In many urban areas throughout the world, the rising mean and peak levels of nitrogen oxides (NO\textsubscript{x}) are a concern. Road traffic and local industry are usually the major NO\textsubscript{x} sources in urban environments, but their relative contributions to the spatial distribution of the NO\textsubscript{x} volumetric mixing ratio is normally unknown. This missing piece of information is required for designing effective abatement measures to reduce ambient NO\textsubscript{x} levels. A new method for estimating the shares of which traffic and industry contribute to the mean ambient NO\textsubscript{x} mixing ratios observed in urban environments is proposed in this paper. The estimation is based on data obtained by routine air pollution monitoring, using a few assumptions about the temporal emission patterns of NO\textsubscript{x} and SO\textsubscript{2} in the area of study. A set of equations is formulated for the unknown industry and traffic contributions to the NO\textsubscript{x} mixing ratios at each monitoring site. These equations are solved using the gradient projection optimization method. The bootstrap technique is used to estimate the errors in the process. Spatial maps of the separate shares of industry and traffic in the total ambient NO\textsubscript{x} levels can be obtained where a sufficient aerial coverage of stations is available. An application of the method to the allocation of NO\textsubscript{x} mixing ratios to traffic and industrial sources in the Haifa Bay area, Israel, is demonstrated. The results are expected to be useful for future planning of traffic thoroughfares and industrial development in the area.

1. Introduction

The rising levels of nitrogen oxides (NO\textsubscript{x}) and their atmospheric transformation products have been a main air pollution worry in major metropolitan areas during the last few decades (1–3). NO\textsubscript{x}, the main component of NO\textsubscript{x}, is one of the five air pollutants identified as being of the greatest concern from health perspectives (4). The need to attain the NO\textsubscript{2} limit values set by the European Commission Air Quality Framework Directive 1999/30/EC prompted many recent studies to facilitate NO\textsubscript{x} abatement programs (e.g., 5–7). The first step in setting forth a NO\textsubscript{x} abatement plan is studying the impact that the current emissions from the various sources has on its present temporal and spatial patterns. Road traffic is in most cases the largest NO\textsubscript{x} source; however, in many areas around the world industrial plants and/or power generation are sources of commensurate magnitude (8, 9).

Estimating the contributions of known sources to the observed levels of air pollutants at specific points is usually termed source apportionment. The most comprehensive approach to source apportionment is by a direct numerical simulation of the coupled meteorological dispersion and chemical reactions of the emissions (10). Simplified dispersion schemes are sometimes used instead of a full three-dimensional dynamical and chemical model (e.g., 7, 6, 11). Running simulations with the different sources switched “on” and “off” can provide an estimation of the impact of different sources on the observed air pollution levels at points of interest in the study area. This is a very appealing approach, as it has the potential of finding the contribution of each identified source to the overall ambient levels of the pollutant at any given point in space and time. However, developing and running such a model is a challenging enterprise, requiring a thorough emissions inventory and much time for the construction and testing of the model. Moreover, the errors accrued in the process might be very large and hard to estimate (7). Fine-tuning the model to minimize its errors can be carried out through an inverse modeling scheme (e.g., 12) but the human and computer resources needed for such a task are not always readily available.

Simple alternatives to a comprehensive direct source apportionment are based on analyses of air pollution measurements. When dealing with pollutants which have identifiable composition signatures that can be tracked to specific sources, the chemical species collected at monitoring sites can be associated with the potential sources through statistical algorithms such as those of the chemical mass balance (13, 14) or the positive matrix factorization (15, 16). The chemical mass balance method requires complete emission composition profile information. The positive matrix factorization method requires a large number of receptor samples and estimation of their errors. Both methods are mainly suitable and have been applied mostly for the apportionment of particulate matter or volatile organic compounds (ref 17 and references therein).

Apportionment of ambient levels of gaseous reactive pollutants can be facilitated based on the ratios of the levels of observed gases. Duncan et al. (18) used NO\textsubscript{x} and SO\textsubscript{2} source concentrations to estimate what percentage of the total ambient NO\textsubscript{x} observed at a monitoring station is attributable to a specific point source. Their method involves knowledge of all the source concentrations and a few assumptions about losses due to dry and wet deposition and gas-phase reactions, which may not always hold. Using a few regression models, Nirel and Dayan (19) studied the role of the SO\textsubscript{2}/NO\textsubscript{x} ratio, encountered at a specific monitoring station, in attributing air pollution to potential sources. Wind directions and information about the structure of the atmospheric boundary layer were also used. Application of their method requires a lot of preliminary data exploration and analysis, which might be difficult to do for a multiple-station data set.

The present study proposes an alternative approach that bypasses all the above-mentioned hurdles, using routinely observed air pollution data (i.e., obviating the need for a complex modeling) in a relatively simple and computationally cheap scheme. The goal is limited to the allocation of observed ambient NO\textsubscript{x} mixing ratios to generic types of sources, namely, traffic and industry. However, the assessment of the traffic and industry NO\textsubscript{x} contributions is a scientifically and practically important issue, which is in the focus of recently published studies (e.g., 6, 8). Our approach is based on an a priori qualitative knowledge of the different temporal emission patterns of the industry and the traffic in the study area. Using this information, we set forth a few general assumptions and derive from them a set of equations.
associating the unknown industry and traffic NO\textsubscript{x} contributions to the ambient NO\textsubscript{x} with the corresponding observed NO\textsubscript{x} mixing ratios at each monitoring site. The system of equations is then solved, and the separate contributions of the source types to the ambient NO\textsubscript{x} levels as measured at the monitoring stations are recovered. Given data from a sufficiently dense monitoring network, the separate contributions at the stations can be interpolated to maps of their spatial patterns. These maps can provide a readily available and worthy insight into the roles that the major sources play in the spatial NO\textsubscript{x} distribution. The proposed methodology is applied at the Haifa Bay area, Israel, where the NO\textsubscript{x} levels are rising and a better understanding of the share of the major NO\textsubscript{x} sources in the area in the ambient NO\textsubscript{x} levels is required for devising optimal management strategies.

2. Methodology

The general approach outlined in the Introduction is specified in this section. While the formulation of the method is made to befit the conditions in the study area of our example, its application to other regions may require only slight modifications that will be discussed later. The basic assumptions which we use are as follows: (a) Industry and traffic are the only sources of NO\textsubscript{x} in the study area. (b) The industry is the only source of SO\textsubscript{2} in the study area. (c) At any point in the study area, the ratio between the mean (over the study period) number of molecules of NO\textsubscript{x} and SO\textsubscript{2} that originate from industrial sources during weekdays is identical to the corresponding ratio during weekends and holidays (henceforth referred to as holidays). Note that no assumptions are made regarding the industrial emission rates and composition except that, on average, the ratio NO\textsubscript{x}/SO\textsubscript{2} in the industrial emissions is similar in weekdays and holidays. In particular, no assumption is made regarding a possible long-term change in this ratio. Assumptions a and b are probably valid in most typical industrialized urban environments (18), especially where nonelectrical space heating and other domestic fuel burning is minimal. (See in the Supporting Information the emissions inventory supporting these assumptions for the case study of this work.) Possible non-negligible background levels of NO\textsubscript{x} and SO\textsubscript{2} are not accounted for, but as will be discussed later, they can be easily incorporated. For assumption c to be valid, the air pollution dispersion conditions and industrial plants’ NO\textsubscript{x} and SO\textsubscript{2} emissions per consumed fuel unit must be, on average, the same during the working days and during the holidays. This may certainly be incorrect for short periods of time due to variations in the working conditions of the industrial furnaces and due to varying dispersion conditions. However, averaged over a long period of time (or over a short period in which the emission characteristics and the meteorological conditions can be considered similar during working days and holidays), assumption c is very reasonable. The validity of assumption c for the demonstrated case study was ascertained by verifying with the major industrial plants that their weekdays and holidays emission characteristics do not differ in any systematic way (i.e., the fuel type and grade and the furnaces’ operating conditions do not depend on the day of the week). Meteorological conditions are certainly blind to the calendar, so it is obvious that averaged over a long enough period of time they are similar for weekdays and holidays.

From assumption a, the following mass balance equations are derived for each monitoring station’s data set:

\[ N_{p}^{w} + N_{t}^{w} = N_{o}^{w} \quad (1) \]

and

\[ N_{p}^{h} + N_{t}^{h} = N_{o}^{h} \quad (2) \]

where \( N_{p}^{w} \) is the mean (over the study period) ambient NO\textsubscript{x} mixing ratio observed at the station during working days and which originated from industrial emissions (point sources), \( N_{t}^{w} \) is the corresponding traffic contribution, and \( N_{o}^{w} \) is the corresponding observed mean ambient NO\textsubscript{x} mixing ratio. \( N_{p}^{h}, N_{t}^{h}, \) and \( N_{o}^{h} \) are the corresponding quantities during holidays. The definitions of weekends, holidays,
and the choice of the hour on which the days change are country/region specific. For example, in Israel, the official weekly rest day is Saturday, and we chose to switch days at 18:00, the average time for day change in the Jewish calendar. These choices optimally capture the weekly and daily business and traffic cycles in the case study used for demonstrating the proposed method. The study period can be any portion of the period for which data records are available (e.g., data from certain seasons, hours of day, etc.) as long as that portion includes sufficient sampling for assumption c to be valid. Assumptions b and c lead, after a simple algebraic manipulation, to a third equation of

\[ N_p^w / N_p^h = S_p^w / S_p^h = S_0^w / S_0^h = F \]  

(3)

where \( S_p^w \) and \( S_p^h \) are the mean (over the study period) SO\(_2\) mixing ratios observed at the monitoring station during working days and holidays, respectively, and F is defined by eq 3 as the ratio between the means of the observed SO\(_2\) levels during weekdays and holidays. In cases where the background NO\(_x\) and/or SO\(_2\) levels are non-negligible, their measured mixing ratios can be incorporated as known constants in eqs 1–3.

Equations 1–3 can be written in the matrix form

\[ Ax = b \]  

(4)

where

\[ A = \begin{pmatrix} 1 & 1 & 0 & 0 \\ 0 & 0 & 1 & 1 \\ -1 & 0 & F & 0 \end{pmatrix} \]

\[ x = [N_p^w, N_p^h, N_0^w, N_0^h] \] \quad and \quad \[ b = [N_0^w, N_0^h, 0, 0] \]

The underdetermined system in eq 4 can be solved for \( x \) by minimization of \(|x|_2^2\) (the Euclidean norm of \( x \)) subject to \( Ax - b = 0 \) and the additional physical constraints

\[
\begin{align*}
0 & \leq N_p^w \leq N_0^w \\
0 & \leq N_p^h \leq N_0^h \\
0 & \leq N_0^h \leq N_0^w \\
0 & \leq N_0^h \leq N_0^w
\end{align*}
\]

Thus, the solution \( x \) contains the study period means of the separate contributions of traffic and industry to the ambient NO\(_x\) levels at a monitoring station, derived from the observed NO\(_x\) and SO\(_2\) levels averaged over the study period and subject to the constraints set on the elements of \( x \). To find the solution to the minimization problem, we employed the gradient projection optimization method (20).

As in any other estimation process, an assessment of the possible errors introduced by the proposed method is a concern. Errors arising due to invalidity of the underlying assumptions are hard to assess but are expected to be small. That is because our use of data averaged over long periods results in mutual cancellation of random observation errors. Biases and systematic data errors should be excluded in the initial data processing, but their complete removal may not always be possible. Applying the method to synthetic data sets (i.e., where the true allocation was known), we noted that slight systematic errors during relatively short durations might sometimes be a source of relatively large allocation errors. Another cause of errors is the inaccuracies introduced by the process of solving eqs 1–3, e.g., the minimization of \(|x|_2\) is not necessarily appropriate in all cases. To estimate the expected solution errors we chose to use the moving blocks bootstrap technique (21). Bootstrapping using moving blocks of data is required due to the temporal autocorrelation in the data. The moving blocks bootstrap involves a division of the data series into blocks. A random selection, with replacement, of the data blocks is then carried out multiple times, yielding many new data sets called bootstrap samples. Due to the random selection, each bootstrap sample is in general slightly different from the others and from the original set. We applied the numerical solution of eqs 1–3 to each of the bootstrap samples and obtained a set of values \( N_p^w, N_p^h, \) \( \bar{N}_p^w, \) \( \bar{N}_p^h, \) \( \bar{N}_0^w, \) \( \bar{N}_0^h, \) which are called the bootstrap replicas of \( N_p^w, N_p^h, \)

\( \bar{N}_0^w, \) \( \bar{N}_0^h, \) respectively. The standard deviations of the replicas \( N_p^w, N_p^h, \) \( \bar{N}_p^w, \) \( \bar{N}_p^h, \) \( \bar{N}_0^w, \) \( \bar{N}_0^h, \) can serve as measures of the errors expected in our real data solutions for \( N_p^w, N_p^h, \) \( \bar{N}_p^w, \) \( \bar{N}_p^h, \) \( \bar{N}_0^w, \) \( \bar{N}_0^h, \) respectively.

An application of the allocation process in all the monitoring stations results in a set of values, associated with the station locations, for each of the variables \( N_p^w, N_p^h, \) \( \bar{N}_p^w, \) \( \bar{N}_p^h, \) \( \bar{N}_0^w, \) \( \bar{N}_0^h, \). Each of these sets can be used to create a corresponding spatial interpolation map. We used for this purpose the ordinary kriging method (22, 23). The accuracy of the interpolation depends on the density of the data points. Where the monitoring stations’ coverage is not sufficiently dense, large interpolation errors may occur. In such a case, only the main features of the interpolation maps should be considered. A detailed discussion of this point, regarding interpolation of data from the same monitoring array used here, can be found in Yuval and Brodsky (24).

3. Case Study Information

Figure 1 shows a general map of the Haifa Bay area. The area includes the city of Haifa and satellite suburbs and towns, totaling nine municipalities and a population of about
500,000. At the heart of the region are a few of Israel’s major industrial plants. An oil-fired power plant and a petroleum refinery together contribute about 80 and 83% of the total industrial NO\textsubscript{x} and SO\textsubscript{2} emissions in the area, respectively. (See the Supporting Information for more details of the emission inventory.) The refinery and a few other large plants in its vicinity work year-round, 24 h a day, with no significant day/night or workday/holiday variations. However, the power plant’s load and emissions vary according to demand. The Haifa District Municipal Association for the Environment (HDMAE) estimated that the mean total industrial emission rates in the study period 2002–2005 were 0.80 tonne/hour NO\textsubscript{x} and 1.74 tonne/hour SO\textsubscript{2} (Supporting Information, Table S1). The roads in the study area experience daily traffic of about 125,000 private cars, 30,000 lorries and light trucks, and a few hundred buses. There are no appreciable seasonal variations in the traffic volumes. However, the weekly rest day (Saturday) and statutory holidays experience a very large reduction of vehicular activity, with the diesel-fueled commercial traffic, responsible for about 75% of the vehicular NO\textsubscript{x} emissions, coming to an almost complete halt. The estimated mean traffic emission rates in the 2002–2005 study period were 1.02 tonne/hour NO\textsubscript{x} and 0.03 tonne/hour SO\textsubscript{2} (Supporting Information, Table S1).

The Persian trough, which dominates the synoptic meteorology of the coastal regions of Israel from mid-May to October, results in ample sunshine and dominant northeasterly winds during the warm-season days. Weak easterly land breezes may occur during the warm-season nights. Due to the Azorean high-pressure system aloft capping the Persian trough at the surface, the mixing height in the warm season is shallow (about 700 m) and stable (\(25^\circ\)). During the cold season (November to April), southeasterly flow dominates, except when subtropical storms pass through the area (mainly from December to February) and result in strong winds veering from southwesterlies to northeasterlies (Figure S2 in the Supporting Information). The cold season is characterized by a lot of mixing except during cold clear nights. Due to the topography of Mt. Carmel on which the city of Haifa is built, the wind directions on the crest and northeasterly slopes of the mountain change counterclockwise from the regional winds (Figure S2 in the Supporting Information).

4. Results

The main goal of the proposed method is to estimate the relative shares of the industry and traffic in the mean observed ambient NO\textsubscript{x} mixing ratios. A way to assess the validity of the method’s results is to test it on subperiods with clear known trends of emissions, dispersion, and chemical transformation. Thus, before we present the final results for the whole study period, we compare the NO\textsubscript{x} allocation during the summers and winters and during the morning and night hours. Winters and summers obviously differ in their light intensity and length of days, which impact indirect photochemical conversion processes of NO\textsubscript{x} to NO\textsubscript{y} species (e.g., reactions of NO\textsubscript{2} with OH and RO\textsubscript{2} radicals). The analysis should also account for other factors such as the differences in the dominant wind directions and the height of the atmospheric mixing layer. The ambient conditions during the morning...
and night hours also differ in many natural meteorological variables, but the largest impact on the ambient NO\textsubscript{x} levels is due to the morning and night rush hour traffic emissions. Qualitative verification of the proposed method can be established if the differences between the winter/summer, morning/night, and the weekend/holidays results reflect the known variations in the corresponding natural and anthropogenic variables.

Figure 2 shows the calculated industry and traffic contributions to the mean observed NO\textsubscript{x} levels in working days and in holidays during winter (December–February) and summer (June–August) in each of the monitoring stations. In agreement with our qualitative knowledge of the traffic emission patterns, in both seasons the calculated traffic contribution to the ambient NO\textsubscript{x} mixing ratios is significantly higher during working days than in holidays (a station-wise average ratio $N^t/N^p$ of 4.12 in the winter and 3.46 in the summer were found). The winter ratio is probably higher because of the later onset of the indirect photochemical NO\textsubscript{x} removal processes of the winter morning rush hour emissions in the weekdays. These weekdays morning emissions have a larger impact on the mean NO\textsubscript{x} values. The NO\textsubscript{x} contribution of industry is also higher during working days but with a much smaller average ratio ($N^i/N^p$ equals 1.17 in the winter and 1.25 in the summer). The ratios between the mean industry and traffic contributions in winter and summer (weighted weekdays and holidays means) are similar (1.27 for traffic and 1.23 for industry). The close similarity in these ratios is expected as the differences between the winter and summer NO\textsubscript{x} mixing ratios is primarily due to the differences in the thickness of the atmospheric mixing layer (25) and in the rate of indirect photochemical NO\textsubscript{x} removal, both having similar effects on traffic and industrial emissions alike.

Figure 3 shows the spatial patterns of the traffic and industry contributions to the mean ambient NO\textsubscript{x} mixing ratios during the summer and the winter. The traffic contribution pattern (parts b and d of Figure 3) show slightly higher mixing ratios than the pattern of the industry contributions (parts a and c of Figure 3). Higher mixing ratios due to both industry and traffic are found in the lower parts of the city of Haifa (in the vicinity of stations 2, 5, and 9, see Figure 1), which is geographically close to the industrial center and to the area where most of the commercial activity takes place.

Figure 4 shows the industry and traffic contributions to the mean observed NO\textsubscript{x} levels in the monitoring stations. Results are shown for the night hours (24:00—04:00) and the mornings (06:00—10:00) in 2002—2005. The error bars indicate 1 standard deviation of the corresponding bootstrap replicas. (a) The contributions in working days (10 944 time points). (b) The contributions in Saturdays and holidays (2205 time points).

FIGURE 4. Contributions of industry and traffic to the mean observed NO\textsubscript{x} levels in the monitoring stations. Results are shown for the night hours (24:00—04:00) and the mornings (06:00—10:00) in 2002—2005. The error bars indicate 1 standard deviation of the corresponding bootstrap replicas. (a) The contributions in working days (10 944 time points). (b) The contributions in Saturdays and holidays (2205 time points).

Figure 5 shows the spatial patterns of the traffic and industry contributions during the summer and the winter. The traffic contribution pattern (parts b and d of Figure 3) show slightly higher mixing ratios than the pattern of the industry contributions (parts a and c of Figure 3). Higher mixing ratios due to both industry and traffic are found in the lower parts of the city of Haifa (in the vicinity of stations 2, 5, and 9, see Figure 1), which is geographically close to the industrial center and to the area where most of the commercial activity takes place. The traffic's contributions to the mean observed NO\textsubscript{x} mixing ratios during the morning and night hours. The morning patterns of the industry and traffic contributions (parts a and b of Figure 5, respectively) are quite similar, except the conspicuous high traffic contributions around station 5 and the general trend of the main traffic contribution pattern toward the east. The onset of the daytime northwesterly wind in summer mornings is around 10:00. Thus, the mean morning industrial contributions pattern is more influenced by the dominating winter easterly winds and the early summer easterly land breeze, which brings industrial NO\textsubscript{x} to the area west of the industry center. The traffic contribution pattern is more influenced by the location of the morning rush hour traffic, which is heavy in the area around station 2 and along the highways leading to the bay of Haifa from the northeast and southeast (see Figure 1). The spatial patterns of the industry and traffic contributions during the night (parts c and d of Figure 5, respectively) are very similar, with the major feature extending from the industrial center toward the west. This reflects the relatively stagnant meteorological conditions in the nocturnal stable boundary layer in which the late evening traffic emissions and the nightly industrial emissions mix and remain in place or advect slowly toward the west with the land breeze.

Figure 6a shows the absolute industry and traffic contributions to the mean observed NO\textsubscript{x} mixing ratios in each of the stations for the whole study period. Figure 6b shows the corresponding relative shares of the total observed NO\textsubscript{x}. Interestingly, the dominance of the traffic’s share during the day hours of the workdays is almost balanced in all the stations by the dominance of the industry’s share during the workdays nights and holidays. In most stations, the differences are smaller than their error estimates. The traffic’s relative shares (Figure 6b) are between 50 and 56% of the

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total observed NOₓ. The most updated emission estimations reported by the HDMAE (Supporting Information, Table S1 and Figure S1) attribute to traffic sources 45% of the emitted NOₓ in the region during 2006. The higher traffic shares that we found are expected as, per unit of emission, the surface-level traffic emissions are yielding a larger contribution to the ground-level ambient NOₓ than the emissions from the tall industrial stacks (6, 8). (Note that the 2002–2005 traffic’s share in the NOₓ emission are higher due to recently acknowledged errors in the HDMAE traffic emission estimations during those years.)

5. Discussion

A new method for allocating ambient NOₓ mixing ratios, observed at monitoring stations, to traffic and industry source types was developed. The method uses readily available monitoring data that are routinely observed and archived throughout the world. Spatial maps of the separate shares of industry and traffic in the total ambient NOₓ levels can be obtained but only where a sufficient aerial coverage of stations is available. While this method does not aim to estimate the absolute emission from each source, it can estimate the contributions from the major source groups (traffic and industry) to the mean observed mixing ratios. The NOₓ allocation results pertain to a specific study period so that intraperiod variations in the emission characteristics and meteorology are averaged out. In cases of long time-scale variations in the emission characteristics (e.g., due to a decrease in the sulfur content in the fuels), the method can be applied separately to homogeneous subperiods of the whole study period to obtain their specific allocation results. The method presented in this study facilitates computation of NOₓ contributions during specific hours of the day. As demonstrated by the night and morning NOₓ allocation results (Figures 4 and 5), large diurnal variations in the traffic...
and industry shares may occur. Thus, for risk assessment studies as well as for NO\textsubscript{2} abatement programs, it is advisable to consider the patterns at the hours of the day when the population at risk tends to be exposed.

The method was applied at the Haifa Bay area, and the results fit well the qualitative known spatial and temporal distribution of NO\textsubscript{2} emissions in the region and their dispersion and transformation by the local meteorological and atmospheric chemistry conditions. This study was motivated by a debate regarding the relative shares of the sources of NO\textsubscript{2} in the study area. With the use of the proposed methodology and the results presented in this paper, abatement efforts and future industry development and highway construction can be better focused and rationally devised.

Acknowledgments

The data were kindly provided by the Haifa District Municipal Association for the Environment and by the Israel Electric Corporation. This work was supported by grants from the Israeli Ministry of Science and Technology, the Seniel Ostrow Research Fund, and the Technion Research and Development Foundation Landau/Ben-David. The authors would like to thank Professor Eldad Haber of Emory University for providing the gradient projection Matlab code.

Supporting Information Available

(a) Table containing the 2002–2006 emission inventories for the Haifa Bay area; (b) figure with pie charts showing the relative shares of the various sources in the NO\textsubscript{2} and SO\textsubscript{2} emissions; (c) winter and summer windroses of wind data from two characteristic stations. This material is available free of charge via the Internet at http://pubs.acs.org.

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Received for review January 30, 2007. Revised manuscript received August 19, 2007. Accepted August 22, 2007.

ES0702317