Concentrations and Allocation of NO$_2$ Emissions to Different Sources in a Distinctive Italian Region after the COVID-19 Lockdown

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Abstract

Nitrogen dioxide concentrations, being short-lived pollutants, are good indicators of changes in emission sources and economic slowdowns. This analysis focuses on the Greater Salento region (Italy) and aims to monitor, by investigating the relative sources, the changes of NO$_2$ tropospheric concentrations notoriously related to vehicular traffic exhausts and, in general, to fossil fuel combustion processes which are now apparently linked to many COVID-19 patients deaths. The principle objective of this paper is to map the tropospheric NO$_2$ local distribution and to extrapolate, from the overall data of average daily concentrations of NO$_2$ as recorded by the ARPA-Puglia ground-based monitoring stations, the single contributions and their mutual relationships of the different diffuse emission sources (motor vehicles and domestic heating systems) by identifying, the environmental background threshold of this pollutant of each geographic area, thanks to the simplified situation determined by the COVID-19 lockdown. The analyzed territory (the so-called “Greater Salento” or Salento Peninsula) is very unusual because there are two provinces with large industrial settlements, Taranto, with the steel area of ex-ILVA, and Brindisi, with petrochemical and thermoelectric power plants, which enclose a territory, the province of Lecce, free of any industrial plants of such sizes and their environmental impacts. From the results of this study, in addition to confirming the obvious and overall decrease of NO$_2$ concentrations (~23.2% compared to previous year) during the lockdown period, interesting and distinctive local allocations of nitrogen dioxide concentrations to different sources have also emerged: heating household systems, and not road traffic, are the main sources of this dangerous pollutant in this region, with an average quota of 44.3%. The studied regional situation is so significant as to allow broader considerations regarding to other similar international areas.
1. Introduction

Investigating, with accuracy, the contribution of the different and numerous sources that determine air pollution is not an easy matter because there are many anthropogenic and natural factors, meteorological dynamics and chemical mechanisms [1]. This is much more difficult when the analyzed territorial area is restricted. However, the blocking of non-essential production activities and the corresponding provisions of self-quarantine and the population’s mobility restriction and transport shutdown—to which, though in different periods, all those countries affected by the COVID-19 global pandemic [2] have adopted in order to slow down and mitigate their SARS-CoV-2 infection [3]—have provided the exceptional opportunity for scientists to study more deeply, and better understand, the complex underlying problems of air pollution and, in particular, of NO2 tropospheric concentrations on a long-term basis.

In all those countries where strict containment measures on the population’s mobility, transport and production activities were adopted, there has been a clear improvement in the tropospheric air quality parameters, including a strong decrease of the dangerous concentrations of nitrogen dioxides [4] [5] [6] [7]. Nitrogen dioxide concentrations, being short-lived pollutants, are good indicators of changes in emission sources and economic slowdowns.

As is known, nitrogen dioxide is a very reactive gas (energetic oxidant and highly corrosive), mostly secondary (as it is the product of NO oxidation in the atmosphere), generated by high temperature combustion processes. It contributes to the formation of photochemical smog as a precursor of other pollutants (ozone, secondary fine particulate matter, nitric acid and nitrous acid); some of which are deposited on the ground, causing eutrophication and acid rain phenomena. Furthermore, it is dangerous to health because it irritates the mucous membrane and is responsible for some diseases of the respiratory system thus causing an increase of early mortality rate [8] [9] [10] [11]. Moreover, recent studies indicate a strong fatal incidence associated with lung ACE-2 overexpression: in fact, in some areas, many COVID-19 patients’ deaths could be linked to a pre-exposure to air pollution, especially NO2 [12] [13] [14].

The NO2 assessment parameters for the protection of health, that is, the daily limit standards, established by the World Health Organization (WHO) Air Quality Guidelines [15], which was implemented by the Italian Legislative Decree nr. 155/2010, are 200 µg/m³ on an average hourly concentration (1-hour mean), and are not to be exceeded, more than 18 times in a year, the threshold hourly alarm (measured over three consecutive hours) of 400 µg/m³ and the an-
nual limit value of 40 µg/m³.

Road traffic exhausts and domestic heating, especially in urban areas, are the greatest direct anthropogenic sources of NO₂. Other causes of emission are industrial fossil fuels combustion processes (e.g. production of nitrogen fertilizers, of nitric acid and ammonia, power generation, steel plants, waste incineration, etc.). For these reasons, the tropospheric concentrations of NO₂ can be correlated, in large part, also with the volume of economic activities of a specific territory [1]. There are, of course, also natural emission sources (lightning, volcanic eruptions, fires, soil processes, etc.) of this pollutant, but they are a minority compared to anthropogenic sources.

The main objectives of this paper are to map the tropospheric NO₂ local distribution and to extrapolate—in a period in which the contribution to emissions from vehicular traffic was greatly reduced from the overall tropospheric concentration of nitrogen dioxide data in the four territories (ex-ILVA, Taranto, Brindisi and Lecce) of Greater Salento, due to the lockdown measures—the contributions of the single emission sources (motor vehicles and domestic heating systems) and their mutual relationships, identifying the background NO₂ pollution for each of these geographical areas. In order to simplify the analysis, the role of meteorological phenomena will not be quantified in this study: weekly means, however, eliminate many effects of short-term weather changes.

2. COVID-19 Italian Regulatory Chronology and a Brief Description of the Analyzed Territories

This is a brief history of the main legislative provisions that characterized the period of the diffusion of the new coronavirus in Italy.

With the Decree of the President of the Council of Ministers (DPCM) of 9th March 2020, the measures of the previous DPCM of 8th March 2020 were extended throughout the Country in order to counter and contain the spread of the COVID-19 virus, and the local restrictive measures adopted with the DPCM of the day before; that is, the prohibition of any movement of persons except those movements justified by necessity, urgency or health reasons, were extended to the entire national territory. From midnight of 9th March, Italy became a “protected area” and the measure is known as the “#iorestoacasa” Decree. On 11th March, the WHO declared the SARS-CoV-2 a pandemic [2]. The above Decree, with subsequent extensions, remained in force until 3rd May 2020, that is until the DPCM of 26th April 2020 was issued stating that, starting from the following 4th May, the end of the lockdown and the resumption of citizens’ movements within their own regions. “Phase 2” began on 18th May. On 3rd June 2020 saw the beginning of “Phase 3”, permitting people to move between Italian regions. So, from 10th March to 3rd May 2020, vehicular traffic throughout the national territory was drastically reduced.

The analyzed territory (the so-called “Greater Salento”) is very unusual because there are two provinces with large industrial settlements (Taranto, with
the steel area of ex-ILVA, and Brindisi, with petrochemical and thermoelectric power plants) that enclose a territory, the province of Lecce, without industrial settlements of these sizes and substantially without high environmental impacts. The NO2 concentrations of this last territory, together with those of the same period of the previous year, and of the periods of 2020 immediately before and after the lockdown, will serve as terms of comparison to try to estimate the contribution of the nitrogen dioxide background pollution (that is, what remains after removing the contribution of vehicular traffic and domestic heating systems) to NO2 concentrations.

The Greater Salento region covers an area of 7127 km2 with a population, at 31.12.2018, equal to 1,764,865 inhabitants [16] and a population density of 247.6 inhabitants per km2.

The province of Taranto covers an area of 2467 km2, with 576,756 inhabitants [16] and a density of 233.8 inhab./km2. According to ANFIA [17], at 31.12.2018, the passenger cars in use in the province of Taranto were 338,163 of which 109,641 were in the province’s capital, and the number of inhabitants per car, respectively, was equal to 1.71 and 1.79. In addition to passenger cars, there were also 32,859 commercial vehicles in the province. This is the Apulian province with the lowest density of active businesses: 49.9 per 1000 inhabitants. 90 companies are present within the industrial area of Taranto, which covers an area of 2.203 hectares [18].

The ex-ILVA steel plant, located in the province of Taranto, and now owned by ArcelorMittal Italia Ltd., is the European main hub of steel manufacturing. The name of “ILVA” derives from the Latin name of the Island of Elba where, since the time of the Etruscans, iron ore was mined. It extends for 1500 hectares, equal to twice the size of the city of Taranto. There are also approximately 15 medium-large sized production plants which produce hydrocarbon and hydrogen.

The province of Brindisi extends over a territorial surface of 1861 km2 has 392,975 inhabitants [16] and with a density of 211.2 inhab./km2. At 31.12.2018, the number of passenger cars in use, in the whole province, were 251.501. In the municipal capital alone they were 52.725. The number of inhabitants per car was equal to 1.56 and 1.65 respectively [17]. In addition to passenger cars, 31,112 commercial vehicles also circulate in this province.

Within the industrial area of Brindisi, which covers a surface of 2700 hectares, there are 239 companies, some of which are of medium-large size and whose production specializations are aeronautics (e.g. GE Avio, Dema, Salver, GSE), chemical (e.g. Lyondell Basell, EniVersalis (ex Polimeri Europa), Chemgas), pharmaceutical (Sanofi Aventis), energy (Enel and Enipower) and metallurgy (e.g. Alfer, LeucciCostruzioni) [18].

In addition to these companies, there are the thermoelectric power plants of North Brindisi and the large “Federico II” coal power plant in Cerano, which is situated less than 15 km away from the city center of Brindisi.

The province of Lecce covers an area of 2799 km2, with 795,134 inhabitants...
and a density of 284.1 inhab./km$^2$. In the province of Lecce, there were 511,480 passenger cars in use as of 31.12.2018, of which 65,961 in the capital, and with a number of inhabitants per car equal to 1.55 and 1.44 respectively [17]. In addition to passenger cars, 65,085 commercial vehicles also circulate in the province.

Lecce, with 67 businesses per 1000 inhabitants, is the Apulian province with the highest density of active micro-companies in the Greater Salento area [18]. Their businesses work in wide range of different production sectors. The main production specializations are wholesale and retail trade, electronic equipment, food products, freight transport, small bodywork, furniture manufacturing, mechanical and thermo-mechanical constructions, logistics, wholesale trade, LPG mixing, and production of transmission parts for motor vehicle. On an industrial level, the larger companies operate in the casting and molding of cast iron and zinc galvanization.

3. Materials and Method

We have chosen to analyze the eight weeks of lockdown period and the population’s mobility restrictions (that is, from Tuesday 10th March to Sunday 3rd May 2020), announced by the Italian Government with DPCM 9th March 2020, the six previous weeks (from Monday 27th January to Monday 9th March) and the subsequent last period (from Monday 4th May to Sunday 14th June). In order to do a comparison with the situation of the previous year, we have taken into consideration the NO$_2$ tropospheric concentrations of the corresponding three periods of 2019, although, they are probably subject to different fluctuations and meteorological conditions: precisely, the 8 weeks of the period from 11th March to 5th May, the six weeks from 28th January to 10th March and, lastly, the other six weeks from 6th May to 16th June 2019.

The sources of the NO$_2$ concentration values were the average daily raw data of the ARPA-Puglia [19] ground-based monitoring stations, taking into consideration both the data of those classified as “urban-traffic stations” and “urban background units”.

We have delimited the field of investigation to the so-called territory of Greater Salento, which includes the provinces of Taranto, Lecce and Brindisi. In detail, for the province of Taranto, there are a total of 11 NO$_2$ active monitoring stations: 6 in the municipal capital and 5 in the province (Grottaglie, Martina Franca, Massafra and two in Statte). Two units are located inside the ex-ILVA plant (Meteo Parchi and Tamburi areas). In the Lecce area, there are 8 ARPA units in operation: 3 in the city and 5 in the province (Campi Salentina, Galatina, Guagnano, Maglie and Surbo). For the Brindisi area, there are a total of 16 functioning stations: 7 in the city and 9 in the province (Ceglie Messapica, Cisternino, Francavilla Fontana, Mesagne, San Pancrazio, San Pietro Vernotico, and three in Torchiarolo).

Generally, the average data of nitrogen dioxide concentration near a specific
detection point must be considered very carefully because the measured values obviously depend on the particular location where the unit is positioned: i.e., it can be placed near a road junction with vehicular congestion, or near to an industrial emission point, or conversely, far from big emission sources.

For this reason, and in order to proceed with a more significant analysis, we have decided to aggregate the daily data of the monitoring stations into 7-days arithmetic means for the area of the ex-ILVA plant, for the three provinces and, within the provinces, for the single urban areas of each capital. Obviously, in cases where the daily data of a specific detection unit were absent, the corresponding value (equal to zero) was not taken into account for the average of the aggregated specific territorial index.

To simplify the analysis, and also because the data are not yet all available within the ARPA-Puglia website, the orography of the different territorial areas (mostly all flat lands) and the respective local meteorological data (wind conditions, average environmental temperatures, atmospheric pressure, precipitation, etc.) were not taken into account. However, using weekly means eliminates many effects caused by short-term weather changes and offers the opportunity to better outline trends.

For the quantification of NO\textsubscript{2} contributions by source, the following assumptions have been made: 1) that NO\textsubscript{2} concentrations are generally attributable to: a portion of background pollution (BP), a portion of pollution from the emissions of vehicular traffic (VT), and a portion attributable to domestic heating systems (HS); 2) that, in the first studied period, all the different components (BP, VT and HS) had a 100% effect; 3) that, in the second period (lockdown), BP had a 100% effect, VT a 30% effect, considering that about 70% (equal to 5% of the total number of vehicles of the whole territory of Greater Salento) of commercial vehicles were in circulation and that only 25% of passenger cars were also circulating, and that HS had an effect of 50% considering the higher environmental temperatures at the end of the second period (average temperatures above 18\textdegree C for about 50% of the days studied); 4) that in the third period, BP was 100%, VT 100% and HS = 0, considering that the heating systems were turned off due to the almost summer average environmental temperatures: above 22\textdegree C from 4\textsuperscript{th} May to 31\textsuperscript{st} May 2020.

Starting from these assumptions, we could set up a simple system of three equations in three unknown variables (BP, VT and HS) to be calculated for each territorial area:

- **First equation (or period):**
  \[
  BP_{100\%} + VT_{100\%} + HS_{100\%} = a; 
  \]
- **Second equation (or period):**
  \[
  BP_{100\%} + VT_{30\%} + HS_{50\%} = b; 
  \]
- **Third equation (or period):**
  \[
  BP_{100\%} + VT_{100\%} + HS_{0\%} = c; 
  \]
of which \(a, b\) and \(c\) are known quantities and correspond to the averages of \(\text{NO}_2\) concentrations for each considered period and within a territorial area (province or capital).

From the above system, we can carry out the following calculations. Because, from the (3),

\[
\text{BP}_{100\%} + \text{VT}_{100\%} = c,
\]

substituting this equivalence in (1), we obtain:

\[
c + \text{HS}_{100\%} = a \quad \text{and then} \quad \text{HS}_{100\%} = a - c
\]

Then, because

\[
\text{VT}_{50\%} \text{ is } 3/10 \text{ VT}
\]

and

\[
\text{HS}_{50\%} \text{ is } 1/2 \text{ HS}
\]

and

\[
\text{HS}_{0\%} \text{ is zero}
\]

from the (2), it follows that

\[
\text{BP}_{100\%} = \text{BP} = b - 3/10 \text{ VT} - 1/2 \text{ HS}
\]

and from the (3), it follows that

\[
\text{VT}_{100\%} = \text{VT} = c - \text{BP}_{100\%} = c - \text{BP}
\]

and then

\[
\text{VT} = c - (b - 3/10 \text{ VT} - 1/2 \text{ HS})
\]

\[
\rightarrow 10\text{VT} = 10c - 10b + 3\text{VT} + 5\text{HS}
\]

\[
\rightarrow 10\text{VT} - 3\text{VT} = 10c - 10b + 3\text{VT} + 5\text{HS}
\]

\[
\rightarrow 7/10 \text{VT} = c - b + 1/2 \text{ HS}
\]

\[
\rightarrow \text{VT} = (c - 1/2 \text{HS} - b) \times 10/7
\]

from the (1), we have

\[
\text{BP}_{100\%} = \text{BP} = a - \text{HS} - \text{VT}
\]

definitely, we obtain that:

\[
\text{HS} = a - c \quad (4)
\]

\[
\text{VT} = (c - \text{HS}/2 - b) \times 10/7 = (c - (a - c)/2 - b) \times 10/7 \quad (5)
\]

\[
\text{BP} = a - \text{HS} - \text{VT} = a - (a - c) - (c - (a - c)/2 - b) \times 10/7 \quad (6)
\]

in this way we were able to calculate, in the following paragraph, the values of the unknown variables, that is the sources allocations of \(\text{NO}_2\) concentrations, for each examined territory.

### 4. Results and Discussion

The scientific results obtained, thanks to the processing of the daily average data available from the 37 ARPA monitoring stations and applying the methods de-
scribed in the previous paragraph, makes it possible to calculate the weekly averages of the concentrations of nitrogen dioxide in the single examined territories (Table 1).

Table 1. Weekly arithmetic means of NO2 concentrations (μg/m³, 2020).

| First period | ex-ILVA | Taranto-City | Taranto-Prov. | Taranto-Tot. | Lecce-City | Lecce-Prov. | Lecce-Tot. | Brindisi-City | Brindisi-Prov. | Brindisi-Tot. | Greater Salento |
|---------------|---------|--------------|---------------|--------------|------------|-------------|------------|--------------|---------------|--------------|----------------|
| 27/01-02/02   | 61.0    | 50.8         | 53.5          | 52.2         | 59.2       | 43.3        | 51.2       | 62.4         | 42.8          | 52.6         | 54.3           |
| 03/02-09/02   | 52.3    | 37.4         | 43.2          | 40.3         | 42.0       | 35.7        | 38.9       | 41.1         | 31.3          | 36.2         | 41.9           |
| 10/02-16/02   | 61.9    | 44.3         | 50.8          | 47.5         | 41.1       | 39.7        | 40.4       | 53.1         | 34.4          | 43.7         | 48.4           |
| 17/02-23/02   | 62.1    | 44.7         | 43.0          | 43.9         | 51.5       | 36.5        | 44.0       | 49.5         | 33.8          | 41.7         | 47.9           |
| 24/02-01/03   | 60.4    | 45.6         | 40.3          | 42.9         | 39.8       | 26.2        | 33.0       | 42.6         | 26.9          | 34.7         | 42.8           |
| 02/03-09/03   | 53.5    | 35.8         | 39.2          | 37.5         | 45.0       | 30.4        | 37.7       | 38.3         | 28.5          | 33.4         | 40.5           |
| Means 27/01-09/03 (A) | 58.5 | 43.1 | 45.0 | 44.0 | 46.4 | 35.3 | 40.9 | 47.8 | 33.0 | 40.4 | 46.0 |
| Standard Deviation | 4.4 | 5.6 | 5.8 | 5.2 | 7.5 | 6.2 | 6.2 | 9.0 | 5.7 | 7.2 | 5.2 |

Lockdown (second) period

| 10/03-15/03 | 58.8    | 39.4         | 42.0          | 40.7         | 29.3       | 24.6        | 26.9       | 32.8         | 21.8          | 27.3         | 38.4           |
| 16/03-22/03 | 62.5    | 44.3         | 39.6          | 42.0         | 48.9       | 37.1        | 43.0       | 50.9         | 32.9          | 41.9         | 47.3           |
| 23/03-29/03 | 30.1    | 16.3         | 20.9          | 18.6         | 18.9       | 16.7        | 17.8       | 27.6         | 15.6          | 21.6         | 22.0           |
| 30/03-05/04 | 43.8    | 23.1         | 18.1          | 20.6         | 16.6       | 15.9        | 16.3       | 26.6         | 13.8          | 20.2         | 25.2           |
| 06/04-12/04 | 48.4    | 25.0         | 24.2          | 24.6         | 21.4       | 17.4        | 19.4       | 29.5         | 17.9          | 23.7         | 29.0           |
| 13/04-19/04 | 40.9    | 23.2         | 23.8          | 23.5         | 13.6       | 12.5        | 13.1       | 26.4         | 15.2          | 20.8         | 24.6           |
| 20/04-26/04 | 34.0    | 16.6         | 20.8          | 18.7         | 13.2       | 10.1        | 11.7       | 27.1         | 12.3          | 19.7         | 21.0           |
| 27/04-03/05 | 39.4    | 21.9         | 21.0          | 21.4         | 10.9       | 9.8         | 10.3       | 30.3         | 13.5          | 21.9         | 23.3           |
| Means 10/03-03/05 (B) | 44.7 | 26.2 | 26.3 | 26.3 | 21.6 | 18.0 | 19.8 | 31.4 | 17.9 | 24.6 | 28.9 |
| Difference: (B - A)/A | -23.6% | -39.1% | -41.6% | -40.4% | -53.5% | -48.9% | -51.5% | -34.4% | -45.7% | -39.0% | -37.2% |
| Standard Deviation | 11.4 | 10.2 | 9.2 | 9.5 | 12.5 | 9.1 | 10.7 | 8.2 | 6.8 | 7.4 | 9.3 |

Third period

| 04/05-10/05 | 38.1    | 23.5         | 26.7          | 25.1         | 15.4       | 12.9        | 14.1       | 24.3         | 12.3          | 18.3         | 23.9           |
| 11/05-17/05 | 32.9    | 22.3         | 24.0          | 23.2         | 16.6       | 12.6        | 14.6       | 29.9         | 12.9          | 21.4         | 23.0           |
| 18/05-24/05 | 50.3    | 29.9         | 28.0          | 28.9         | 17.1       | 15.5        | 16.3       | 31.8         | 15.0          | 23.4         | 29.7           |
| 25/05-31/05 | 47.9    | 24.5         | 17.3          | 20.9         | 16.0       | 17.1        | 16.6       | 26.3         | 11.0          | 18.7         | 26.0           |
| 01/06-07/06 | 35.1    | 21.1         | 22.3          | 21.7         | 20.0       | 13.4        | 16.7       | 28.5         | 14.4          | 21.4         | 23.7           |
| 08/06-14/06 | 40.1    | 25.5         | 25.3          | 25.4         | 20.2       | 14.9        | 17.5       | 33.6         | 16.9          | 25.3         | 27.1           |
| Means 04/05-14/06 (C) | 40.7 | 24.5 | 23.9 | 24.2 | 17.5 | 14.4 | 16.0 | 29.1 | 13.7 | 21.4 | 25.6 |
| Difference: (C - B)/B | -8.9% | -6.7% | -9.0% | -7.8% | -18.7% | -20.0% | -19.3% | -7.4% | -23.2% | -13.1% | -11.4% |
| Standard Deviation | 7.0 | 3.1 | 3.8 | 2.9 | 2.0 | 1.8 | 1.3 | 3.4 | 2.1 | 2.7 | 2.5 |
| Total Average | 47.7 | 30.8 | 31.2 | 31.0 | 27.8 | 22.1 | 25.0 | 35.6 | 21.2 | 28.4 | 33.0 |
| Total Standard Deviation | 11.0 | 10.9 | 11.4 | 11.0 | 15.2 | 11.0 | 13.0 | 10.8 | 9.6 | 10.1 | 10.9 |

Source: our own processing of data from ARPA Puglia (http://www.arpa.puglia.it).

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First of all, we would like to specify that, for none of the air quality monitoring stations of the examined areas and for all the time periods analyzed (both for 2020 and for the previous year), the limits established by the current Italian legislation (Legislative Decree 155/2010) have been exceeded.

Some considerations can be drawn examining Table 1. First of all, during the lockdown period, the NO$_2$ average concentrations decreased ($-37.2\%$) for Greater Salento, but much less for ex-ILVA ($-23.6\%$), because it is obviously much less affected by vehicle emissions. Conversely, the province of Lecce, overall, is the area with the greatest reduction ($-51.5\%$); this is a clear indication that, in this area, vehicles emissions are the main sources of nitrogen dioxide.

This general decreasing trend also continued during the third period following the lockdown, with an average of $-11.4\%$ for Greater Salento, $-23.2\%$ for the province of Brindisi (excluding the capital), and $19.3\%$ for the entire province of Lecce. This is probably due to the shutdown of the domestic heating systems following the rise of average seasonal temperatures. In particular, between the third and second periods, the province of Lecce shows the greatest decrease ($-19.3\%$), followed by the province of Brindisi (excluded capital) with its $-13.1\%$, ex-ILVA ($-8.9\%$), Brindisi ($-7.4\%$) and Taranto ($-6.7\%$). The areas near the large industrial plants have recorded the lowest decreases. Away from these production sites, the decreases of NO$_2$ concentrations are higher because they are less dependent on industrial emissions and much more on emissions of heating systems. This can be seen very clearly by the corresponding graphic representation (Figure 1).

As we can see, the NO$_2$ concentration values of ex-ILVA are considerably higher for all the three periods, compared to the other territories and also compared to the average of Greater Salento, with differences ranging from $30\%$, before lockdown, to around $60\%$, during the lockdown, and in the following third

![Figure 1. NO$_2$ average concentrations during the analyzed three periods of 2020.](image-url)
period. Immediately after ex-ILVA, the territory which has the highest average concentration values for all the analyzed periods, due to the presence of its chemical and thermoelectric plants, is the city of Brindisi: 47.8 μg/m³ (46 μg/m³ for Greater Salento) in the first period, 31.4 μg/m³ (28.9 μg/m³ for Greater Salento) in the second period, 29.1 μg/m³ (25.6 μg/m³ for Greater Salento) in the third period.

The subsequent Copernicus Sentinel-5P satellite images by European Space Agency (ESA) [20], relating to the following three 14-days periods of 2020: 24.02 - 09.03 (Figure 2(a)); 30.03 - 13.04 (Figure 2(b)); 04.05 - 18.05 (Figure 2(c)), are confirmations of the above statements. We can visually see the progressive downsizing of the red zones, which represent the tropospheric concentration of NO₂, both in the second and third periods, especially near the industrial area of ex-ILVA and of a large part of the province of Taranto.

The standard deviations, for all the regions, are considerably higher during the lockdown period (9.3 μg/m³ for Greater Salento) compared to the previous period (5.2 μg/m³) and to the following period (2.5 μg/m³). This is probably due to the fact that, during the second period, the emission quota of the household heating systems, whose operation is fluctuating because it is a function of the
average daily temperatures, has become predominant compared to the other polluting sources. Over the three considered periods, the highest standard deviation variations are those of Lecce (15.2 μg/m³), while the province of Brindisi has the lowest variations (9.6 μg/m³).

In order to have an overview of the many data included in Table 1, we can represent the weekly average values of NO₂ concentrations in the corresponding graph below (Figure 3).

For the entire territory of Greater Salento, the anomalous data of the weekly averages for the period 16/03-22/03, that is, a week after the declaration of restriction of people’s movement, is clearly evident. This could be due to the average temperatures of the following days, 17th, 18th, 20th and 21st March (according to the ARPA Puglia weather data) [19], that were exceptionally much lower than the average seasonal ones and therefore led to a much longer functioning time of the household heating systems. Conversely, after this period, there is also a drastic reduction of NO₂ concentrations starting from the week of 23/03-29/03.

As already mentioned above, and as graphically highlighted in Figure 1, the average of NO₂ concentrations of ex-ILVA and Brindisi are significantly and almost always constantly above the lines of the corresponding weekly average data of the other territories.

Table 2 and the related Figure 4 illustrate the calculation of the correlation coefficients between the pollutant concentration values of ex-ILVA and Brindisi, with their surrounding regions and with Greater Salento.

The average concentration values of the ex-ILVA area are highly correlated with those of Taranto (0.947) and with those of the territory of the province of Lecce (excluding the city of Lecce); less correlated with the province of Brindisi (0.818) and, in particular, with those of the city of Brindisi (0.799). Conversely, the weekly average values of Brindisi are highly correlated with those of its province (0.963) and also with those of the province of Lecce (0.937), much less with those of the province of Taranto (0.899) and with those of ex-ILVA (0.799).
This means that the contribution to the nitrogen dioxide concentrations of the threshold pollution values of ex-ILVA and Brindisi are predominant compared to other sources (vehicles and heating systems emissions) and that both partly influence the air quality of the province of Lecce (see Figure 2).

In Table 3, the territorial differences, based on the number of inhabitants,
Table 2. Territorial correlation of the weekly average NO$_2$ concentrations and standard deviation during the period 27/01-14/06 2020.

|               | ex-ILVA | Taranto-City | Taranto-Prov. | Taranto-Tot. | Lecce-City | Lecce-Prov. | Lecce-Tot. | Brindisi-City | Brindisi-Prov. | Brindisi-Tot. | Greater Salento* |
|---------------|---------|--------------|---------------|--------------|------------|-------------|------------|--------------|----------------|---------------|----------------|
| Correlation with ex-ILVA | 1.000   | 0.947        | 0.840         | 0.905        | 0.828      | 0.854       | 0.846      | 0.799        | 0.825          | 0.818         | 0.872          |
| Correlation with Brindisi | 0.799   | 0.908        | 0.899         | 0.917        | 0.928      | 0.930       | 0.937      | 1.000        | 0.963          | 0.992         | 0.963          |
| Standard deviation | 11.0    | 10.9         | 11.4          | 11.0         | 15.2       | 11.0        | 13.0       | 10.8         | 9.6            | 10.1          | 11.2           |

*without ex-ILVA; Source: our own processing on data of ARPA Puglia data (http://www.arpa.puglia.it).

Table 3. Passenger cars and other variables (31.12.2018).

|               | Taranto-City | Taranto-Prov. | Taranto-Tot. | Lecce-City | Lecce-Prov. | Lecce-Tot. | Brindisi-City | Brindisi-Prov. | Brindisi-Tot. | Greater Salento |
|---------------|--------------|---------------|--------------|------------|-------------|------------|--------------|----------------|---------------|----------------|
| Inhabitants (nr.) | 196,702      | 380,054       | 576,756      | 95,269     | 699,865     | 795,134    | 86,812       | 306,163        | 392,975       | 1,764,865     |
| Surface (km$^2$)  | 250          | 2217          | 2467         | 239        | 2560        | 2799       | 333          | 1528           | 1861          | 7127           |
| Cars (nr.)       | 109,641      | 228,522       | 338,163      | 65,961     | 445,519     | 511,480    | 52,725       | 198,776        | 251,501       | 1,101,144      |
| Inhabitants (%)  | 11.1%        | 21.5%         | 32.7%        | 5.4%       | 39.7%       | 45.1%      | 4.9%         | 17.3%          | 22.3%         | 100.0%         |
| Surface (%)      | 3.5%         | 31.1%         | 34.6%        | 3.4%       | 35.9%       | 39.3%      | 4.7%         | 21.4%          | 26.1%         | 100.0%         |
| Cars (%)         | 10.0%        | 20.8%         | 30.7%        | 6.0%       | 40.5%       | 46.4%      | 4.8%         | 18.1%          | 22.8%         | 100.0%         |
| Inhab./surface (nr./km$^2$) | 787.1        | 171.4         | 233.8        | 398.8      | 273.4       | 284.1      | 260.7        | 200.4          | 211.2         | 247.6           |
| Cars/inhab. (*100) | 55.7         | 60.1          | 58.6         | 69.2       | 63.7        | 64.3       | 60.7         | 64.9           | 64.0          | 62.4            |
| Cars/surface (nr./km$^2$) | 438.7        | 103.1         | 137.1        | 276.1      | 174.0       | 182.7      | 158.3        | 130.1          | 135.1         | 154.5           |

Source: our own processing of data from ANFIA (http://www.anfia.it).

territorial surface area, circulating cars, population density and number of cars per inhabitant and per km$^2$, are reported.

Taranto has the highest population density compared to the corresponding territorial surface (787.1 inhab./km$^2$, 247.6 for Greater Salento) and the highest concentration of passenger cars (438.7 cars/km$^2$, 154.5 for Greater Salento). Lecce is in second place, at a considerable distance, based on the same indexes. Last is the province of Brindisi. Basically, this could mean that the high NO$_2$ concentrations of the cities of Taranto and Lecce would be largely due both to population density, which presupposes a massive use of domestic heating systems, and to the significant number of cars per km$^2$. Further on, we will check if this hypothesis is correct.

As we can easily see both from Table 1 and from the above corresponding graphic representation (Figure 3), there is an apparently abnormal trend: in the third period, the average emissions are lower than those in the lockdown period. This is probably due to the massive shutdown of the household heating systems. We will verify this hypothesis by analyzing what happened in the corresponding periods of 2019 (Table 4).

In the second period of 2019, the average of NO$_2$ concentrations, for the entire Greater Salento, including ex-ILVA, was 37.1 μg/m$^3$, 28.4% higher than the
Table 4. Weekly arithmetic means of NO\textsubscript{2} concentrations (μg/m\textsuperscript{3}, 2019).

| First period | ex-ILVA | Taranto-City | Taranto-Prov. | Taranto-Tot. | Lecce-City | Lecce-Prov. | Lecce-Tot. | Brindisi-City | Brindisi-Prov. | Brindisi-Tot. | Greater Salento |
|--------------|---------|--------------|---------------|--------------|------------|-------------|------------|--------------|---------------|--------------|----------------|
| 28/01/19-03/02/19 | 58.7    | 35.6         | 45.7          | 40.7         | 43.4       | 30.1        | 36.7       | 48.4         | 34.4          | 41.4         | 44.4           |
| 04/02/19-10/02/19 | 51.6    | 36.0         | 41.6          | 38.8         | 42.4       | 33.6        | 38.0       | 44.9         | 31.4          | 38.2         | 41.6           |
| 11/02/19-17/02/19 | 50.4    | 36.3         | 30.1          | 33.2         | 30.7       | 19.5        | 25.1       | 28.1         | 18.5          | 23.3         | 33.0           |
| 18/02/19-24/02/19 | 68.9    | 50.5         | 57.1          | 53.8         | 54.7       | 30.2        | 42.5       | 55.7         | 36.7          | 46.2         | 52.8           |
| 25/02/19-03/03/19 | 66.8    | 45.9         | 40.4          | 43.1         | 46.8       | 32.8        | 39.8       | 54.8         | 31.8          | 43.3         | 48.3           |
| 04/03/19-10/03/19 | 62.6    | 37.5         | 43.1          | 40.3         | 44.5       | 23.6        | 34.0       | 46.9         | 28.2          | 37.5         | 43.6           |

Means 28/01-10/03 (A) | 59.9 | 40.3 | 43.0 | 41.6 | 43.8 | 30.2 | 36.0 | 46.5 | 30.2 | 38.3 | 44.0 |

Second period
| 11/03/19-17/03/19 | 62.0    | 36.7         | 36.2          | 36.4         | 45.3       | 28.6        | 37.0       | 46.7         | 31.0          | 38.8         | 43.5           |
| 18/03/19-24/03/19 | 58.4    | 31.7         | 27.5          | 29.6         | 26.7       | 23.0        | 24.9       | 33.9         | 19.6          | 26.7         | 34.9           |
| 25/03/19-31/03/19 | 64.6    | 34.9         | 30.9          | 32.9         | 36.5       | 21.0        | 28.7       | 37.6         | 21.1          | 29.3         | 38.9           |
| 01/04/19-07/04/19 | 62.4    | 37.1         | 41.5          | 39.3         | 44.5       | 26.1        | 35.3       | 47.4         | 26.9          | 37.1         | 43.5           |
| 08/04/19-14/04/19 | 60.0    | 33.9         | 34.3          | 34.1         | 51.2       | 21.5        | 36.4       | 41.1         | 25.8          | 33.4         | 41.0           |
| 15/04/19-21/04/19 | 62.9    | 41.7         | 35.7          | 38.7         | 33.2       | 22.0        | 27.6       | 36.2         | 21.9          | 29.0         | 39.6           |
| 22/04/19-28/04/19 | 39.1    | 23.4         | 26.6          | 25.0         | 25.0       | 13.3        | 19.1       | 27.5         | 16.8          | 22.1         | 26.3           |
| 29/04/19-05/05/19 | 40.6    | 25.7         | 29.7          | 27.7         | 26.2       | 17.2        | 21.7       | 33.3         | 22.1          | 27.7         | 29.4           |

Means 11/03-05/05 (B) | 56.3 | 33.1 | 32.8 | 33.0 | 36.1 | 21.6 | 28.8 | 38.0 | 23.1 | 30.5 | 37.1 |

Difference: (B − A)/A | −6.0% | −17.8% | −23.7% | −20.8% | −17.5% | −23.7% | −20.0% | −18.3% | −23.3% | −20.3% | −15.5% |

Third period (C)
| 06/05/19-12/05/19 | 48.4    | 26.9         | 34.1          | 30.5         | 21.5       | 15.5        | 18.5       | 28.6         | 18.4          | 23.5         | 30.2           |
| 13/05/19-19/05/19 | 38.9    | 22.5         | 27.2          | 24.8         | 26.1       | 15.8        | 20.9       | 34.9         | 16.5          | 25.7         | 27.6           |
| 20/05/19-26/05/19 | 43.2    | 27.8         | 32.6          | 30.2         | 24.5       | 14.4        | 19.4       | 39.2         | 20.5          | 29.9         | 30.7           |
| 27/05/19-02/06/19 | 31.4    | 21.1         | 22.3          | 21.7         | 21.1       | 12.8        | 16.9       | 22.4         | 13.9          | 18.1         | 22.0           |
| 03/06/19-09/06/19 | 54.1    | 40.4         | 42.3          | 41.4         | 29.9       | 17.5        | 23.7       | 36.0         | 22.3          | 29.2         | 37.1           |
| 10/06/19-16/06/19 | 50.0    | 38.3         | 35.5          | 36.9         | 35.3       | 19.4        | 27.3       | 40.3         | 22.9          | 31.6         | 36.4           |

Means 06/05-16/06 (C) | 44.3 | 29.5 | 32.3 | 30.9 | 26.4 | 15.9 | 21.1 | 33.6 | 19.1 | 26.3 | 30.7 |

Difference: (C − B)/B | −21.2% | −10.9% | −1.5% | −6.2% | −26.9% | −26.4% | −26.7% | −11.6% | −17.6% | −13.8% | −17.4% |

Source: our own processing of data from ARPA Puglia (http://www.arpa.puglia.it).

The corresponding average value of 2020. The corresponding decrease, compared to the previous period of the same year, was −15.5% (−37.2% in 2020). The difference, however, between the third period and the second, was, for 2019, −17.4% (−6.4 μg/m\textsuperscript{3}) compared to the corresponding lowest mean of 2020 (−11.4%, 3.4 μg/m\textsuperscript{3}). This could probably be attributed to the higher average environmental temperatures during the second period of 2020 compared to the same period of 2019.

Considering the concentration means of this pollutant of ex-ILVA, in the three periods of 2020 (Table 1), and comparing them with the corresponding means of Greater Salento, excluding ex-ILVA, it is noted that, in the first period,
the difference is 16.8 μg/m³ (+40.1%), in the second period it is 21.2 μg/m³ (+89.8%) and in the third period it is 20.2 μg/m³ (+98.4%). In the previous year, however, the corresponding differences (Table 4) were greater in the first period (+54.9%), but lower in the second (+82.8%) and significantly more contained in the third period (+69.9%). This is probably a sign that the decrease of vehiculars circulation during the lockdown period persist even after it, likely because there is a reduction in commercial activity.

In Table 5 the inter-periods differences of the NO₂ concentrations during 2020 compared to the previous year are shown in detail.

For Greater Salento, the greatest biannual variations occurred, obviously, in the second period and were equal to −8.3 μg/m³ (−22.3%). The territorial area with the highest decreases was the province of Lecce (−31.3%); in particular, for Lecce, the decreases compared to the previous year were even greater: with −40.2%, in the second period, and −33.5%, in the third period. The situation of Lecce can be explained by the fact that its NO₂ concentrations depend largely on vehicular traffic and domestic heating systems and very little on pollutants of industrial origin; this fact will be verified later. The province of Brindisi, on the other hand, had the smallest decreases (−19.3%). This could be explained by the fact that in the district adjacent to the city of Brindisi there is a consistent concentration of highly polluting industries, while, for the province of Lecce, the low biannual decrease can be explained considering that the concentration means of 2019 were already very low.

Excluding the values of ex-ILVA, whose NO₂ concentrations are, to a predominant extent, presumably dependent on internal dynamics, from the second period of the analyzed two-year period, the decrease is substantially the same as before (−23.4%, for 2020, compared to a −22.3%, for 2019).

Next, we have deduced the contribution of emission sources to the average weekly concentrations of this pollutant. Starting from the considerations and hypotheses outlined in the previous paragraph, set and solve, for the entire Greater Salento, the system of 3 equations in 3 unknowns, as illustrated above (see formulas [4] [5] [6]), obtaining the results shown in the following Table 6, by inserting the NO₂ average concentrations (Table 1) of the three analyzed periods.

### Table 5. Differences between the three periods (μg/m³, 2020-2019).

|               | ex-ILVA | Taranto-City | Taranto-Prov. | Taranto-Tot. | Lecce-City | Lecce-Prov. | Lecce-Tot. | Brindisi-City | Brindisi-Prov. | Brindisi-Tot. | Greater Salento |
|---------------|---------|--------------|---------------|--------------|------------|-------------|------------|--------------|----------------|---------------|-----------------|
| **First period** | −1.3    | 2.8          | 2.0           | 2.4          | 2.7        | 7.0         | 4.8        | 1.4          | 2.8            | 2.1           | 2.0             |
|               | −2.2%   | 7.0%         | 4.6%          | 5.8%         | 6.1%       | 24.7%       | 13.4%      | 3.0%         | 9.2%           | 5.4%          | 4.5%            |
| **Second period** | −11.5   | −6.9         | −6.5          | −6.7         | −14.5      | −3.6        | −9.0       | −6.6         | −5.2           | −5.9          | −8.3            |
|               | −20.5%  | −20.8%       | −19.9%        | −20.3%       | −40.2%     | −16.5%      | −31.3%     | −17.3%       | −22.7%         | −19.3%        | −22.3%          |
| **Third period** | −3.6    | −5.0         | −8.4          | −6.7         | −8.8       | −1.5        | −5.2       | −4.5         | −5.3           | −4.9          | −5.1            |
|               | −8.1%   | −17.0%       | −26.0%        | −21.7%       | −33.5%     | −9.3%       | −24.4%     | −13.4%       | −28.0%         | −18.7%        | −16.6%          |

Source: our own processing on data of ARPA Puglia (http://www.arpa.puglia.it).
As expected, for the area of ex-ILVA the traffic exhaust component has the lowest share (12%) and the background pollution component is the highest (57.6%). Also for Brindisi the share of the background pollution is preponderant referred to the other components (39.6%), contrary to what happens in all the remaining regions of Greater Salento, where the preponderant share must be ascribed to the household heating systems (44.3%, for the entire territory of Salento, in contrast to a smaller quota of 21.5% of vehicular traffic and 34.2% of basic pollution). The aforementioned considerations are much more easily inferred by the corresponding graphic representation (Figure 5).

5. Conclusions

What clearly emerges from our processing and interpretation of the ARPA-Puglia data is the significant decrease of the NO₂ concentration weekly averages during the Italian lockdown period. It is also evident that a further decrease occurred, for

| Components | ex-ILVA | Taranto-City | Taranto-Prov. | Taranto-Tot. | Lecce-City | Lecce-Prov. | Lecce-Tot. | Brindisi-City | Brindisi-Prov. | Brindisi-Tot. | Greater Salento |
|------------|---------|--------------|---------------|--------------|------------|-------------|------------|--------------|---------------|--------------|----------------|
| HS         | 17.8    | 18.6         | 21.1          | 19.8         | 28.9       | 20.9        | 24.9       | 18.8         | 19.2          | 19.0         | 20.4           |
| VT         | 7.0     | 10.8         | 11.7          | 11.2         | 14.8       | 9.8         | 12.3       | 10.1         | 7.8           | 8.9          | 9.9            |
| BP         | 33.7  | 13.7         | 12.3          | 13.0         | 2.7        | 4.6         | 3.7        | 19.0         | 6.0           | 12.5         | 15.7           |
| HS %       | 30.4%   | 43.2%        | 46.8%         | 45.0%        | 62.2%      | 59.2%       | 60.9%      | 39.2%        | 58.3%         | 47.0%        | 44.3%          |
| VT %       | 12.0%   | 25.1%        | 25.9%         | 25.5%        | 32.0%      | 27.7%       | 30.1%      | 21.1%        | 23.6%         | 22.1%        | 21.5%          |
| BP %       | 57.6%   | 31.8%        | 27.2%         | 29.4%        | 5.8%       | 13.2%       | 9.0%       | 39.6%        | 18.1%         | 30.8%        | 34.2%          |

Legend: HS: household heating systems; VT: vehicular traffic; BP: background pollution. Source: our own processing of data from ARPA Puglia (http://www.arpa.puglia.it).

Figure 5. Shares of NO₂ concentrations from different sources in the provinces of Greater Salento during the three periods.
all the analyzed provinces, during the last period, that is during the post-lockdown phase and the resumption of movements of people. This is probably attributable, as written above, to the shutdown of the domestic heating systems, subsequent the significantly higher average environmental temperatures.

During the lockdown period, the province of Lecce, in its entirety, had the largest decreases in NO2 concentrations, referred to the surrounding territories and to the immediately previous weeks. This is attributable to the fact that—as also shown by the last table and the last graph concerning the contribution of the different component sources to the overall average values of nitrogen dioxide concentration—in this specific territory, the emission sources of vehicular traffic and household heating systems are, by far, predominant referred to the back-ground pollution, which is instead relevant for both the provinces of Taranto and Brindisi.

This paper contributes to the awareness that at least regarding NO2 emissions, the containment measures concerning the blocking of traffic, adopted so far by European and Italian governments and by those municipalities congested by circulating vehicles are not sufficient to lower the concentration peaks of nitrogen dioxide, because other sources of emissions (such as the household heating systems, above all), especially in cities with high demographic density or high industrialized sites, contribute to the emissions of this pollutant. It is therefore necessary to reconsider the problems relating to NO2 pollution and air quality of our cities in order to take more incisive and effective, global and local, containment measures identifying new strategies focused on multiple emission sources and multi-sectorial actions.

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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.

Authors’ Contributions

The contribution is to be attributed, in equal parts, to the authors. All authors have read and approved the final manuscript.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.
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