A Prediction Model of the Build-up of Volatile Organic Compounds on Urban Roads

Parvez Mahbub1*, Ashantha Goonetilleke1, Godwin A. Ayoko2

1School of Urban Development, Queensland University of Technology, GPO Box 2434, Brisbane 4001, Queensland, Australia
2Chemistry Discipline, Queensland University of Technology, GPO Box 2434, Brisbane 4001, Queensland, Australia
s.mahbub@qut.edu.au; a.goonetilleke@qut.edu.au; g.ayoko@qut.edu.au

*Corresponding Author: Parvez Mahbub; Tel: 61 7 3138 9945; Fax: 61 7 3138 1170; email: s.mahbub@qut.edu.au

Abstract

A model to predict the build-up of mainly traffic generated volatile organic compounds or VOCs (toluene, ethylbenzene, ortho-xylene, meta-xylene and para-xylene) on urban road surfaces has been presented in this paper. The model required three traffic parameters namely Average daily Traffic (ADT), Volume to Capacity Ratio (V/C) and Surface Texture Depth (STD) and two chemical parameters namely Total Suspended Solid (TSS) and Total Organic Carbon (TOC) as predictor variables. Principal Component Analysis as well as Two Phase Factor Analysis was performed to characterise the model calibration parameters. Traffic congestion was found to be the underlying cause of traffic related VOC build-up on urban roads. The model calibration was optimised using orthogonal experimental design. Partial Least Square Regression was used for model prediction. It was found that a better optimised orthogonal design could be achieved by including the latent factors of the data matrix into the design. The model performed fairly accurately for three different land uses as well as five different particle size fractions. The relative prediction errors were 10%-40% for the different size fractions and 28%-40% for the different land uses while the coefficients of variation of the predicted inter-site VOC concentrations were in the range of 25% to 45% for the different size fractions. Considering the sizes of the data matrices, these coefficients of variation were within the acceptable inter-laboratory range for analytes at ppb concentration levels.

Keywords: Volatile organic compounds, experimental design, partial least square regression
Introduction

Volatile organic compounds (VOCs) are amongst the major pollutants generated by transport activities in urban areas (1-3). Several studies have characterised the influence of traffic on the generation of VOCs in ambient urban atmosphere (4-6). Toluene, ethylbenzene, ortho-xylene, meta-xylene and para-xylene are amongst the most common species of VOCs (boiling points ranging from 50 °C -260 °C). These are termed as gasoline range organics (3, 7, 8). The International Agency for Research on Cancer has listed them as carcinogenic to humans (9).

Urban stormwater runoff acts as the primary medium for the transport of pollutants from urban road surfaces (10-13). Consequently, the risks to urban water resources from toxic and mutagenic pollutants generated by traffic activities have increased due to the rapid urbanisation witnessed in most parts of the world. Though, the investigation of pollutant build-up processes on urban roads has received significant attention in recent years, there has been very limited work undertaken with regards to the characterisation of the build-up of VOCs on actual road surfaces. Olson et al. (14) studied VOC concentrations in the near-road environment and found that the concentrations decreased exponentially with increasing distance from the roadway. However, they collected air samples from various locations near roadway. Limited information on the spatial variability of VOCs has led researchers to propose prediction methods for certain VOCs such as toluene, benzene, ethylbenzene, ortho-xylene, meta xylene and para xylene in the atmosphere (e.g., 15). Their approach was based on six GIS based ancillary variables which included distance to nearest border crossing, elevation, population density, distance to roads, traffic intensity and distance to nearest petroleum facility. These ancillary variables may be irrelevant elsewhere mainly due to lack
of in-depth understanding of the VOC build-up characteristics on road surfaces as well as lack of knowledge on identifying the influencing parameters on such processes.

This paper presents a robust model to predict VOCs (toluene, ethylbenzene, ortho-xylene, meta-xylene and para-xylene) on urban road surfaces using three traffic parameters and two chemical parameters. This build-up model excludes the VOCs in the ambient air which may not necessarily get deposited on roads. To the best of our knowledge, there has not been any previous research which has been based on the physical collection of VOCs from the actual road surfaces. The closest is the work undertaken by Olson et al (14) where sample collection was performed at some distances from the roads.

The selection of the model parameters was based on the characterisation of the build-up of VOCs on urban roads. Different land uses, namely, residential, industrial and commercial as well as different particulate and dissolved fractions are incorporated into the model. The model differentiates the VOC build-up on urban roads from that of ambient urban atmosphere. It reduces the degree of complexity involved in the prediction of traffic generated VOC build-up on urban road surfaces. This prediction model will be useful to environmental planners in terms of reliable prediction of VOCs on urban roads in changing traffic conditions and the use of the model structure as a guide to predict other traffic generated pollutants on urban roads.

**Materials and Methods**

**Site Selection**

It is hypothesised in this study that vehicle congestions due to increased traffic volumes in the urban areas would directly affect the pollutant build-up on urban roads. Hence, eleven road
sites were selected as study sites for the build-up sample collection in three suburbs in the
Gold Coast region in South-East Queensland, Australia. The sites selected for sample
collection for VOC build-up covered three types of land uses including residential,
commercial and industrial which provided a cross-section of traffic activities on road surfaces
within the Gold Coast region. The selected suburbs were also representative of the transport
infrastructure developed in the Gold Coast region in the past decade.

Average daily traffic (ADT), volume to capacity ratio (V/C) and surface texture depth (STD)
were selected as the primary traffic parameters for this study. ADT and congestion as
described by V/C are two important traffic parameters in terms of characterising urban roads.
Whilst ADT refers to the traffic count at a fixed point of the road section, V/C describes the
congestion on the stretch of road during the peak hour (16). Additionally, the STD refers to
the deviation of the surface profile of a pavement from a true planar surface often known as
macro-texture of the pavement (17).

Organic matter can act as binding agents between solids and other pollutants such as heavy
metals in source, transport as well as deposit phases (18). Hwang and Foster (19) have
reported 68%-97% enrichment of polycyclic aromatic hydrocarbons in particulate phase in
urban runoff. In another study, Moilleron et al. (20) have shown that 85% of the total
aliphatic hydrocarbon content in urban runoff was attached to particulate phase. In this
context, surface roughness becomes an issue because of the potential trapping of organics to
the particulate matter. Smooth surface reduce trapping but provide a more efficient transport
mechanism, and vice versa. While describing a physically based urban wash-off model, Shaw
et al. (21) have addressed the role of surface roughness of asphalt pavement in capturing and
attenuating particulate matter. The road texture also affects the interactions between tires and
driving surface. Kreider et al. (22) have found that the characterisation of roadway particles, tire wear particles and tread particles are totally different during their build-up and attributed friction between tire and the road surface as one of the principal causes of such characterisation. Hence in this study, surface texture depth (STD) has been selected as a pavement parameter affecting the build-up of VOCs on the roads.

A transport model called ‘ZENITH’ (23), which is currently being used by the Gold Coast City Council for pavement infrastructure planning and design purposes, was used to predict the current ADT and V/C of the selected study sites whilst STD was directly measured in the field. The selected study sites with their traffic and pavement characteristics are given in the Figure S1 in the supporting information.

**Build-up Sample Collection**

A method referred to as ‘Wet and Dry Vacuuming’ (24) was employed for the collection of pollutant build-up samples. This consisted of the use of a domestic vacuum cleaner with a water filtration system for the collection of road dust from a $2 \times 1.5 \text{ m}^2$ plot area in 25L plastic containers. Immediately thereafter deionised water was sprayed at 2 bar pressure on the plots and vacuuming was undertaken again to collect any remaining build-up matter. Samples were then transported to the laboratory for sub-sampling and further analyses.

The pollutant build-up process has been characterised as four main functional forms such as, linear, power, exponential, and Michaelis-Menton (25). Amongst these, the non-linear asymptotic form proposed by Sartor et al. (26) has been most often cited and also used in several stormwater quality models such as, DR3-QUAL, FHWA, SWMM (25). Huber (25) also suggested that build-up limits of paved surfaces influence the accumulation of solids.
especially due to the fact that surface materials on highways tend to remain in a state of
suspension due to eddies created by traffic. In this context, Egodawatta (27) noted that
pollutant build-up on road surfaces asymptote to an almost constant value after a seven day
antecedent dry period. Hence, seven dry days were allowed at each site prior to any sample
collection in conformity with the findings of Egodawatta (27).

The samples collected in the field were transferred into the splitting chamber of the churn
splitter in the laboratory. The total composite sample volume was 8 L, of which 6 L was used
to prepare 500 mL sub-samples for particulate fraction analyses of the VOCs. The remaining
sample in the churn splitter was used for the dissolved fraction analyses. The particle size
distributions of the solids in the sub-samples were determined using a Malvern Mastersizer S
Particle Size Analyser capable of analysing particles between 0.05 and 900 µm diameters.
The particle size distributions of the sub-samples were compared with each other and used as
a guide for homogeneity maintained in the sub-sampling process. The total particulate
analytes were then fractioned into four size ranges, namely, >300 µm, 150-300 µm, 75-150
µm, 1-75 µm using wet sieving. The filtrate passing through a 1 µm membrane filter was
considered as the potential total dissolved fraction. In each case, the prepared sub-samples
were stored in 500 mL amber glass bottles with PTFE seals, preserved at 4°C in the
laboratory and analysed within 14 days of collection. A total of 55 build-up samples were
prepared for the 11 selected sites with each site having 5 samples for the five size fractions
mentioned above. Seven replicate samples for each sub-sample at each fraction were also
prepared and means, standard deviations as well as inter-site percentage coefficient of
variations have been reported in the supporting information.

Sample Testing
The samples were tested for volatile organics such as, toluene, ethylbenzene, ortho-xylene, meta-xylene and para-xylene according to the USEPA method 5035 and 8260B (7) using purge and trap extraction followed by gas chromatography/mass spectrometry (GC/MS). A brief description of the quality control measures are given in the supporting information.

The total and dissolved organic carbon (TOC and DOC) concentrations and total and dissolved suspended solids (TSS and TDS) concentrations were also determined for the samples (28).

**Data Analysis**

The data matrices consisted of eleven objects and nine variables for each of the five particle size fractions. The results for the different volatile organics, suspended solids and organic carbons due to the traffic attributes, ADT, V/C and STD were defined as ‘build-up values’. The eleven traffic objects were designated with object identifiers R1, R2, R3, I4, I5, C6, C7, R8, C9, C10 and R11 where the prefix C, I and R represented commercial, industrial and residential sites, respectively. After initial observation of the probability distribution of the objects and variables, standardisation of each variable was undertaken as a pre-treatment measure.

The data analysis was designed to investigate the build-up process of volatile organics generated from traffic on urban roads and then to use the findings from the initial investigations to develop a prediction model for volatile organics build-up. Multivariate chemometrics methods such as principal component analysis (PCA), two phase factor analysis (FA) and partial least squares regression (PLS) were employed for the data analysis.
A brief description on these techniques with relevant references is outlined in the supporting information.

Results and Discussion

Exploratory PCA

In the six build-up data matrices (five for different size fractions described above and one as total particulate), the eleven traffic objects were considered having object attributes such as average daily traffic (ADT), volume to capacity ratio (V/C) and surface texture depth (STD) that were responsible for generating the VOCs, TSS and TOC. PCA was performed on each of the pre-treated build-up data matrices. Figure 1 shows the PCA biplot of the total particulate build-up data matrix.

Insert Figure 1

In Figure 1, VOCs form a group (A) consisting of toluene (TOL), ethylbenzene (ETB), meta and para-xylene (MPX) and ortho-xylene (OX) that are very strongly correlated with each other on both PCs. This suggests that they originated from similar or even the same sources. The negative loading of ADT and the strong positive correlation between V/C and the group (A) volatiles on PC1 suggest that a highly congested traffic lane of a road is more likely to cause the volatile organic build-up than the traffic count (ADT) on the road. The texture of the road surface could also affect the capture of particulate matters that might be associated with organics during the build-up phase. Patra et al. (29) have illustrated that vehicle induced turbulence caused faster resuspension of coarser particulate matter than the finer particles on road surfaces. Mahbub et al. (30) have shown that coarser particles from 75 µm to >300 µm exhibit rapid resuspension and redistribution due to traffic activities in commercial and
industrial areas. Hence, in this study, the VOCs that were associated with coarser particulate matters from 75 µm to >300 µm were getting quickly redistributed and caused the inverse relationship with the free flowing traffic.

None of the objects except R1 had any noteworthy positive scores on PC1. Only four out of eleven objects (I5, R2, I4, and R8) had negative scores along with negative loadings from group (A) organics on PC2 which accounts for only 11.5% variance. This meant that the group (A) VOCs were slightly present in two industrial sites (I4 and I5) and three residential sites (R1, R2 and R8). However, these VOCs were not present in any of the commercial sites (C6, C7, C9, and C10) or in the two remaining residential sites (R3 and R11). This suggests that land use (commercial, residential and industrial) did not have any clear-cut significant influence on the build-up of traffic generated VOCs, even though partial influence from some residential and industrial areas could be observed. Similar findings for the four particulate and the potential dissolved fractions strengthened the above conclusions relating to the measured variables. Modelled traffic data as well as small sample size might have contributed to such facts. Strong correlations between group (A) VOCs and TSS and TOC were observed in all size fractions. Organics’ association with solids during build-up could be attributed to such phenomenon. PCAs for each size fractions are provided in the supporting information.

Wang and Zhao (3) reported that the target VOCs were mostly found in the gaseous phase due to their high vapour pressure. This might have caused re-emission of the VOCs and hence the resulting weak correlations between land use and VOC concentrations in build-up. The deposition of VOCs on the pervious and impervious surfaces has been related to various meteorological conditions such as temporal fluctuations in wind activities (31) as well as wind velocity and vertical diffusivity with the height above ground (32). Furthermore,
Johnson and Belitz (33) have reported statistically significant correlation between VOC concentrations with urban land uses whilst investigating the groundwater quality near water supply wells. In another study, Pitt et al. (34) illustrated high detection frequencies of organic toxicants in particulate sample fractions originating from vehicle parking and service areas. Therefore, this study incorporated the different land uses into the model calibration even though weak correlations existed between land use and VOC concentrations at the study sites.

Factor Analysis

In order to extract the independent factors latent in the data structure, factor analysis (FA) was performed in two phases, namely, PCE for factor extraction followed by orthogonal VARIMAX rotation for the data matrices of all size fractions. The rotated component matrices for the total particulate fractions as well as all fractions including the dissolved fractions showed that two independent factors effectively separated the nine measured variables in Table 1.

Table 1 shows that in both matrices TSS, ADT, TOC and STD are associated with one factor whilst all the target volatile organics and V/C are associated with the other. These two independent factors were extracted based on the initial eigenvalue criteria $\geq 1$. Out of eleven road sites, three were chosen to establish a calibration set and eight were chosen for the validation set for the PLS prediction model. Both, the calibration and the validation sets consisted of a mix of residential, commercial and industrial sites from both suburbs.

Experimental Design
The calibration set for the PLS model was optimised using a two level orthogonal experimental design (35, 36). As the levels of the extracted factors were unknown, the two levels were set for high and low (37). The design incorporated fifteen experiments altogether with twelve individual experiments and three replicate experiments at centre. The calibration set for the experiment was chosen from the five different size fractions with each contributing three experiments. At each size fraction, the residential, commercial and industrial sites contributed one experiment each. The central or replicate experiments are chosen in order to identify any curvature present on the response surface. However, these replicate experiments were chosen from the same site at different fractions in order to minimise the intra-site variability. The optimised calibration set is provided in the supporting information. The PCA biplot of the calibration matrix is presented in Figure 2.

In Figure 2, ADT, TOL, ETB, MPX, OX and VC have positive loadings and STD has negative loading on PC1. Only four experiments (E1, E4, E5 and E7) have either positive or negative scores on PC2 while none of the target volatile organics have any significant loadings on PC2. ADT, V/C and the target VOCs are well presented in at least 6 of the experiments (E2, E3, E5, E6, E8 and E11) whilst TSS, TOC and STD are presented in the rest (E1, E4, E7, E9, E10 and E12) on PC1. Moreover, the three central experiments (C1 to C3) are very close to the origin and have no significant scores on any of the PCs. This means that these central experiments are re-runs of the same experiments and do not need to be included as part of the main experiments. The curvatures of the two factor response surfaces for toluene, ethylbenzene, meta and para xylene and ortho-xylene were also checked by
comparing their mean response values with the means of the three central experiments and no curvature was found for the target volatile organics in the calibration set.

**PLS Model Validation**

In the PLS regression, the target volatile organics TOL, ETB, MPX and OX were considered as dependent or measured variables and TSS, TOC, ADT, VC and STD were considered as the predictor variables. A cross validation method (38), leaving one experiment out at a time from the calibration set was used to measure the standard error in cross validation (SECV). The following three criteria were used to decide the required number of PLS components for regression:

- The SECV<1
- 15% maximum difference between the percentage variance explained by the predictor and the measured variables
- There is no dramatic increase in the percentage variance explained by the predictor with the inclusion of an additional PLS component.

Table 2 shows the outcome of the PLS regression based on the above criteria.

Insert Table 2

As a final step in model validation, data matrices were constructed from the eight remaining road sites. Each matrix consisted of the five predictor variables from the calibration set and eight objects. As the TSS and TOC were measured for the five different size fractions mentioned above, the PLS model was then applied to each of them in order to predict the target volatile organics in the five fractions. A 95% confidence limit was set to deal with the
uncertainty in the prediction (39). The model was able to predict the target volatile organics for each of these fractions at most study sites with very few exceptions. Figure 3 presents the prediction results for each size fraction which includes at least one of the target volatile organics.

In Figure 3a, at around 50% of the study sites, the predicted measurements of toluene are very close to their observed measurements for >300 µm. In Figure 3c, around 88% of the predicted measurements of ethylbenzene are very close to their observed measurements for 75-150 µm. Acceptable results were achieved for other target volatile organics in Figures 3b, 3d and 3e except for toluene at only one study site for >300 µm in Figure 3a. In order to achieve a better perception of the overall PLS model performance, five different error statistics were measured based on the relative prediction error (40). The relative prediction errors (RPE) for each of five size fractions were evaluated as follows:

\[
RPE_f = \left( \frac{\sum_{i=1}^{f} (X_{i,\text{observed}} - X_{i,\text{predicted}})^2}{\sum_{i=1}^{f} (X_{i,\text{observed}})^2} \right)^{0.5}
\]

and the total prediction error at the study sites was expressed as follows:

\[
RPE_{\text{Total}} = \left( \frac{\sum_{i=1}^{f} \sum_{j=1}^{n} (X_{ij,\text{observed}} - X_{ij,\text{predicted}})^2}{\sum_{i=1}^{f} \sum_{j=1}^{n} (X_{ij,\text{observed}})^2} \right)^{0.5}
\]

where \( n \) and \( f \) are number of sites and number of fractions respectively, and \( X_{ij} \) denotes the concentration of the volatiles at \( i \)th fraction at the \( j \)th study site. Figure 4 gives the error statistics.
In Figure 4, the RPE for the five size fractions ranged between 10% to 40% whilst the total RPE at all study sites ranged between 28% to 40% for the different target volatile organics using single component PLS prediction.

Ni et al. (41) reported up to 36% RPE for the prediction of nitrobenzene and nitro-substituted phenols using the single component PLS method. The calibration set for their model could be prepared in the laboratory under stringent quality control measures, whereas in this study, the preparation of the calibration set was beyond laboratory control as measurements of some variables such as ADT, V/C, STD could not be performed otherwise. Nevertheless, this study produced a robust PLS prediction model by exhibiting sufficient confidence interval (95%) in uncertainty of prediction. The main advantage of this model was the identification of a better calibration set for the orthogonal design by considering the independent factors latent in the original data matrix using the two phase factor analysis. This approach provided the calibration set underpinned by the factor optimisation technique whereas the earlier study by Ni et al. (41) used the number of measured variables as the factors in the optimisation of the calibration matrix. Bias could be introduced into the calibration if all dependent variables are considered as factors.

The inter-site coefficients of variation (CV) for the VOC concentrations at ppb level at five different size fractions as well as the total sample were calculated and shown in Figure 5.
In Figure 5, the percentage CV of the predicted VOC concentration varied between 37% to 45% for toluene, 25% to 34% for ethylbenzene, 24% to 36% for meta and para xylene and 36% to 45% for ortho xylene for the five different size fractions as well as the total sample at three different land uses. Regardless of the test methods and the analytes, Horwitz (42) suggested the acceptable inter-laboratory coefficients of variation at the ppb level concentration to be ±45%. Thus, in this study, the abovementioned CV(%) suggest that the prediction model performed fairly accurately in three different land uses as well as for the five different size fractions.

List of Figures

Figure 1 PCA biplot of total particulate volatile organic build-up on urban roads; objects are indicated by labels with the prefix I, C or R for industrial, commercial and residential sites respectively

Figure 2 PCA biplot of the calibration set shows the scores of fifteen experiments and the loadings of nine measured variables

Figure 3 Prediction results of (a) Toluene; (b) & (c) Ethylbenzene; (d) Meta & Para Xylene; (e) Ortho xylene at different size fractions showing most predictions were within 95% confidence limit

Figure 4 Relative Prediction Error (RPE) at each size fractions and total RPE at all study sites

Figure 5 Coefficients of variation (CV) at each size fractions and total sample at all study sites
Figure 1
Figure 2
Figure 3
Figure 4
Figure 5
Table 1. Correlation coefficients matrix after VARIMAX factor rotation with bold coefficients showing variables strongly associated with corresponding factors

<table>
<thead>
<tr>
<th>Measured variables</th>
<th>Correlation coefficients after rotation for total particulate fraction</th>
<th>Correlation coefficients after rotation for all fractions taken together</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Factor 1</td>
<td>Factor 2</td>
</tr>
<tr>
<td>TSS</td>
<td>.954</td>
<td>.253</td>
</tr>
<tr>
<td>ADT</td>
<td>-.952</td>
<td>-.267</td>
</tr>
<tr>
<td>TOC</td>
<td>.889</td>
<td>.450</td>
</tr>
<tr>
<td>STD</td>
<td>.719</td>
<td>.682</td>
</tr>
<tr>
<td>VC</td>
<td>.051</td>
<td>.872</td>
</tr>
<tr>
<td>OX</td>
<td>.540</td>
<td>.811</td>
</tr>
<tr>
<td>MPX</td>
<td>.578</td>
<td>.803</td>
</tr>
<tr>
<td>TOL</td>
<td>.579</td>
<td>.799</td>
</tr>
<tr>
<td>ETB</td>
<td>.528</td>
<td>.792</td>
</tr>
</tbody>
</table>

Table 2 PLS Regression parameters for the predictor variables

<table>
<thead>
<tr>
<th>Measured variables</th>
<th>PLS components</th>
<th>variance explained by predictor variables, %</th>
<th>variance explained by measured variables, %</th>
<th>Coefficient of Determination, $r^2$</th>
<th>SECV</th>
<th>Regression Coefficients for predictor variables</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>TSS</td>
</tr>
<tr>
<td>TOL</td>
<td>1</td>
<td>65.42</td>
<td>67.84</td>
<td>0.7</td>
<td>0.66</td>
<td>-</td>
</tr>
<tr>
<td>ETB</td>
<td>2</td>
<td>68.28</td>
<td>53.26</td>
<td>0.6</td>
<td>0.99</td>
<td>0.00001</td>
</tr>
<tr>
<td>MPX</td>
<td>1</td>
<td>65.51</td>
<td>59.97</td>
<td>0.69</td>
<td>0.73</td>
<td>0.00003</td>
</tr>
<tr>
<td>OX</td>
<td>1</td>
<td>65.64</td>
<td>71.14</td>
<td>0.71</td>
<td>0.60</td>
<td>0.00001</td>
</tr>
</tbody>
</table>
Acknowledgements

This study was undertaken as a part of an Australian Research Council funded linkage project (LP0882637). The first author gratefully acknowledges the postgraduate scholarship awarded by the Queensland University of Technology to conduct his doctoral research. Contributions from Vince Alberts at Queensland Health for VOC analysis and the support from and Gold Coast City Council are also gratefully acknowledged.

Supporting Information Available

Detailed description of quality control measures in sample testing were presented in the supporting information section. The data analysis techniques used in this study were also discussed. An additional figure (Figure S1) was provided to present the study sites and the relevant traffic and pavement data. This information is available free of charge via the internet at http://pubs.acs.org.

Brief

This study has presented a model to predict the traffic generated VOC concentrations on urban roads and hence differentiated the prediction of VOCs on urban roads from that of ambient urban atmosphere.

References


movement and dispersion due to traffic on an urban road. *Atmos. Environ.* **2008**, *42*, 3911-3926.


