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TECHNICAL PAPER

Development of outcome-based, multipollutant mobile source indicators

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Multipollutant indicators of mobile source impacts are developed from readily available CO, NO_x, and elemental carbon (EC) data for use in air quality and epidemiologic analysis. Two types of outcome-based Integrated Mobile Source Indicators (IMSI) are assessed. The first is derived from analysis of emissions of EC, CO, and NO_x such that pollutant concentrations are mixed and weighted based on emission ratios for both gasoline and diesel vehicles. The emission-based indicators ($IMSI_{EB}$) capture the impact of mobile sources on air quality estimated from receptor models and their uncertainty is comparable to measurement and source apportionment uncertainties. The $IMSI_{EB}$ have larger correlation between two different receptor sites impacted by traffic than single pollutants, suggesting they are better indicators of the local impact of mobile sources. A sensitivity analysis of fractions of pollutants in a two-pollutant mixture and the inclusion in an epidemiologic model is conducted to develop a second set of indicators based on health outcomes. The health-based indicators ($IMSI_{HB}$) are weighted combinations of CO, NO_x, and EC pairs that have the lowest P *value in their association with cardiovascular disease emergency department visits, possibly due to their better spatial representativeness. These outcome-based, multipollutant indicators can provide support for the setting of multipollutant air quality standards and other air quality management activities.*

Implications: Integrated mobile source indicators (IMSI) were developed and assessed for use in air quality and epidemiologic analysis. IMSI contribute to fill the gap in the path towards a multipollutant air quality approach in two aspects: IMSI represent an innovative way to identify mixtures of pollutants based on outcomes and constitutes an alternative approach to assess multipollutant health effects. IMSI developed for mobile sources can be easily applied to other sources. Results can support the setting of multipollutant air quality standards.

Supplemental Material: Supplemental materials are available for this article. Go to the publisher's online edition of the *Journal of the Air & Waste Management Association* for materials showing the estimation of uncertainties using propagation of errors, comparison of source impacts from CMB and PMF and wind direction and speed for the Jefferson Street monitoring location in Atlanta.

Introduction

Air quality standards, such as the National Ambient Air Quality Standards (NAAQS) in the United States, have traditionally focused on setting maximum limits to ambient concentrations of individual pollutants. The NAAQS, and air quality standards in general, are developed from available studies, both mechanistic and epidemiological, that seek to deduce the impacts to human health from air pollution. To date, most air pollution epidemiologic work has examined associations between health outcomes and individual pollutants. However, human exposure to air pollution occurs in a multipollutant setting. Thus, a multipollutant approach may be more realistic to understanding risks and regulating urban air pollution.

Multipollutant approaches have been extensively applied in controlling emissions of pollutants to the atmosphere. Pollutants are rarely emitted in isolation by a source, and control devices for one pollutant can usually modify emissions of all of the compounds. For example, a three-way catalytic converter for gasoline vehicles is able to control nitrogen oxides, carbon monoxide, and unburned hydrocarbons at the same time (Takahashi et al., 1996; Westerholm et al., 1996). Furthermore, multipollutant control has been demonstrated to be cost-effective (U.S. Environmental Protection Agency, 2007a).

From a regulatory viewpoint, multipollutant regulations already exist for emission standards. For example, heavy- and light-duty fleets are required to meet nitrogen oxide (NO_x), carbon monoxide (CO), particulate matter (PM), hydrocarbon

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(HC), nonmethane HC (NMHC) standards (U.S. Environment Protection Agency, 2011a). In addition, U.S. Environmental Protection Agency (EPA) recently proposed the aquatic acidification index (AAI), a multipollutant index developed based on analysis of ecological effects, to be used as part of a potential combined NAAQS standard considering the combined effects of NO_x and sulfur oxide (SO_x) deposition on aquatic ecosystems (EPA, 2011b). In the past years, substantial progress has been made to move towards a result-oriented, risk-based, multipollutant approach in air quality management (NARSTO, 2010). A consistent limitation of adopting this multipollutant approach has been the identification of mixtures of pollutants in the atmosphere and the health effects of such mixtures (Hidy and Pennell, 2010; National Research Council, 2004; EPA, 2007). Statistical tools, such as factor analysis (FA), have been suggested to overcome this limitation (EPA, 2011c). Receptor models have also been used to combine pollutants in source categories and source impacts have been associated with health outcomes (Sarnat et al., 2008; Thurston et al., 2005). However, these techniques rely on an abundant amount of air quality data, including availability of specific components that are not routinely measured.

Multipollutant models in epidemiologic analysis have generally included two or more pollutants at a time within a model, with the goal of identifying confounders in associations with health rather than the effects of a mixture of pollutants (Bell et al., 2011; Dominici et al., 2010; Mauderly and Samet, 2009; Mauderly et al., 2010). Multipollutant models are subject to exposure measurement error (e.g., when surrogate measurements from central monitoring sites are used to assess human exposure), but can also have differential errors (e.g., where the pollutant measured with the least amount of error is the one with the strongest signals) and reduced statistical power (when more than one pollutant at a time is included) (Vedal and Kaufman, 2011). Moreover, the mixtures included in multipollutant models do not always represent an actual or unique source of emissions, which complicates designing effective measures to improve public health (Franklin et al., 2008; Hart et al., 2011; Lenters et al., 2010; Metzger et al., 2004; Peng et al., 2009).

Mobile source emissions have been identified as a key urban air pollution component adversely affecting public health (Beelen et al., 2008; Tonne et al., 2007). In the Atlanta area, elevated NO_2, CO, PM with an aerodynamic diameter <2.5 μm ($PM_{2.5}$), organic carbon (OC), and elemental carbon (EC) concentrations, pollutants traditionally related to traffic, have been associated with emergency department (ED) visits for cardiovascular disease (CVD) (Health Effects Institute, 2010; Metzger et al., 2004). Results from using receptor models in epidemiologic analysis provide further support that combustion-related sources are associated with CVD (Sarnat et al., 2008).

The adverse impact of mobile sources on health is due to the magnitude of these sources in the Atlanta area, where traffic emissions are estimated to account for 30% of the $PM_{2.5}$, 84% of the NO_x, and 97% of the CO emissions (EPA, 2007b). Results from source apportionment indicate that the contribution of tailpipe mobile source emissions to ambient $PM_{2.5}$ varies from 17% to 26%, and the total impact from mobile sources is likely larger considering that a significant amount of crustal material (i.e., Al, Si, Ca, Fe, K) originates from the resuspension of dust due to vehicles (Lee et al., 2008b; Liu et al., 2005; Kim et al., 2003, 2004).

Our objective in this work is to develop and assess outcome-based, multipollutant indicators for mobile sources here called integrated mobile source indicators (IMSI). We aim to investigate if more refined indicators of mobile sources are more likely to explain associations with health outcomes. IMSI are simple to construct and calculate from readily available data, and can be used in air quality and epidemiologic analyses. The species considered are CO, NO_x, and EC available from routine air quality monitoring networks. Two sets of IMSI are developed: the first is based on outcomes from analysis of pollutant emissions and observed concentrations (here called $IMSI_{EB}$). A sensitivity analysis is used to refine the indicators based on two-pollutant mixtures of NO_x&EC, NO_x&CO, and CO&EC and develop a second set of indicators based on health outcomes (here called $IMSI_{HB}$). Temporal and spatial variability of IMSI are assessed and compared with source impacts from receptor models.

Methods

IMSI development and assessment follows four steps: (i) selection of pollutants and analysis of emission inventories; (ii) development of the emission-based integrated indicators ($IMSI_{EB}$); (iii) comparison of air pollutant impact analysis using indicators with results from receptor models; and (iv) associations with acute health responses in Atlanta and development of health-based indicators ($IMSI_{HB}$).

Pollutant selection and analysis of emission inventories

Traditionally, CO and NO_x have been used as gaseous indicators of vehicular activity. CO is emitted primarily by gasoline-fueled engines, whereas both gasoline and diesel engines have substantial NO_x emissions. Mobile source–based $PM_{2.5}$ is generated not only via combustion processes but also mechanical grinding and secondary formation (i.e., formation in the atmosphere from $PM_{2.5}$ precursors under photochemical conditions). Because $PM_{2.5}$ can have several sources, it is preferable to use components that are better indicators of $PM_{2.5}$ from combustion sources. $PM_{2.5}$ EC and OC are formed during combustion, with OC being produced in early stages of combustion and EC at later stages and higher temperatures (Watson et al., 2001). OC is also formed from other processes, including secondary formation from biogenic emissions. Gasoline vehicles (GVs) usually have a higher OC/EC ratio than diesel vehicles (DVs), with values around 3.0–4.0 for GVs and below 1.0 for DVs (Lee and Russell, 2007; Zheng et al., 2007). Because diesel exhaust contains much higher EC concentrations than gasoline exhaust, EC has been used as a tracer for diesel impacts on PM (Marmur et al., 2005).

Other $PM_{2.5}$ components, including heavy metals such as zinc (Zn), nickel (Ni), vanadium (V), copper (Cu), and lead (Pb), have also been used as tracers to identify mobile source impacts on air quality, and specifically to split calculated impacts

between gasoline and diesel vehicles (Lee et al., 2008a). Zn is used as a tracer of GV, and is a component of additives in lubricating oil, Pb and Cu are produced from brake wear and road traffic, and Ni and V are found in diesel exhaust. Organic compounds, such as hopanes and polycyclic aromatic hydrocarbons (PAHs), are used as tracers of traffic-related PM impacts as well (Brook et al. 2007; Zheng et al. 2002). Although these organic compounds are very useful in the identification of specific source impacts, their measurement is more resource intensive and their concentrations are not as widely available.

We chose CO, NO_x, and EC to develop the traffic-related IMSI because these species are ubiquitous to monitoring stations in the United States and emission inventories. A detailed analysis of CO, NO_x, and EC emissions and ambient air concentrations in downtown Atlanta (Fulton County) was conducted for the period 1999–2007. Emissions from mobile sources (on-road and non-road) were obtained from the EPA National Emission Inventory (NEI) (E.P.A., 2007) and the Visibility Improvement State and Tribal Association (VISTAS) project (Georgia-DNR, 2007). The 2002 EPA NEI was consulted for emission estimates, but estimates from earlier years are expected to be used in the development of this inventory.

Additionally, we applied the EPA Motor Vehicle Emissions Simulator (MOVES 2010) to identify the fraction of emissions from on-road GVs and DVs (EPA, 2010). Both NEI and MOVES use nationwide information of vehicle miles traveled (VMT) to estimate on-road emissions, but emissions factors used in MOVES 2010 have been revised from those used in NEI.

Ambient air quality data were obtained from the Jefferson Street monitoring location (JST), a site operated by the Southeastern Aerosol Research and Characterization Study (SEARCH). Description of the measurement methods is found elsewhere (Edgerton et al., 2005; Hansen et al., 2003). Briefly, elemental carbon (EC) is measured on 24-hr $PM_{2.5}$ samples using quartz filters from a particle composition monitor (PCM) and analyzed by the thermal-optical reflectance (TOR) method at the Desert Research Institute (DRI) following the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol (Chow et al., 1993). CO, NO, and NO_2 are measured every minute and averaged to the hour. CO is measured using nondispersive infrared spectrophotometry. NO_2 is measured via photolytic conversion to NO, followed by chemiluminescence. NO and NO_2 are summed and reported as NO_x. For the period 1999–2004, a total of 1701 days were selected for use after removing days with missing data or data with high uncertainty.

An additional site, the South DeKalb (SD) monitoring station from the EPA's Speciation Trends Network (STN) located 15.3 km to the southeast of JST (Figure 1) was examined to assess the spatial variability of $IMSI_{EB}$. JST site is located within 3.2 km of interstate I-85 and 8.0 km of interstate I-20, and in close proximity to the intersection of these two highways. SD is located within 3.2 km of interstate I-285, and in close proximity to the intersection of this highway and I-20. Both JST and SD are heavily impacted by traffic emissions. The air quality monitoring is conducted at street level in both sites. At SD, NO_x was available in the same number of days at JST. For CO, SD did not record measurements before 2003, and therefore, only 562 days were available. EC is measured every third day at SD, therefore only 462 days were available.

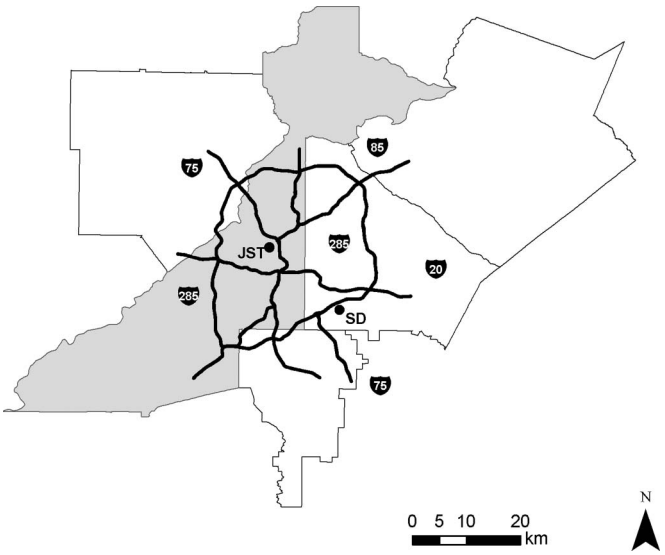

Figure 1. Location of Jefferson Street (JST) and South DeKalb (SD) monitoring stations in Atlanta. Area in gray is Fulton County in Atlanta.

Carbon monoxide (CO)

The NEI reports total CO emissions of approximately 295,000 tons/year for Fulton County in 2002, of which 97% are from mobile sources (75% on-road and 22% non-road). The on-road CO emissions estimated with MOVES are lower (approximately 190,000 tons/year) due to revisions in the emission factors from 1992 to 2002. MOVES estimates that 98% of the on-road CO emissions are from GVs and 2% are from DVs.

On a daily basis, on-road CO emission estimates are 20% higher during weekdays than weekends, indicating a decrease in GV travel during weekends. On a monthly basis, CO emissions from GVs have two periods of increase during the year (Figure 2a): June through August due to the use of air conditioning in summer, and December through February as result of cold start emissions and lower ambient temperatures (EPA, 2008).

Ambient CO concentrations are the lowest during the summer months (Figure 2a), especially during June and July when dispersion of contaminants is favored and CO photochemical destruction is faster. The highest concentrations are found from October through December when wind speeds are slower (Figure S1 in the Supplemental Material) and thermal inversion episodes trap pollutants nearer the ground. From January through March, inversions are still present, but wind speeds are significantly higher than prior months resulting in greater dispersion of pollutants.

On an annual basis, there is a clear trend between reductions in ambient air concentrations of CO and emission reductions from 1999 to 2007 (Figure 2b). Comparison of CO emission

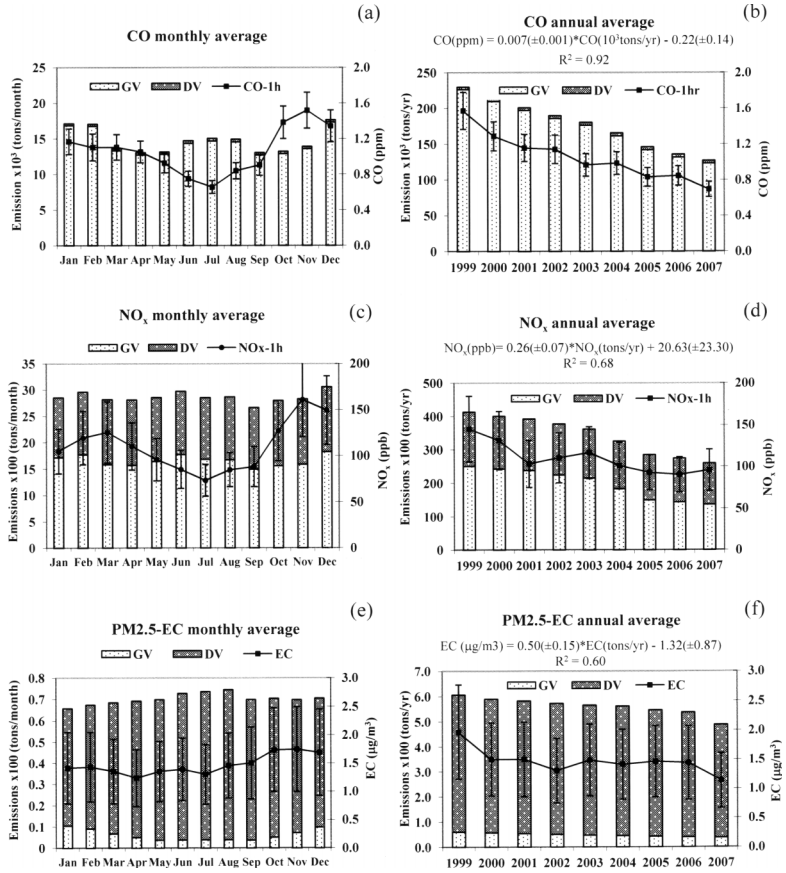

Figure 2. Monthly and annual trends of CO, NO_x, and EC. Bars represent emissions estimates from MOVES in tons/month (a, c, e) or tons/yr (b, d, f). Bold line represents ambient air concentrations of CO (ppm), NO_x (ppb), and EC ($\mu g/m^3$) on right vertical scale. Error bars are the root mean square (RMS) error of daily uncertainties from measurements. R^2 denotes the correlation between annual emissions and annual average concentrations.

estimates from MOVES and NEI in 1999, 2002, and 2005 shows good agreement between both methods. The reduction of CO emissions in 2007 with respect to 1999 was 48% in NEI and 45% in MOVES.

These results support using CO concentrations as an indicator of GV impacts, though this indicator is limited to local sources and can lead to potential biases in a regional assessment.

Nitrogen oxides (NO_x)

The NEI reports total NO_x emissions of approximately 47,000 tons/year for Fulton County in 2002, of which 87% are from mobile sources (72% on-road and 15% non-road). NO_x emissions estimated with MOVES are lower than the NEI estimates (approximately 38,000 tons/year) due to lower exhaust emissions from heavy-duty vehicles (U.S. Environmental Protection Agency, 2010). MOVES estimates that 60% of the on-road NO_x emissions are from GVs and 40% are from DVs. Although diesel engines emit more NO_x than spark ignition engines per mile traveled, the gasoline fleet is significantly larger (about 10 times) than the diesel fleet for Fulton County (Blanchard et al., 2010). Other sources contributing to NO_x emissions are classified as area and point sources, in particular fuel combustion in electrical generating utilities (EGUs) and biomass burning. It is expected, however, that most of the NO_x impacting the receptor stations come from mobiles sources, because EGUs have high effective stack heights such that pollutants are better dispersed before impacting the monitor station at the surface. Further, NO_x emissions from point sources were 13% of the total emissions in 1999, but only 2% in 2007. EPA has estimated that nationwide reductions of NO_x are the result of emissions reductions at electrical utilities and among on-road mobile sources (EPA, 2008).

On a weekly basis, NO_x ambient air concentrations are 24% higher on weekdays than on weekends, consistent with a larger reduction of DV than GV traffic during weekends. On a monthly basis, NO_x follows a similar trend to CO, with higher concentrations in winter and lower concentrations during summer (Figure 2c) when NO_x is more rapidly removed by photochemical reactions. NO_x emissions from DVs are relatively constant throughout the year, whereas NO_x emissions from GVs have a similar trend to CO emissions, increasing in summer months due to the use of air conditioning (A/C) systems and in winter months due to cold start emissions and lower ambient temperatures (EPA, 2008).

On a yearly basis, NO_x ambient concentrations decreased from 1999 to 2001, increased during 2002 and 2003, and decreased again until 2007 (Figure 2d). The significant reduction during the period 1999–2001 (from 154 to 103 ppb) is likely a result of the implementation of the EPA acid rain program and stationary controls to reduce ozone, combined with mobile

source reductions (EPA, 2005). From 2002 to 2007, reductions in ambient NO_x are attributed largely to decreases in on-road NO_x emissions. NO_x emissions from on-road sources have a stronger correlation with ambient NO_x during the period 2002–2007 ($R^2 = 0.65$) than 1999–2007 ($R^2 = 0.36$).

These results indicate that mobile source NO_x emissions have a large impact on ambient NO_x concentrations, but are not as dominant as mobile source CO emissions on ambient CO concentrations.

Elemental carbon (EC)

VISTAS estimates of EC emissions for 2002 is 92% from mobiles sources (on-road and non-road) and 8% from other sources, such as biomass burning. From the on-road fraction, MOVES estimates 91% from DVs and 9% from GVs. On a weekly basis, EC concentrations are 30% higher during weekdays than weekends, due to the higher fraction of diesel vehicle traffic on weekdays. On a monthly basis, EC concentrations are lowest in spring and summer as compared with the October–December period. During the cooler months, dispersion of pollutants is not favored due to increased thermal inversions and reduced wind speeds (Figure 2e).

EC emissions from DVs increase in summer due to an increase in VMT and greater construction activity with the subsequent increase in non-road emissions. Although not as large as emissions from DVs, EC emissions from GVs can be an important source during winter time because of cold starts and lower ambient temperatures (U.S. Environmental Protection Agency, 2008).

On an annual basis, EC concentrations decreased from 1.97 $\mu g/m^3$ in 1999 to 1.13 $\mu g/m^3$ in 2007 (Figure 2f) as a result of changes in fuel composition and controls on mobile sources, such as the introduction of low- (50 ppm) and ultra-low- (15 ppm) sulfur diesel in 2002 and 2006, respectively (EPA, 2000). Controls in point and open fires might also have helped on this reduction, such as the open burning ban implemented in the 13-county metro Atlanta area in 1996. These results indicate that EC emissions are dominated by mobile sources for Fulton County, particularly by DVs.

In summary, data from Atlanta using a number of emissions and monitoring databases show that CO and EC are likely good indicators of GVs and DVs, respectively. NO_x appears to be an indicator of overall mobile sources and cannot be easily used to discern between GVs and DVs. Because only 20% of the total OC emissions are from mobiles sources (on-road and non-road) (Georgia-DNR, 2007) and a fraction between 26% and 47% of ambient OC can be formed secondarily (Pachon et al., 2010), our development of $IMSI_{EB}$ does not use OC, though the indicators can be used to estimate the OC from mobile sources.

Development of the emission-based integrated indicators

We propose a multipollutant indicator of CO, NO_x, and EC to assess mobile source impacts on air quality. In this work, the highest 1-hr values at each day for CO and NO_x were chosen because the 1-hr metric has been found more associated with health outcomes than other metrics (Metzger et al., 2004). The $IMSI_{EB}$ uses ratios of mobile-source-to-total emissions for each pollutant as weighting coefficients (eq 1). Mobile source fractions of each pollutant can be estimated by multiplying these ratios by the ambient air concentrations. For example, the fraction of ambient EC from mobile sources is estimated here as the total EC concentration multiplied by the ratio ($E_{EC,mobile}/E_{EC,total}$), where $E_{i,j}$ represents the emission of pollutant i for source category j, in units of tons/year estimated at Fulton County in Atlanta. Pollutant concentrations for EC are in units of $\mu g/m^3$, which differ from the units of ppm used for CO and NO_x. Therefore, a normalization of ambient air concentrations by the standard deviation of 1701 samples for EC, CO, and NO_x was performed (Bevington and Robinson, 2003). The normalization is also useful in this case because each pollutant has a different range of measurements.

$$IMSI_{EB} = \frac{\frac{E_{EC,mobile}}{E_{EC,total}} \times C'_{EC} + \frac{E_{NOx,mobile}}{E_{NOx,total}} \times C'_{NOx} + \frac{E_{CO,mobile}}{E_{CO,total}} \times C'_{CO}}{\frac{E_{EC,mobile}}{E_{EC,total}} + \frac{E_{NOx,mobile}}{E_{NOx,total}} + \frac{E_{CO,mobile}}{E_{CO,total}}} \quad (1)$$

where $C'_i = C_i/\sigma_i$ denotes the normalized concentrations (C_i and σ_i represent the average concentration and standard deviation of pollutant i in the period 1999–2007) at the Jefferson Street receptor site. The $IMSI_{EB}$ uses normalization by the sum of the emission ratios in such a way that the indicator can be easily compared with other IMSIs.

The ratios between mobile emissions ($E_{i,mobile}$) and total emissions ($E_{i,total}$) in eq 1 are defined as the weighting coefficients. These weighting coefficients for NO_x and CO, obtained from the NEI database, are 0.84 ± 0.03 and 0.97 ± 0.01, respectively. Uncertainties in the NO_x and CO mobile/total ratios are obtained as 1 standard deviation of the estimated ratios in the period 1999–2007. The weighting coefficient for EC was estimated from VISTAS in 2002 to 0.92 ± 0.04. Uncertainty in the EC mobile/EC total ratio is obtained as one standard deviation of the monthly values during 2002. After replacing the values of the weighting coefficients into eq 1, this can be expressed as

$$IMSI_{EB} = 0.33 \times C'_{EC} + 0.31 \times C'_{NOx} + 0.36 \times C'_{CO} \quad (2)$$

We were also interested in differentiating impacts from gasoline and diesel exhaust emissions, because the contribution at the receptor site can be quite different and the control mechanisms are specific to each type of vehicle. Therefore, we define integrated indicators for gasoline vehicles ($IMSI_{EB,GV}$) and diesel vehicles ($IMSI_{EB,DV}$) using specific emission ratios from gasoline and diesel emissions estimated with MOVES.

The ratio of gasoline-to-mobile emissions was used as a weighting coefficient for each species, being 0.58 ± 0.02 for NO_x and 0.98 ± 0.01 for CO, obtained from the application of MOVES. For EC, the ratio of gasoline-to-mobile emissions is more seasonally dependent, with a summer value of 0.06 ± 0.01 and a winter value of 0.12 ± 0.04. These results indicate more weight on the CO and NO_x than EC. Therefore, $IMSI_{EB,GV}$ was defined as a weighting mixture of CO and NO_x only:

$$\mathrm{IMSI_{EB,GV}} = \frac{\frac{E_{\mathrm{NOx,GV}}}{E_{\mathrm{NOx,total}}} C'_{\mathrm{NOx}} + \frac{E_{\mathrm{CO,GV}}}{E_{\mathrm{CO,total}}} C'_{\mathrm{CO}}}{\frac{E_{\mathrm{NOx,GV}}}{E_{\mathrm{NOx,total}}} + \frac{E_{\mathrm{CO,GV}}}{E_{\mathrm{CO,total}}}} \quad (3)$$

where $\frac{E_{\mathrm{NOx,GV}}}{E_{\mathrm{NOx,total}}} = \frac{E_{\mathrm{NOx,GV}}}{E_{\mathrm{NOx,mobile}}} \times \frac{E_{\mathrm{NOx,mobile}}}{E_{\mathrm{NOx,total}}}, \frac{E_{\mathrm{CO,GV}}}{E_{\mathrm{CO,total}}} = \frac{E_{\mathrm{CO,GV}}}{E_{\mathrm{CO,mobile}}} \times \frac{E_{\mathrm{CO,mobile}}}{E_{\mathrm{CO,total}}}$.

After replacing the values of the weighting coefficients into eq 3, this can be expressed as

$$\mathrm{IMSI_{EB,GV}} = 0.37 \times C'_{\mathrm{NOx}} + 0.63 \times C'_{\mathrm{CO}} \quad (4)$$

Similarly, the ratios of diesel-to-total mobile emissions obtained from MOVES, used as weighting coefficients, for NO_x was 0.42 ± 0.02 and for CO 0.02 ± 0.01. For EC, the ratio of diesel-to-total mobile emissions was more seasonally dependent, with a summer value of 0.94 ± 0.01 and a winter value of 0.88 ± 0.04. These results imply more weight on EC and NO_x than CO. Therefore, $\mathrm{IMSI_{EB,DV}}$ was defined as a weighting mixture of EC and NO_x only:

$$\mathrm{IMSI_{EB,DV}} = \frac{\frac{E_{\mathrm{NOx,DV}}}{E_{\mathrm{NOx,total}}} C'_{\mathrm{NOx}} + \frac{E_{\mathrm{CO,DV}}}{E_{\mathrm{CO,total}}} C'_{\mathrm{CO}}}{\frac{E_{\mathrm{NOx,DV}}}{E_{\mathrm{NOx,total}}} + \frac{E_{\mathrm{CO,DV}}}{E_{\mathrm{CO,total}}}} \quad (5)$$

where $\frac{E_{\mathrm{NOx,DV}}}{E_{\mathrm{NOx,total}}} = \frac{E_{\mathrm{NOx,DV}}}{E_{\mathrm{NOx,mobile}}} \times \frac{E_{\mathrm{NOx,mobile}}}{E_{\mathrm{NOx,total}}}, \frac{E_{\mathrm{CO,DV}}}{E_{\mathrm{CO,total}}} = \frac{E_{\mathrm{CO,DV}}}{E_{\mathrm{CO,mobile}}} \times \frac{E_{\mathrm{CO,mobile}}}{E_{\mathrm{CO,total}}}$.

After replacing the values of the weighting coefficients into eq 5, this can be expressed as

$$\mathrm{IMSI_{EB,DV}} = 0.31 \times C'_{\mathrm{NOx}} + 0.69 \times C'_{\mathrm{EC}} \quad (6)$$

Specification of the $\mathrm{IMSI_{EB}}$ has limitations. First, the emission fraction for a pollutant is translated to an ambient fraction assuming that the average source and receptor fractions are the same. This assumption has been tested in the past with good results: gas-to-$PM_{2.5}$ emission ratios were used to optimize source profiles in Atlanta, leading to a more accurate apportionment of $PM_{2.5}$ from mobile sources and coal-fire power plants (Marmur et al., 2005). A second limitation is the assumption that emission ratios are constant on a daily basis and equal to the annual average. However, daily variations in the indicators are captured by the use of ambient air concentrations.

Comparison of air pollutant impact analysis using indicators with results from receptor models

The $\mathrm{IMSI_{EB}}$ were compared with source apportionment from receptor models. The chemical mass balance method CMBv.8.2 (Watson et al., 1984) and the positive matrix factorization method PMFv3.0 (Norris and Vedantham, 2008) were applied to the same period of time as the indicators. For PMF, strong species (i.e., species with signal/noise ratio >2.0) were NO_3, SO_4, NH_4, EC, four OC thermal fractions, Al, Si, Fe, K, Ca, Br, Mn, and Zn, and weak species (i.e., species with signal/noise ratio <2.0) were $PM_{2.5}$, Cu, Pb, and Se. The temperature-resolved OC fractions were chosen to improve the separation between gasoline and diesel source impacts (Kim et al., 2004; Liu et al., 2005). Missing data were replaced by their geometric mean to conserve the original number of samples for better performance of the PMF algorithm (Reff et al., 2007). For CMB, optimized source profiles were chosen from a previous study in Atlanta and sources, such as coal-fire power plant and cement production, were eliminated to avoid negative source impacts (Marmur et al., 2005).

Associations between pollutant mixtures and acute health responses in Atlanta

We have assumed that pollutants mix based on emission ratios, but this mixture can be influenced by meteorological conditions. The sensitivity analysis aims to assess different fractions on which pollutants can combine once in the atmosphere. Based on the combination of pollutants in the $\mathrm{IMSI_{EB,GV}}$ and $\mathrm{IMSI_{EB,DV}}$, a sensitivity analysis was performed between two-pollutant mixtures. For $\mathrm{IMSI_{EB,GV}}$ mixtures of CO and NO_x were evaluated and for $\mathrm{IMSI_{EB,DV}}$ mixtures of EC and NO_x were chosen. In addition, mixtures of EC and CO were also evaluated. The sensitivity analysis was performed as follows.

$$\mathrm{NO_x\&EC} = \alpha \times C'_{\mathrm{NOx}} + (1-\alpha) \times C'_{\mathrm{EC}} \quad (7)$$

$$\mathrm{NO_x\&CO} = \alpha \times C'_{\mathrm{NOx}} + (1-\alpha) \times C'_{\mathrm{CO}} \quad (8)$$

$$\mathrm{CO\&EC} = \alpha \times C'_{\mathrm{CO}} + (1-\alpha) \times C'_{\mathrm{EC}} \quad (9)$$

where C'_{EC}, C'_{CO}, and C'_{NOx} represent the normalized concentrations and α is a parameter that varies from 0 to 1, in such a way that allows comparing pollutants individually (when α is equal to 0 or 1) versus two-pollutant mixtures. In particular, the combination of NO_x&CO at $\alpha = 0.4$ (i.e., $0.4 \times C'_{NOx} + 0.6 \times C'_{CO}$) is equivalent to $\mathrm{IMSI_{EB,GV}}$ (eq 4) and the combination of NO_x&EC at $\alpha = 0.3$ (i.e., $0.3 \times C'_{NOx} + 0.7 \times C'_{EC}$) is equivalent to $\mathrm{IMSI_{EB,DV}}$ (eq 6).

The association of mobile source multipollutant metrics with health effects was assessed in an epidemiologic analysis. CVD ED visits were chosen as the health endpoint for this analysis, given that those have been found to be associated with combustion-related activities in Atlanta (Metzger et al., 2004; Sarnat et al., 2008). Our epidemiologic models only assess short-term effects of these pollutants (i.e., we look at the daily measure of the indicator and see if it predicts that same day's CVD count). Assessing relationships between chronic exposures to the indicators (i.e., long-term averages of the exposures) is beyond the scope of analysis. Briefly, data were collected for ED visits related to CVD from 41 hospitals in metro Atlanta from 1999 to 2004. Daily ED counts are regressed with air pollution indicators using a Poisson generalized linear model (GLM). We used a similar epidemiologic model to the model applied in previous analyses of CVD outcomes in these data; this Poisson generalized linear model is described in more detail elsewhere (Metzger et al., 2004; Sarnat et al., 2008). However, for this analysis, as well as other more recent analyses of these data, we included more stringent control for temperature (Strickland et al., 2010).

Unlike traditional multipollutant models that solve for different regression coefficients, our approach solves for only one β for a multipollutant indicator. The values of α at which the two-

pollutant mixtures show the strongest association with CVD define the health-based integrated indicators ($IMSI_{HB}$).

Results

The assessment of the $IMSI_{EB}$ was conducted as follows: (i) analysis of $IMSI_{EB}$ trends; (ii) comparison with mobile source impacts from receptor models; (iii) uncertainties in the estimation of the indicators; (iv) comparison with $IMSI_{HB}$ derived from associations with CVD ED visits; and (v) implication for multipollutant air quality standards.

$IMSI_{EB}$ trends

$IMSI_{EB}$ exhibit similar temporal trends as ambient EC, CO, and NO_x, with a decrease during summer and increase during fall and reduction in annual averages from 1999 to 2007 (Figure 3). On a monthly basis, $IMSI_{EB,GV}$ showed a larger reduction in concentrations during summer than $IMSI_{EB,DV}$ consistent with less commuting from light-duty traffic during the summer months. On an annual basis, there is a larger decrease in $IMSI_{EB,GV}$ than $IMSI_{EB,DV}$ explained by a more rapid introduction of new control technologies in the gasoline fleet than diesel vehicles. The comparison of $IMSI_{EB}$ annual averages with reduction in emissions of CO, NO_x, and EC with respect to 1999 shows a similar trend, suggesting a good agreement between indicators estimated with ambient air concentrations and emissions from mobile sources.

On a daily basis, $IMSI_{EB}$ is 22% larger during weekdays than weekends, capturing the increase in traffic during the week. The $IMSI_{EB,DV}$ trend has a large decrease during weekends (30%) than $IMSI_{EB,GV}$ (14%), and is explained by reduced diesel traffic during the weekend.

Comparison with results from receptor models

CMB and PMF methods yield similar estimates for PM air quality impacts for the chosen sources (Table S1 in the Supplemental Material). The correlation between gasoline and diesel source impacts resolved by CMB and PMF (Table 1) was substantially lower than the correlation of the combined fractions into one mobile source (R^2 = 0.75/0.51 in winter/summer), which demonstrates the difficulties receptor models have in adequately capturing the split between gasoline and diesel daily contributions. The proposed $IMSI_{EB}$ correlates more strongly with total mobile source impacts from CMB (R^2 = 0.73/0.53 in winter/summer) than PMF (R^2 = 0.72/0.48 in winter/summer). The $IMSI_{EB,DV}$ correlates stronger with diesel source impacts from CMB and PMF than the corresponding correlation of $IMSI_{EB,GV}$ with gasoline source impacts from the receptor models. This is due to both CMB and PMF using EC as a fitting species to solve for diesel contributions, and $IMSI_{EB,DV}$ is heavily weighted by EC, whereas $IMSI_{EB,GV}$ uses CO, a species that is typically not used to fit the CMB or PMF gasoline source categories.

In the southeastern United States, biomass burning can also be a significant source of EC, CO, and NO_x to ambient air (Lee et al., 2008a). $IMSI_{EB}$ does not seem to be influenced by daily impacts from this source. On selected days with biomass burning activity over 4.0 $\mu g/m^3$ of $PM_{2.5}$ identified by both CMB and PMF, the integrated indicators were more strongly correlated with source impacts from mobile sources than wood burning. On those days, the correlation between $IMSI_{EB}$ and mobile source impacts from CMB (R^2 = 0.86) or PMF (R^2 = 0.83) were significantly higher than the correlation between $IMSI_{EB}$ and source impacts from biomass burning from CMB (R^2 = 0.13) or PMF (R^2 = 0.34). This result supports the emissions estimates and analyses detailed above that found emissions of EC, CO, and NO_x in Atlanta predominantly from mobile sources.

Uncertainties in mobile source indicators

Uncertainties are involved in several steps of the calculations (e.g., emission estimates, ambient measurements, receptor models), and we estimate uncertainties in $IMSI_{EB}$ and compare them with uncertainties from singles species and receptor model source contributions. Uncertainties in ambient measurements (EC, CO, NO_x) were estimated as one-third of the detection limit ($\mu g/m^3$ or ppm) plus the product of analytical uncertainty (percentage) and concentration (Polissar et al., 1998). Both detection limit and instrument uncertainty were obtained from the SEARCH study (Hansen et al., 2003). Uncertainties in CMB source impacts are estimated using the effective variance method, which considers uncertainties in both source profiles and ambient concentrations (Lee and Russell, 2007). In PMF,

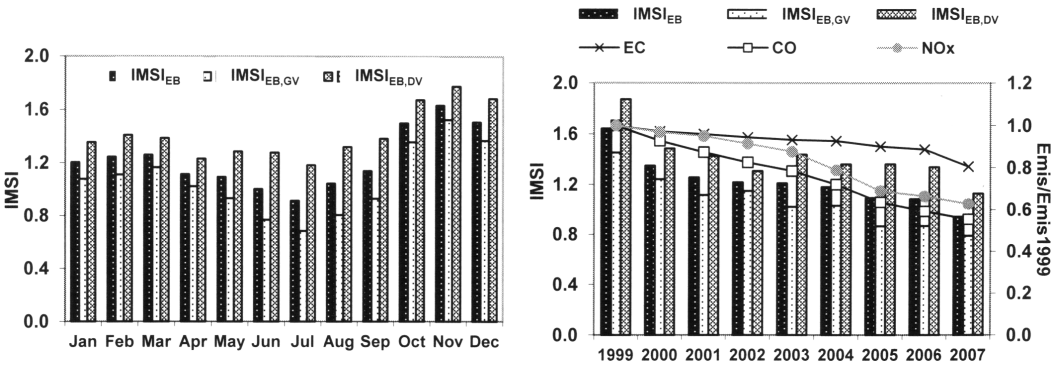

Figure 3. Temporal trends (a) monthly (b) annual of $IMSI_{EB}$, $IMSI_{EB,GV}$, and $IMSI_{EB,DV}$ (unitless). The indicators are normalized such as they have a standard deviation of 1. Annual trend is compared with reduction in emissions of CO, NO_x, and EC with respect to 1999 (on right *y*-axes).

Table 1. Correlations between $IMSI_{EB}$, $IMSI_{EB,GV}$, and $IMSI_{EB,DV}$ with single species and daily source impacts from CMB and PMF

	(R^2)											
	EC	NO_x, 1-hr	CO, 1-hr	PMF_{DV}	PMF_{GV}	PMF_{mob}	CMB_{GV}	CMB_{DV}	CMB_{mob}	$IMSI_{EB}$	$IMSI_{EB,GV}$	$IMSI_{EB,DV}$
EC	**1.00**	0.46	0.57	0.49	0.56	0.72	0.43	0.88	0.86	0.77	0.64	0.9
NO_x, 1-hr	0.37	**1.00**	0.53	0.32	0.40	0.50	0.26	0.38	0.45	0.79	0.76	0.75
CO, 1-hr	0.28	0.49	**1.00**	0.41	0.43	0.58	0.39	0.42	0.58	0.87	0.94	0.67
PMF_{DV}	0.34	0.23	0.17	**1.00**	0.20	0.75	0.46	0.31	0.57	0.49	0.44	0.50
PMF_{GV}	0.25	0.11	0.11	0.00	**1.00**	0.69	0.28	0.47	0.51	0.55	0.49	0.59
PMF_{mob}	0.55	0.31	0.27	0.63	0.44	**1.00**	0.51	0.53	0.75	0.72	0.64	0.75
CMB_{GV}	0.17	0.10	0.08	0.20	0.01	0.17	**1.00**	0.13	0.78	0.43	0.41	0.43
CMB_{DV}	0.86	0.27	0.20	0.22	0.26	0.44	0.01	**1.00**	0.58	0.62	0.49	0.78
CMB_{mob}	0.77	0.32	0.23	0.39	0.16	0.51	0.62	0.45	**1.00**	0.73	0.64	0.81
$IMSI_{EB}$	0.66	0.81	0.77	0.32	0.19	0.48	0.15	0.52	0.53	**1.00**	0.97	0.92
$IMSI_{EB,GV}$	0.39	0.76	0.92	0.23	0.13	0.34	0.11	0.29	0.33	0.92	**1.00**	0.81
$IMSI_{EB,DV}$	0.91	0.67	0.43	0.37	0.23	0.56	0.18	0.76	0.72	0.86	0.62	**1.00**

Notes: Values in the upper right represent correlations during winter time (October–March); values in the lower left represent correlations during summer time (April–September). PMF = positive matrix factorization model; CMB = chemical mass balance method; GV = gasoline vehicles; DV = diesel vehicles; mob = mobile sources = GV+DV.

uncertainties in factor contributions are not given explicitly by the model, so a bootstrapping procedure is used (Norris and Vedantham, 2008). Uncertainties in $IMSI_{EB}$ are propagated from individual uncertainties taking into account that CO, NO_x, and EC are correlated between each other and, therefore, covariance terms need to be included (International Organization for Standardization, 1993). Application of the propagation of errors in the estimation of the $IMSI_{EB}$ uncertainties is explained in detail in the Supplemental Material.

Among single species, CO and NO_x have lower uncertainties than EC because gas species are typically more precisely measured than filter-based $PM_{2.5}$ species measurements (Table 2). The $IMSI_{EB}$ show uncertainties larger than the ambient measurements, because uncertainties in emissions ratios are involved in addition to the ambient measurement uncertainties. Furthermore, the calculation of uncertainties in $IMSI_{EB}$ includes the uncertainty provided by correlated quantities, as it has been demonstrated they can impact uncertainty results (Espinosa et al., 2010). Here, the uncertainty in $IMSI_{EB}$ is primarily driven by the uncertainties in the covariance relationships between EC, CO, and NO_x. For $IMSI_{EB,GV}$, the uncertainties are driven by the covariance relationships of CO and NO_x. For $IMSI_{EB,DV}$, uncertainties are mostly driven by uncertainties in EC measurements and emissions ratios and the covariance relationships of EC and NO_x.

The range of uncertainties of $IMSI_{EB}$ (0.51–0.68) is comparable to the range of uncertainties in PMF (0.26–0.67), which is a typical method to estimate contributions from mobile sources. The high range of uncertainties in CMB (1.00–1.48) is mostly explained by uncertainties in the source profiles (Lee and Russell, 2007).

Table 2. Comparison of uncertainties between indicators

Indicator	Indicator Value[a]	Standard Deviation[b]	Uncertainty[c]	Relative Uncertainty
CO, 1-hr (ppm)	1.16	1.00	0.16	0.14
NO_x, 1-hr (ppm)	0.12	0.10	0.03	0.25
EC ($\mu g/m^3$)	1.53	0.97	0.64	0.42
$IMSI_{EB}$	1.31	0.90	0.72	0.55
$IMSI_{EB,GV}$	1.17	0.96	0.80	0.68
$IMSI_{EB,DV}$	1.48	0.94	0.76	0.51
PMF_{mob}	2.94	2.30	1.11	0.38
PMF_{GV}	1.37	1.21	0.36	0.26
PMF_{DV}	1.57	1.65	1.05	0.67
CMB_{mob}	2.54	1.70	2.53	1.00
CMB_{GV}	1.34	1.15	2.00	1.48
CMB_{DV}	1.27	1.02	1.60	1.26

Notes: [a]Average of daily values from 1999 to 2004. [b]For daily values from 1999 to 2004. [c]Uncertainties are estimated as the RMS average $\left(\overline{\sigma_i} = \sqrt{\frac{1}{N}\sum \sigma_i^2}\right)$ of daily uncertainties from 1999 to 2004.

$IMSI_{HB}$ derived from associations with CVD ED visits

The epidemiologic model was implemented with time series of pollutant concentrations (CO, NO_x, EC), sources impacts (from CMB and PMF), $IMSI_{EB}$, $IMSI_{EB,GV}$ $IMSI_{EB,DV}$ and the two-pollutant mixtures from the sensitivity analysis. A total of 40 metrics were evaluated and compared for the daily association between the metric and corresponding ED visits for CVD in the period 1999–2004. Within the single species, NO_x, 1-hr, was the most strongly associated with CVD, followed by CO, 1-hr; EC was found borderline significant (Table 3). Pollutants that are better measured such as CO and NO_x typically have stronger associations in epidemiologic models (Vedal and Kaufman, 2011), and NO_x has been proposed as indicator of toxic species emitted by traffic (Brook et al., 2007). When the three pollutants are combined to form the $IMSI_{EB}$, the strength of association is only slightly greater than either for EC or CO, 1-hr, separately, but less than NO_x, 1-hr. Using $IMSI_{EB,GV}$ and $IMSI_{EB,DV}$ as predictors of CVD-related ED visits in the model resulted in statistically significance associations. Gasoline and diesel source impacts from CMB and PMF were not found to be significantly associated with CVD. The lack of significance may be due to the use of EC as a fitting species in both CMB and PMF. Differences in time periods, health outcomes, and analytic methods (e.g., tighter controls in the epidemiologic analyses) may explain the significant association between mobile source daily contributions and CVD found in other studies in the Atlanta area (Metzger et al., 2004; Sarnat et al., 2008).

The inclusion of the two-pollutant mixtures (NO_x&EC, NO_x&CO, CO&EC) in the epidemiologic model is represented in three curves with a minimum point where the association with CVD is strongest (Figure 4). The minimum point suggests that the combinations of pollutants at specific fractions are more likely to explain associations with health outcomes than individual species. The minimum point in the curves occurs at $\alpha = 0.6$ for NO_x&CO, at $\alpha = 0.7$ for NO_x&EC, and at $\alpha = 0.5$ for CO&EC. It would seem reasonable that a large fraction of NO_x in the NO_x&CO and NO_x&EC pairs would give a stronger significance to the association with CVD outcomes, but this is not the case. Similarly, for the CO&EC pair one could also expect that a larger fraction of CO in the mixture would give greater significance. The minimum occurs when both pollutants are approximately equally weighted.

The points where the association of the two-pollutant mixtures and CVD is strongest defined the health-based indicators. That is, we define $IMSI_{HB,NC}$ at $\alpha = 0.6$ for NO_x&CO, $IMSI_{HB,NE}$ at $\alpha = 0.7$ for NO_x&EC, and $IMSI_{HB,CE}$ at $\alpha = 0.5$ for CO&EC. Pollutants used in the two-pollutant $IMSI_{HB}$ are denoted by a subscript, e.g., "NE" for NO_x and EC mixture. The $IMSI_{HB}$ hold different fractions of pollutants than the $IMSI_{EB}$, suggesting that other sources, such as biomass burning, may be contributing in the association with CVD though the *P* values are relatively constant between both integrated indicators. The advantage of using $IMSI_{EB}$ is that specific control mechanisms can be suggested to mobile sources facilitating the work of policy makers.

The IMSI and the two-pollutant mixtures showing different associations with CVD outcomes at different α values can be

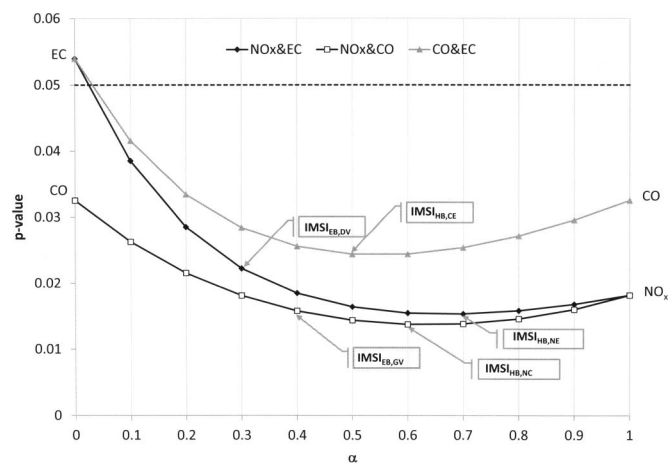

Figure 4. Sensitivity analysis of the association between pairs of pollutants and CVD outcomes; the dashed line represents *P* value = 0.05.

Table 3. Results for the associations of ED for CVD with mobile source impacts metrics (sorted by *P* value)

Indicator	IQR[a]	RR[b] per IQR	95% CI[c]	*P* Value
CMB-DV	1.0 $\mu g/m^3$	1.005	0.997–1.014	0.206
PMF-GV	2.3 $\mu g/m^3$	1.005	0.997–1.012	0.206
PMF-DV	2.3 $\mu g/m^3$	1.006	0.998–1.014	0.168
CMB-GV	1.0 $\mu g/m^3$	1.006	0.999–1.012	0.079
EC	1.0 $\mu g/m^3$	1.008	1.000–1.017	0.054
CO, 1-hr	0.9 ppm	1.007	1.001–1.014	0.033
$IMSI_{EB}$	0.9	1.007	1.001–1.014	0.029
$IMSI_{EB,DV}$	0.7	1.010	1.001–1.018	0.022
NO_x, 1-hr	0.1 ppm	1.008	1.001–1.015	0.018
$IMSI_{EB,GV}$	0.8	1.009	1.002–1.017	0.018

Notes: [a]Interquartile range; [b]risk ratio; [c]confidence interval. CMB = chemical mass balance method; PMF = positive matrix factorization model; GV = gasoline vehicles; DV = diesel vehicles; $IMSI_{EB}$ = emission-based integrated mobile source indicator; $IMSI_{EB,GV}$ = $IMSI_{EB}$ estimated for gasoline vehicles; $IMSI_{EB,DV}$ = $IMSI_{EB}$ estimated for diesel vehicles.

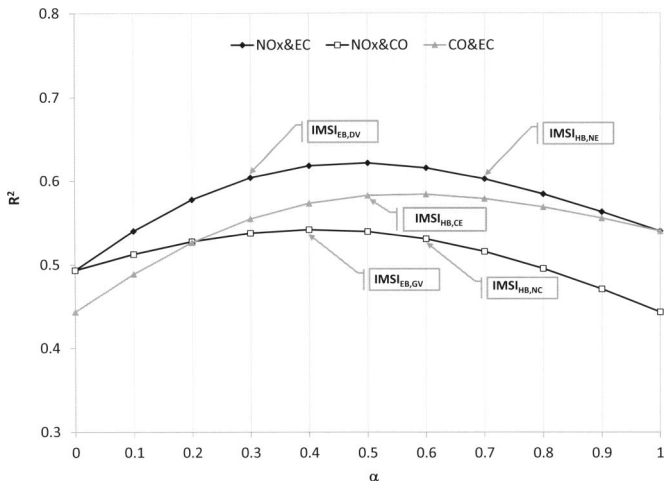

Figure 5. Correlation (R^2) between binary mixtures and the pollutant not included in the mixture (NO_x&EC vs CO; NO_x&CO vs EC; CO&EC vs NO_x). Vertical scale starts at 0.3 to emphasize correlations.

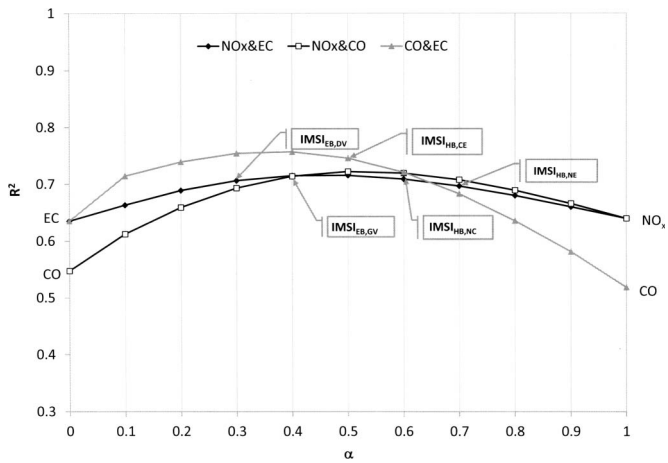

Figure 6. Correlation (R^2) between binary mixtures at two different receptor sites JST and SD. Vertical scale starts at 0.3 to emphasize correlations. Overlapped days at JST and SD: NO_x&EC ($n = 398$), NO_x&CO ($n = 560$), and CO&EC ($n = 173$).

partially explained by the correlations between individual pollutants and the two-pollutant mixture and more clearly by the correlation between indicators at two different receptor sites.

With respect to the first explanation, the correlation of each pair of pollutants and the third pollutant not included in the pair changes with the α fraction (Figure 5). For example, the correlation between the NO_x&EC mixture and CO has a maximum value at $\alpha = 0.5$ ($R^2 = 0.63$), which is larger than the correlation between CO and EC ($R^2 = 0.49$) or CO and NO_x ($R^2 = 0.55$). Similarly, the binary mixtures NO_x&CO and CO&EC have stronger correlations with EC and NO_x at $\alpha = 0.4$ and $\alpha = 0.6$, respectively.

A higher correlation of a binary mixture with a third pollutant more strongly associated with CVD, may explain the higher association of that mixture with the health outcome. For example, the strongest association of the CO&EC mixture with CVD at $\alpha = 0.5$ (Figure 4) might be explained by the higher correlation of the CO&EC pair and NO_x at that particular alpha (Figure 5). However, this is not the case for NO_x&EC and NO_x&CO pairs, which already include the statistical power of NO_x in the health association.

A more clear explanation can be found in the correlation between indicators at different receptor sites. We observe that the correlations between pairs of pollutants estimated at JST and the corresponding pairs calculated at SD are stronger at certain value of α than others (Figure 6). These values are $\alpha = 0.5$ for NO_x&EC ($R^2 = 0.72$) and NO_x&CO ($R^2 = 0.72$) and $\alpha = 0.4$ for CO&EC ($R^2 = 0.76$). The correlations between pair of pollutants at JST and SD are stronger than the correlations between single pollutants at both receptor sites ($R^2 = 0.64$ for EC; $R^2 = 0.55$ for CO; $R^2 = 0.59$ for NO_x). Previous studies have found that for pollutants with large spatial error, health associations are likely attenuated (Goldman et al., 2010). Pollutant mixtures having a stronger correlation between sites than single pollutants may offer a better explanation of the stronger association with health.

As a result of the greater correlation between sites, $IMSI_{EB,GV}$ and $IMSI_{EB,DV}$ constitute better indicators for mobile sources, which are expected to be ubiquitous in the area. This result complements a previous analysis in which mobile source impacts were classified as having "intermediate" spatial representativeness of the Atlanta area due to the lack of a unique marker (EC was used as the only mobile tracer) (Marmur et al., 2006). Using $IMSI_{EB}$, mobile source impacts show a more robust representativeness in the area.

Finally, the association of pollutant mixtures with health outcomes could be related to interaction between pollutants as has been shown in laboratory studies (Mauderly and Samet, 2009). This hypothesis needs further study.

Implications for multipollutant air quality standards

$IMSI_{EB}$ and $IMSI_{HB}$ can provide support to the setting of multipollutant air quality standards in a manner similar to that used in the development of the aquatic acidification index (AAI) (EPA, 2011b). The AAI was designed to take into account the combined effects of NO_x and SO_x in the acidification of aquatic ecosystems, given that these two species are linked from atmospheric chemistry. Similarly, $IMSI_{HB}$ assess the effects of mixtures of pollutants associated with mobile sources on health and $IMSI_{EB}$ assess mixtures representing the gasoline and diesel vehicle impacts on air quality. Because mobile sources and their composition are ubiquitous, it is expected that the integrated indicators can be applied in other cities. IMSI are simple to construct and calculate and can be estimated at any monitoring site where EC, CO, and NO_x concentrations are available.

Conclusions

This study proposed an approach to develop multipollutant indicators based on analysis of emissions inventories and health outcomes. The $IMSI_{EB}$ are simple to construct and calculate and demonstrate advantages over the use of single species: $IMSI_{EB}$ have stronger correlation between two different receptor sites

than single pollutants, suggesting they are better indicators of the local impact of mobile sources, they agree well with the observed trends of traffic, and they have stronger associations with observed health effects, possibly due to their stronger correlation between sites. Uncertainties in $IMSI_{EB}$ are similar to uncertainties in ambient measurements and receptor models. A sensitivity analysis of fractions in $IMSI_{EB}$ led to the development of $IMSI_{HB}$, suggesting mixtures of pollutants more strongly associated with CVD outcomes.

The use of IMSI in epidemiologic modeling constitutes an alternative approach to assess the health impact of pollutant mixtures. Although the approach presented in this article was developed for mobile sources, this work can be extended to other sources. IMSI can support the setting of multipollutant air quality standards because they represent the impact of traffic on health.

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