Air quality management of multiple pollutants and multiple effects

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ABSTRACT

Multiple complex relationships between several pollutants and effects on human health, visibility, climate, odours, material, and ecosystems are demonstrated. There is no simple way to manage multiple effects of multiple pollutants, but consideration of these relationships will enhance the value of air quality management, add benefits to counteract the costs of emission reduction measures, and increase the probability that measures to improve one effect will not result in other adverse effects. Simple first steps include: going beyond compliance in air quality monitoring and source characterization, evaluating the effects of precursor emissions reductions on both O_3 and PM2.5 through modelling, and estimating the values of co-benefits for multiple effects.

Keywords; pollutants, management, emissions, reduction, multiple, modelling.

INTRODUCTION

Since the first recognition of and efforts to improve adverse air quality in London (Halliday 1961; Brimblecombe 1976; 1978), air quality management has involved multiple pollutants and multiple effects. Visible black smoke emanating from coal-burning chimneys, along with the acrid taste and smell of sulfur dioxide (SO_2), were easily associated with observations of black deposits on buildings and respiratory distress, respectively. Recognition of these causal relationships led to curtailments in the use of coal and improved burning practices. Visual plumes and odours were used as the primary methods to identify sources; controls on excessive emissions (Griebling 1952; Hemeon 1971) were applied only to large industrial sources until the latter part of the 20th century (Bachmann 2007; Chow *et al.* 2007b). Improved measurement methods for gases and particles (Katz 1980; Lodge 1989; Chow 1995; Watson *et al.* 1995; Kerker 1997; Spurny 1998; McMurry 2000; Chow *et al.* 2008; Chow and Watson 2011a), along with quantitative measures of adverse health effects (Vedal 1997; Pope, III and Dockery 2006; Beggs and Bennett 2011; Knibbs *et al.* 2011; Mejia *et al.* 2011), led to the definition and regulation of ambient concentrations for individual pollutants with multiple sources.

The focus on single pollutants and emphasis on their adverse health effects has been an effective air quality management strategy, as shown by downward trends in ambient concentrations of these pollutants and concomitant improvements in respiratory and cardiovascular health (Malm *et al.* 2002; Bachmann 2007; Dominici *et al.* 2007; Bahadur *et al.* 2011; Murphy *et al.* 2011). However, as ambient pollutant concentrations decrease and our knowledge concerning these emissions improves, the atmospheric transformations and synergies among pollutants and their effects are becoming evident, as illustrated in Figure 1.

Simultaneous environmental management of several pollutants is not a new concept (Baasel 1985), and examples given below demonstrate that it has been practiced for decades. Recent interest in these practices has been sparked by recommendations from the US National Academy of Sciences (NRC 2004), followed by several conceptual frameworks (Brook *et al.* 2009; Longhurst *et al.* 2009; Dominici *et al.* 2010; Greenbaum and Shaikh 2010; Hidy and Pennell 2010; Mauderly *et al.* 2010; Chow *et al.* 2010b). Little has been written concerning practical methodology and application.

In a comprehensive critical review of the topic, Hidy and Pennell (2010) proposed a risk-based approach that would limit combinations of pollutants that adversely affect human health. However, the authors were vague on the methodologies for establishing and enforcing thresholds for these combinations (Chow *et al.* 2010b). Hidy and Pennell (2010) acknowledged a lack of information for formulating guidance for this approach, especially regarding transfer functions that relate concentration to exposure, exposure to dose, and dose to adverse health effects. Mauderly and Samet (2009) argue that adverse health effects can be greater than the sum of their parts, citing animal experiments that demonstrate increased toxicity of whole diesel exhaust as compared to the sum of the gas and particle phases by themselves.

The emphasis of most multi-pollutant concepts has been on human health. Multiple benefits have been addressed, mostly through economic analyses of co-benefits, especially those associated with climate-forcing pollutants (Markandya *et al.* 2009; He *et al.* 2010; Rive and Aunan 2010; Rive 2010; Bambrick *et al.* 2011), as illustrated in Figure 2. Good economic cases can be made that greenhouse gases (GHG) emission reductions are usually accompanied by reductions in other pollutants from Figure 1, providing benefits above and beyond those needed to reduce global warming. Recognizing benefits for all adverse effects establishes greater value for a given emission reduction project.

POLLUTANTS AND MULTIPLE EFFECTS

Criteria Pollutants

Criteria pollutants are regulated in different countries, such as in Australia and New Zealand (Australian Government 2011; New Zealand Government 2011), by ambient air quality standards. The indicators nearly always include nitrogen dioxide (NO_2), SO_2, carbon monoxide (CO), ozone (O_3), suspended particulate matter (PM), and lead (Pb, Bachmann 2007; Chow *et al.* 2007b). PM is regulated according to the mass of its size fraction in the categories of Total Suspended Particles (TSP), PM10, and PM2.5 (*i.e.*, PM with aerodynamic diameters (d_p) <30 µm, 10 µm, and 2.5 µm, respectively). O_3 and PM are considered "indicators" of a broader class of pollutants. Neil Frank of the U.S. EPA (Watson *et al.* 1995) defined an indicator as "…a surrogate…to represent the agents of concern….In the case of PM, this indicator is PM mass concentration in a specified size fraction. In order to treat the regulated community fairly and to provide a uniform level of health protection across the nation, the indicator must be consistently defined in terms of stable, reproducible measurements." Air quality management of excessive O_3 and PM2.5 levels addresses multi-pollutant issues, as indicated by the relationships in Figure 1. Although primarily managed to attain benefits to public health, O_3 and PM2.5 control strategies also decrease other adverse environmental effects.

HAZARDOUS AIR POLLUTANTS (HAPS) AND TOXIC ELEMENTS

HAPs (U.S. EPA 2004; 2006), defined by the U.S. Environmental Protection Agency (EPA), include a subset of volatile organic compounds (VOCs; *e.g.*, benzene, toluene, xylene, etc.) and PM (*e.g.*, arsenic, selenium, cadmium, etc.) that are related to adverse health effects through risk factors (Lester *et al.* 2007; Chen *et al.* 2008; Craig *et al.* 2008; Linkov *et al.* 2009). Diesel particulate matter (DPM), with elemental carbon (EC) as an ambient air indicator, has been deemed a toxic air pollutant in California (Saricks *et al.* 2000; Chow 2001; Lloyd and Cackette 2001; SCAQMD 2010), with similar declarations in Australia (Marquez and Salim 2007; Nelson *et al.* 2008). HAPs are regulated by emission limits rather than ambient standards (U.S. EPA 2001).

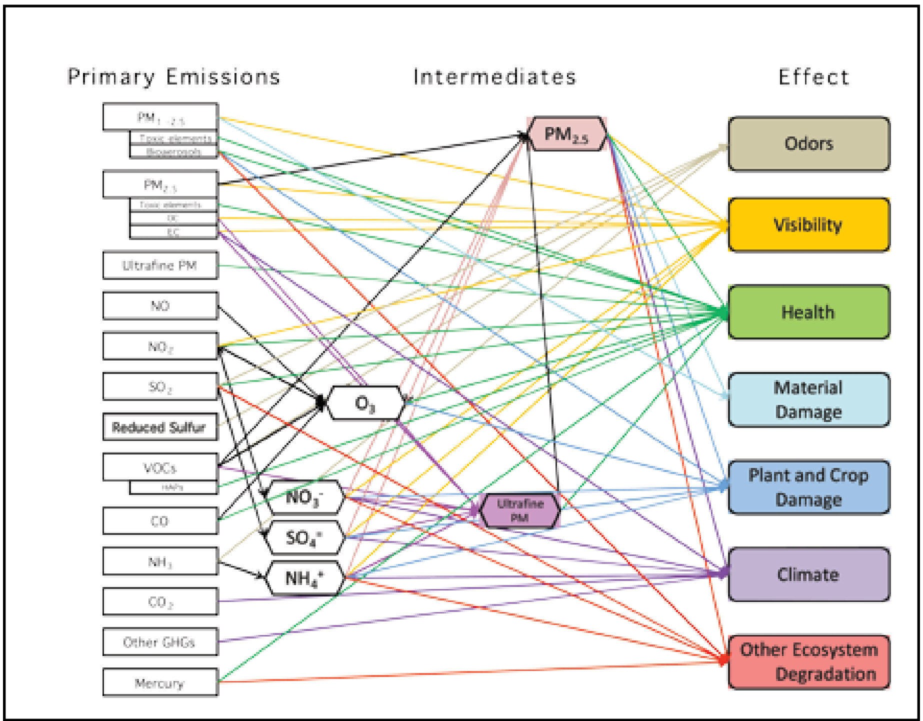

Figure 1. Simplified relationships among pollutant emissions, their transformation products, and their effects. Not illustrated are the lifetimes of these pollutants or the lag times and durations for their effects. These range from minutes to hours for ultrafine particles, to more than one-hundred years for CO_2 and some greenhouse gases (GHG). (VOCs: Volatile Organic Compounds; PM2.5: fine particulate matter [PM]; PM10-2.5: coarse PM; OC: organic carbon; EC: elemental carbon; SOA: Secondary Organic Aerosol; HAPs: Hazardous Air Pollutants).

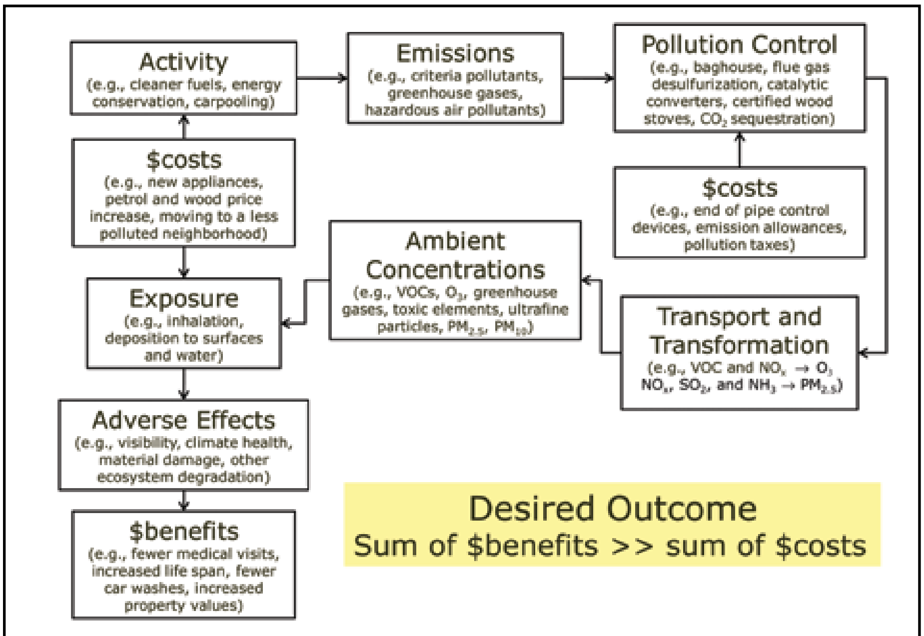

Figure 2. Illustration of the monetary inputs and outputs for a co-benefit analysis. Investments are made in modifying activities, exposure, and emission reduction technologies. Benefits include the value of money saved by avoiding adverse effects such as climate adaptation measures, health-care and associated costs, crop damage, etc.

GHG AND SHORT-LIVED CLIMATE FORCERS (SLCFS)

Carbon dioxide (CO_2), methane (CH_4), and certain halocarbons are long-lived (*i.e.*, ten to a hundred years) emittants and major climate forcing constituents (Forster *et al.* 2007; MacCracken 2008). Although SLCFs such as O_3 (Gauss *et al.* 2006; Wallack and Ramanathan 2009) and EC (Koch and del Genio 2010; Ramana *et al.* 2010; Chow *et al.* 2010a; Frieler *et al.* 2011) have shorter lifetimes (*i.e.*, hours to days), they are more amenable to rapid concentration reductions and their regulation could result in immediate mitigation of global warming and other adverse effects.

OTHER POLLUTANTS

Figure 1 shows several non-regulated pollutants that are important contributors to adverse effects. Ammonia (NH_3) interacts with nitric acid and sulfuric acid to create ammonium nitrate (NH_4NO_3) and ammonium sulfate ($(NH_4)_2SO_4$), respectively, which contribute to PM2.5 mass. These components have a great influence on visibility and ecosystems. NH_3 can be a limiting factor in $(NH_4)_2SO_4$ formation (Watson *et al.* 1994). As SO_2 levels are reduced to control sulfate levels, NH_3 may be made available to form more NH_4NO_3 (West *et al.* 1999).

Ultrafine particles (*i.e.*, d_p <0.1 μm) result from primary emissions near roadways (Morawska *et al.* 1998; Hitchins *et al.* 1999; Holmes *et al.* 2005; Ristovski *et al.* 2005; Dahl *et al.* 2006; Morawska *et al.* 2008; Knibbs *et al.* 2009; Knibbs and de Dear 2010; Knibbs *et al.* 2011) and photochemical nucleation (Holmes 2007; Chow and Watson 2007; Lee *et al.* 2008; Suni *et al.* 2008; Modini *et al.* 2009; Cheung *et al.* 2011), with deep lung penetration (Amis *et al.* 1990; Choi *et al.* 2007) and subsequent adverse health consequences (Biswas and Wu 2005; Chow *et al.* 2005) becoming recognized.

Organic carbon (OC) is often associated with EC in the form of soot from incomplete combustion, for which EC is a good indicator. OC also derives from secondary organic aerosol (SOA), which results from the oxidation of multiple VOCs as they age in the atmosphere. Only a small number of VOC and OC compounds have been characterized (Watson *et al.* 2001; Chow *et al.* 2007a; Mauderly and Chow 2008), and even smaller numbers have been related to atmospheric reactivity and HAPs. Mercury (Hg), mostly from coal combustion and biomass burning (Nelson 2007; Friedli *et al.* 2009), is not hazardous in air, but it deposits in lakes and streams where it then accumulates in fish.

EXAMPLES OF MULTI-POLLUTANT AIR QUALITY MANAGEMENT

Even within the constraints of a single-pollutant system, many air quality management strategies involve several pollutants. The following illustrate a small selection of successful approaches:

- Establishment and enforcement of vehicle emission standards (Ministry of Transport 2007; Department of Infrastructure and Transport 2011): These set limits on nitrogen oxides (NO_x), CO, and VOC emissions for both petrol- and diesel-fuelled vehicles, with additional limits for DPM.
- Elimination of Pb additives to petrol: Tetraethyl lead, originally added as an octane-booster (Kitman 2000), contaminated catalytic converters needed to attain NO_x and CO emission standards. This Pb removal provided the co-benefit of reducing ambient and blood serum Pb level (Simpson and Xu 1994; Menkes and Fawcett 1997; Cook and Gale 2005), along with lowering NO_x, CO, and VOC emissions.
- Enforcement of the US Clean Air Visibility Rule (Chow *et al.* 2002; Watson 2002): The Rule estimates chemical extinction from a weighted sum of PM2.5 chemical components and coarse particle mass, as well as detectable NO_2 levels (Pitchford *et al.* 2007) and requires continuous reductions intended to restore natural visibility conditions in the national parks and wilderness areas by 2065. This is a good example of a multi-pollutant metric.
- Promotion of clean energy development projects in China (NRC 2008): While mostly undertaken to reduce GHG emissions, energy efficiency and renewable energy efforts often reduce the use of dirtier fossil fuels that have co-benefits for other pollutants such as NO_x, SO_2, and PM.
- Monitoring the Hong Kong Hedley Environmental Index (Hedley 2011): Hedley *et al.* (2008) have established a weighted sum of NO_2, SO_2, O_3, and PM10 concentrations that relate to adverse health effects. Economic values are assigned to the adverse effects, and a running tally is broadcast on the website.

FUTURE STEPS TOWARD MULTI-POLLUTANT AIR QUALITY MANAGEMENT

There are major challenges for restructuring existing air quality networks and changing traditional methods for source characterization. Large investments have been made in establishing infrastructure for the existing networks; however, overlapping networks (*e.g.*, photochemical vs. PM) with different operations disguise real costs. To redesign or modify existing air quality management frameworks without allocating additional resources (*e.g.*, instruments, expertise) will inevitably increase the workload for local air pollution agencies. There are also deficiencies in collaboration among different disciplinary areas in seeking alternative/cost effective monitoring methods (*e.g.*, multi-tiered approach from less to more costly technology, reducing averaging times, expanding concentration ranges). Long-term commitments from government and other agencies are needed to support multi-pollutant air quality management.

Several practical measures can be phased in over time. Priority should be given to: 1) conducting advanced ambient and source monitoring beyond compliance, 2) focusing on multi-pollutant emissions reductions, and 3) evaluating co-benefits on multi-pollutant and multi-effects. Examples are as follows:

- Improve ambient monitoring (Chow and Watson 2008). The current approach to criteria pollutant monitoring requires a separate instrument for each pollutant and a costly air conditioned shelter. Advanced microsensors (Oto *et al.* 2001; Ohira and Toda 2005; Pejcic *et al.* 2007; Cho *et al.* 2008) hold promise for the design of more portable, less costly monitors that measure a variety of pollutants. For integrated PM filter samples, more information could be obtained on the relationship between ambient composition and effects through additional analyses beyond mass assessment (Chow and Watson 2011b). More detailed measurements of ultrafine particles and size distributions at higher time resolutions could enhance data already being generated by existing stations (Wang *et al.* 2009; Watson *et al.* 2011).
- Conduct real-world emission tests that are comparable to ambient measurement methods. The old single-pollutant compliance stack test methods (U.S.EPA 1996; 1997; 2008) using heated filters and glass impingers in ice buckets should be replaced by a dilution chamber integrating the same instruments used for ambient sampling (England *et al.* 2007a; 2007b). In-plume samplers with fast-response instruments could ascertain real-world emission distributions from on-road vehicles to determine the fraction of emissions from high emitters (Jayaratne *et al.* 2005; Morawska *et al.* 2007; Johnson *et al.* 2008; Nussbaum *et al.* 2009; Zhu *et al.* 2011).
- Evaluate the effects of O_3 and PM2.5 control strategies concomitantly. Because changes in NO_x and VOC emissions affect O_3 levels, as well as the nitrate and SOA portions of PM2.5, models (Zawar-Reza *et al.* 2005; Holmes and Morawska 2006; Zawar-Reza and Sturman 2008) should be capable of addressing both pollutants. It may be that excessive PM2.5 occurs during the winter, while the highest O_3 levels occur during summer. Because emission reductions are likely to be constant in all seasons, it is necessary to evaluate their effects throughout the year.
- Calculate co-benefits for reductions in multiple pollutants on multiple effects. This type of integrated assessment is still primitive, but rudimentary tools are available (IIASA 2011; U.S. EPA 2011; UrbanEmissions.info 2011) to evaluate costs and benefits when used with a proper understanding of their limitations and uncertainties.

SUMMARY AND CONCLUSION

The management of air quality for single pollutants with an emphasis on adverse human health effects has been effective for the development and application of emission reduction technologies. In the future, however, pollutants that are subsets of, or additions to, the standard criteria pollutants (*e.g.*, NO_2, SO_2, CO, O_3, PM, and Pb) should be considered not only regarding their effects on human health, but also as they relate to odours, visibility, climate, material damage, plant and crop damage, and other ecosystem degradation. Subsets of pollutants include ultrafine particles, as well as the chemical composition of primary PM, especially the carbonaceous fraction. Non-criteria gases such as NH_3 and Hg, as well as a larger subset of VOCs beyond those that are considered to be atmospherically reactive or toxic, need to be added to the standard criteria pollutants considered. Considering additional effects encourages the economic estimation of co-benefits, which further justifies and provides greater impetus for improving ambient air quality.

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