

# Predicting odorous emissions from bitumen collected at 13 refineries: A combined GC-MS and supervised learning approach

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## ABSTRACT

During asphalt paving operations, bitumen emissions occasionally give rise to unpleasant odours attributed to volatile organic compounds. While infrequent, these odours can significantly disrupt community well-being, local air quality, and workers' productivity. Predicting odours from a bitumen source before its use in the field is an ideal strategy to address these challenges proactively.

This study introduces a novel Linear Discriminant Analysis (LDA) method, utilising data from headspace gas chromatography-mass spectrometry (HS-GC-MS) of bitumen samples to forecast the likelihood of odours in bituminous road binders.

The LDA model, developed using HS-GC-MS results from sixteen straight-run binders of known odour status collected globally, demonstrates high accuracy in odour prediction through two cross-validation techniques. This

accuracy enables the rapid identification of odorous bitumen samples using GC-MS data. Furthermore, our method suggests a substantial contribution to odour from alkanes and arenes.

The proposed approach provides a simple and practical tool, offering the potential for selective use or pre-treatment of bitumen, thereby reducing the introduction of highly odorous binders into paving projects.

This methodology presents an innovative step towards proactive odour management in asphalt paving, contributing to community well-being, environmental quality, and the efficiency of paving operations.

### Keywords:

Bitumen  
Odour  
Discriminate analysis  
Emissions  
Pollution

## 1. Introduction

Bitumen used as a binder in paving projects can occasionally produce significant malodours that can lead to disruption of local communities or the shutdown of a paving project.[1–3] Odour pollution is a substantial and often underestimated factor influencing air quality in local communities.

The disruptive odours that can emanate from bitumen not only pose challenges to workers on paving sites but also have far-reaching effects on the overall well-being of nearby residents and businesses.

[4,5] Although not necessarily indicative of harmful emissions, these odours can cause substantial discomfort and distress among local inhabitants, potentially impacting their quality of life and daily activities.

[1,6] Consequently, many countries have recognized the need for stringent air quality and odour emissions regulations.

These regulations serve as essential measures to safeguard the health and comfort of residents and maintain the air quality of local environments.[7]

There is significant literature on the emissions and toxicological aspects of bitumen.[8–32]

However, odour is a largely unresolved issue in bitumen materials research, occupying little space in the literature relative to the bulk of bitumen research.[1–3]

Odour in relation to bitumen within this work refers to a distinctly poor odour that is significant enough to cause disruptions and complaints from personnel.[1,6]

This poor odour is distinctively different from fuming where large volumes of fume are emitted from bitumen. [33,34]

While many analytical measures of bitumen properties can be assessed quantitatively, odour is more subjective and susceptible to environmental conditions making its metrics largely qualitative.[35–39]

Specific chemicals that are known to be odorous can be quantified, but given the range of chemicals present in the emissions of bitumen, quantification of all these chemicals is a monstrous task and may still struggle to describe a clear picture of the odour produced from such a complex substance.

Consequently, there are many challenges in identifying odour in bitumen products due to this range of chemicals and, the techniques available to analytically assess odour.







[1,5,6,38,39]

In attempts to quantify odour from bitumen, some studies have sought to identify correlations between particular chemicals and odour, [1,6] the literature on odour from bitumen emissions is notably constrained.

These works have aimed to refine the list of chemicals influencing odour, however, the range of compounds contributing to poor odour is vast and encompasses hydrocarbons, aromatics, and heterocycles. Moreover, it must be noted that these chemical lists of

odorous compounds are yet to undergo thorough validation through additional research.[1,6]

Compelling methods to predict odour are yet to be presented in relation to bitumen emission odour. [1–3,6,40] Determining the odour potential of a bitumen before it is deployed in road construction operations may reduce the need for odour reducing or masking additives.[1]

A predictive method could reduce negative effects on air quality and the delays that a highly odorous bitumen may cause due to disruption to workers or communities.

Assessing odour from complex mixtures often requires human participants to form an audit panel, thereby limiting its application.[41,42]

Methods that consider odour by statistical means present a more simplified approach to considering complex mixtures of chemicals.

[2,3,40,43–46] Statistical analysis of bitumen emissions and odour has been investigated previously with limited application.[2,3,40]

The study by Autelitano and Giuliani[3] focused on characterising bitumen emissions and odours using handheld equipment capable of measuring the concentration of specific chemical classes from gases but not identifying specific chemicals. Their approach involved considering complex odorous mixtures by studying the relative response of various chemicals. The researchers incorporated principal component analysis to reduce dimensionality and separate samples based on the first two principal components, enabling the differentiation of samples based on the relative responses of a range of chemical classes.

This statistical treatment and analysis of bitumen emissions exemplify methods that can address the intricacies of odours within complex gaseous mixtures.

Importantly, it is worth noting that the study did not extend its research to predicting odorous characteristics in bitumen, highlighting a notable

gap in the literature that our work aims to fill.

Discriminant analysis is a statistical technique used to identify distinguishing characteristics in datasets that segregate samples into distinct classes. When using Linear Discriminate Analysis (LDA), often referred to as Fisher's LDA,[47]  $p$  variables are used to construct discriminate axes.

The  $p$  variables are the initial set of features within a

dataset, such as the measurements taken for each sample. An axis is constructed using the  $p$  variables to maximise the separation between two or more data classes.[48] This is done by maximising the distance between the means of each group for a given variable while minimising the spread of values within each group.[49]

The application of LDA tailored specifically for odour assessment has proven successful in evaluating malodours associated with food products.[45,50–54]

LDA has emerged as a powerful tool for discerning complex odour profiles in these well-established areas, providing valuable insights into sensory perceptions, quality control, and product development.

Applying LDA to evaluate odours from bitumen emissions represents a largely uncharted territory[2,3,40] with potential for innovation in terms of predictive analysis. The perception of odour in bitumen can be considered a multifaceted phenomenon influenced by intricate mixtures of chemicals.[35,36,41,42,55] Considering not only chemicals known for their malodorous properties but all the chemicals that constitute the emission profile becomes pivotal when assessing odours within complex mixtures.

The amalgamation of seemingly inconspicuous chemicals may give rise to distinctly odorous mixtures. Consequently, a comprehensive model tasked with evaluating odour risk must also consider chemicals that may initially appear inconsequential to the overall odour potential of bitumen.

Although bitumen samples share many of the same chemicals in their emissions, the concentrations of these chemicals can exhibit significant variations among different samples.[56]

Here we examine chemicals present in bitumen emissions to predict odours based on well-established historic bitumen samples with a record of malodour. By adopting a comprehensive approach that considers various chemicals and their intricate mixtures within bitumen and its fumes, this research aims to provide valuable insights into predicting odour attributes linked to bitumen emissions.

## 2. Methods and materials

To prepare the sample,  $1.00 \pm 0.01$  g of bitumen was placed in a sealed headspace vial with an incubation time of 90 min to establish equilibrium.

The method was carried out on an Agilent 8890 GC/5977B GC/MSD instrument fitted with CTC PAL RSI 120 attachments, including a headspace sampler with sample heating and agitation.

Headspace sampling was completed automatically with the CTC PAL systems 8010–0265 2500  $\mu$ L headspace sampler. The samples were heated at 150 °C for 90 min in the agitator before gas was extracted with the automated syringe. A headspace





sample volume of 500 µL was used.

The injection mode was set to split (1:10). The injection port was set to 250 °C and was fitted with an Agilent 5190–3983: 800 µL (single taper) linear. The GC was fitted with a J&W DB-624 GC Column (30 m, 0.25 mm, 1.40 µm).

Flow was 1.525 mL/min, and pressure was 12.685 psithrough the column.

This method was used for both scan and selective ion monitoring (SIM) runs. Using a scan mode with mass ranges between 33 and 300 m/z a range of potential compounds that could be used to predict odour were analysed (Table S1) and the responses of the integrated molecular ion were recorded.

Using SIM, the analytes previously found to be predictive of odour were analysed, the molecular ions were integrated, this is found in Table 1.

The selected analytes were confirmed based on library matching from scan data and analytical standards two analytical standards: ASTM surrogate base gasoline (RGO-711–1) and benzene, toluene, ethylbenzene and m, p, o-xylenes (BTEX) standard (BTX-2000 N), both purchased from Agilent.

Sixteen unique bitumen samples were used in this work, they were provided by Puma Energy (Australia)

Bitumen Pty Ltd and were taken from a library of samples with known performance spanning two decades.

The 16 samples were received from 13 different refineries across various international regions (Table 2). All samples were collected from a sealed tin containing the bulk sample, all samples had been stored indoors in controlled conditions since decanting on the scale of millilitres to litres.

None of the bitumen samples were modified bitumen.

The sample set contained 8 samples with no odour complaints and 8 samples that had confirmed odour status. The samples were acquired from a range of countries and were composed of a range of penetration grades (Table 3).

These were assigned to 'No odour' and 'Odour' groups by Puma as part of a survey reviewing problematic bitumen products, the complaints were assessed and substantiated by Puma Energy Bitumen.

The results were supplied for this work as qualitative binary data of 'No odour' and 'Odour'. All samples were run in duplicates with sample 44771 being run four times to measure variance. The duplicates were not averaged but entered as two individual data points.

**Table 1**  
Molecular ions of selected analytes using the SIM method.

No.	Analyte	M <sup>+</sup> ion
1	Pentane	72
2	Hexane	86
3	Benzene	78
4	Heptane	100
5	Octane	114
6	m, p-Xylene	106
7	Nonane	128
8	o-Xylene	106
9	1,3,5-Trimethylbenzene	120
10	1,2,4-Trimethylbenzene	120
11	1,2,3-Trimethylbenzene	120

**Table 2**  
Bitumen sample source, names and status used this this study.

Refinery	Name	Penetration grade	Group
A	207	160/220	No odour
B	44,771	80/100	Odour
C	23,762	70/100	Odour
D	22,707	70/100	Odour
E	46,558	160/220	Odour
F	47,272	50/70	Odour
G	198	50/70	No odour
A	204	50/70	No odour
H	227	35/50	No odour
I	237	40/60	Odour
J	25,249	70/100	No odour
F	33,248	70/100	No odour
K	180	10/20	No odour
L	C170	70/100	No odour
M	49,585	70/100	Odour
M	49,586	160/220	Odour

**Table 3**

Leave-one-out cross-validation accuracy and kappa for LDA model.

Cross-validation accuracy rate	1.0000
Cross-validation error rate	0.0000
Kappa	1.0000

This was done using the HSGC- MS method, producing 34 data points, 16 'No odour' and 18 'Odour'. To prepare a sample  $1.00 \pm 0.01$  g of bitumen was collected using a metal spatula without heating so any emissions could be minimised from the bulk sample. This was then added to a headspace vial and sealed.

The reproducibility of the samples was very high despite the collection of cold bitumen.

The data used in this study consisted of the integration values of the molecular ions for each of the 11 analytes in Table 1, this was done using Agilent MS quantitative analysis build 10.1.733.0. Once processed the dataset comprised 30 data entries and 11 predictor variables.

The integration values for each analyte were computed as a fraction of the net area of the 11 peaks (Eq. S1). After this, the data was then normalised using vector normalisation (Eq. S2). Next, a series of feature ratios were computed from the dataset. These feature ratio values were calculated to capture the relationships between the features in the dataset, which represents pairs of analytes in the emissions.

This enables a greater focus on the relationships between features. The dataset consists of 11 normalized vectors, each corresponding to an analyte. These features are systematically divided by each other, yielding a total of 121 values.

Among these, 11 values result from identical divisions that are equal to 1, reducing the count to 110. Half of these values are reciprocals, such as  $1/2$  and  $2/1$ , which convey the same information in this work. Consequently, this process culminates in the extraction of 55 variables, labelled V1-V55. This

refined dataset was then integrated into the LDA model for further analysis.

All data pre-processing, model training, and statistical analyses were performed using the R Studio programming language (version 2023.06.0 + 421).[57] The 'MASS' package[58] was utilised for the implementation of the LDA model and permutation test.

The corrplot package[59] was used for generating correlation matrix plots. And the ggplot2 package was used for graphical manipulation.[60] To differentiate between the odour and non-odour groups, we employed LDA.

LDA is a supervised classification technique that finds linear combinations of the predictor variables to maximize the separation between predefined groups. The LDA model was trained using the LDA function from the MASS package in R Studio.

To evaluate the performance and generalization ability of the LDA model, leave-one-out cross-validation (LOOCV) was conducted. LOOCV is a technique that iteratively trains the model using all but one observation, and then evaluates the model's performance on the left-out observation.

This process is repeated for each observation in the dataset, resulting in an accurate estimate of the model's predictive performance.

To determine the statistical significance of the model's performance two cross-validation methods were used. LOOCV and 5-fold crossvalidation tuning the shrinkage hyperparameter for shrinkage linear discriminate analysis (SLDA) with the lambda and diagonal tune grid set

between 0 and 1 with increments of 0.1. The 'caret' package[61] was used for the cross-validation (CV) methods.

The collinearity of variables was assessed using a correlation matrix generated using the corrplot package.

Collinearity refers to the correlation or interdependence between predictor variables, which can affect the reliability and interpretability of statistical models.[49,62–64]

### 3. Results and discussion

In the initial phase of selecting predictor variables, a comprehensive assessment was conducted on 32 analytes to evaluate their ability to predict odour in bitumen samples (Table S1).

These analytes were chosen not only for their potential to produce odour but based on their presence in all samples to varying extents.

Thiols and disulfides were excluded from this evaluation as they were present in only a subset of the samples, and when they did appear it was in both odorous and nonodorous samples.

Consequently, despite their known poor odour[65,66] no definitive conclusions could be drawn regarding their contribution to odour in these bitumen samples. During the assessment of the model and its prediction capabilities many of these compounds did not contribute to the separation of the samples into odour and non-odour samples.

Therefore, among these 32 analytes, a subset of 11 compounds were selected (Table 1), there included alkanes, benzene, and benzene derivatives, they demonstrated a high level of efficacy in predicting odour using LDA.

The analysis identified collinearity among the predictor variables. Collinearity indicates a linear relationship between pairs of predictor variables, which can be positive or negative. High positive or negative values of collinearity can obscure the individual effect that a predictor has on the prediction

of the samples. Vector normalisation had some effect on reducing the collinearity but not significantly.

This collinearity therefore carried over into the feature ratios dataset (Fig. S1). This observation is not unexpected due to the underlying relationships between the chemicals that produce these variables.

Given that bitumen is derived from crude oil, it is reasonable to expect the concentration of an alkane to be relative to other alkanes in the mixture, and the same can be expected for benzene and its derivatives. While overall there is observed collinearity, when considering the GC–MS data there are still significant variations in the ratios of analytes present in samples. Typically, collinearity raises concerns in statistical modelling due to its potential to obscure individual variable impacts.[62]

Rather than seeking to isolate the effects of individual analytes, our primary focus lies in evaluating the model's efficacy. As a result, we have consciously chosen to retain these collinear variables in our analysis to pursue a pragmatic approach by prioritising the model's practical utility.

The analysis conducted with the LDA model unveiled a clear and significant separation between groups, as depicted by the linear discriminant value (LD1) in Fig. 1. This outcome underscores the model's ability in distinguishing between the odorous and non-odorous groups.

Notably, the limited variability observed within each group suggests that the predictor variables exhibit minimal fluctuations.

Given the smaller size of the sample set CV needs to be carefully considered. Two methods for CV were carried out and compare to assess the performance of the LDA model. Firstly, a LOOCV method was used to maximise the size of the training data set. Secondly, A 5-fold crossvalidation method was used to assess the model more pessimistically. demonstrated an excellent ability to distinguish between the odorous

and non-odorous groups through LOOCV. Demonstrating a 100% accuracy rate in LOOCV, as detailed in Table 3. This high accuracy helps to validate the model's competence in classifying the bitumen samples and capturing vital data associations.

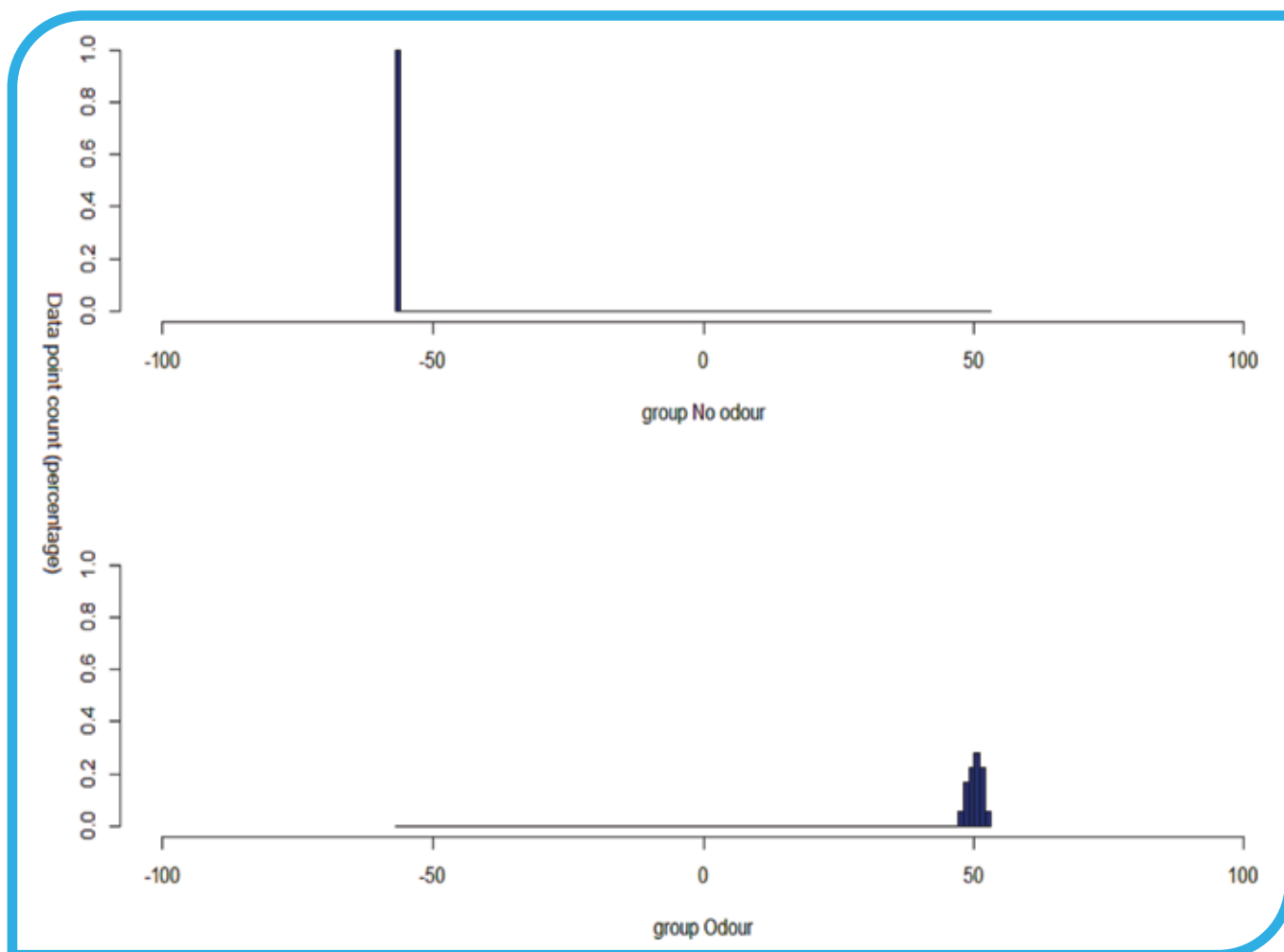


Fig. 1. LDA histogram of No odour and odour groups plotted on the first linear discriminate axis (LD1). The groups are separated along the LD1 axis, the group means are - 56.4 and 50.2 for No odour and Odour respectively, with minimal spread within each group where variance is 0.037 and 1.8 for No odour and Odour, respectively. The odour groups contain 18 samples, and the No odour group contains 16 samples.

Kappa values range from - 1 to 1 and where a kappa close to 1 indicates a good agreement between the model and the true labels of the samples, the obtained kappa value of 1 provides statistically solid evidence that the observed accuracy rate is significant and not due to chance.

Assessing the model when tuning shrinkage in LDA using a 5-fold cross-validation method yielded an equally high accuracy and kappa (Table 4).

that the LDA model demonstrated robust discriminatory power in predicting the groups. The

high accuracy rate and kappa value support the notion that the observed predictions are unlikely to result from random chance.

**Table 4**

Five-fold cross-validation accuracy and kappa for SLDA model including optimisation metrics diagonal and lambda.

Cross validation accuracy rate	1.0000
Cross-validation error rate	0.0000
Kappa	1.0000
Diagonal	0
Lambda	0.1



Thus, the LDA model exhibits reliable predictive capabilities for distinguishing between the odour and no odour groups.

Implementing methods for detecting odorous bitumen before deployment may minimise the occurrence of worker complaints, interventions by environmental regulatory agencies, and the need for deodorising and masking treatments.

These methods can result in cost savings and smoother project execution while reducing odour pollution in surrounding areas.

Given the intricate nature of odorous emissions in complex organic mixtures, it is essential to employ methods considering a range of organic chemicals and their interactions contributing to odour.

Statistical methods like LDA, in conjunction with analytical instruments, present a methodology capable of considering the complexity of odour in bituminous materials.

These approaches are particularly valuable when a training dataset is accessible, as they allow for a comprehensive examination of the complex relationships involved.

While the response of arenes and alkanes in bitumen do not correlate with odour directly based on their relative responses from GC-MS, when considering the ratios of these chemicals collectively with appropriate statistical methods, odorous samples can repeatably be distinguished from non-odorous samples using an LDA model.

This suggests that the 11 selected alkanes and arenes are significant contributors to poor odour in bitumen. While other VOCs that contain nitrogen, oxygen and sulfur and have various levels of saturation are likely to contribute to bitumen odour (Table S1), these findings suggest a previously unexplored association between bitumen odour and alkanes and arenes. Although novel in the context of bitumen, such

association are not unprecedented in other industries, where these chemical classes have been contributors in the generation of poor odour.[67]

While isolated alkanes or benzene derivatives may exhibit relatively benign scents,[68] their presence in bitumen, combined with other volatile compounds, can give rise to complex odour profiles.

Their role as potential odour predictors in bitumen extends beyond their individual scent profiles.[67,69] The perception of odour is highly dependent on the mixtures' specific chemical composition.

Some mixtures may elicit the perception of distinct individual odours, exhibiting varying degrees of dominance and blending. Conversely, certain mixtures may present a homogeneous blend where only one component predominates the perceived odour. Furthermore, mixtures can generate a unique, collective odour attributed to the overall composition of the mixture.[55] Research has illustrated how mixtures of chemicals with each component below its respective odour threshold can still be perceptible by participants when combined. These mixtures can exhibit an additive or hyper-additive effect in regards to detectability and odour intensity, enhancing detectability beyond the contributions of the individual components alone.[41]

In this context, the alkanes and arenes measured in this work contribute significantly to the complex odour profile of bitumen where the synergy of various compounds, rather than the inherent odour of individual components, ultimately defines the odorous characteristics of bitumen. By considering these compounds not as direct sources of malodour but as contributors to a complex odour profile, we gain a more nuanced understanding of how bitumen emissions generate distinct odours. This perspective encourages the exploration of predictive models that consider the combinations of different chemical constituents, providing valuable insights into the factors contributing to odorous bitumen products.

#### 4. Conclusion

This study employed LDA to predict odorous bitumen using chemical information from bitumen emissions. The proposed methodology demonstrated a high level of effectiveness in discriminating among eight odorous and eight non-odorous globally sourced samples, achieving a 100% accuracy rate when assessed using LOOCV and 5-fold crossvalidation SLDA. Our findings highlight the substantial contribution of alkanes and arenes to the manifestation of poor odour in bitumen emissions. Integrating statistical tools, such as LDA, with analytical instruments provides a comprehensive approach to understanding the intricate odour profiles in bituminous materials. While individual compounds may not exhibit a direct correlation with odour, their collective interactions play a significant role in shaping distinct odorous characteristics. Further research is imperative to precisely understand how these compounds influence odour production in bituminous materials.

Implementing pre-deployment methods for detecting odorous bitumen can be a practical solution to alleviate worker complaints, mitigate regulatory interventions, and reduce the need for additional treatments. These measures can result in cost savings and contribute to smoother project execution and a notable reduction in odour pollution within the surrounding areas of paving projects. By prioritising the prediction of odorous bituminous materials, we can effectively limit the entry of poor-quality products into the market. This proactive perspective not only ensures the delivery of high-quality bitumen but also contributes to a substantial reduction in odour pollution from bitumen.

As our work provides a foundation for future investigations, it is evident that ongoing research efforts are crucial to advancing our understanding of the complexities involved in bitumen odour and refining predictive models for enhanced industry practices.

#### Author Contributions.

Zachary Deller did the conceptualization and writing of the original manuscript draft. Filippo Giustozzi

and Subashani Maniam did the supervision, funding acquisition and project development. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

CRedit authorship contribution statement Zachary Deller: Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. Filippo Giustozzi: Writing – review & editing, Supervision, Project administration. Subashani Maniam: Writing – review & editing, Supervision, Project administration. Declaration of competing interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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