

Diesel passenger vehicle shares influenced COVID-19 changes in urban nitrogen dioxide pollution

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Abstract. Diesel-powered vehicles emit several times more nitrogen oxides than comparable gasoline-powered vehicles, leading to ambient nitrogen dioxide (NO₂) pollution and adverse health impacts. The COVID-19 pandemic and ensuing changes in emissions provide a natural experiment to test whether NO₂ reductions have been starker in regions of Europe with larger diesel passenger vehicle shares. Here we use a semi-empirical approach that combines *in-situ* NO₂ observations from urban areas and an atmospheric composition model within a machine learning algorithm to estimate business-as-usual NO₂ during the first wave of the COVID-19 pandemic in 2020. These estimates account for the moderating influences of meteorology, chemistry, and traffic. Comparing the observed NO₂ concentrations against business-as-usual estimates indicates that diesel passenger vehicle shares played a major role in the magnitude of NO₂ reductions. European cities with the five largest shares of diesel passenger vehicles experienced NO₂ reductions ~ 2.5 times larger than cities with the five smallest diesel shares. Extending our methods to a cohort of non-European cities reveals that NO₂ reductions in these cities were generally smaller than reductions in European cities, which was expected given their small diesel shares. We identify potential factors such as the deterioration of engine controls associated with older diesel vehicles to explain spread in the relationship between cities' shares of diesel vehicles and changes in NO₂ during the pandemic. Our results provide a glimpse of potential NO₂ reductions that could accompany future deliberate efforts to phase out or remove passenger vehicles from cities.

40 *Keywords:* Urban air quality, machine learning, environmental modeling, atmospheric
41 chemistry, nitrogen dioxide, COVID-19, diesel
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43 1. Introduction

44 Ambient nitrogen dioxide (NO₂) pollution is a global concern for public health,
45 particularly in urban areas, and is linked with decreased lung function, cardiopulmonary
46 and respiratory disease, and pediatric asthma, among other adverse health effects
47 [1, 2, 3, 4]. Traffic emissions are often the dominant source of urban NO₂, followed
48 by emissions from industrial sources and energy production and usage [5]. As such,
49 NO₂ is an effective surrogate for the broad traffic-related mix of pollutants.

50 Reductions in urban NO₂ during the pandemic (hereafter “ Δ NO₂”) varied greatly
51 across the world [e.g., 6, 7, 8, 9]. Direct comparisons of Δ NO₂ among cities are
52 inherently complicated by different meteorological patterns [10], stay-at-home measures,
53 and levels of adherence to these measures in each city. However, even after accounting or
54 normalizing for these important moderating factors, differences in Δ NO₂ likely remain.
55 With all else equal, one cause of these differences is vehicle fuel type. Reductions in
56 NO₂ have purportedly been larger in regions dominated by diesel vehicles [11]. While
57 a large body of literature has documented NO₂ changes during the pandemic, a smaller
58 portion has explored reasons for intercity differences in NO₂ changes. None, to the best
59 of our knowledge, has specifically examined the role of different vehicle fuel types in
60 causing these intercity differences.

61 Diesel-powered passenger vehicles emit substantially greater emissions of nitrogen
62 oxides (NO_x \equiv NO + NO₂) than comparable petrol- (or gasoline-)powered vehicles [12].
63 For example, real-world measurements indicate that Euro 6 diesel vehicles emit ten
64 times more NO_x than Euro 6 gasoline vehicles [13]. Since the late 1990s, European
65 nations experienced a “diesel boom,” where diesel passenger vehicles were intentionally
66 promoted as an alternative to petrol-powered passenger vehicles on the premise they
67 emit less CO₂ [14]. However, diesel and petrol vehicles have both produced similar
68 real-world CO₂ emissions since the early 2000s [15]. The proportion of diesel-powered
69 passenger vehicles to the total number of passenger vehicles (henceforth “diesel shares”)
70 steadily increased until the Volkswagen emissions scandal was brought to light in 2015.
71 Since then, diesel shares of new car registrations have declined in Europe [16]. Diesel
72 NO_x, including emissions in excess of certification limits, has contributed to high NO₂
73 pollution in Europe [e.g., 17, 18, 19, 20, 21] and adverse health impacts [e.g., 22, 14, 23].
74 In several countries outside of Europe such as the United States, Canada, and China,
75 diesel shares are much smaller, and petrol is the primary fuel consumed by passenger
76 vehicles [e.g., 24].

77 In this study, we examine how the COVID-19 pandemic can reveal the fingerprint
78 of diesel passenger vehicles on NO₂ pollution in urban areas. The pandemic, which

79 largely affected the transportation sector due to stay-at-home measures, provides an
 80 unprecedented natural experiment that allows us to tease out the relationship between
 81 urban vehicle fleets and ΔNO_2 . Additionally, we discuss ways that additional air quality,
 82 emissions, and traffic data would strengthen future efforts to study clean transit and air
 83 quality.

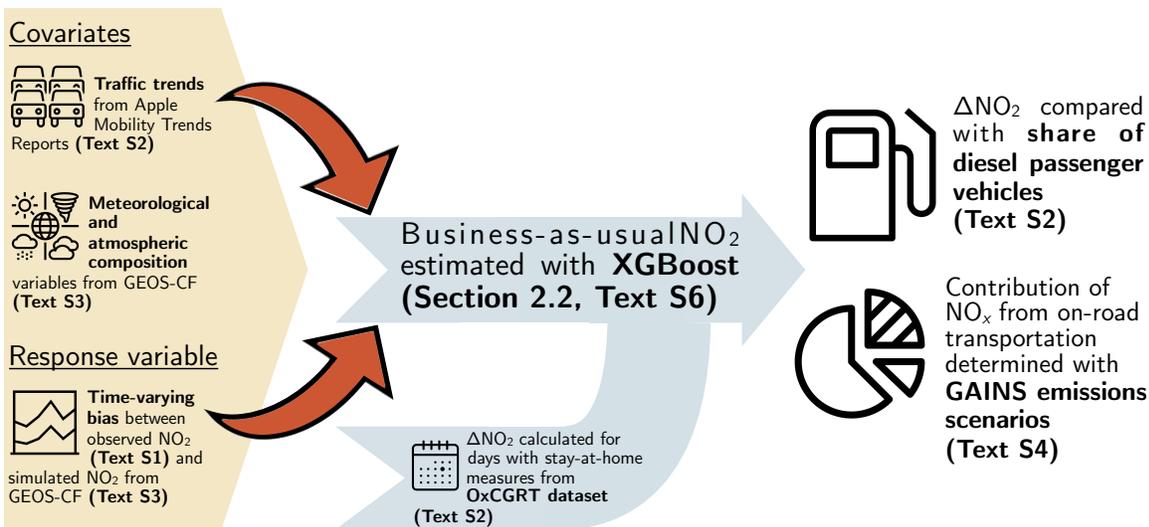


Figure 1. Process diagram showing the materials and methods used to quantify influence of diesel passenger vehicle shares on changes in NO_2 during COVID-19. GEOS-CF = GEOS Composition Forecast Modeling System; XGBoost = eXtreme Gradient Boosting; OxCGRT = Oxford Covid-19 Government Response Tracker; GAINS = Greenhouse gas-Air pollution Interactions and Synergies.

84 2. Materials and methods

85 2.1. Materials

86 We select 22 focus cities spanning 17 European countries based on the availability of
 87 *in-situ* NO_2 monitors (Text S1, Figure S1), city- or country-level traffic trends during
 88 the pandemic (Text S2, Figure S2), and country-level diesel shares (Text S2, Table
 89 S1). Publicly-available data on diesel shares at a subnational level do not exist to our
 90 knowledge, so we choose only 1-2 cities per country in our analysis (Text S1). Traffic data
 91 come from Apple Mobility Trends Reports [25] and represent traffic volumes relative
 92 to baseline volumes. This dataset began in 2020, and we form synthetic traffic data
 93 for 2019 using day-of-the-week proxies (Text S2, S5). As discussed in Section 1, diesel
 94 shares represent the proportion of diesel-powered passenger vehicles to the total number
 95 of passenger vehicles.

96 The NASA GEOS Composition Forecast Modeling System [GEOS-CF; 26] provides
 97 hourly, high-fidelity estimates of meteorology and atmospheric composition at $0.25^\circ \times$
 98 0.25° (~ 25 km) horizontal resolution globally (Text S3). The model's emissions

inventories do not account for the impact of COVID-19 on anthropogenic emissions, thus representing a counterfactual, business-as-usual scenario for the COVID-19 period [7]. We sample the surface-level (lowest model level) meteorological fields and pollutant concentrations from GEOS-CF at grid cells colocated with each *in-situ* NO₂ monitor. Both observed and modeled NO₂ concentrations are obtained for 1 January 2019 to 30 June 2020.

We also leverage emissions scenarios from the Greenhouse gas-Air pollution INteractions and Synergies (GAINS) model to explore how the contribution of light-duty vehicles to total anthropogenic NO_x emissions varies across cities (Text S4). Figure 1 illustrates how these data sources are combined within our methodological framework.

2.2. Methods

To isolate the influence of emissions changes on NO₂ reductions during COVID-19 for each city, we develop bias-corrected, business-as-usual NO₂ concentrations from GEOS-CF and compare them to observed concentrations. We then aggregate NO₂ observations and colocated GEOS-CF output to city-averaged daily mean values (Text S5).

We first bias correct NO₂ concentrations simulated with GEOS-CF using eXtreme Gradient Boosting (XGBoost) (Text S6). Briefly, XGBoost corrects the bias in GEOS-CF NO₂ against observed NO₂ as a time-varying function of air pollutants, meteorology, and traffic (Table S2). We build and test this XGBoost algorithm during our 2019 training period, with substantially improved model-observation agreement (Figure S3). We then apply the XGBoost bias correction algorithm to modeled NO₂ concentrations in 2020 to estimate business-as-usual NO₂ from 1 January to 30 June 2020. This approach accounts for differences in local meteorology, atmospheric composition, and traffic between 2019 and 2020, as these factors influenced NO₂ concentration independently of fuel type [27]. This approach builds on previous work to estimate business-as-usual pollutant concentrations during the pandemic [28, 29, 30, 31, 7, 32].

We characterize ΔNO_2 as

$$\frac{\text{NO}_{2, \text{observed}} - \text{NO}_{2, \text{business-as-usual}}}{\text{NO}_{2, \text{business-as-usual}}} \times 100\%. \quad (1)$$

When calculating ΔNO_2 in a particular focus city, we average over all dates where stay-at-home measures (Text S2, Figures 1 and S2) are either recommended or required through 30 June 2020 and refer to this period as “lockdown”.

3. Results

GEOS-CF captures daily NO₂ variability in our focus cities (Figure S7), reinforcing its ability to aid in understanding lockdown-related NO₂ changes. We highlight London to further illustrate GEOS-CF’s capabilities and our methods (Figure 2a). The temporal correlation (r) between modeled and observed NO₂ in 2019 for London is 0.78 ($r = 0.60$ averaged over all cities; Figure S3b). Despite the good correlation, there

136 is a negative model bias relative to observations in many of our focus cities (mean
 137 fractional bias = -0.60 averaged over all cities; Figure S3a). GEOS-CF’s negative bias
 138 is well-documented, especially in Europe and North America where there are publicly
 139 available observations [26]. This bias may stem from model resolution; uncertainties in
 140 atmospheric transport, boundary layer height, vertical mixing, emissions, and chemistry;
 141 and monitor interference with other nitrogen-containing compounds [33, 34, 7].

142 Correcting the bias in modeled NO_2 with XGBoost leads to substantially better
 143 agreement against observations than the native GEOS-CF concentrations, and the
 144 aforementioned negative model bias is greatly reduced. Figure 2a illustrates the excellent
 145 agreement between business-as-usual and observed NO_2 in 2019 prior to the lockdown.
 146 In this example for London, the mean fractional bias in 2019 is reduced from -0.41 with
 147 the native GEOS-CF concentrations to -0.02 with the bias-corrected concentrations,
 148 and we note similar improvements in other focus cities (Figure S7).

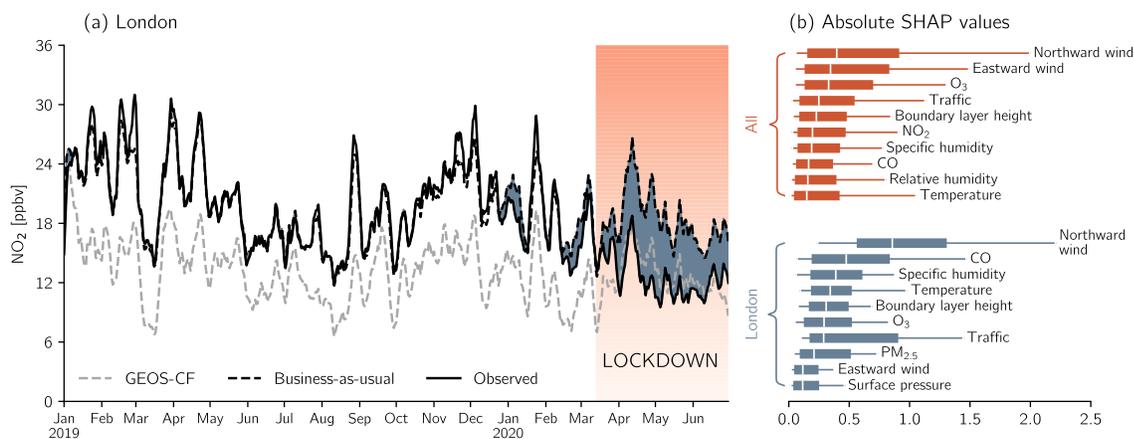


Figure 2. Illustration of XGBoost-inferred business-as-usual concentrations and drivers of these predictions. (a) Observed, GEOS-CF, and business-as-usual NO_2 concentrations in London. Time series represent the daily average of all *in-situ* monitors or their colocated model grid cells in London. The shaded red band denotes the 2020 lockdowns in the United Kingdom, and blue shading corresponds to days where observed NO_2 is less than business-as-usual NO_2 to highlight the COVID-19 lockdowns. (b) SHAP value distributions for the ten most important meteorology-, composition-, and traffic-related XGBoost input variables for all focus cities (top) and London (bottom) are ranked by their median value, here indicated by vertical white lines. XGBoost input variables are provided by GEOS-CF and Apple Mobility Trends Reports (Table S2). Boxes show the interquartile range, and whiskers extend to the 10th and 90th percentiles.

149 We characterize the relative contribution of input variables in generating the
 150 business-as-usual NO_2 concentrations with SHapley Additive exPlanations (SHAP)
 151 values (Figure 2b, Text S6). The absolute SHAP values illustrate the global importance
 152 of input variables, and a larger SHAP value for a particular variable means that
 153 that variable has a more important impact on the bias correction. Assessing feature
 154 importance via SHAP values indicates that local atmospheric transport and species

155 related to basic ozone (O_3) chemistry are the most important variables for inferring
 156 business-as-usual NO_2 concentrations for both London and all focus cities (Figure 2b).
 157 The partial dependence plots in Figure S6 show how XGBoost’s bias correction is
 158 affected by individual input variables. This analysis reveals a nonlinear relationship
 159 between the input variables and the bias correction, and the predicted bias is largest for
 160 meteorological, traffic, or chemical conditions at anomalously high or low extremes.

161 Traffic emerges as one of the most influential variables in estimating business-as-
 162 usual concentrations (Figure 2b). The relative contribution of traffic in London ranks
 163 lower than for the aggregation of SHAP values over all focus cities, but the distribution
 164 has right-skew with a wide range for large SHAP values (Figure 2b). This result indicates
 165 that intraweek traffic variations in London are one of the most important variables in
 166 correcting the bias and producing business-as-usual NO_2 concentrations for certain days
 167 in our measuring period and particular folds of the k-fold cross validation.

168 Observed NO_2 concentrations begin to diverge from business-as-usual concentra-
 169 tions in London around mid-February 2020, slightly preceding the United Kingdom’s
 170 declaration of recommended stay-at-home measures (compare Figures 2a and S2). When
 171 averaged over the lockdowns, ΔNO_2 between the observed and business-as-usual con-
 172 centrations is -28.5% in London. Observed NO_2 concentrations exhibit departures from
 173 business-as-usual concentrations in spring 2020 in other cities as well but with varying
 174 magnitudes (Figure S7). Contemporaneous studies have found NO_2 reductions of sim-
 175 ilar magnitudes in London and our other focus cities using complementary methods
 176 [35, 36, 37, 32].

177 Our focus cities span a spectrum of pre-lockdown NO_2 pollution levels and diesel
 178 shares ranging from 8.1% in Athens, Greece to 69.2% in Vilnius, Lithuania (Figure 3,
 179 Table S1). Mean 2019 NO_2 in all 22 focus cities exceeded the recently-revised World
 180 Health Organization annual mean NO_2 guideline value of $10 \mu g m^{-3}$ (~ 5.3 ppbv,
 181 assuming an ambient temperature of 298.15 K and pressure of 1013.25 hPa). Even
 182 Helsinki, which had the lowest 2019 NO_2 concentration (~ 8.4 ppbv) of all focus cities,
 183 exceeded this guideline value by 60% (colors in Figure 3).

184 The change in NO_2 during the pandemic (ΔNO_2 , Equation 1) averaged across
 185 cities is -23.8% (standard deviation = 16.0%), and the precise magnitude ranges by
 186 approximately 60% across cities. We next compare ΔNO_2 with cities’ diesel shares and
 187 see a clear pattern emerge: cities with larger diesel shares tend to have larger ΔNO_2 ,
 188 while ΔNO_2 is smaller in cities with smaller diesel shares ($r = -0.50, p = 0.02$; Figure
 189 3). For example, the average change in NO_2 ($\overline{\Delta NO_2}$) in cities with the top five largest
 190 diesel shares ($\overline{\Delta NO_2} = -38.1\%$) is ~ 2.5 times larger than the change in cities with the
 191 five smallest shares ($\overline{\Delta NO_2} = -15.0\%$). The slope of the linear regression fit between
 192 ΔNO_2 and diesel shares provides a succinct summary of our results (Figure 3). This
 193 slope indicates that the larger shares of diesel passenger vehicles have stronger impact
 194 on the ΔNO_2 during the pandemic; specifically, ΔNO_2 decreased by 5.3% for every 10%
 195 increase in diesel shares (Figure 3).

196 The intercept of the linear regression in Figure 3 suggests a very small change in

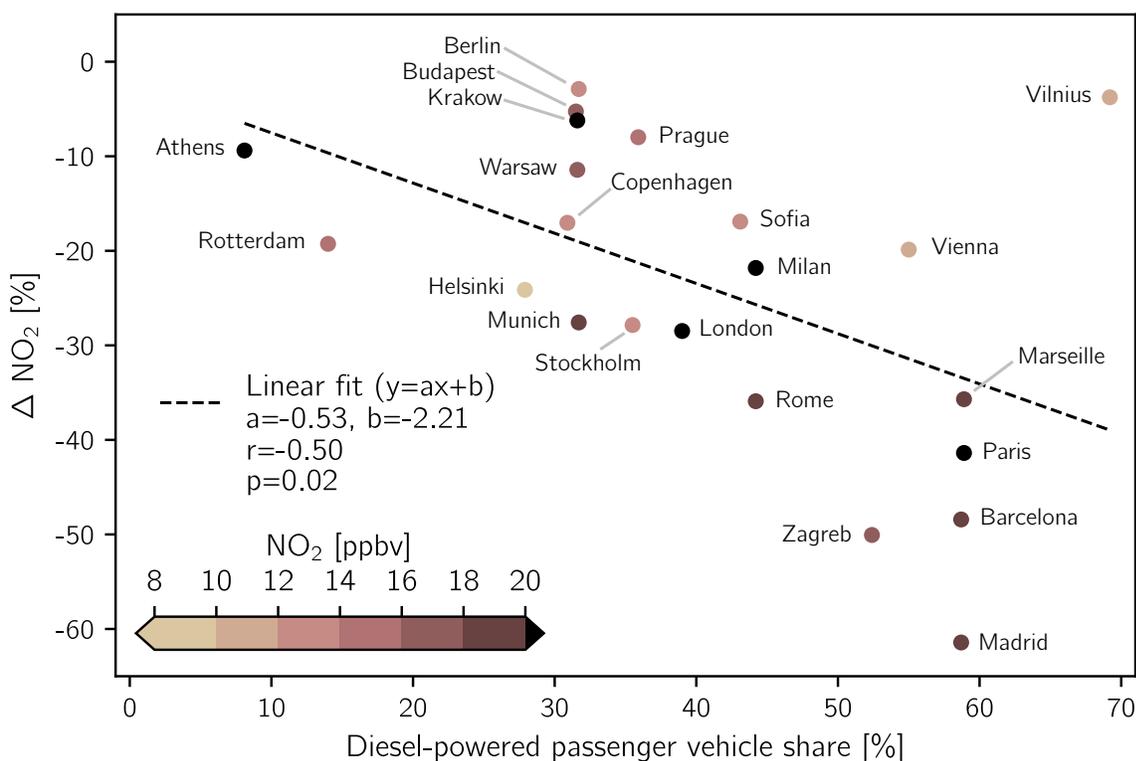


Figure 3. Association of passenger vehicle diesel share with changes in NO₂ (ΔNO_2) during the pandemic. Points are colored by annual mean NO₂ concentrations in 2019. Dashed line shows the linear regression of ΔNO_2 on diesel shares. Inset text indicates the slope, intercept, correlation coefficient and p -value of this regression.

197 NO₂ for cities whose shares of diesel passenger vehicles are close to 0%. Even cities
 198 with these small shares, such as those in North America with mostly gasoline-powered
 199 passenger vehicles, experienced substantial decreases in NO₂. For example, Goldberg
 200 and colleagues [10] found a median NO₂ decrease of $\sim 22\%$ in major North American
 201 cities during spring 2020 after adjusting for seasonality and meteorology. In all cities,
 202 other sources of urban NO_x beyond diesel passenger vehicles (e.g., heavy-duty vehicles,
 203 power plants, maritime activity, industry) not accounted for in our experimental design
 204 contributed to ΔNO_2 , regardless of the diesel passenger vehicle share.

205 We next describe sensitivity analyses that speak to the robustness of our results.
 206 Testing whether traffic volumes from Apple Mobility Trends Reports can capture
 207 weekday-weekend differences in traffic patterns affirms the ability of this dataset to
 208 serve as a proxy for the day of the week and XGBoost to capture these intraweek
 209 variations (Figure S8). The OxCGRT lockdown dates represent country-level dates for
 210 stay-at-home measures if at least some region of a given country has the restrictions
 211 [38]. Responsibility for COVID-related restrictions was often delegated to state or local
 212 governments; however, to the best of our knowledge, no globally consistent database with
 213 city-specific lockdown dates exists. Given uncertainties associated with these dates, we

214 recalculate ΔNO_2 for a uniform time period extending from 15 March 2020 to 15 June
215 2020 and find substantively similar results (compare Figures 3 and S9). We examine
216 the extent to which ΔNO_2 varied between recommended versus required stay-at-home
217 measures shown in Figure S2 and the impacts of restriction type on the diesel share-
218 ΔNO_2 relationship. Again, we observe no substantive changes (compare Figures 3 and
219 S10).

220 We test whether including a cohort of additional cities outside of Europe (Mexico
221 City, Los Angeles, Auckland, and Santiago; Text S1) from the C40 Cities network
222 leads to consistent conclusions regarding the relationship between diesel shares and
223 ΔNO_2 . C40 Cities is a network of the world’s megacities committed to addressing
224 climate change, and the four additional cities included in our study provided data to C40
225 (see Acknowledgements) after expressing interest in learning from lockdowns to design
226 post-COVID recovery measures that may further support air quality improvements and
227 reductions in NO_2 . These additional cities specifically allow us to test whether our
228 findings are generalizable to cities with different cultural and behavioral practices (e.g.,
229 reliance on public transit, adherence to COVID-19 containment measures) and lower
230 diesel shares compared to the European cohort focused on elsewhere in this study.

231 Given the small diesel shares in these cities (cohort-averaged share= 4.0%; Table
232 S1), we expect they would experience small to modest NO_2 reductions. This is indeed the
233 case, and the cohort-averaged ΔNO_2 of -14.8% is markedly smaller than the reduction
234 in many European cities with larger diesel shares (Figures S9-S10). This cohort of C40
235 Cities also demonstrates some of the challenges associated with inferring business-as-
236 usual NO_2 . For example, Los Angeles has one of the smallest diesel shares of all cities
237 examined (Table S1) but experienced markedly larger NO_2 reductions than other cities
238 with small diesel shares. NO_x emissions related to the Ports of Los Angeles and Long
239 Beach, one of the largest ports in North America, might inflate ΔNO_2 compared to
240 cities without ports or other large point sources of NO_x . The topic of unconsidered
241 moderating influences is further discussed in Section 4.

242 Despite the strong, statistically significant relationship between diesel shares and
243 ΔNO_2 (Figure 3), ΔNO_2 does not increase monotonically as the share of diesel passenger
244 vehicles grows. There are several cities with similar diesel shares, yet different ΔNO_2 ,
245 and we next explore key factors that could explain the spread among cities’ ΔNO_2 given
246 their diesel shares.

247 One factor to explain the spread in ΔNO_2 is vehicle age. NO_x emission rates are
248 not stable over diesel passenger vehicles’ lifetimes and increase linearly with age [39].
249 This increase may result in “effective diesel shares” that are larger than the ones used in
250 our study, especially for focus cities with older passenger vehicle fleets (Table S1). With
251 all else equal, we hypothesize that cities with older passenger vehicles would experience
252 larger ΔNO_2 than cities with newer vehicles.

253 For brevity, we discuss this role of vehicle age for a few cities: Vienna, Austria;
254 Paris, France; and Madrid, Spain. These cities have among the largest, yet very similar,
255 diesel shares of all focus cities in our study, but there is a spread of $\sim 40\%$ in ΔNO_2

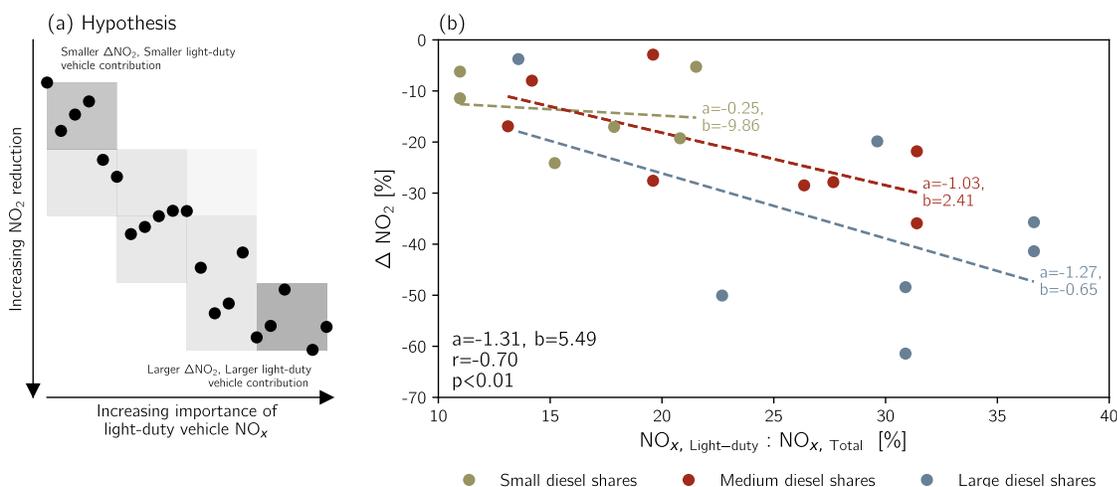


Figure 4. (a) The hypothesized association between the contribution of light-duty vehicle NO_x emissions to total NO_x emissions and NO₂ reductions during the pandemic illustrated with synthetic data. (b) ΔNO₂ versus light-duty vehicle to total NO_x emissions from GAINS (Text S4) where each scatter points represents a focus city. Points are colored by their diesel shares, which are discretized into tertiles (small ≤ 33rd percentile, medium = 33rd – 66th, large ≥ 66th). For each group of diesel shares, colored lines show the linear regression and colored text the slope (a) and intercept (b) of this regression. Inset text in the lower left denotes the slope, intercept, correlation coefficient and *p*-value for the regression of ΔNO₂ on the light-duty NO_x contribution using the full dataset.

256 among these cities. For the aforementioned three cities with large diesel shares, our
 257 hypothesis regarding vehicle age is valid: passenger vehicles in France and Spain are 1.9
 258 and 4.4 years older on average, respectively, than those in Austria (Table S1). Vehicle
 259 age provides a plausible, evidence-based hypothesis to explain some of the intercity
 260 spread in our results, although we note it cannot explain all variability. The results of
 261 previous studies [e.g., 40, 39] imply that future policies to preferentially remove older
 262 diesel passenger vehicles from cities may have outsized impacts compared to removing
 263 newer diesel vehicles.

264 Another factor to explain variability in intercity ΔNO₂ is the contribution of light-
 265 duty vehicles to overall NO_x emissions. On average, road transportation contributes
 266 47% of total NO_x emissions in European cities but ranges from approximately 20% to
 267 70% depending on the city [5, 41]. We hypothesize that cities with similar diesel shares
 268 would likely have different ΔNO₂ if their light-duty vehicle sectors have different-sized
 269 contributions to total anthropogenic NO_x emissions (Figure 4a). To test this hypothesis,
 270 we leverage emissions scenarios from GAINS to find the contribution of NO_x emissions
 271 from light-duty vehicles to total NO_x emissions for each focus city (Text S4).

272 Unsurprisingly, diesel shares are correlated with the contribution of light-duty
 273 NO_x emissions to total NO_x emissions ($r = 0.57, p < 0.01$; not shown), meaning
 274 that cities with a larger share of diesel passenger vehicles tend to have a larger
 275 proportion of NO_x emissions from light-duty vehicles. ΔNO₂ also increases as the overall

276 contribution of light-duty NO_x emissions to total NO_x emissions grows in all focus cities
 277 ($r = -0.70, p < 0.01$; Figure 4b).

278 Since our original hypothesis posits that cities with *similar* diesel shares might
 279 have different ΔNO_2 if their light-duty vehicle sector contributes differently to total
 280 NO_x emissions, we partition cities into groups with similar diesel shares and investigate
 281 how ΔNO_2 varies within these groups. We find that ΔNO_2 increases as the light-duty
 282 NO_x emissions contribution increases among cities with similar diesel shares (Figure
 283 4b). For example, cities with “medium diesel shares” (Figure 4b) have diesel shares
 284 that range from 31.7% to 44.2%. Among these cities, cities where light-duty vehicles
 285 contribute a larger proportion to total NO_x indeed experienced larger ΔNO_2 during the
 286 pandemic, thus affirming our original hypothesis.

287 The analysis in Figure 4 can also shed light on cities with outlying ΔNO_2 values
 288 in Figure 3. In Vilnius, GAINS indicates that NO_x emissions from light-duty vehicles
 289 only constitute 13.6% of total NO_x emissions, one of the smallest contributions of all
 290 our focus cities (Figure 4b). It follows that a small ΔNO_2 might be expected in Vilnius
 291 even given the large diesel share. For simplicity, we have chosen tertiles to group similar
 292 diesel shares, but we have also tested a larger number of groups (e.g., quartiles, quintiles)
 293 and found similar results.

294 4. Discussion

295 Major strengths of our analysis include our semi-empirical approach that leverages
 296 air quality data from monitoring networks as well as our use of a machine learning
 297 algorithm, XGBoost, to establish the relationship between NO_2 and local meteorology,
 298 atmospheric composition, and traffic trends. By combining XGBoost with GEOS-
 299 CF to infer business-as-usual NO_2 during the COVID-19 pandemic, we have further
 300 demonstrated how this methodology can be used for emergent research questions for
 301 which relying on observations or atmospheric models alone would be challenged by
 302 moderating influences, incomplete spatial coverage, and inaccuracies.

303 Several factors and limitations of our data and methods may impact our results.
 304 GEOS-CF’s use of 2010 anthropogenic emissions for all following years may under- or
 305 overestimate NO_2 , especially in areas undergoing rapid changes in emissions. More up-
 306 to-date emissions are under development and slated to be included in future versions
 307 of GEOS-CF [26]. Our framework does not consider intercity differences in the type of
 308 passenger vehicles (i.e., gasoline versus diesel) that remained parked and off the road
 309 during the pandemic due to lack of data. The use of national-level diesel shares (Text
 310 S2) and national-level light-duty vehicle and total NO_x emissions (Text S4, Figure 4) is
 311 a simplification when examining individual cities but an important first step to estimate
 312 how the passenger vehicle traffic fleet contributes to urban NO_2 . There have been efforts
 313 to provide gridded (not national-level) inventories for specific types of vehicles and
 314 vehicle fuels for regions outside the European Union [e.g. 42, 43, 44]. Future research on
 315 urban transportation and air quality will benefit from the inclusion of these inventories.

316 While our study incorporated changes in traffic into our machine learning approach,
317 the pandemic impacted many forms of urban activity besides on-road traffic. NO_x
318 emissions from the aviation, rail, and maritime sectors plummeted during COVID-19
319 [e.g. 45]. We have not accounted for trends in these activities within XGBoost as we
320 are challenged by a lack of city-specific time series data. While these other activities
321 can be important contributors to urban NO_x emissions, we find a strong relationship
322 of passenger vehicle fuel type on ΔNO_2 , meaning that the impact of fuel type on NO_2
323 is strong enough to observe through our methodological approach even despite these
324 other sectors. Moreover, recent studies point to on-road traffic, particularly passenger
325 vehicles, as the primary driver of NO_2 reductions during the pandemic [46, 8]. An
326 analysis of ΔNO_2 against changes in traffic from the Apple Mobility Trends Reports
327 in our 22 focus cities reveals a positive, albeit weak, relationship between ΔNO_2 and
328 changes in traffic (Figure S11). Comparing traffic data from Apple’s dataset against
329 *in-situ* traffic counts and the impact of traffic dataset choice on ΔNO_2 further justifies
330 our use of the Apple’s dataset in our study (Text S7, Figure S12).

331 We investigated whether the location of *in-situ* NO_2 monitors or the stringency
332 of mobility restrictions are correlated with diesel shares such that they would bias the
333 observed association between diesel shares and ΔNO_2 in Figure 3 towards or away from
334 the null. We did not detect a statistically significant relationship between diesel shares
335 and these factors (Figure S13), indicating they are not a major contributor to the diesel
336 share- ΔNO_2 relationship.

337 The number and distribution of *in-situ* monitors vary from city to city (Figure
338 S1), and monitors may be sited in different environments (e.g., traffic, industrial,
339 background). Monitor siting could impact our results if monitors are disproportionately
340 sited in neighborhoods where ΔNO_2 substantially differed from the true city-averaged
341 value. For example, Berlin, Germany stands out given the small ΔNO_2 during the
342 pandemic (Figure 3). Less than half of Berlin’s monitors are located near traffic
343 (Figure S13b), and a recent study showed the statistical significance of pandemic-
344 related NO_2 reductions varied across different environments for NO_2 monitors [47]. We
345 explored whether ΔNO_2 within individual cities varied across traffic and non-traffic NO_2
346 monitors, expecting to find a larger decrease at traffic sites. Although we did not find
347 a significant difference for ΔNO_2 calculated with traffic versus non-traffic monitors, the
348 magnitude of the diesel share- ΔNO_2 relationship was nearly double when ΔNO_2 was
349 estimated using only traffic monitors (9.7% decrease for every 10% increase in diesel
350 shares using traffic NO_2 monitors compared to the 5.3% decrease in Figure 3 using all
351 monitors), and $\overline{\Delta\text{NO}_2}$ for different monitors types was suggestive of a difference (Figure
352 S14). While non-uniform changes in NO_2 *within* cities are interesting and have been the
353 subject of other studies [e.g., 48], the primary goal of our study is to reconcile differences
354 *among* cities’ ΔNO_2 in light of their different diesel shares.

5. Conclusion

Our study demonstrates that diesel shares played a major role in the magnitude of ΔNO_2 experienced by cities during the COVID-19 natural experiment. The magnitude of ΔNO_2 varies from approximately -3% to -61% across cities, and ΔNO_2 is a factor of ~ 2.5 times larger in European focus cities with the top five diesel shares compared to cities in the bottom five. The relationship between diesel shares and COVID-related NO_2 reductions deduced from a sensitivity analysis that considers C40 member cities outside of Europe is in reasonable agreement with our results from Europe and suggests the generalizability of our findings.

By leveraging this unique natural experiment, we are able to observe the relationship between NO_2 and diesel shares. Previous observational and modeling studies have documented the impact of diesel fuel on pollution and health, and our study is the first to investigate the impact of diesel fuel on NO_2 pollution during this natural experiment. The relationship between ΔNO_2 and diesel shares gives an indication of the changes in NO_2 that could be expected if cities decrease their diesel shares through policy, economic forces (e.g., increased affordability of electric passenger vehicles), or social forces (e.g., diesel passenger vehicles viewed unfavorably as a result of “Dieselgate”). Our results will also aid in understanding why ΔNO_2 varied among urban areas given their different diesel shares.

Our key findings are relevant for present-day and future policies. The temporary NO_2 reductions during the COVID-19 pandemic could be sustained through long-term policies to reduce the number of passenger vehicles in urban areas through, for example, policies such as congestion pricing or those that promote active transportation (e.g., cycling, walking). Should these policies be implemented, our results suggest that cities with larger diesel shares would experience larger NO_2 reductions. Beyond decreasing NO_2 and the associated public health damages, these types of policies would also slow climate change, decrease concentrations of other harmful pollutants such as particulate matter and O_3 , and encourage healthier lifestyles if active forms of transportation replace passenger vehicles [e.g., 49]. Focus cities such as Paris and Berlin are poised to ban most or all diesel passenger vehicles in the near future [50]. We expect that our results will reinforce these efforts in Paris and Berlin and could catalyze other cities to implement similar policies.

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