pope and Dockery, 2006) used similar $PM_{2.5}$ inlets, and $PM_{2.5}$ was selected as the fine particle indicator for the U.S. $PM_{2.5}$ NAAQS (U.S. EPA, 1997a). Arguments were made for a 1.0 μm cut-point, since the $PM_{2.5–1.0}$ fraction is composed of geological material, which was also indicated by measurements (Lundgren *et al.*, 1996). Fig. 2 shows that normal and heavy nose-breathing lower the 50% cut-point closer to 1.0 μm. However, because 2.5 μm is near the minimum in the particle size distribution (See Fig. 1), shifts in the cut-point cause only minor (~5%) differences in mass concentration (Chow, 1995). In addition, the growing data base for $PM_{2.5}$ (Chow and Koutrakis, 1998) with associated epidemiological studies, and the production of sampler inlets with these size-selective properties (Watson and Chow, 2011), resulted in $PM_{2.5}$ mass as the selected health indicator. While most of the $PM_{2.5}$ health relationships have been established in North America and Europe, there is increasing evidence of similar associations in China (e.g., Venners *et al.*, 2003; Perera *et al.*, 2005; Kan *et al.*, 2007; Guo *et al.*, 2009; Huang *et al.*, 2009; Jia *et al.*, 2011; Xie *et al.*, 2011; Cao *et al.*, 2012b, 2012; Langrish *et al.*, 2012).

SIZE-SELECTIVE PM MEASUREMENTS

Much has been written on aerosol sampling and analysis from North American and European perspectives (e.g., Owens, 1922; Chow, 1995; Spurny, 1998; Landsberger and Creatchman, 1999; McMurry, 2000; Burtscher, 2002; Wilson *et al.*, 2002; Chow *et al.*, 2007a; Chow and Watson, 2012; Watson and Chow, 2011), and these are still areas of active research. The trend is toward less intrusive and less-costly portable samplers, continuous monitoring devices, and samplers capable of greater chemical speciation (Chow *et al.*, 2006c, d, 2008a, b, 2009, 2010a; Chow and Watson, 2008). As noted above, inhalable particles were first detected and counted by optical microscopy in the 18th century (Rammazini, 1703), but optical microscopy is of limited value when the particle size approaches the wavelength of light (~0.5 μm), so mostly the coarse fraction was detected. Large particles (> 10 μm) were characterized in wet and dry deposition (Ducros, 1815; Smith, 1852; Russell, 1885) in the 19th century. Aitken (1888a, b, 1890) measured particle numbers (dominated by UP) at the end of the 19th century, while the British Smoke method from the 1920s quantified the darkening caused by particles drawn through a filter by measuring the attenuation of light reflected from the aerosol deposit (Hill, 1936). PM mass concentrations determined by drawing air through filter paper, measuring the volume sampled, and weighing the filter before and after sampling to quantify the deposit were first reported for London by Russell (1885). This evolved into the high-volume (hivol) sampler in the late 1940s to sample airborne radioactivity after atmospheric testing of nuclear weapons (Robson and Foster, 1962), as illustrated in Fig. 3(a). This was further modified into the peaked-roof hivol (Fig. 3(b)) for total suspended particles (TSP, in the size range 0 to ~30–50 μm) that were operated throughout the U.S. from the 1960s to the 1980s (Jutze and Foster, 1967) and are still being used to determine attainment of the U.S. TSP lead (Pb) NAAQS (U.S. EPA, 2008).

As knowledge increased about PM inhalation properties (Fig. 2), and as measurement methods were developed to determine the size-selective properties of the hivol and other inlets, it became apparent that the ill-defined hivol cut-point for TSP mass: 1) varied by wind speed and orientation into the wind; 2) collected non-inhalable, as well as inhalable particles; and 3) allowed for substantial particle deposition during passive sampling periods (Chow, 1995). Based on health effects and inhalation studies available at the time, and the development and testing of new inlets, the PM_{10} NAAQS was promulgated by the U.S. EPA in 1987 as the best indicator of excessive PM exposure. PM_{10} inlets (Fig. 3(c)) with multiple impaction jets replaced the peaked roof hivol inlets for compliance monitoring. Part of the motivation in retro-fitting existing hivol samplers with a new inlet was the large TSP network already in place and the excessive cost associated with replacing that equipment and re-training operators.

While adequate for the 1980s, the hivol sampler is not optimal for size-selective sampling or for obtaining more than mass measurements from the filter. The hivol blower has a limited amount of suction, and cannot use membrane filters that are more appropriate to accommodate several chemical analyses. The high flow rates (~1000 L/min) could load up the inlets, thereby changing the 50% cut-points or re-entraining collected dust back into the air flow (Chow, 1995). Hivol samplers are also bulky and have high line current requirements, thereby limiting the locations in which they can be conveniently deployed. The U.S. rules also provided for a 50% cut-point of 10 ± 0.5 μm (U.S. EPA, 1987), which engendered a race to a lower cut point, toward a $PM_{9.5}$ fraction, to minimize the odds of exceeding the PM_{10} NAAQS.

In developing U.S. PM_{10} State Implementation Plans (SIPs) during the late 1980s, it was soon discovered that more chemical-specific information was needed from the PM_{10} sample than could be obtained from the hivol glass-fiber filters. Low- (e.g., ~10 L/min) and medium- (e.g., ~100 L/min) volume filter samplers with single or multiple channels were developed for this purpose to facilitate receptor model source apportionment studies which proved successful in developing effective emission reduction strategies (Mathai and Stonefield, 1988; Watson, 1989; Chow and Ono, 1992). These studies identified uninventoried sources such as road dust, residential wood combustion, and high emitting engines that were not considered in previous control efforts. Through careful network design

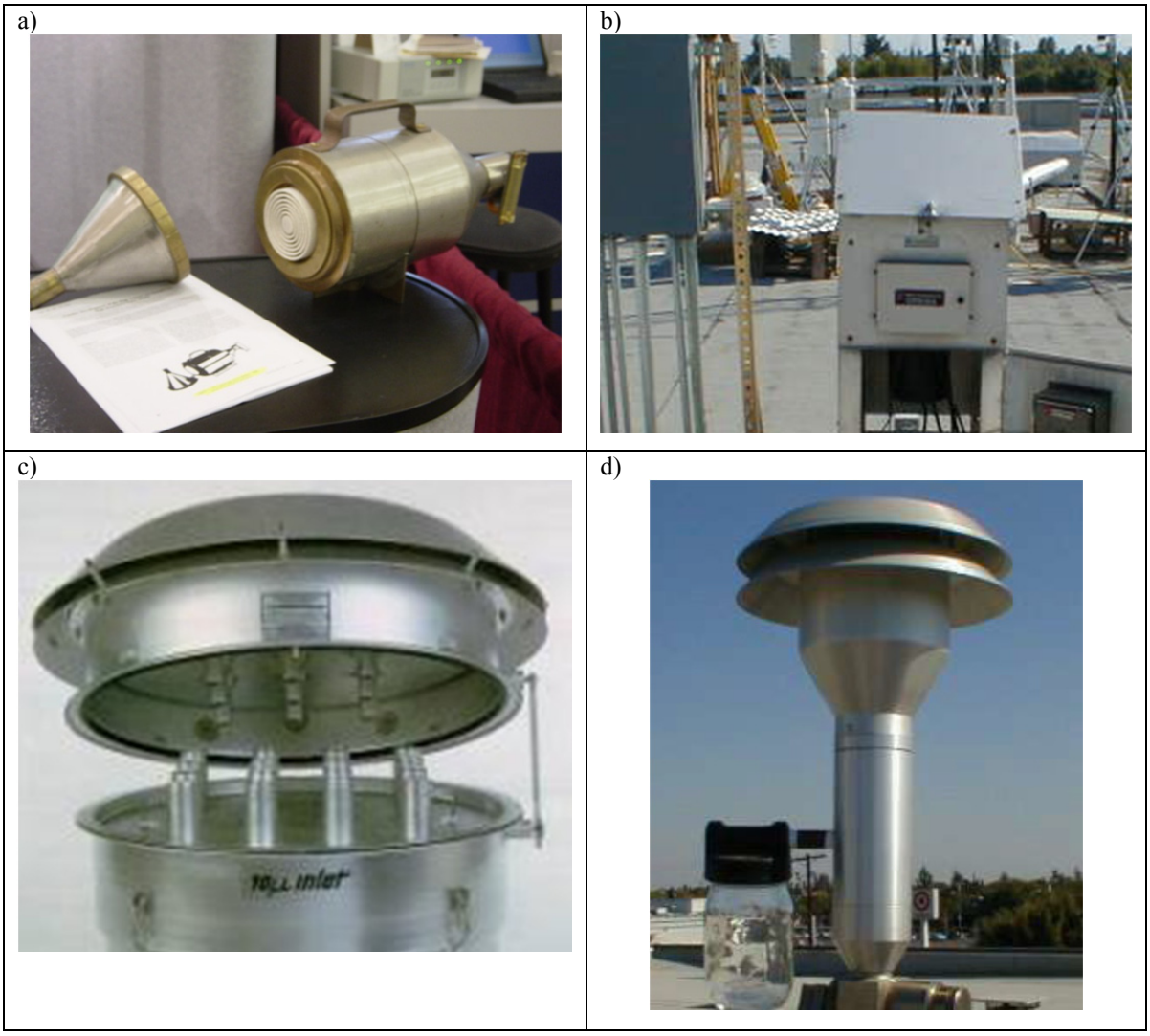

Fig. 3. Examples of: a) the original hivol sampler for total suspended particles (TSP) with pleated filter for radiation measurements; 2) the peaked-roof hivol inlet for TSP (~1130 L/min); c) the PM_{10} hivol retrofit (1130 L/min), showing impaction jets; and d) the PM_{10} inlet for low-volume (16.7 L/min) samplers. See Watson and Chow (2011) for a complete listing of PM_{10} inlet options.

and deployment, source apportionment studies also helped to separate local from regional source contributions to PM and expanded the geographical areas that needed pollution control (Chow *et al.*, 2002). More recent PM_{10} monitors use the low-volume inlet (Fig. 3(d)) with 47 mm diameter ringed Teflon-membrane filters.

The "fine" PM fraction was considered in the mid-1980s, but it took until 1997 to justify $PM_{2.5}$ as the new indicator (U.S. EPA, 1997a). Several commonly used $PM_{2.5}$ inlets are illustrated in Fig. 4. Instead of specifying performance criteria for the $PM_{2.5}$ inlet, U.S. EPA specified the Well-Impactor Ninety-Six (WINS), illustrated in Fig. 4(a), as the design standard. This inlet uses a filter soaked in oil as an impaction surface to remove particles. It was found that the oil did not perform well in cold weather or a highly polluted environment and that the impacted particles would accumulate in a conical pile, the tip of which could break off and be transmitted to the sampled filter. With additional development and testing, the Very Sharp Cut Cyclone (VSCC, Fig. 4(b)) was accepted as an equivalent inlet and has replaced the WINS in most U.S. compliance monitoring applications. Intercomparison studies (e.g., Chow *et al.*, 2008a) show that alternatives to the U.S. EPA inlets (Chan and Lippman, 1977; Kenny and Gussman, 1997; John and Reischl, 1980; Kenny *et al.*, 2000; Kenny and Gussman, 2000) provide comparable $PM_{2.5}$ mass measurements in most situations and may be more applicable to non-U.S. compliance situations.

EVOLUTION OF NATIONAL AMBIENT AIR QUALITY STANDARDS IN THE U.S. AND CHINA

NAAQS (Bachmann, 2007; Chow *et al.*, 2007b) for PM and other ambient air contaminants were first established in the U.S. and have since been implemented in other countries. NAAQS consist of an indicator (e.g., PM_{10} or

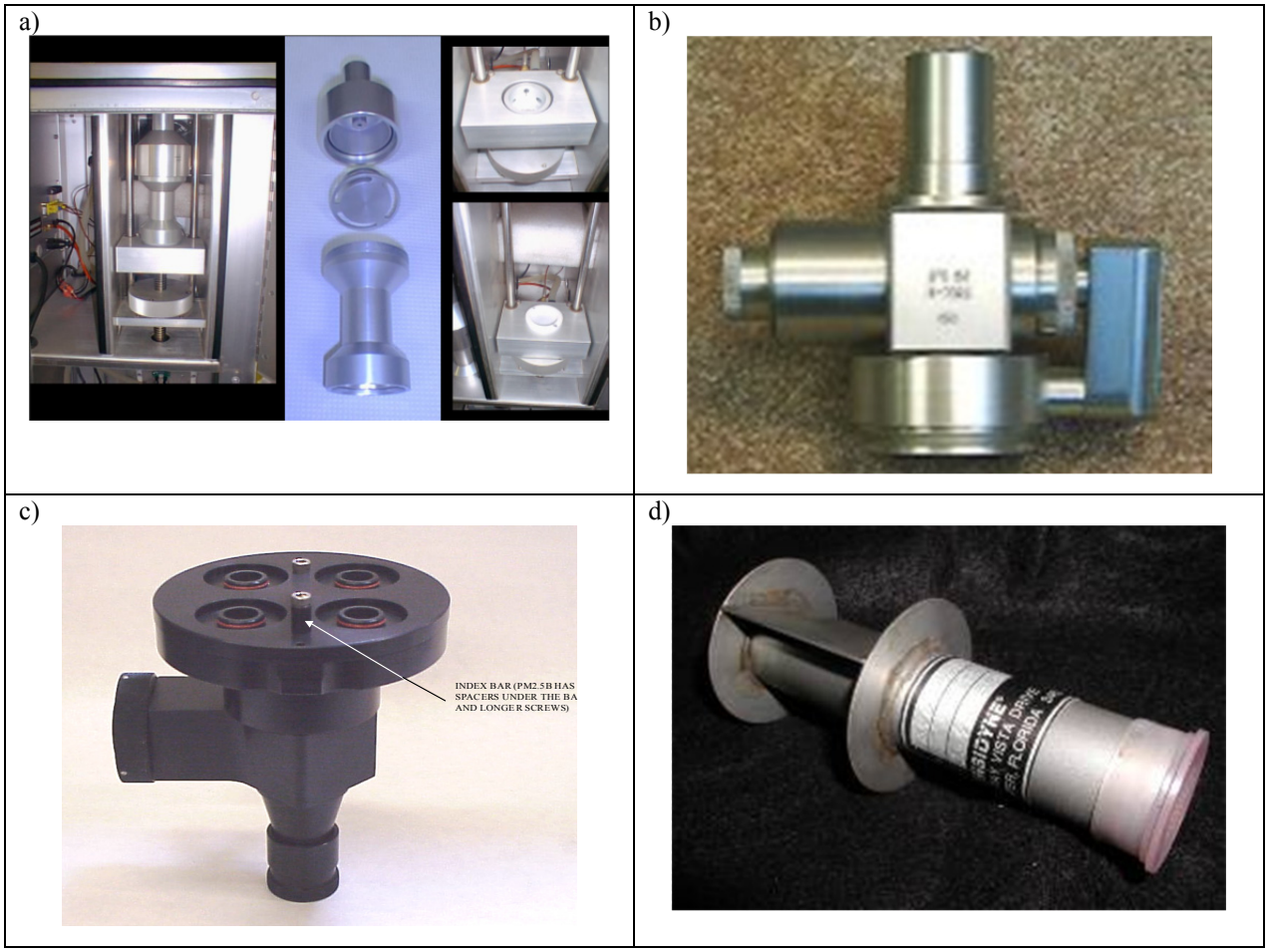

Fig. 4. Examples of $PM_{2.5}$ inlets for: a) Well Impactor Ninety-Six (WINS; 16.7 L/min) initially specified for U.S. compliance monitoring (Peters *et al.*, 2001; Vanderpool *et al.*, 2001a, b; 2007); b) the Very Sharp Cut Cyclone (VSCC; 16.7 L/min) that has demonstrated equivalence to the WINS (Kenny *et al.*, 2004) and is now implemented in compliance networks; c) the Air Industrial Hygiene Laboratory (AIHL; John and Reischl, 1980) cyclone (22.8 L/min) used in the U.S. IMPROVE regional $PM_{2.5}$ network (Watson, 2002); and d) Bendix/Sensidyne 240 cyclone (113 L/min) used for parallel sampling on multiple channels for chemical speciation measurements (Chan and Lippmann, 1977; Chow *et al.*, 1993). See Watson and Chow (2011) for a complete listing of $PM_{2.5}$ inlet options.

$PM_{2.5}$ mass), an averaging time (e.g., 5 min, 1-hr, 24-hr, or 1-year), a concentration level, and a statistical form (Watson *et al.*, 1995). In the U.S., the 1970 Clean Air Act and its Amendments require the U.S. EPA to re-evaluate the NAAQS every five years to determine the extent to which they provide an adequate margin of safety for the most susceptible populations, typically children, the elderly, and people with respiratory problems. As noted in Table 1, a more stringent level (12 $\mu g/m^3$) has been promulgated by the U.S. EPA for the annual $PM_{2.5}$ NAAQS while retaining the indicator, averaging time, and statistical form (U.S. EPA, 2013). Bachmann (2007) describes air quality management as an iterative circle in which: 1) the best information is used to set ambient air quality standards; 2) air is monitored to identify areas that exceed the standards; 3) source contributions are identified through source and receptor modeling; 4) emission reduction strategies are formulated and implemented, and 5) long-term measurements are used to evaluate the successes and failures of those strategies. The key component for air quality management (i.e., the center of the circle in Fig. 2 of Bachmann (2007)) is an active research component that is continually developing new measurement methods, exploring atmospheric interactions, establishing relationships between concentrations and adverse effects, and inventing less polluting technologies.

China's Environmental Protection Office (EPO) was established in 1973, evolved into the Chinese State Environmental Protection Administration (SEPA) in 1984, and was promoted to the Ministry of Environmental Protection (MEP) in 2008 (NRC, 2008). In 1987, the Committee of the People's Congress Council enacted the Law of the People's Republic of China (PRC) on the Prevention and Control of Atmospheric Pollutants (revised in 1995 and 2000) along with the PRC Environmental Protection Law in 1989 (MEP, 2013).

Table 2 traces the evolution of Chinese PM NAAQS. The approach was, and still is, to set different limits for different environments. Class I areas were designated by the

Table 1. Summary of particulate matter (PM) National Ambient Air Quality Standards (NAAQS) implemented by the U.S. Environmental Protection Agency (U.S. EPA).

Year of Implementation	Indicator[a]	24 hr Average ($\mu g/m^3$)	Statistical Form for 24 hr Average	Annual Average ($\mu g/m^3$)	Statistical Form for Annual Average
1971	TSP	260	Not to be exceeded more than once per year	75	Annual geometric mean
1987	PM_{10}	150	Not to be exceeded more than once per year on average over a three-year period	50	Annual arithmetic mean averaged over three years
1997a	$PM_{2.5}$	65	98th percentile averaged over three years	15	Annual arithmetic mean averaged over three years
2006	PM_{10}	150	Same as 1987 NAAQS	None	Annual average was vacated
2006	$PM_{2.5}$	35	98th percentile averaged over three years	15	Same as 1997 NAAQS
2013	PM_{10}	150	Same as 1987 NAAQS	None	Annual average was vacated in 2006
2013	$PM_{2.5}$	35	Same as 2006 NAAQS	12	Annual arithmetic mean averaged over three years.

[a] See Fig. 1 for definition of the particle size mass fraction indicators.

Table 2. Evolution of ambient air quality standards for particulate matter (PM) in China.

Implementation Document[a]: EPO (1982)

Standard	Daily Average[b] ($\mu g/m^3$)			Maximum Not to Exceed[c] ($\mu g/m^3$)		
	Class I[d]	Class II[e]	Class III[f]	Class I[d]	Class II[e]	Class III[f]
TSP	150	300	500	300	1000	1500
Airborne Particles[g]	50	150	250	150	500	700

Implementation Document: SEPA and STSA (1996)

Standard	Daily Average ($\mu g/m^3$)			Annual Arithmetic Mean ($\mu g/m^3$)		
	Class I[d]	Class II[e]	Class III[f]	Class I[d]	Class II[e]	Class III[f]
TSP	120	300	500	80	200	300
Airborne Particles[g]	50	150	250	40	100	150

Implementation Document: MEP (2012)

Standard	24 hr Average ($\mu g/m^3$)			Annual Arithmetic Mean ($\mu g/m^3$)		
	Class I[d]	Class II[h]		Class I[d]	Class II[h]	
TSP	120	300		80	200	
PM_{10}	50	150		40	70	
$PM_{2.5}$	35	75		15	35	

[a] EPO: Environmental Protection Office; SEPA: State Environmental Protection Administration; STSA: State Technology Supervision Administration; MEP: Ministry of Environmental Protection.
[b] The daily average for 1982 and 1996 was redefined as a 24-hr average in 2012.
[c] Replaced by annual standards as of 1996.
[d] Applies to national parks, conservation areas, and designated historical sites.
[e] Applies to residential and commercial areas.
[f] Applies to industrial and heavy traffic areas.
[g] Defined as airborne particles with $d_p < 10$ μm; redefined as inhalable particulate matter (PM_{10}).
[h] Applies to residential, commercial, cultural, industrial, and heavily trafficked areas. Previous Class III areas are included in Class II.

central government, while the Class II and III designations were assigned by provincial and local governments (Siddiqi and Zhang, 1984; Florig, 2002). The first ambient air quality standards issued by the EPO in 1982 included both TSP ($d_p < 100$ μm) and "airborne particles" ($d_p < 10$ μm, somewhat equivalent to PM_{10}) by gravimetric mass (EPO, 1982; Siddiqi and Zhang, 1984). Measurement methods for PM were not specified until 1986 (SEPA, 1986), at which time a 50% cut point of 10 ± 1 μm and geometric standard deviation (σ_g) of ≤ 1.5 were defined for "airborne

particles", which were renamed "inhalable PM" (PM_{10}) in 1996 (SEPA and STSA, 1996). PM_{10} samplers were later specified with a 50% cut point of 10 ± 0.5 μm and σ_g = 1.5 ± 0.1; $PM_{2.5}$ was introduced with a 50% cut point of 2.5 ± 0.2 μm and σ_g of 1.2 ± 0.1 (MEP, 2011).

EPO (1982) regulated daily average and maximum not-to-exceed concentrations of TSP and PM_{10}. A minimum sampling time of one hour was required to determine the not-to-exceed concentration. To compute a daily average, at least two intervals with total sample durations > 6 hr were needed for TSP (SEPA, 1988), and at least four intervals with each sample duration > 1 hr required for airborne particles (SEPA, 1986).

SEPA and STSA (1996) replaced the 1982 approach with daily and annual arithmetic average limits for TSP and airborne particles. A sample duration of 12 to 24 hrs was required to compute a daily average. For the annual average, each month was represented by five or more daily averages, and these 12 monthly averages were averaged for the year, with the provision of at least 60 daily samples distributed throughout the year.

Replacement of manual filter samplers (SEPA, 2005a) with contiuous Beta Attenuation Monitors (BAM) and Tapered Element Oscillating Microbalances (TEOM) was permitted as of 2005 (SEPA, 2005b, 2007), requiring averaging of 12 to 24 hourly values to compute the daily average, and 12 monthly averages (derived from at least 21 daily averages) to determine the annual average.

MEP and AQISQ (2012) added $PM_{2.5}$ standards, requiring 20 to 24 valid hourly concentrations for the daily average and at least 324 daily averages with ≥ 27 days per month (≥ 25 days for February) to compute the annual average.

Measurement methods for the Chinese NAAQS evolved, but documentation on this evolution is not as complete as that for the U.S. Federal Reference Methods and Federal Equivalent Methods (U.S. EPA, 2012a). "Super-fine glass-fiber filters" were specified by EPO (1982) for gravimetric analysis. Glass-fiber and perchloroethylene filters with ≥ 90% collection efficiency were specified by SEPA (1988). SEPA (2005a) specified flow rates (1050 L/min or 100 L/min for high- and medium-/low-volume sampling, respectively) with a 70 mm diameter filter. Sample volumes were to be adjusted to standard temperature and pressure (STP at 273°K and 101.3 kPa, respectively) using "superfine glass-fiber" or "organic-fiber" filters with collection efficiencies of ≥ 99% for 0.3 μm particles and post-sampling storage at ≤ 4°C (SEPA, 2005a). Additional filter substrates (i.e., glass-fiber, quartz-fiber, perchloroethylene, polypropylene, and cellulose-fiber) were authorized by MEP (2011).

SEPA (1986) had filters placed in a desiccator for 24 hr, followed by gravimetry with a 0.1 mg sensitivity balance, followed by an additional hour of desiccation and a second weighing. The difference between the two gravimetric analyses was to be within ± 0.4 mg (SEPA, 1986). MEP (2011) specified a 0.01 mg sensitivity balance for filter weighing after equilibration at 15–30°C temperature and 45–55% relative humidity. A standard filter is used to evaluate weighing precision that is equilibrated for 24 hr, and weighed 10 times with variations less than ± 5 mg and ± 0.5 mg for high- and medium-/low-volume samplers, respectively (MEP, 2011).

AIR QUALITY MANAGEMENT IN THE FUTURE

Air quality management (AQM) and the reduction of ambient air pollution levels require extraordinary scientific and technical efforts and commitment from both the government and the community at large. The U.S. experience (Bachmann, 2007; Chow *et al.*, 2007b) provides a basis for China and other countries to develop their AQM strategies.

With a continuous race between economic development and environmental pollution, clean air is a major challenge in China that requires substantial efforts in pollutant measurement and control (Zhang *et al.*, 2012). While the history of establishing and implementing NAAQS in the U.S. has proven to be effective without hindering economic growth (U.S. EPA, 2012b), there are important shortfalls that should not be repeated in other countries. For example, single pollutant monitoring strategies (U.S. EPA, 2005) for determining NAAQS compliance are insufficient. Air quality networks in China and other countries should specify multiple objectives including: 1) examination of long-term trends, source zones of influence, and source apportionment modeling, and 2) evaluation of relationships between ambient measurements and adverse effects. Instead of focusing on PM abatement at a local level, monitoring networks should include the measurement of precursor gases such as sulfur dioxide, nitrogen oxides, and carbon monoxide. Fig. 5 shows some of the complex interactions among air pollution sources, ambient pollutants, and effects that will need to be addressed in the future.

Of particular note is the emergence of UP and black carbon (BC) as potential future indicators of adverse effects on health and climate. The European Union is evaluating test methods for certifying engine exhaust UP emissions (Wang *et al.*, 2010; Giechaskiel *et al.*, 2012), and the California Air Resources Board (CARB, 2011) has proposed methods for both UP and BC as engine exhaust indicators. Research is progressing in continuous monitors and microsensors that can more cost-effectively monitor multipollutant observables in source emissions and ambient air (Chow *et al.*, 2008a, Wang *et al.*, 2012).

China has made a good start in adopting $PM_{2.5}$ as an indicator of adverse health effects. Although the Class II levels are higher than those in the U.S. and other countries, a more frequent revision cycle would allow them to be adjusted in the future to conform with emerging science on the multiple adverse effects of excessive $PM_{2.5}$. The new standards should provide motivation to city and provincial Environmental Protection Bureaus (EPBs) to increase their efforts in pollution monitoring and control. Enforcement of NAAQS needs to be stringent and centralized by the government, with specified siting requirements, measurement methods (e.g., instrument type, calibration, filter media, sampling duration, and data validation), attainment deadlines, consequences, and penalties. A "track and evaluation" system is needed to assess accountability and long-term control measure effectiveness.

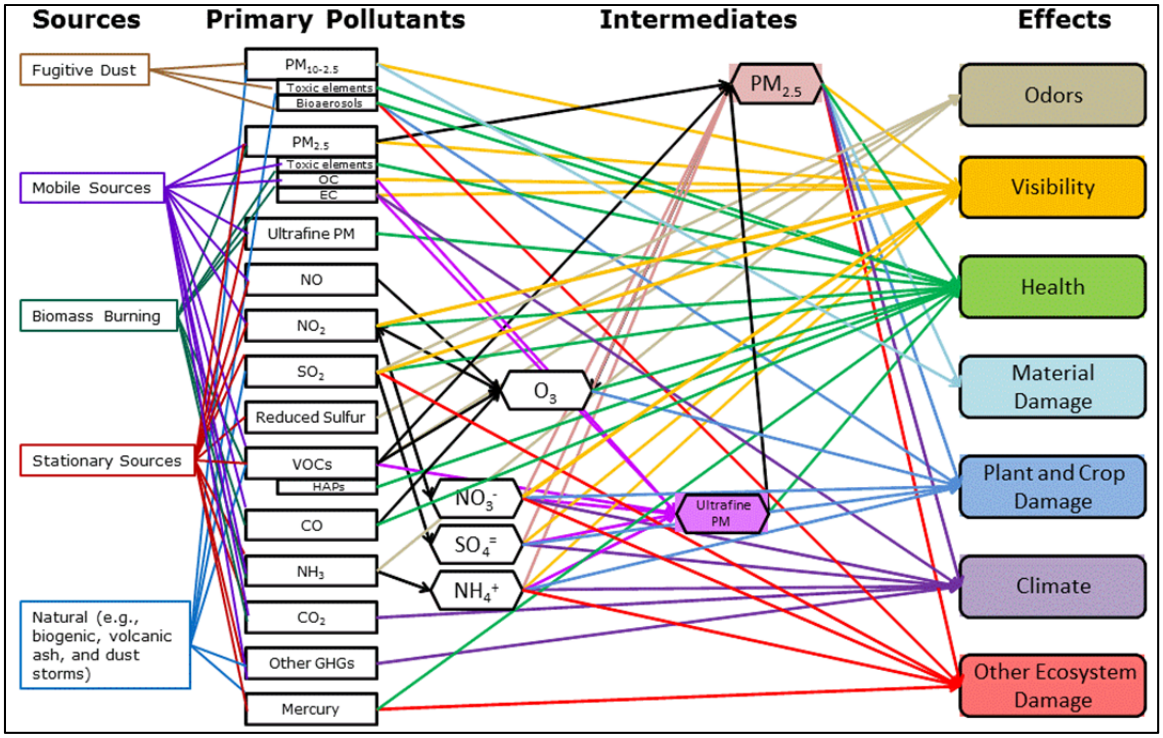

Fig. 5. Multiple pollutants and their multiple adverse effects (Baasel, 1985; Chow *et al.*, 2010a; Hidy and Pennell, 2010; Mauderly *et al.*, 2010; Chow and Watson, 2011; Hidy *et al.*, 2011). This graph shows simplified relationships and the interactions among various emission sources, their primary pollutants, the transforming product (Intermediates) in the atmosphere, and the effects by most dominant pollutants. It should be noted that the residence time of each pollutant varies (e.g., from minutes to hours for ultrafine PM (UP) to over 100 years for CO_2 and some greenhouse gases [GHGs]). In addition, the exposed concentration level, lag times, and duration dictate the extent of adverse effects. Challenges for $PM_{2.5}$ air quality management will involve its interaction with these different pollutants and effects.

China-specific guidance documents need to be produced that can be used by EPB staff to design monitoring networks, select, operate, and maintain monitoring instruments, implement quality control and quality assurance systems, and interpret the results (U.S. EPA, 1997b, 1998a, b; 2007). With consistent and healthy economic growth, China can leapfrog (Chow *et al.*, 2010c) ahead of other countries in $PM_{2.5}$ monitoring and control.

China and all other countries need to devote their resources and effort to improving air quality and continue investing in the scientific research that provides knowledge for future standards and serves as a training ground for future generations of air quality professionals.

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