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NO₂ levels inside vehicle cabins with pollen and activated carbon filters: A real world targeted intervention to estimate NO₂ exposure reduction potential



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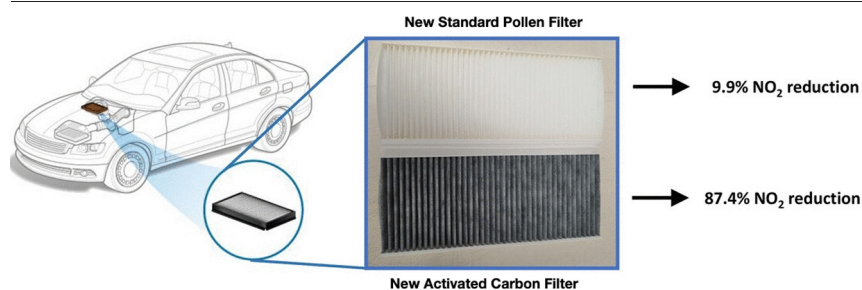
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HIGHLIGHTS

- NO₂ levels were examined inside cars while driving with new pollen and activated carbon filters.
- Implementing new activated carbon filters showed significant in-car NO₂ reduction by 87.4 %.
- Activated carbon filter reduction efficiency drops by 6.8 % per month of use.
- Implementation of activated carbon filters should be considered to reduce in-vehicle exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

Traffic related nitrogen dioxide (NO₂) poses a serious environmental and health risk factor in the urban environment. Drivers and vehicle occupants in general may have acute exposure to NO₂ levels. In order to identify key controllable measures to reduce vehicle occupant's exposure, this study measures NO₂ exposure inside ten different vehicles under real world driving conditions and applies a targeted intervention by replacing previously used filters with new standard pollen and new activated carbon cabin filters. The study also evaluates the efficiency of the latter as a function of duration of use. The mean in-vehicle NO₂ exposure across the tested vehicles, driving the same route under comparable traffic and ambient air quality conditions, was $50.8 \pm 32.7 \mu\text{g}/\text{m}^3$ for the new standard pollen filter tests and $9.2 \pm 8.6 \mu\text{g}/\text{m}^3$ for the new activated carbon filter tests. When implementing the new activated carbon filters, overall we observed significant ($p < 0.05$) reductions by 87 % on average (range 80 - 94.2 %) in the in-vehicle NO₂ levels compared to the on-road concentrations. We further found that the activated carbon filter NO₂ removal efficiency drops by $6.8 \pm 0.6 \%$ per month; showing a faster decay in removal efficiency after the first 6 months of use. These results offer novel insights into how the general population can control and reduce their exposure to traffic related NO₂. The use and regular replacement of activated carbon cabin air filters represents a relatively inexpensive method to significantly reduce in-vehicle NO₂ exposure.

1. Introduction

Nitrogen dioxide (NO₂) is a pollutant of major concern to urban air quality. Ambient NO₂ not only causes acute threats to public health, but also contributes to an economic burden (Eum et al., 2019; Fenech and Aquilina, 2020; Kaufman et al., 2016; Renzi et al., 2018; Schwartz et al.,

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2018), establishing NO₂ as a leading environmental risk factor for health globally. Exposure to NO₂ is known to be associated with reduced lung function and increased risk of cancer (Adam et al., 2015; Hamra et al., 2015; WHO, 2013), and is responsible for tens of thousands of premature deaths each year across Europe (EEA, 2019; RCP, 2016; COMEAP, 2018).

In the urban atmosphere of most developed cities, the dominant source of NO₂ is vehicle combustion where NO₂ is either emitted directly from the exhaust or formed in the atmosphere when nitric oxide reacts with ozone (Matthaios et al., 2019). Vehicle interiors represent microenvironments where human exposure to traffic-related NO₂ occurs. Vehicle cabins can be considered as confined spaces where air quality can be improved compared to on-road or ambient air pollution by a combination of ventilation settings and route choice (Matthaios et al., 2020). By regulating the indoor:outdoor exchange rate via the vehicle's ventilation system or by opening and closing windows, vehicle occupants have the ability to self-control their exposure to both indoor and outdoor pollutants.

Another important intervention to improve in-vehicle exposure to air pollution is to apply and appropriately maintain suitable cabin air filters. Most cabin air filters have a pleated filtering medium that consists of natural and/or synthetic fibers, where the passage of air through the filter creates an electrostatic charge that helps attract and trap small particles (Wang, 2001). Quality filters have several layers of filtering media that use different methods of filtration (impaction, diffusion or interception) and are embedded with an activated carbon layer (Chan et al., 2021). Carbon filters trap gases through a process called adsorption, which occurs when molecules bind to surfaces (Lee and Davidson, 1999; Abumaizar et al., 1998). The more porous the activated carbon, the better, as this will increase the amount of surface area available for pollutant gases to bind onto when air passes through the filter (Przepiorski, 2006). These activated carbon filters can collect pollutant gases and particles and significantly reduce occupants' exposure to diesel exhaust emissions (Muala et al., 2014).

Knowing that in Europe 56 % of the population use their cars as the main transportation option, it is essential to understand controllable exposure reduction mechanisms for better management of occupants' exposure to air pollutants. Aiming to demonstrate the effectiveness of an intervention to improve NO₂ exposure inside cars, this study examines: 1) the NO₂ levels inside different vehicles as a function of air filter choice under real world driving conditions and 2) develops a model to estimate the cabin filter efficiency in reducing in-vehicle NO₂ exposure as a function of months in use.

2. Methods

2.1. Setting

Measurements were performed in Birmingham, UK, the second largest city in the UK with 1.15 million inhabitants. The climate of the city is considered temperate marine with average daily temperatures ranging between 18.6 and 21.4 °C in the summer and 6.6–7.3 °C during winter. Birmingham city is served by the M5, M6, M40 and M42 motorways, with M6 also passing through the city. Birmingham City Council introduced a Clean Air Zone from 1 June 2021, which charges polluting vehicles for traveling into the city centre. The study was conducted from June to December 2021, incorporating the UK summer and autumn. The driving schedule for the campaign took place on the same route, during normal daytime traffic outside of peak periods (11:00–15:00), for weekdays only.

2.2. Experimental set-up and filter details

Ten vehicles were used in the study, ranging in year of manufacture from 2004 to 2020, type: petrol, diesel, hybrid, electric and cabin size (volume) 1.26–2.72 m³. In-vehicle measurements were performed with 1) mechanical ventilation: full fan power setting, air conditioning (AC) on, recirculation off and all windows fully closed, or 2) with full fan power setting, AC and recirculation on and all windows fully closed, or 3) no mechanical ventilation and front windows half-open. The effect of

air filter type and age was explored: each vehicle was tested three times with the original (pollen or charcoal) filter installed (filter already in place for regular use), and then with either a new standard pollen or a new activated charcoal cabin air filter. Three of the vehicles were then re-tested after 3 months of use of the activated charcoal filter. Each vehicle mobile testing measurement lasted between 2.5 and 3 h, with a total of 89.3 h of collection data. New filters from the same manufacturer (Bosch) were used for all vehicles to minimize any variations between manufacturers.

Standard pollen cabin filters typically reduce the burden of particles (i.e. fine particulate matter (PM_{2.5})) entering the vehicle's interior, effectively capturing also pollen, dust, mould spores and debris. Activated carbon filters, besides filtering particulates, are chemically treated with activated carbon that gives them a grey appearance and helps remove a wide variety of gases and odors. These include exhaust gases such as ammonia, sulfur dioxide, nitrogen oxides, hydrogen sulfide and aromatic compounds from incomplete combustion. Table 1 shows the characteristics of the cars and filters.

2.3. Equipment and quality assurance of measurements

The NO₂ measurements were performed using two Thermo Scientific chemiluminescent (EN14211:2012 NO_x) analysers – models 42i-TL (outside) and 42i (inside). The instruments were mounted within the cabin of each vehicle, with two equivalent inlets (to ensure the same losses in the tube walls) with one protruding into ambient air, and the other into the cabin space in the driver's breathing zone. The instruments were serviced and calibrated by the manufacturer before the campaign and prior to the start of the campaign they were co-located next to a reference NO₂ instrument for 2 days in the University of Birmingham Air Quality Supersite (which in turn is calibrated against National Physical Laboratory (NPL) reference standards) for inter-calibration. Both instruments were co-located at the same location after the measurement campaign to examine potential shifts in the inter-calibration of measurements. Fig. S1 in the appendix shows that the inter-calibrations between the two NO_x monitors and the reference NO_x instrument were similar before and after the campaign (R² > 0.97 in both cases) giving confidence in the quality of the measurements.

2.4. Data analysis

We determined the overall mean NO₂ exposure using descriptive statistics. We estimated the mean and max percentage reduction of on-road NO₂ exposure inside the car cabin as follows: (the arithmetic mean concentration on-road – arithmetic mean concentration in-vehicle) / (arithmetic mean concentration on-road) × 100. For the comparison of cases with new pollen and new activated carbon filters in different cars we used descriptive statistics and applied two tailed Welch *t*-test and Wilcoxon test to estimate the in-vehicle: on-road reduction significance at 0.95 confidence intervals. We also used a linear model to calculate the change in the activated carbon cabin filter efficiency with months in use. The performance of the filter with months in use was assessed by the combination of the time since the filter (already in place for regular use during the first experiments) was last changed during a vehicle service and the 3 month follow-up measurements.

3. Results

Of the collected data, 80.3 % were collected during June, July, August and September, while of the remaining 19.7 %, 12.1 % was collected during October and 3.2 and 4.4 % during November and December, respectively. Overall, in-vehicle NO₂ concentrations were on average 1.6 times lower when the windows were closed with air recirculation compared to windows opened. In-vehicle NO₂ concentrations were almost unchanged (factor of 1.03 times lower) between closed windows with fresh air coming into the cabin through the ventilation system and windows open, with new standard pollen filters. However, with new activated carbon filters fitted, in-vehicle NO₂ levels were on average 14.3 times lower with closed windows

Table 1
Vehicle and filter characteristics tested in the study.

Vehicle	Fuel	Cabin size (m ³)	Filter Pleats	Filter size (W × L × H) (cm)	Activated carbon filter weight (g)	Pollen filter weight (g)
Skoda Fabia 2010	Gasoline	2.52	33	24.6 × 21.6 × 3.2	310	130
Saab 9–3 Aero 2004	Gasoline	2.72	40	35.1 × 16.1 × 3	280	100
Ford Focus 2007	Gasoline	2.56	32	44.8 × 20.3 × 3.5	320	180
Ford Transit 2009	Diesel	1.26	28	23.5 × 23.5 × 2	180	100
Ford Puma 2020	Hybrid	2.69	23	20 × 25 × 2	219	65
Ford Fiesta 2010	Gasoline	2.40	28	24 × 19 × 3.5	201	90
Nissan Leaf 2014	Electric	2.62	25	25.5 × 14.5 × 2.5	103	44
Nissan Qashqai 2010	Diesel	2.62	30	20.5 × 19.2 × 2	180	120
Peugeot 108 2016	Gasoline	2.04	25	19.5 × 14.5 × 3.1	160	90
Hyundai i30 2020	Hybrid	2.57	24	20 × 23.9 × 2.2	150	120

and air recirculation, compared with windows open. In-vehicle NO₂ concentration were a factor of 6.6 times higher between windows open, and closed windows with fresh air coming into the cabin through the ventilation system. Fig. 1 shows a time series example of in-vehicle, on-road NO₂ levels while driving a Saab with either a new pollen or a new activated carbon cabin filter.

To examine the impact of new activated carbon and new pollen filters, Table 2 shows the in-vehicle and on-road NO₂ concentrations (µg/m³) measured during mobile experiments with different cars and under mechanical ventilation only. The separate analysis of recirculation mode and fresh air showed a similar reduction with minor differences in the mean NO₂ reduction (1–3%), hence they were included jointly as overall mechanical ventilation. The overall arithmetic mean in-vehicle NO₂ concentrations across all measurements (under mechanical ventilation) were 9.2 µg/m³ (s.d. = 8.6 µg/m³) and 50.8 µg/m³ (s.d. = 32.7 µg/m³) for new activated carbon and new pollen cabin filters, respectively. For old activated carbon filters (filters already in use) the overall in-vehicle NO₂ levels were 34.8 µg/m³ (s.d. = 31.0 µg/m³). The overall highest in-vehicle NO₂ concentrations were 55 µg/m³, 163 µg/m³ and 325 µg/m³ for new activated carbon, old activated carbon and new pollen filters, respectively, and were measured during periods of traffic congestion, when waiting at traffic lights or approaching roundabouts. Lowest concentrations were observed when driving through the clean air zone and in trip segments with clear road ahead. When driving with a new activated carbon filter installed, the

overall mean in-vehicle NO₂ levels were significantly ($p < 0.05$) reduced compared to on-road levels by 87.4% (range 80–94.2%). Similar significant ($p < 0.05$) reductions were observed in the maximum NO₂ levels, where the overall reduction was 91.2% (range from 75.2 to 95.2%). When driving with a new pollen filter in use, the overall mean in-vehicle NO₂ reduction compared to on-road NO₂ concentrations was 9.9% (range from 2.5 to 12.5%) while the overall reduction in the maximum concentration was 4.6% (range from 1.8 to 15.5%). When the cars were measured with their old activated carbon the reductions had larger variations (from 8.6 to 72.2%), that depended on the age of the filter.

Fig. 2 shows the in-vehicle NO₂ reduction as a function of months in use of the activated carbon filter. Greater (nearly constant) reductions were observed in the in-vehicle NO₂ during the first 3 months of the implementation of the activated carbon filter, followed by an approximately linear reduction during months 4–8, with a more rapid reduction after 8 months of use. A significant ($p < 0.05$) inverse relationship was found between the NO₂ in cabin reduction and filter months in use, where the activated carbon filter efficiency was found to fall by $6.8 \pm 0.6\%$ per month ($R^2 = 0.93$). The filter degradation is very small during the first months and there is evidence for a slope change between 6 and 8 months. When taking into account the data only for the first 6 months and comparing them to the data after 6 months, there is evidence for a change in the decay rate (25% change from 68% to 43%), however more data are needed to confirm this change in filter performance.

4. Discussion

The present study has investigated the effectiveness of new pollen and activated carbon cabin filters in reducing NO₂ concentrations inside vehicle cabins under real world driving conditions. The in-vehicle NO₂ levels with the use of new activated carbon were below the current (2021) daily and annual WHO guidelines (World Health Organization, 2021), however, in the case of new pollen filters, the levels were above the WHO NO₂ guideline level of 10 µg/m³ (annual average) and 25 µg/m³ (24-h mean).

On a repeated real-world experimental set up, implementation of new Bosch standard (non-activated carbon) pollen filters, showed no statistically significant changes in the NO₂ levels inside, relative to on-road concentrations, with the actual reductions ranging from 2.5 to 12.5% (Table 2). These results are in agreement with Matthaios et al. (2020), where the ratio of indoor:outdoor NO₂ levels was reduced due to minimizing the air exchange rate in vehicles equipped with pollen filters. In addition, a small amount of in-vehicle NO₂ maybe lost due to reactions with volatile organic compounds and vehicle surface area (Mendez-Jimenez et al., 2021). For the same experimental set-up, the installation of new Bosch activated carbon filters showed much greater and statistically significant ($p < 0.05$) reductions in the in-vehicle NO₂ concentrations varying from 80 to 94.2%, relative to on-road ambient air levels (Table 2). In a real-world study conducted in Volvo Heavy Goods Vehicles, in Sweden, researchers tested 6 activated carbon filters in city and highway driving and stationary in a road tunnel and found 4 of them achieved 80% reductions in the in-cabin NO₂ compared to on-road levels (Moldanova et al., 2019). Similarly, a study in Germany also showed up to 90% reduction to the

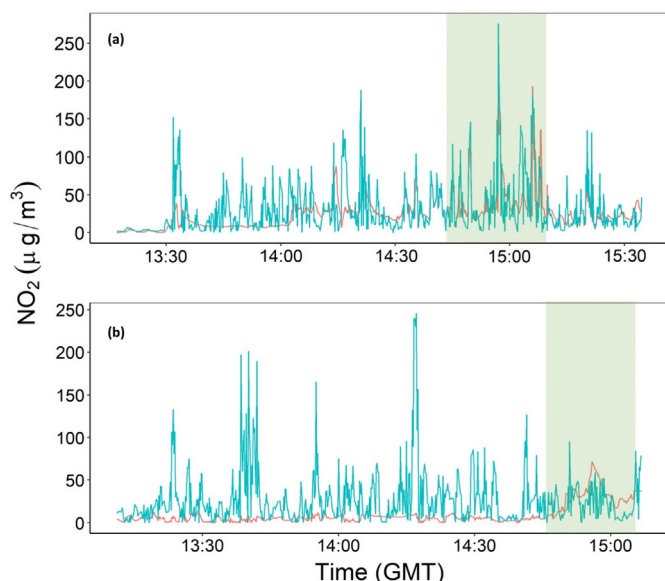


Fig. 1. Time series of in-vehicle (red solid line) and on-road (blue solid line) NO₂ concentrations in a Saab 2004 model while driving on urban roads in Birmingham, UK; (a) test with new pollen filter; (b) test with new activated carbon filter. Unshaded area: windows closed and mechanical ventilation settings on. Green shaded area: windows open and mechanical ventilation off.

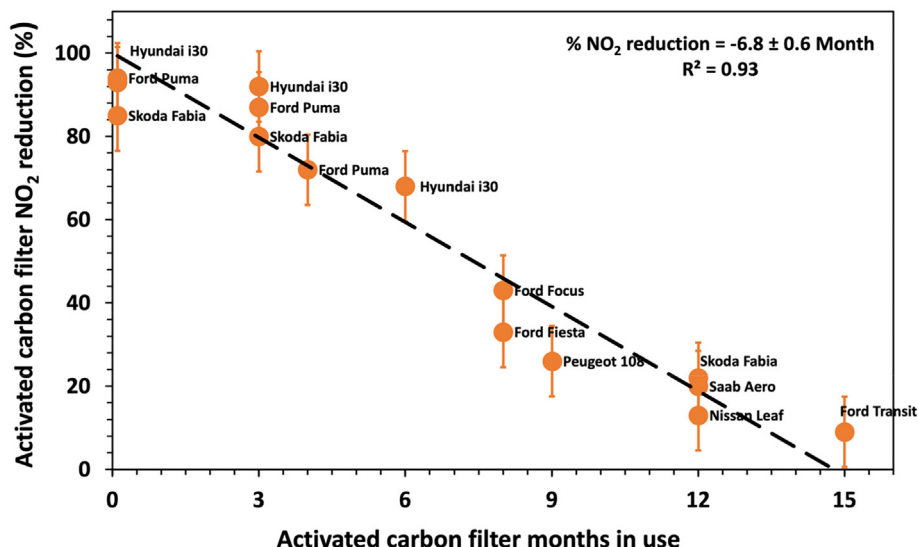


Fig. 2. Relationship between mean in-vehicle NO₂ reductions under windows closed and mechanical ventilation settings on and months in use of the activated carbon filter. The bars indicate the standard error of the mean reduction in each car.

in-vehicle NO₂ compared to on-road concentrations when implementing new activated carbon filters (Pohler et al., 2018). Other studies typically tested cabin filters following the manufacturer laboratory tests and the

ISO TS 11155–2 “Road vehicles - Air filters for passenger compartments - Test for gaseous filtration” standard. Testing in these conditions consists mainly of the measurement of adsorption of several single gases in humid

Table 2

Descriptive statistics of 10 s NO₂ concentrations in-vehicle and on-road (directly outside the vehicle cabins) under mechanical ventilation with new activated carbon, new standard pollen filters, old activated carbon filters (already in use) and 3-month old activated carbon filters. SD: Standard Deviation. g: gasoline car; d: diesel car; h: hybrid; e: electric.

Cabin Filter	Vehicle	Filter Age	On-road NO ₂ (µg/m ³)		In-vehicle NO ₂ (µg/m ³)		Change in Mean (%)	Change in Max (%)
			Mean(±SD)	Max	Mean(±SD)	Max		
New activated carbon	Skoda Fabia 2010 ^g	New	83.5 ± 75.2	294	12.4 ± 9.2	57	-85.1	-89.7
	Saab 9–3 Aero 2004 ^g	New	51.7 ± 61.5	462	10.4 ± 6.2	35	-80.0	-92.4
	Ford Focus 2007 ^g	New	47.4 ± 26.8	529	8.3 ± 12.3	52	-82.5	-90.0
	Ford Transit 2009 ^d	New	86.3 ± 81.2	423	8.5 ± 7.0	57	-81.6	-86.4
	Ford Puma 2020 ^h	New	86.0 ± 102.0	991	11.0 ± 12.7	46	-87.1	-95.2
	Ford Fiesta 2010 ^g	New	47.2 ± 48.7	367	4.5 ± 6.5	48	-90.3	-87.0
	Nissan Leaf 2014 ^e	New	109.6 ± 118.4	1053	7.8 ± 12.0	71	-92.6	-93.2
	Nissan Qashqai 2010 ^d	New	124.0 ± 145.0	1564	19.2 ± 14.2	92	-84.8	-94.1
	Peugeot 1082016 ^g	New	53.5 ± 19.2	301	7.3 ± 10.3	74	-86.3	-75.2
	Hyundai i30 2020 ^h	New	42.8 ± 27.6	187	2.5 ± 1.9	12	-94.2	-93.4
	Overall		73.2 ± 70.3	617	9.2 ± 8.6	55	-87.4	-91.2
	New Pollen	Skoda Fabia 2010 ^g	New	51.9 ± 50.0	360	55.4 ± 49.3	353	-2.5
Saab 9–3 Aero 2003 ^g		New	53.8 ± 61.0	519	49.5 ± 48.5	494	-7.9	-4.8
Ford Focus 2007 ^g		New	52.0 ± 54.5	421	45.4 ± 38.2	404	-12.5	-4.0
Ford Transit 2009 ^d		New	106.5 ± 63.7	418	103.0 ± 44.4	406	-3.3	-3.0
Ford Puma 2020 ^h		New	63.4 ± 85.0	301	57.0 ± 40.6	281	-9.5	-6.7
Ford Fiesta 2010 ^g		New	63.0 ± 54.5	338	55.1 ± 30.8	286	-12.5	-15.3
Nissan Leaf 2014 ^e		New	44.6 ± 41.0	313	40.6 ± 20.0	309	-8.9	-1.2
Nissan Qashqai 2010 ^d		New	52.4 ± 44.7	280	49.3 ± 26.5	268	-5.9	-4.2
Peugeot 1082016 ^g		New	44.7 ± 50.8	440	41.0 ± 23.3	372	-8.3	-15.5
Hyundai i30 2020 ^h		New	32.3 ± 9.7	45	25.6 ± 5.5	42	-10.7	-6.8
Overall			56.4 ± 32.6	340	50.8 ± 32.7	325	-9.9	-4.6
Old activated carbon		Skoda Fabia 2010 ^g	12	39.4 ± 23.1	131	26.5 ± 7.5	103	-24.2
	Saab 9–3 Aero 2003 ^g	12	38.0 ± 36.8	442	30.8 ± 10.1	360	-18.9	-18.4
	Ford Focus 2007 ^g	8	27.4 ± 30.1	223	15.3 ± 12.4	185	-44.2	-37.1
	Ford Transit 2009 ^d	15	24.4 ± 18.4	199	22.3 ± 17.8	190	-8.6	-4.6
	Ford Puma 2020 ^h	4	52.2 ± 44.7	220	14.5 ± 6.8	38	-72.2	-82.5
	Ford Fiesta 2010 ^g	8	22.0 ± 23.7	251	14.7 ± 6.8	147	-33.2	-41.4
	Nissan Leaf 2014 ^e	12	25.2 ± 22.2	166	22 ± 14.2	161	-12.6	-3.3
	Nissan Qashqai 2010 ^d	-	-	-	-	-	-	-
	Peugeot 1082016 ^g	9	34.1 ± 31.8	251	25.3 ± 20.1	168	-25.8	-32.9
	Hyundai i30 2020 ^h	6	50.8 ± 48.3	471	34.6 ± 15.7	116	-68.1	-75.3
	Overall		34.8 ± 31.0	262	22.9 ± 12.4	163	-34.2	-37.6
	3 month old activated carbon	Skoda Fabia 2010 ^g	3	54.7 ± 48.7	240	10.8 ± 4.9	55	-80.0
Hyundai i30 2020 ^h		3	46 ± 10.2	160	3.8 ± 2.2	26	-91.7	-84.0
Ford Puma 2020 ^h		3	52.9 ± 40.9	213	7.1 ± 5.6	51	-86.6	-78.6
Overall			51.2 ± 33.3	204	7.3 ± 4.2	44	-86.1	-79.8

air (50 % relative humidity) as a carrier gas through the cabin air filters at 23 °C. Test gases are often toluene or nitrogen dioxide, while the testing concentrations given in the standards vary between 30 ppmV to 80 ppmV. Then the capacity of each filter type for single noxious gases can be calculated by mass balance using breakthrough curves. Sager (2012) tested this procedure with 4 ppmV of NO₂ in several artificially aged activated carbon filters and found that NO₂ adsorption dropped by 12 % after 90 min. The author also noted that cabin filters used in cars exhibit very different adsorption behaviour dependent upon their loading history and the environment in which the car was driven, and highlighted that the artificial aging of a filter with diesel exhaust nearly depleted its adsorbing capacity. Given that filter manufacturers advertise that their filters equally capture all the gases, a relevant experimental study, Heo et al. (2019), in Korea, tested 15 commercially available activated carbon cabin filters for their gas removal performance for toluene and n-butane. The study found that only one filter out of the 15 satisfied the local Korean standards. The authors highlight that given the wide range performance of the filters, there is an urgent need to establish legally enforced certification parameters, and recommend that more attention should be given to the selection of commercially available car cabin filters. It should be noted that, as far as the authors are aware, currently in the UK and in EU there are no such standards.

The work reported here has some important limitations. The tested cars may not be representative of the entire vehicle fleet as only specific manufacturers and models were tested. Additionally, we only tested the performance of the cabin filters from one manufacturer, and different cabin filter brands might have different performance not only in reducing in-vehicle NO₂ exposure but also the lifetime of the filter might be different. Since measurements were only conducted in Birmingham UK, the NO₂ exposure inside the car cabins may not be representative of exposures in other locations (with different on-road pollution levels). The results presented here should be interpreted with these limitations in mind.

5. Implications for vehicle occupant NO₂ exposure

The intervention of changing the vehicle cabin air filter type from pollen to activated carbon significantly reduced NO₂ concentrations by 87.4 % relative to on-road levels on average in the car interior under mechanical ventilation settings. However, the performance of the activated carbon filter in reducing NO₂ dropped significantly with time, by 6.8 % per month on average, showing change in decay rate after the first 6 months of use. The majority of the cabin air filter manufacturers recommend filter change for air cabin filters annually or for every 15 K Km; however, our results provide evidence that suggests that earlier replacement may be appropriate. It should be noted that the degradation rate of the filter is impacted by its cumulative exposure to pollutants, based on the amount of driving and pollutant levels in the atmosphere. In Europe, a majority of the population use cars as their main means of transportation on a daily basis (DfT, 2019; Fiorello et al., 2016), therefore, utilizing appropriate cabin filters could significantly reduce NO₂ exposures and hence the risk of related adverse health effects. Installing vehicle activated carbon cabin air filters represents a simple, inexpensive and effective method to reduce in-car exposure to NO₂, relative to on-road ambient levels. Their use should be considered to reduce NO₂ exposure, particularly for population groups who spend significant periods in vehicles, which may include professional drivers. The efficiency of such filters for NO₂ removal decreases with time, indicating that frequent replacement is necessary to maximise the benefit.

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CRediT authorship contribution statement

Vasileios N. Matthaios: Conceptualization, Methodology, Formal analysis, Writing - Original Draft, Funding acquisition. Daniel Rooney: Experimental, Methodology. Roy M. Harrison: Validation, Writing- Reviewing

and Editing. Petros Koutrakis: Validation, Writing - Review & Editing. William J. Bloss: Writing - Review & Editing, Funding acquisition.

Data availability

The dataset from the in-vehicle and on-road NO₂ measurements is available at <https://doi.org/10.5281/zenodo.7388363>.

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.

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