



Published in final edited form as:

J Air Waste Manag Assoc. 2012 April ; 62(4): 431–442.

Development of Outcome-based, Multipollutant Mobile Source Indicators

Jorge E. Pachon^{1,3}, Sivaraman Balachandran¹, Yongtao Hu¹, James A. Mulholland¹, Lyndsey A. Darrow², Jeremy A. Sarnat², Paige E. Tolbert², and Armistead G. Russell¹

¹Georgia Institute of Technology, School of Civil and Environmental Engineering, Atlanta, GA, USA

²Emory University, Rollins School of Public Health, Atlanta, GA, USA

³Universidad de La Salle, Programa de Ingeniería Ambiental, Bogota, Colombia

Abstract

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.

INTRODUCTION

Air quality standards, such as the National Ambient Air Quality Standards (NAAQS) in the US, 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.

Please address correspondence to: Ted Russell, School of Civil and Environmental Engineering, Georgia Institute of Technology, 331 Ferst Dr., Atlanta, GA, 30332; phone +1-404-894-3079; fax +1-404-894-8266; ted.russell@ce.gatech.edu.

Jorge Pachon is assistant professor in the School of Environmental Engineering at Universidad de La Salle in Bogota, Colombia. Sivaraman Balachandran is graduate research assistants, Yongtao Hu is a research scientist, Jim Mulholland is a professor, and Ted Russell is the Georgia Power Professor of Environmental Engineering in the School of Civil and Environmental Engineering at the Georgia Institute of Technology. Lyndsey Darrow and Jeremy Sarnat are assistant professors and Paige Tolbert is the chair of the Department of Environmental Health at the Rollins School of Public Health at Emory University.

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.¹⁻² Furthermore, multipollutant control has been demonstrated to be cost-effective.³

From a regulatory viewpoint, multipollutant regulations already exist for emission standards. For example, heavy and light-duty fleets are required to meet NO_x, CO, PM, HC, NMHC standards.⁴ In addition, 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 SO_x deposition on aquatic ecosystems.⁵

In the past years, substantial progress has been made to move towards a result-oriented, risk-based, multipollutant approach in air quality management.⁶ 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.^{3, 7-8} Statistical tools, such as factor analysis (FA), have been suggested to overcome this limitation.⁹ Receptor models have also been used to combine pollutants in source categories and source impacts have been associated with health outcomes.¹⁰⁻¹¹ 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.¹²⁻¹⁵ 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).¹⁶ 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.¹⁷⁻²¹

Mobile source emissions have been identified as a key urban air pollution component adversely affecting public health.²²⁻²³ In the Atlanta area, elevated NO₂, CO, PM_{2.5}, organic carbon (OC) and EC concentrations, pollutants traditionally related to traffic, have been associated with Emergency Department (ED) visits for cardiovascular disease (CVD).^{18, 24} Results from using receptor models in epidemiologic analysis provide further support that combustion-related sources are associated with CVD.¹⁰

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 NO_x emissions and 97% of CO emissions.²⁵ 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 re-suspension of dust due to vehicles.²⁶⁻²⁹

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. IMSIs 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 IMSIs 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 IMSIs 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, while 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). Since 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.³⁰ OC is also formed from other processes, including secondary formation from biogenic emissions. Gasoline vehicles (GV) usually have a higher OC/EC ratio than diesel vehicles (DV), with values around 3.0–4.0 for GV and below 1.0 for DV.^{31–32} Because diesel exhaust contains much higher EC concentrations than gasoline exhaust, EC has been used as a tracer for diesel impacts on PM.³³

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.²⁹ 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.^{34–35} While 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 US and emissions 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)²⁵ and the Visibility Improvement State and Tribal Association (VISTAS) project.³⁶ 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 GV and DV.³⁷ Both NEI and MOVES use

nation-wide 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.^{38–39} Briefly, elemental carbon (EC) is measured on 24-hour 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 NO₂ are measured every minute and averaged to the hour. CO is measured using non-dispersive infrared spectrophotometry. NO₂ is measured via photolytic conversion to NO, followed by chemiluminescence. NO and NO₂ 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 (Fig. 1) was examined to assess the spatial variability of IMSI_{EB}. JST site is located within two miles of interstate I-85 and five miles of interstate I-20, and in close proximity to the intersection of these two highways. SD is located within two miles 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.

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 GV and 2% are from DV.

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 GV have two periods of increase during the year (Fig. 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.⁴⁰

Ambient CO concentrations are the lowest during the summer months (Fig. 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 (Fig. S1 in the SI) 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 (Fig. 2b). Comparison of CO emission 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.³⁷ MOVES estimates that 60% of the on-road NO_x emissions are from GV and 40% are from DV. 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.⁴¹ Other sources contributing to NO_x emissions are classified as area and point sources, in particular fuel combustion in electrical generating utilities (EGU) and biomass burning. It is expected, however, that most of the NO_x impacting the receptor stations come from mobile sources, because EGU 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 nation-wide reductions of NO_x are the result of emissions reductions at electrical utilities and among on-road mobile sources.⁴²

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 (Fig. 2c) when NO_x is more rapidly removed by photochemical reactions. NO_x emissions from DV are relatively constant throughout the year, whereas NO_x emissions from GV have a similar trend to CO emissions, increasing in summer months due to the use of A/C systems and in winter months due to cold start emissions and lower ambient temperatures.⁴⁰

On a yearly basis, NO_x ambient concentrations decreased from 1999 to 2001, increased during 2002 and 2003, and decreased again until 2007 (Fig. 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.⁴³ From 2002–2007, 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 mobile sources (on-road and non-road) and 8% from other sources, such as biomass burning. From the on-road fraction, MOVES estimates 91% from DV and 9% from GV. 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 to the October–December period. During the cooler months, dispersion of pollutants is not favored due to increased thermal inversions and reduced wind speeds (Fig. 2e).

EC emissions from DV 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 DV, EC emissions from GV can be an important source during winter time because of cold starts and lower ambient temperatures.⁴⁰

On an annual basis, EC concentrations decreased from 1.97 $\mu\text{g}/\text{m}^3$ in 1999 to 1.13 $\mu\text{g}/\text{m}^3$ in 2007 (Fig. 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, respectively⁴⁴. 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 DV.

In summary, data from Atlanta using a number of emissions and monitoring databases show that CO and EC are likely good indicators of GV and DV, respectively. NO_x appears to be an indicator of overall mobile sources and cannot be easily used to discern between GV and DV. Since only 20% of the total OC emissions are from mobile sources (on-road and non-road)³⁶ and a fraction between 26–47% of ambient OC can be formed secondarily⁴⁵, 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 1hr values at each day for CO and NO_x were chosen because the 1hr metric has been found more associated with health outcomes than other metrics.¹⁸ 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 ($E_{\text{EC,mobile}}/E_{\text{EC,total}}$), 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 $\mu\text{g}/\text{m}^3$, 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.⁴⁶ The normalization is also useful in this case since each pollutant has a different range of measurements.

$$\text{IMSI}_{\text{EB}} = \frac{\frac{E_{\text{EC,mobile}}}{E_{\text{EC,total}}} * C'_{\text{EC}} + \frac{E_{\text{NO}_x,\text{mobile}}}{E_{\text{NO}_x,\text{total}}} * C'_{\text{NO}_x} + \frac{E_{\text{CO,mobile}}}{E_{\text{CO,total}}} * C'_{\text{CO}}}{\frac{E_{\text{EC,mobile}}}{E_{\text{EC,total}}} + \frac{E_{\text{NO}_x,\text{mobile}}}{E_{\text{NO}_x,\text{total}}} + \frac{E_{\text{CO,mobile}}}{E_{\text{CO,total}}}} \quad (1)$$

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 St 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 IMSI.

The ratios between mobile emissions ($E_{i,\text{mobile}}$) and total emissions ($E_{i,\text{total}}$) 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 one 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:

$$IMSI_{EB} = 0.33 * C_{EC}' + 0.31 * C_{NOx}' + 0.36 * C_{CO}' \quad (2)$$

We were also interested in differentiating impacts from gasoline and diesel exhaust emissions, since 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:

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

where $\frac{E_{NOx,GV}}{E_{NOx,total}} = \frac{E_{NOx,GV}}{E_{NOx,mobile}} * \frac{E_{NOx,mobile}}{E_{NOx,total}}$, $\frac{E_{CO,GV}}{E_{CO,total}} = \frac{E_{CO,GV}}{E_{CO,mobile}} * \frac{E_{CO,mobile}}{E_{CO,total}}$. After replacing the values of the weighting coefficients into eq 3, this can be expressed as:

$$IMSI_{EB,GV} = 0.37 * C_{NOx}' + 0.63 * C_{CO}' \quad (4)$$

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, $IMSI_{EB,DV}$ was defined as a weighting mixture of EC and NO_x only:

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

where $\frac{E_{NOx,DV}}{E_{NOx,total}} = \frac{E_{NOx,DV}}{E_{NOx,mobile}} * \frac{E_{NOx,mobile}}{E_{NOx,total}}$, $\frac{E_{CO,DV}}{E_{CO,total}} = \frac{E_{CO,DV}}{E_{CO,mobile}} * \frac{E_{CO,mobile}}{E_{CO,total}}$. After replacing the values of the weighting coefficients into eq 5, this can be expressed as:

$$IMSI_{EB,DV} = 0.31 * C_{NOx}' + 0.69 * C_{EC}' \quad (6)$$

Specification of the $IMSI_{EB}$ 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.³³ 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 $IMSI_{EB}$ were compared with source apportionment from receptor models. The chemical mass balance method CMBv.8.2⁴⁷ and the Positive Matrix Factorization method PMFv3.0⁴⁸ 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, Zn and weak species (i.e., species with signal/noise ratio < 2.0) were $PM_{2.5}$, Cu, Pb, Se. The temperature-resolved OC fractions were chosen to improve the separation between gasoline and diesel source impacts.^{27–28} Missing data were replaced by their geometric mean to conserve the original number of samples for better performance of the PMF algorithm.⁴⁹ 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.³³

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 $IMSI_{EB,GV}$ and $IMSI_{EB,DV}$, a sensitivity analysis was performed between two-pollutant mixtures. For $IMSI_{EB,GV}$, mixtures of CO and NO_x were evaluated and for $IMSI_{EB,DV}$ 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.

$$NO_x \& EC = \alpha * C_{NO_x}' + (1-\alpha) * C_{EC}' \quad (7)$$

$$NO_x \& CO = \alpha * C_{NO_x}' + (1-\alpha) * C_{CO}' \quad (8)$$

$$CO \& EC = \alpha * C_{CO}' + (1-\alpha) * C_{EC}' \quad (9)$$

where C_{EC}' , C_{CO}' and C_{NO_x}' 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., $0.4 * C_{NO_x}' + 0.6 * C_{CO}'$) is equivalent to $IMSI_{EB,GV}$ (eq. 4) and the combination of NO_x & EC at $\alpha=0.3$ (i.e., $0.3 * C_{NO_x}' + 0.7 * C_{EC}'$) is equivalent to $IMSI_{EB,DV}$ (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 as those have been found to be associated with combustion-related activities in Atlanta.^{10, 18} 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.^{10, 18} However, for this analysis, as well as other more recent analyses of these data, we included more stringent control for temperature.⁵⁰

Unlike traditional multipollutant models that solve for different regression coefficients, our approach solves for only one β for a multipollutant indicator. The values of alpha 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 (Fig 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 SI). 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 US, biomass burning can also be a significant source of EC, CO and NO_x to ambient air.⁵¹ $IMSI_{EB}$ does not seem to be influenced by daily impacts from this source. On selected days with biomass burning activity over $4.0 \mu\text{g}/\text{m}^3$ 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 single species and receptor model source contributions. Uncertainties in ambient measurements (EC, CO, NO_x) were estimated as one-third of the detection limit ($\mu\text{g}/\text{m}^3$ or ppm) plus the product of analytical uncertainty (percentage) and concentration.⁵² Both detection limit and instrument uncertainty were obtained from the SEARCH study.³⁸ Uncertainties in CMB source impacts are estimated using the effective variance method, which considers uncertainties in both source profiles and ambient concentrations.³¹ In PMF, uncertainties in factor contributions are not given explicitly by the model, so a bootstrapping procedure is used⁴⁸. 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.⁵³ Application of the propagation of errors in the estimation of the IMSI_{EB} uncertainties is explained in detail in the Supplemental Information (SI).

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 since 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.⁵⁴ 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.³¹

IMSI_{HB} Derived from Associations with CVD ED Visits

The epidemiologic model was implemented with time series of pollutant concentrations (CO, NO_x, EC), source 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 -1hr was the most strongly associated with CVD, followed by CO-1hr; 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,¹⁶ and NO_x has been proposed as indicator of toxic species emitted by traffic.³⁵ 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-1hr separately, but less than NO_x -1hr. Using IMSI_{EB,GV} and IMSI_{EB,DV} as predictors of CVD-related ED visits in the model resulted in statistically significant 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.^{10, 18}

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 (Fig. 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 $\text{IMSI}_{\text{HB,NC}}$ at $\alpha=0.6$ for NO_x &CO, $\text{IMSI}_{\text{HB,NE}}$ at $\alpha=0.7$ for NO_x &EC, and $\text{IMSI}_{\text{HB,CE}}$ 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 IMSIs and the two-pollutant mixtures showing different associations with CVD outcomes at different α values can be 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 (Fig. 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 $\alpha=0.5$ (Fig. 4) might be explained by the higher correlation of the CO&EC pair and NO_x at that particular alpha (Fig. 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 (Fig. 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.⁵⁵ 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).⁵⁶ 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.¹² 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).⁵ 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. Since mobile sources and their composition are ubiquitous, it is expected that the integrated indicators can be applied in other cities. IMSIs 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 IMSIs in epidemiologic modeling constitutes an alternative approach to assess the health impact of pollutant mixtures. Although the approach presented in this manuscript was developed for mobile sources, this work can be extended to other sources. $IMSI$ can support the setting of multipollutant air quality standards since they represent the impact of traffic on health.

Acknowledgments

This project is funded by the Environmental Protection Agency of the United States of America under grants Number R83362601 and R83386601, Georgia Power, the Southern Company, and the Colombian Institute for the Development of Science and Technology COLCIENCIAS. We thank Dr. Eric Edgerton at ARA, Inc for access to the SEARCH data.

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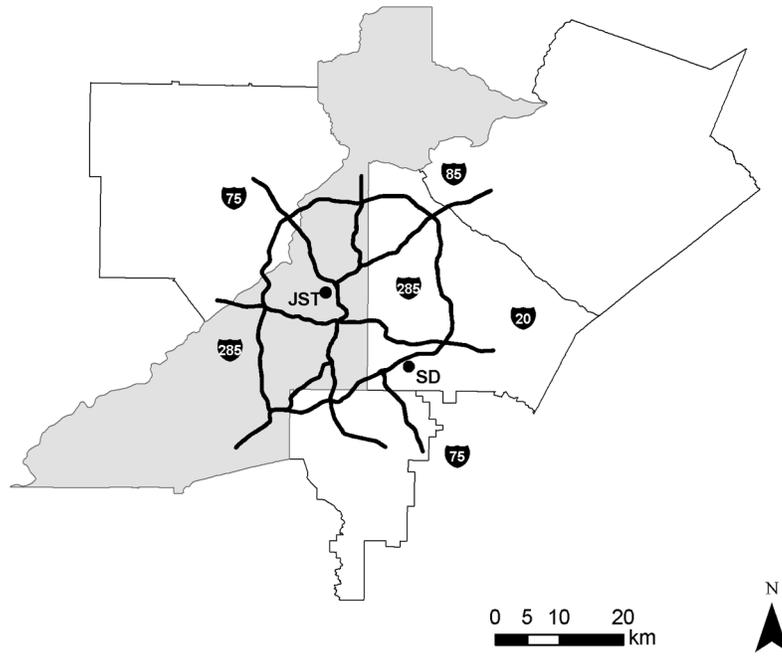


Figure 1.
Location of Jefferson Street (JST) and South DeKalb (SD) monitoring stations in Atlanta.
Area in gray is Fulton County in Atlanta.

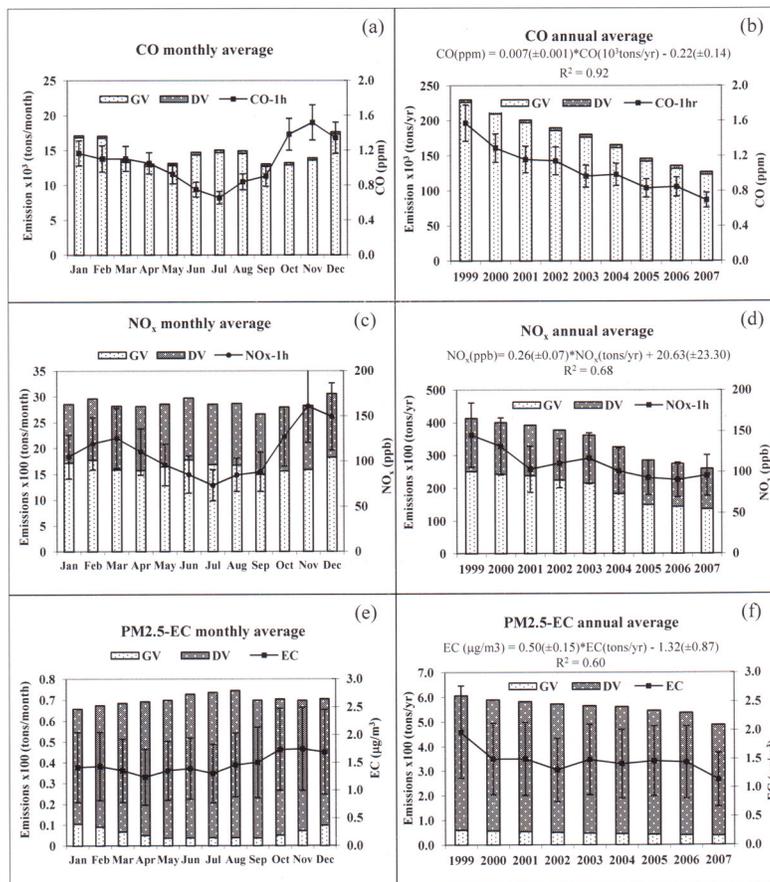


Figure 2. Monthly and annual trends of CO, NO_x and EC. Bars represent emissions estimates from MOVES in tons/month (a, c, e) or tons/yr (b, d, f). Bold line represents ambient air concentrations of CO (ppm), NO_x (ppb) and EC (µg/m³) on right vertical scale. Error bars are the root mean square (RMS) error of daily uncertainties from measurements. R² denotes the correlation between annual emissions and annual average concentrations.

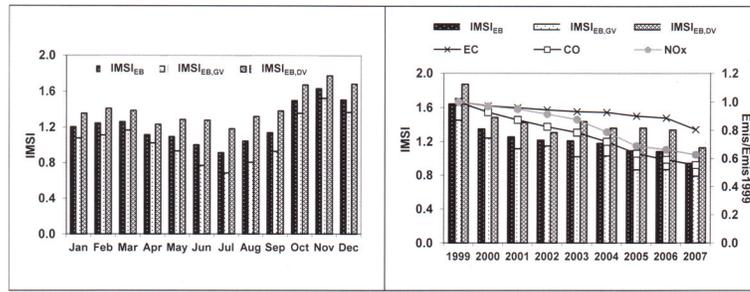


Figure 3. Temporal trends (a) monthly (b) annual of $IMSI_{EB}$, $IMSI_{EB,GV}$ and $IMSI_{EB,DV}$ (unitless). The indicators are normalized such as they have a standard deviation of one. Annual trend is compare with reduction in emissions of CO, NO_x and EC with respect to 1999 (on right y axes).

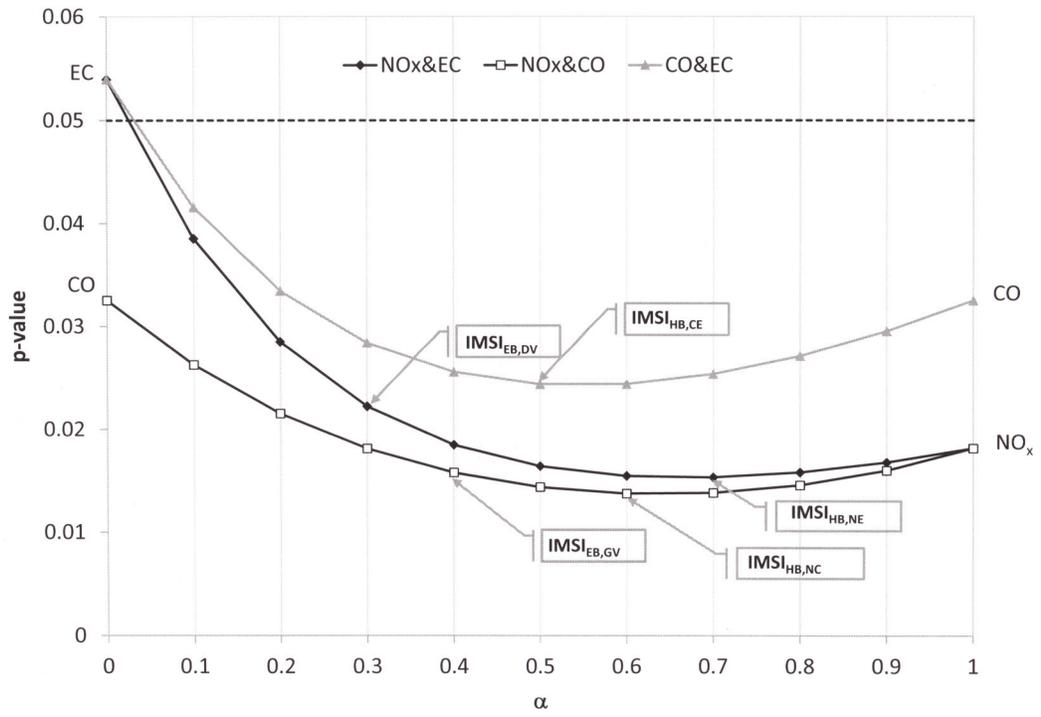


Figure 4. Sensitivity analysis of the association between pairs of pollutants and CVD outcomes; the dashed line represents p-value = 0.05.

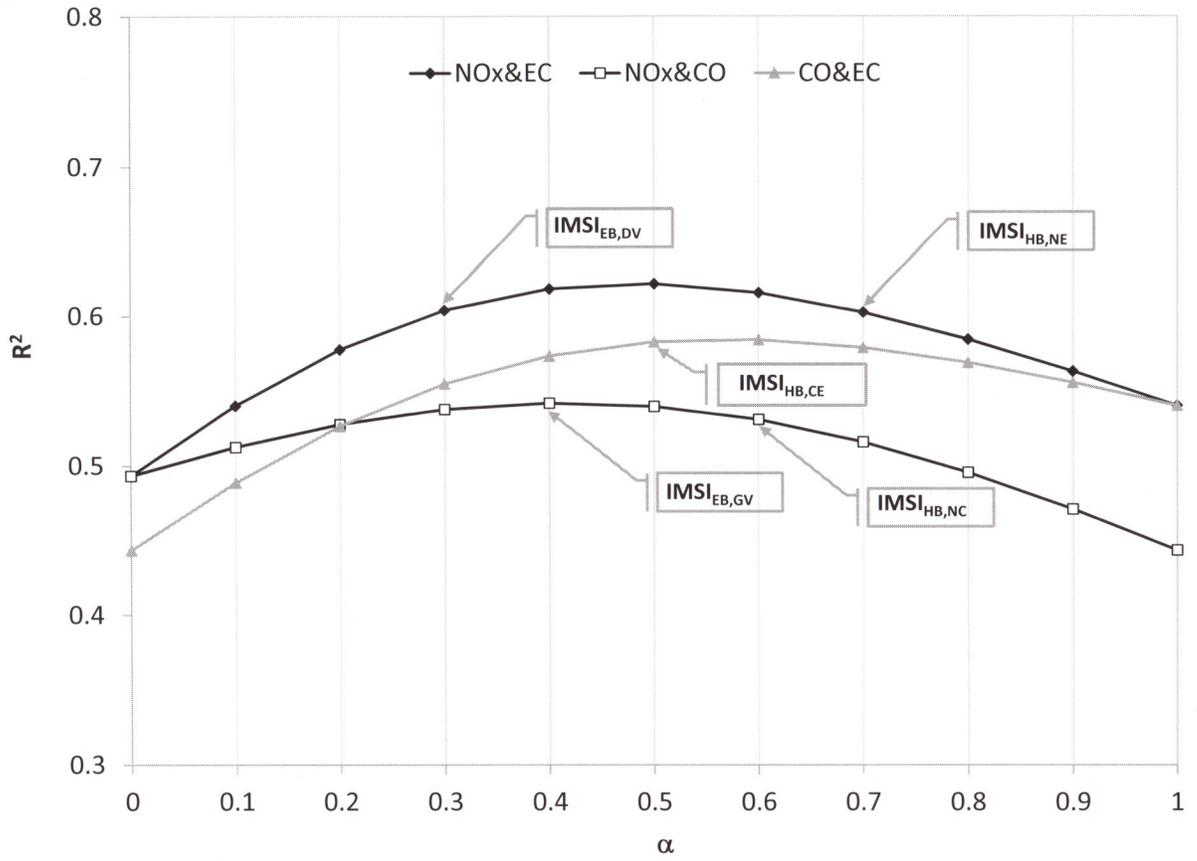


Figure 5. Correlation (R^2) between binary mixtures and the pollutant not included in the mixture ($\text{NO}_x\&\text{EC}$ vs CO ; $\text{NO}_x\&\text{CO}$ vs EC ; $\text{CO}\&\text{EC}$ vs NO_x). Vertical scale starts at 0.3 to emphasize correlations.

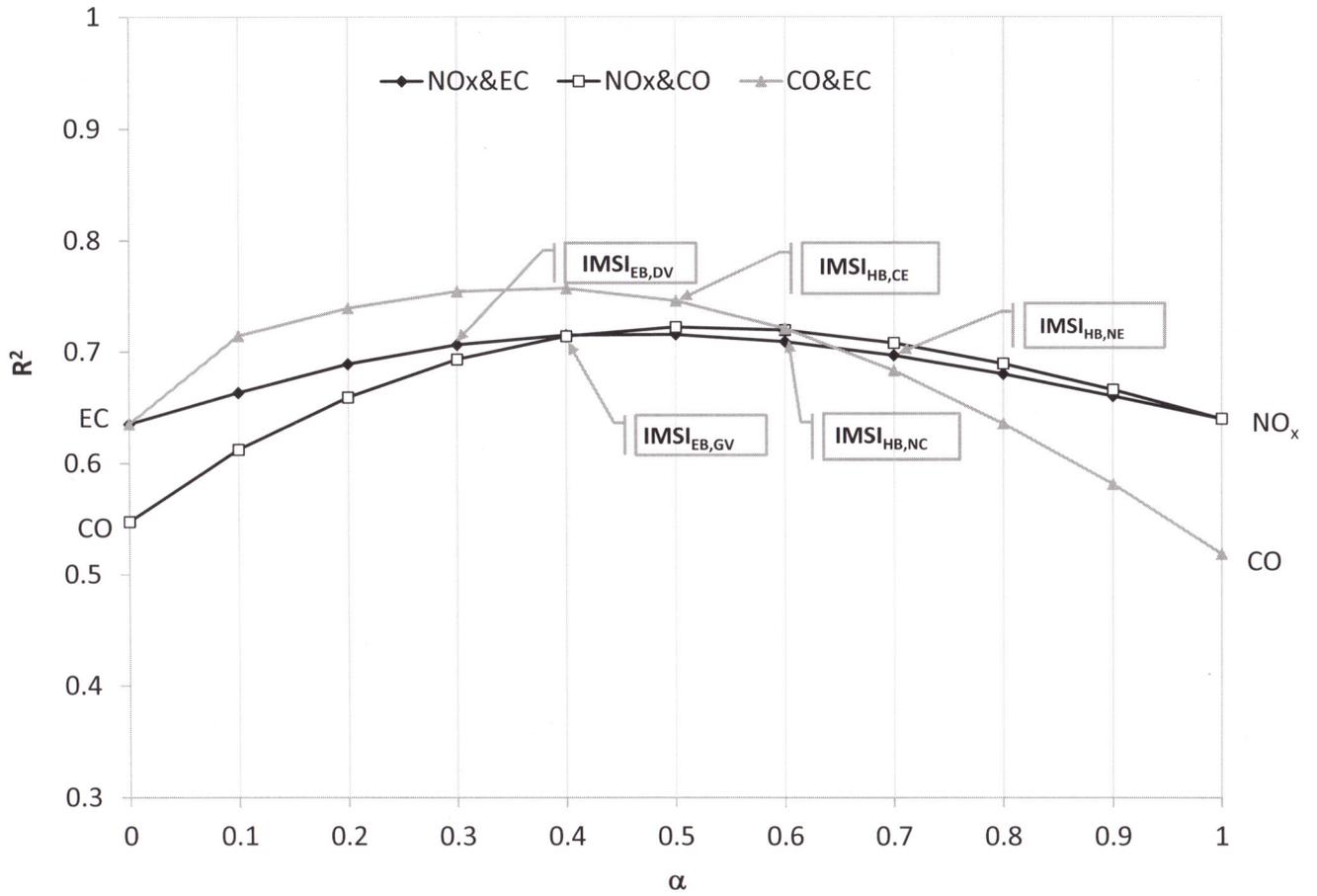


Figure 6. Correlation (R^2) between binary mixtures at two different receptor sites JST and SD. Vertical scale starts at 0.3 to emphasize correlations. Overlapped days at JST and SD: $\text{NO}_x\&\text{EC}$ ($n=398$), $\text{NO}_x\&\text{CO}$ ($n=560$) and $\text{CO}\&\text{EC}$ ($n=173$).

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	NOx 1h	CO 1h	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	0.91
NOx 1h	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 1h	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 (Oct–Mar); values in the lower left represent correlations during summer time (Apr–Sep). PMF stands for the Positive Matrix Factorization; CMB stands for the Chemical Mass Balance method; GV stands for gasoline vehicles; DV stands for diesel vehicles; mob stands for mobile sources=GV+DV.

Table 2

Comparison of uncertainties between indicators

Indicator	Indicator value [€]	Standard deviation [£]	Uncertainty [¥]	Relative uncertainty
CO 1h (ppm)	1.16	1.00	0.16	0.14
NO _x 1h (ppm)	0.12	0.10	0.03	0.25
EC (µg/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:

[€] Average of daily values from 1999–2004;

[£] For daily values from 1999–2004;

[¥] Uncertainties are estimated as the RMS average $\left(\overline{\sigma_i} = \sqrt{\frac{1}{N} \sum \sigma_i^2}\right)$ of daily uncertainties from 1999–2004.

Table 3

Results for the associations of ED for CVD with mobile source impacts metrics (sorted by p-value)

Indicator	IQR [€]	RR [£] per IQR	95% CI [¥]	p-value
CMB-DV	1.0 µg/m ³	1.005	0.997 – 1.014	0.206
PMF-GV	2.3 µg/m ³	1.005	0.997 – 1.012	0.206
PMF-DV	2.3 µg/m ³	1.006	0.998 – 1.014	0.168
CMB-GV	1.0 µg/m ³	1.006	0.999 – 1.012	0.079
EC	1.0 µg/m ³	1.008	1.000 – 1.017	0.054
CO-1h	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 -1h	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:

€ Interquartile range;

£ Risk ratio;

¥ Confidence Interval; CMB is the Chemical Mass Balance method; PMF is the Positive Matrix Factorization model; GV stands for gasoline vehicles; DV stands for diesel vehicles; IMSI_{EB} is the emission-based integrated mobile source indicator; IMSI_{EB,GV} is the IMSI_{EB} estimated for gasoline vehicles; IMSI_{EB,DV} is the IMSI_{EB} estimated for diesel vehicles.