https://doi.org/10.5194/acp-2020-600
Preprint. Discussion started: 27 July 2020

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Sudden changes in nitrogen dioxide emissions over Greece due to lockdown after the outbreak of COVID-19

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Abstract: The unprecedented order, in modern peaceful times, for near-total lockdown of the Greek population, as means of protection against the Severe Acute Respiratory Syndrome CoronaVirus-2, commonly known as COVID-19, infection, has brought unintentional positive side-effects to the country's air quality levels. S5P/TROPOMI monthly mean tropospheric nitrogen dioxide (NO_2) observations show an average decrease of -3% to -26% [-1% to -27%] with an average of -22% [-11%] for March and April 2020 respectively, compared to the previous year, over the six larger Greek metropolitan areas, attributable mostly to vehicular emission reductions. Furthermore, significant effects for shipping emissions over the Aegean Sea as well as the areas surrounding major Greek ports were observed, of approximately -12% [-5%]. For the capital city of Athens, weekly analysis was possible and it revealed a marked decline in NO_2 load between -8% and -43% for seven of the eight weeks studied. Chemical transport modelling, provided by the LOTOS-EUROS CTM, shows that the magnitude of these reductions cannot solely be attributed to the difference in meteorological factors affecting NO_2 levels during March and April 2020 and the equivalent time periods of the previous year. Taking this factor into account, the resulting decline was estimated to range between 0% and -37% for the five largest cities, with an average of ~ -10%. As transport is the second largest sector that affects Greece's air quality, this occasion may well help policy makers in enforcing more targeted measures to aid Greece in further reducing emissions according to international air quality standards.

Keywords: Air quality; nitrogen dioxide; NOx; emissions;Sentinel-5P; TROPOMI; LOTOS-EUROS; COVID-19; pandemic; Athens; Greece

1. Introduction

In this work we aim to show the quantifiable and beyond doubt decline in tropospheric nitrogen dioxide (NO_2) levels over Greece during the ongoing Severe Acute Respiratory Syndrome CoronaVirus-2, commonly known as *COVID-19*, pandemic, as sensed by the space-borne S5P/TROPOMI, here after TROPOMI, instrument. By comparing the relative levels for the months of March and April for years 2020 and 2019, while properly accounting for the differences in meteorology using the simulations of a Chemical Transport Model, CTM, we enumerate the improvement in local and regional air quality due to the reduced nitrogen oxides (NOx) emissions.

In the following sections, we provide basic information on tropospheric NOx, we focus on current knowledge of the nominal NOx emissions over Greece, we then present a brief overview of the capabilities of current and past satellite instruments in sensing abrupt atmospheric content changes and enumerate the dates when the different lockdown measures were enforced in Greece.

1.1. Nitrogen oxides in the troposphere

Nitrogen dioxide (NO_2) and nitrogen oxide (NO), referred to more commonly as nitrogen oxides (NOx), are important trace gases in the Earth's troposphere. NOx are emitted as a result of both anthropogenic activities, such as fossil fuel combustion and biomass burning, and natural processes, such as microbiological processes in soils, wildfires and lightning. In the presence of sunlight, the photochemical cycle of tropospheric ozone (O_3) converts NO into NO_2 on a timescale of minutes and so NO_2 is considered a robust measure for concentrations of nitrogen oxides (Jacob, 1999). For typical levels of the OH radical, the lifetime of NOx in the lower troposphere is less than a day, normally a few hours depending on the season and the rates of the photochemical reactions [see for e.g. Beirle et al., 2011; Mijling and van der A, 2012]. As a result, it is well accepted that NO_2 fluxes will remain relatively close to their source which, first of all, makes it possible for NOx emissions to be well detected from space [see for e.g. Stavrakou et al., 2008; Lamsal et al., 2010; van der A et al., 2008, among others] but also precludes any issues of transboundary pollution effects which might otherwise hinder this study.

In the troposphere, NO_2 plays a key role in air quality issues, as it directly affects human health [WHO, 2016]. In the European Union, the evidence of NO_2 health effects has led to the establishment of air quality standards for the protection of human health. Limit values for NO_2 are set at 200 μg m^{-3} for 1 h average concentrations (with 18 exceedances permitted per year), and 40 μg m^{-3} for annual average concentrations (European Council Directive 2008/50/EC, 2008). Concentrations above the annual limit value for NO_2 are still widely registered across Europe, even if concentrations and exposures continue to decrease [EEA, 2019]. In Greece in particular, the annual average standard of 40 μg m^{-3} has not been exceeded for years 2007 to 2017 when assuming all in situ stations; the traffic stations of Athens and Thessaloniki however show annual levels up to 45 μg m^{-3} for years 2015 to 2017. It hence follows logically that monitoring closely abrupt changes in NOx emissions for diverse locations plays a key role in shaping future environmental policies and directives.

1.2. Nitrogen dioxide emissions over Greece

According to the EEA Report No 8/2019, updated by the EU 2019 Environmental Implementation Review for Greece [EU, 2019], the country's NOx emissions by sector come from road transport, industry (which mainly covers the energy production and distribution sector), non-road transport, household and agriculture. The relative percentages for NOx air emissions, separated by sector, as extracted from the 2018 Air Emission Account 2015 report by the Hellenic Statistical Authority[1] [HAS, 2018], are: industry 48%, transport 22%, energy supply 18%, manufacturing 6%, central heating 4%, agriculture 1% and others 1%. Based on the European Environmental Agency, EEA, European Pollutant Release and Transfer Register (E-PRTR)[2], 77% of the reported industrial NOx/NO_2 emissions over Greece came from thermal power stations and other combustion installations. The monthly energy balance reports[3], composed by the Independent Power Transmission Operator of the Hellenic Electricity Transmission System (HETS), show that the total energy requested for March 2020 [4.152GWh] was -2.1% lower than 2019 [4.224GWh] , whereas for April 2020 [3.527GWh] was -9.8% lower than 2019 [3.527]. These reductions are quite typical of the seasonality of the energy consumption in Greece which peaks in December and January, due to heating needs, and in July and August, due to cooling needs, with seasonal lows in spring (April and May) and autumn (October and November). Furthermore, in the work of Fameli and Asimakopoulos, 2016, it is reported that the annual mean NOx emissions for Greece for years 2006 to 2012 can be attributed as follows, in order of relevance: industry, 45±3%, road transport, 35±8%, shipping 11±3%, non-road transport, 10±4%, central heating, 5±2%, with agriculture and aviation showing an average of around 1± each. If we assume that years 2019 and 2020 were not exceptional in their temperature levels for the spring months, then it follows that changes in central heating emissions will not bare a significant of the emission changes observed.

1.3. Sensing abrupt emission changes from space-borne sensors

Abrupt emission changes have already been reported using space-borne observations for a number of recent local and continental circumstances. Castellanos and Boersma, 2012, reported significant reductions in nitrogen oxides over Europe driven by environmental policy and the economic recession based on OMI/Aura observations between 2004 and 2010. Vrekoussis et al., 2013 and Zyrichidou et al., 2019, report strong correlations between pollutant levels and economic indicators showing that the 2008 economic recession has resulted in proportionally lower levels of pollutants in large parts of Greece. The latter, for years 2008 to 2015, showed surprisingly that, while the wintertime tropospheric NO_2 trends were negative, significant positive formaldehyde trends were observed from space, shown to be due to increased usage of affordable indoor heating methods (e.g. fireplaces and wood stoves). Space-sensed reduction in emissions, on a shorter time scale, have also been attributed to strict measures enforced for benign reasons. Mijling et al., 2009, calculated, using OMI/Aura and CTM results, reductions in NO_2 concentrations of approximately 60% above Beijing during the 2008 Olympic and Paralympic Games. Ding et al.,

[1] https://www.statistics.gr/en/statistics/env
[2] https://prtr.eea.europa.eu/#/home
[3] http://www.admie.gr/en/market-statistics/montlhy-energy-balance/

2015, showed a ~30% reduction in OMI/Aura columns, which was translated into a ~25% in actual emission levels during the Nanjing 2014 Youth Olympic Games.

Numerous first reports suggesting an improved air quality in after the COVID-19 lockdown was enforced have already been seen in major media outlets. Here we note the findings of Liu et al., 2020, who report, based on both OMI/Aura and TROPOMI, a 48% drop in tropospheric NO_2 from the 20 days averaged before the 2020 Lunar New Year to the 20 days after, which is 20% larger than that from recent years, and relate the increase in decline to the date of each Chinese province lockdown. Bauwens et al., 2020, based on the same sensors, also report an average NO_2 column drop over all Chinese cities of -40% relative to the same period in 2019, while the decreases in Western Europe and the U.S. were found to range between -20 to -38%.

1.4. The COVID-19 situation over Greece

A short review on the COVID-19 situation over Greece is given here, mainly focusing on providing the dates in March 2020 of the successive restrictive measures that affected NOx emissions. The country's General Secretariat for Civil Protection, GSVP[4], reacted quickly to the emerging situation in the neighbouring country Italy and long before the first causalities were reported, major festivities for the Carnival season planned for the 28th of February to the 2nd of March were cancelled, followed by cancellation of all other cultural and sporting activities on March 8th. On the 11th of March, all levels of education were suspended, when also a first wave of workplace closures begun and culminated on Monday 16th when all restaurants, cafes with sitting facilities, and in general the food [apart from supermarkets] and hospitality industry were shut down. In the following two days, all remaining retail activities were suspended apart from pharmacies. On Monday 16th restrictions on the size of public gatherings were announced and the public transport section [buses, trams, underground, trains] started to reduce capacity. On the same day, international travel controls begun banning incoming flights from high risk countries allowing flights only into Athens International airport, while the four inland borders were shut by the neighbouring countries at the end of March. On Monday 23rd of March, full restrictions on the people's movements were imposed with strict stay-at-home mandates, with exceptions for essential working personnel. The country remained in full lockdown mode until May 4th, including all religious-related congregations, with a complete and without exception restriction around the Greek Orthodox Easter holidays of the 19th of April.

These discreet dates of expected change in NO_x emissions permit the analysis and discussion of the observations during this unprecedented period, presented in the results section.

2. Materials and Methods

In this section we introduce the TROPOMI tropospheric NO_2 observations, the CTM LOTOS-EUROS simulations and the proposed methodology to account for the different meteorological conditions between the nominal period of March-April 2019 and the disrupted one of March-April 2020.

2.1. TROPOMI NO_2 observations.

The recently launched TROPOMI instrument on the Sentinel-5 Precursor (S5P) mission [Veefkind et al., 2012] has been providing global observations of ozone, nitrogen dioxide, carbon monoxide, sulfur dioxide, formaldehyde, and methane, as well as aerosol and cloud properties[5] since early 2018. Its very high spatial resolution of 3.5 × 7 km^2, 3.5 × 5.5 km^2 since August 2019, and improved signal-to-noise ratio compared to previous space-borne instruments, permits the detection of tropospheric pollution from small-scale emission sources and the estimation of very localized emissions from anthropogenic activities, such as industrial points sources, as well as regional fires. Lorente et al., 2019, have already reported updated emissions over the Paris metropolitan area using TROPOMI observations, while Ialogno et al., 2020, have assessed the capabilities of this instrument in evaluating city-wide air quality levels compared to the more traditional ground-based and in situ NO_2 monitoring methods.

In this work we use the publicly available TROPOMI offline v1.2 and v1.3 for March-April 2019 and for March-April 2020 tropospheric NO_2 data accessed via the Copernicus Open Data Access Hub[6]. The algorithm producing

[4] https://www.civilprotection.gr/en
[5] http://www.tropomi.eu/
[6] https://scihub.copernicus.eu/

https://doi.org/10.5194/acp-2020-600
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this data is described by van Geffen et al. (2019); it is based on the approach used for processing OMI/Aura NO_2 data within DOMINO and the FP7 Quality Assurance for Essential Climate Variables, QA4ECV[7], projects (Boersma et al, 2011; 2018). Routine validation is being carried out by the Validation Data Analysis Facility[8], VDAF, who also provide quarterly the Validation Report of the Copernicus Sentinel-5 Precursor Operational Data Products, ROCRV. Near-real-time validation can also be accessed via the S5P Mission Performance Center Data Validation Server[9]. Here we note that, within the ROCVC #06, reported on the 30th of March 2020, the TROPOMI tropospheric NO_2 columns show a negative bias of roughly -30% compared to global ground-based columnar data, mostly over background regions. Since in this work the relative differences between different time periods will be examined, absolute differences to standard instruments do not affect our findings, as the stability of the TROPOMI datasets is assured.

For the purposes of this work, orbital files over Greece, between 19° and 30°E and 34° and 42°N, were gridded onto a 0.10x0.05° grid using the Atmospheric Toolbox®[10] for different temporal scenarios discussed below. The data have been filtered, as recommended, using the quality flag indicator ≥ 75 assuring the data under this flag is restricted to cloud-free (cloud radiance fraction < 0.5) and snow-ice free observations. An example figure is presented in Section 3.1 (Figure 2) where the major NOx emitting sectors around Greece are prominent, the capital city Athens and the second largest city Thessaloniki, in the North, as well as emissions by one of the two largest thermal power plants, in Ptolemaida, also in the North. As in the following we will further discuss the effect on shipping emissions in the Aegean Sea, in the major shipping track that emerges from the Strait of Bosporus in Istanbul moving SW towards Athens before turning Westwards towards the Mediterranean Sea, the domain also shows emissions from known locations in the Turkish Asia Minor coast, both cities and power plants.

For details on the algorithm, as well as the suggested data usage, we refer the reader to the official TROPOMI Algorithm Theoretical Basis Document[11], the Product User Manual[12] and the Product Readme File[13].

2.2. LOTOS-EUROS CTM simulations.

The open source chemical transport model LOTOS—EUROS[14] v2.2.001 is used for the purposes of this study to simulate NO_2 columns over the Greek domain for March and April for years 2019 and 2020. The CTM model is originally aimed at air pollution studies and simulates gases (O_3, NOx, SO_2 etc) as well as aerosols (sulfate, nitrate, PM10, PM2.5 etc.) in the troposphere. The gas-phase chemistry of the model is a modified version of CBM-IV (Gery et al., 1989). The Isorropia II module (Fountoukis and Nenes, 2007) is used for the aerosol chemistry, while further information on the model and its activity can be found in Manders et al., 2017. LOTOS-EUROS is the national air quality model for the Netherlands (Vlemmix et al., 2015), and has been used for distinct studies as well to investigate NO_2 values (Timmermans et al., 2011; Curier et al., 2012; 2014). LOTOS-EUROS also participates at the operational Copernicus Atmosphere Monitoring Service (CAMS) consisting one of the seven CTMs that provide the official ensemble air quality forecasting service[15].

Vlemmix et al., 2015, compared LOTOS-EUROS NO_2 tropospheric columns with MAX-DOAS measurements and found a good agreement between the two datasets, with a correlation coefficient between the daily averaged columns equal to 0.72. Schaap et al., 2013, compared the LOTOS-EUROS NO_2 simulations with OMI/Aura retrievals. They also showed that the model captures the NO_2 spatial distribution satisfactorily and is able to explain 91% of the OMI signal variation across Europe, while the systematic difference was attributed to the summer period. Detailed evaluation of the LOTOS-EUROS NO_2 simulations over Greece have showed that, compared to in situ concentration measurements, the correlation ranges between 0.42 to 0.55 for the different air quality stations studied, while compared to MAXDOAS columns the correlation spans between 0.41 and 0.55 for the urban load and between 0.58 and 0.64 for comparisons over remote directions (Skoulidou et al., 2020.)

In this study the domain spans from 19° to 30° East and 34° to 42° North with a grid resolution of 0.1°×0.05° (longitude × latitude), as seen in Figure 1, where an example model NO_2 column output for March 2020 at 12:00

7 http://www.qa4ecv.eu/
8 http://mpc-vdaf.tropomi.eu/
9 https://mpc-vdaf-server.tropomi.eu/
10 https://atmospherictoolbox.org/
11 http://www.tropomi.eu/document/atbd-nitrogen-dioxide
12 http://www.tropomi.eu/document/product-user-manual-nitrogen-dioxide-0
13 http://www.tropomi.eu/document/product-readme-file-nitrogen-dioxide
14 https://lotos-euros.tno.nl/
15 https://www.regional.atmosphere.copernicus.eu/

https://doi.org/10.5194/acp-2020-600
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UTC is shown. The locations of expected emission sources [urban areas, power plants, road transport and shipping plumes] can be seen for the region. In the vertical, the model distinguishes 10 levels which extend from the surface to about 175hPa. The height of these levels refer to the levels of the ECMWF (European Centre for Medium-range Weather Forecasts) meteorological input data that are used to drive the model with an horizontal resolution of 7km×7km. The initial and boundary conditions are constrained from a coarser run of LOTOS-EUROS that is performed over the larger European domain (15° W to 45° E and 30° – 60° N) with a resolution of 0.25°×0.25°. The anthropogenic emissions come from the CAMS-REG (CAMS Regional European emissions) inventory for the year 2015 with a horizontal resolution of 0.1°×0.05° (Kuenen et al., 2014).

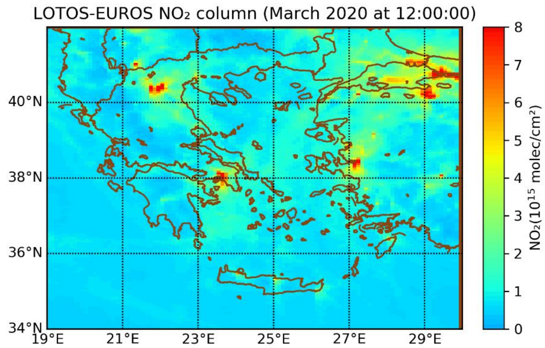

Figure 1. Mean LOTOS-EUROS NO_2 tropospheric column in March 2020, at 12Q00 UTC, the approximate overpass time of TROPOMI over the region.

2.3. Comparative Methodology.

While it would make sense to simply compare the NO_2 levels over Greece for the two periods, assuming that the emission sources have not changed dramatically between 2019 and 2020, one should not discard the effects that various meteorological parameters have on NO_2 levels. Meteorological conditions, such as wind speeds, temperature inversions and the depth of the boundary layer, often play pivotal roles in local air quality levels (Jacob and Winner, 2009). The ambient levels of secondary NO_2 pollution are determined through the accumulation or dispersion of pollutants, low or high solar irradiances, regional transport of clear or polluted air and atmospheric chemistry for the formation of secondary species, in this case via the chemical coupling of NO_x with O_3 (for e.g. Seo et al., 2017).

To ensure that the observed decrease in NO_2 levels was not due to diverse meteorological conditions between one year and the next, relative differences on NO_2 columns provided by the LOTOS-EUROS model are calculated and their average magnitude is set as the expected contribution by the different meteorology. This forms a standard level above which we expect COVID-19 related, i.e. emission-related, reductions. The premise of this thinking is as follows: differences in the satellite observations will contain the intertwined effect of differences in meteorology on concentrations and of differences in emissions. For the model we keep the emissions constant for the two periods but use the meteorology of 2019 and 2020 so that we can isolate the impact of meteorology on concentrations. We cannot of course exclude the possibility that the LOTOS-EUROS model has biases in the resulting NO_2 column depending on the meteorological conditions, for example due to uncertainties in mixing under stable conditions, however we expect those to smear out for the spatiotemporal scales studied.

In this point we should stress that the satellite observations are more often than not gap-ridden, since in the suggested screening all but clear-skies remain. Spring-time months are rainy months, even for typically sunny Greece, which means that a one-to-one comparison of the satellite observations for the two periods, even on a weekly basis, is usually impossible. During our analysis it was found that, for e.g. the last week of March of 2020, the first week of full lockdown, was fully cloudy for the Northern Greece even though the equivalent week in March 2019 was all sunny. As a result, weekly comparisons were only possible for the major NO_2 hotspot over Greece, the city of Athens, while the rest of the domain was examined on a monthly basis.

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In technical terms, the LOTOS-EUROS simulations were performed as discussed in Section 2.2 but for the averaging were restricted, on a daily basis, to the TROPOMI pixels that actually provided an observation. Even though a direct comparison of the CTM results to the satellite data is not the focus of this paper, we imposed this filter to make sure that the same days with the same meteorological conditions were viewed by both methods. Similarly, since we compare the relative changes of the two datasets, and not one against the other, we did not apply the TROPOMI averaging kernels to the CTM profiles, as we would do if we were to directly compare the two.

3. Results

In the following section, we first show the effect on monthly NO_2 levels over the entire domain, the six Greek cities with the largest number of inhabitants, and on shipping tracks in the Aegean Sea. We then present a more in depth analysis, on a weekly basis, for the city of Athens.

3.1. Lockdown effects on monthly NO_2 levels

In Figure 2 the monthly mean tropospheric NO_2 levels over Greece, the Northern neighboring countries, the Aegean Sea as well as the coast of Turkey and the Istanbul area, are shown for year 2019 in the left and year 2020 in the right, with the month of March presented in the upper panel and the month of April in the lower panel. Even though the hotspots appear strong for year 2019, with discrete shipping tracks and ground-tracks over Turkey showing clearly, the different meteorological conditions between March and April obviously affect both the location of the maxima as well as the absolute level of those maxima. Since Greece entered full lockdown mode within the first three weeks of March, while Turkey imposed intermittent movement restrictions from the beginning of April onwards, the NO_2 hotspot around the megacity of Istanbul and the Bosporus Strait is still pronounced in March 2020 [upper left] while in Greece most of the smaller urban emission points are missing, and Athens is shown in sharp decline. In April 2020, the Turkish hotspots are also reduced in magnitude, as expected. In the following sections we focus on specific hotspot locations and introduce numerical findings.

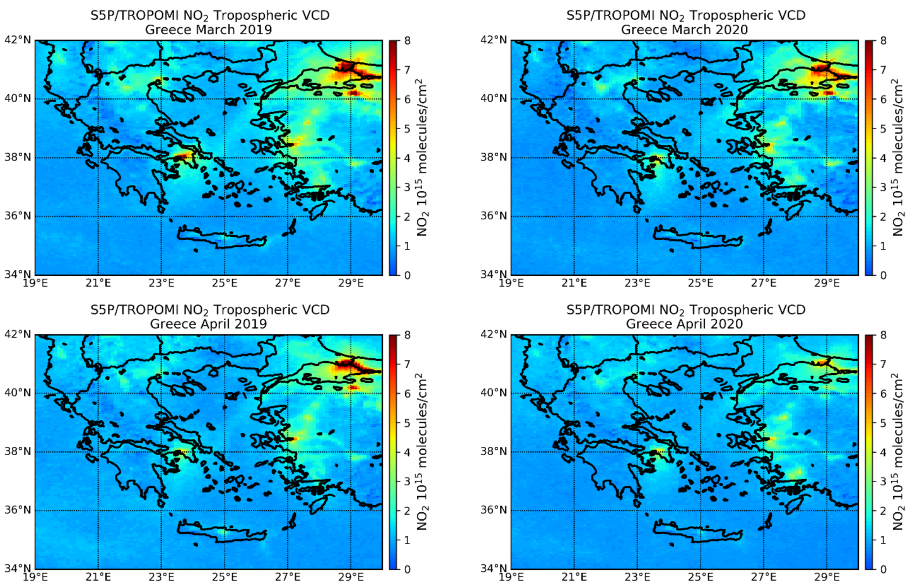

Figure 2. Monthly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, for March [upper] and April [lower] for the 2019 [left] and 2020 [right].

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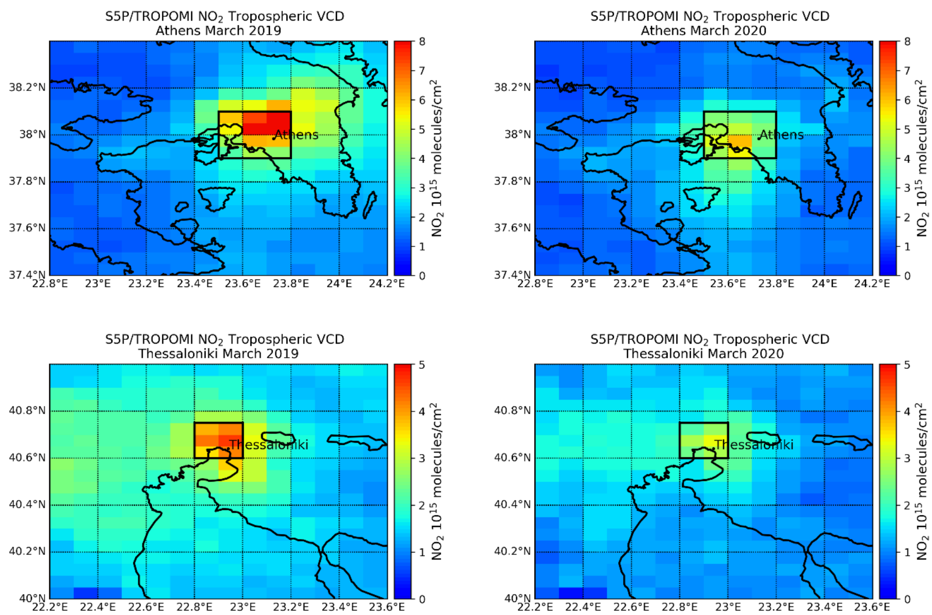
Table 1. Geographical distribution and population of the six main cities discussed in this work, as per the 2011 Greek Census.

City	Region of	Latitude	Longitude	Population
Athens	Attica	37.98° N	23.72° E	3.218.218
Thessaloniki	Central Macedonia	40.64° N	22.94° E	789.191
Larisa	Thessaly	39.63° N	22.41° E	144.651
Volos	Thessaly	39.36° N	22.95° E	118.707
Patra	Western Greece	38.24° N	21.73° E	168.202
Heralkio	Crete	35.33° N	25.14° E	153.653

In Figure 3 the monthly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, are depicted for March 2019 [left] and 2020 [right] for six major cities in Greece top to bottom, namely, Athens [37.98° N, 23.72° E], Thessaloniki [40.64° N, 22.94° E], Larisa [39.63° N, 22.41° E], Volos [39.36° N, 22.95° E], Patra [38.24° N, 21.73° E] and Heraklio [35.33° N, 25.14° E]. We focus on the locations where the major transport emissions are expected. These six cities, according to the HAS 2011 census[16], host 4.45 out of the 10.8 million of the Greek population (Table 1). Even though the NO_2 levels are low over the four smaller cities, we were interested in examining the ability of TROPOMI in sensing both the load and expected changes for these, relatively clean, cities [numeric results are given in Table 2].



[16] https://www.statistics.gr/2011-census-pop-hous

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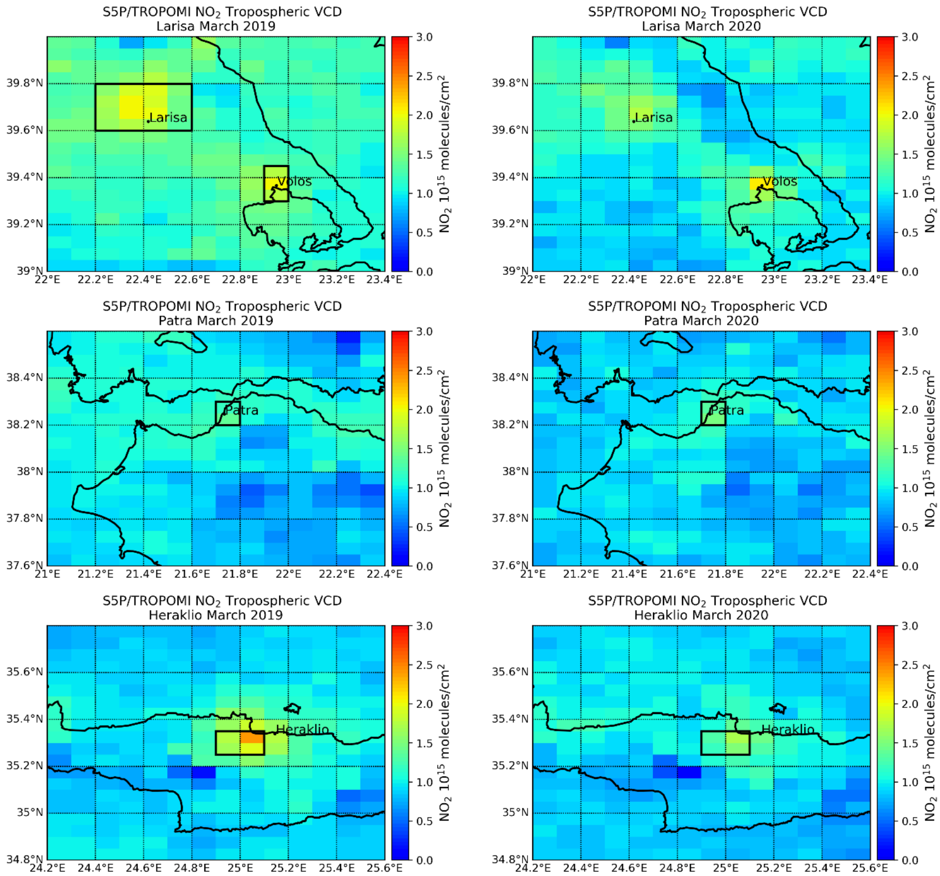

Figure 3. Monthly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, for March 2019 [left] and March 2020 [right] for the five major cities in Greece. First row, Athens; second, Thessaloniki; third, Larisa and Volos; fourth, Patras and fifth, Heraklio. The boxes mark the pixels used in the numerical analysis.

In Figure 4 the monthly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, for March [upper] and April [lower] for the 2019 [blue] and 2020 [orange] for the six major cities in Greece, from left to right, Athens, Thessaloniki, Larisa, Volos, Patra and Heraklio. Overall, the NO_2 levels are proportional to the city population, with Athens and Thessaloniki showing the highest levels while the remaining four present similar conditions. In Table 2, the full statistics that relate to Figure 4 are given, where it can firstly be noted that for the month of March, the relative differences in NO_2 loading sensed by the satellite sensor between 2019 and 2020 range from -3 to -34%, whereas for the month of April from -1% to -27%, apart from the case of Volos which shows an increase of 5%. In general, as was already seen in Figure 3, the NO_2 levels are higher in all cases for both March months, than the equivalent April ones. Recall as well that, due to the cloudiness conditions, the representability of the months (effective day) is not the same for all cities.

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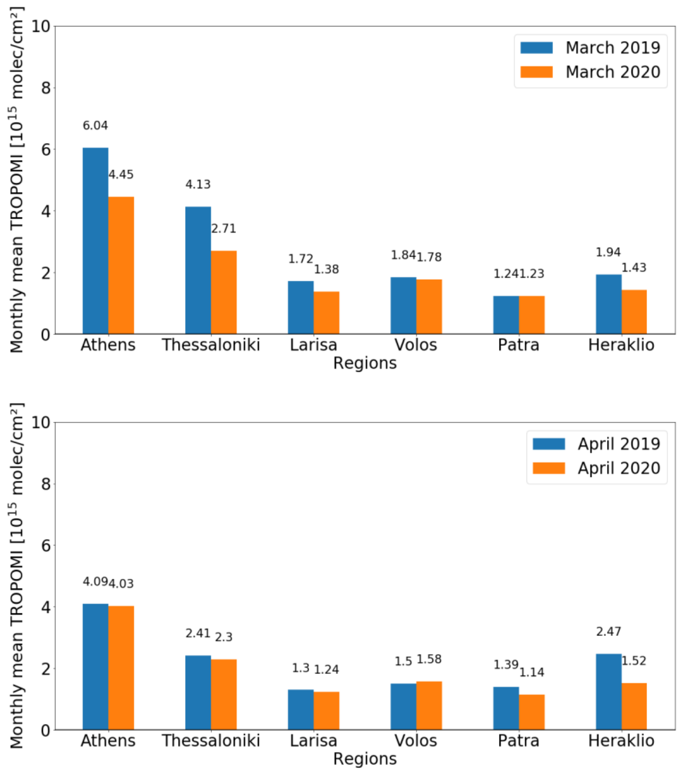

Figure 4. Monthly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, for March [upper] and April [lower] for the 2019 [blue] and 2020 [orange] for the five major cities in Greece, from left to right, Athens, Thessaloniki, Larisa, Volos, Patra and Heraklio.

Table 2. Monthly mean TROPOMI NO_2 levels [10^{15} molecules/cm^2] over major cities in Greece for March [left block] and April [right block] for year 2019 and 2020 and their relative difference, standard error and number of pixels [in brackets].

Location	**03.2019**	**03.2020**	**% diff**	**04.2019**	**04. 2020**	**% diff**
Athens [12]	6.04±0.48	4.45±0.22	-26%	4.09±0.14	4.03±0.11	-1%
Thessaloniki [6]	4.13±0.14	2.71±0.16	-34%	2.41±0.09	2.30±0.11	-5%
Larisa [6]	1.72±0.06	1.38±0.04	-19%	1.30±0.03	1.24±0.02	-5%
Volos [3]	1.84±0.08	1.78±0.12	-3%	1.50±0.08	1.58±0.11	+5%
Patra [2]	1.24±0.07	1.23±0.06	-	1.39±0.01	1.14±0.01	-18%
Heralkio [4]	2.18±0.14	1.61±0.10	-26%	2.24±0.15	1.64±0.07	-27%

As expected, the *in situ* air quality monitoring stations around Athens showed this decrease in a more pronounced way. Measurements reported in the European Environmental Agency Air Quality monitoring service[17] were downloaded for stations that reported for both months of March and April 2019 and 2020, namely: two traffic urban stations (Aristotelous and Patision), two background urban stations (Marousi and Peristeri), an industrial urban station (Elefsina) and a background suburban station (Agia Paraskeui). The monthly mean surface concentration [μgr/m^3] for March [blue] and April [orange] 2019 and March [grey] and April [yellow] 2020 are

[17] https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm

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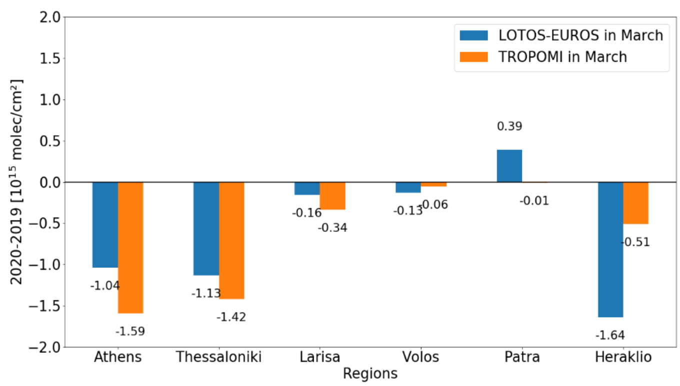
shown in Figure S1. The average decrease ranges between ~ -10 and -30% for the entire two month lockdown period, with the lowest variability observed for the case of Elefsina, a station which is mainly affected by the oil refineries and heavy industry. For Thessaloniki, a reduction on surface NO_2 concentrations between -11% and -58% between years 2019 and 2020 was reported by the local governing body of the Region of Central Macedonia for the period March 11th to April 20th by the five in situ air quality monitoring stations depending on the type of station, urban suburban and industrial [pers. comm.].

In Figure 5, upper, the monthly mean absolute differences in tropospheric NO_2 columns (10^{15} molecules/cm^2) between 2020 and 2019 are shown for TROPOMI [orange] and LOTOS-EUROS [blue] for the five major cities in Greece, for Athens, Thessaloniki, Larisa, Volos, Patra and Heraklio. We opted to show absolute differences here, and not percentage ones as might be expected, since a small relative change on a low NO_2 abundance would result in the erroneous image of a large reduction, as is shown for the case of the city of Patras [second to the right] which also has the lowest reported monthly concentrations, between 1.14±0.01 and 1.39±0.01x10^{15} molecules/cm^2 for the months shown. In the lower panel of Figure 5, the emission changes are quantified in the following manner: the percentage differences for LOTOS-EUROS between 2019 and 2020 are calculated, as the equivalent ones seen by TROPOMI. By subtracting the two percentage differences, and not directly comparing the two, the actual NO_2 reduction may be quantified. This is roughly be -12% for Athens, -9% for Thessaloniki, -12% for Larisa, near zero for Volos [as was already seen by the TROPOMI observations, Figure 3, third panel], -37% for Patra and unexpectedly positive, +15%, for Heraklio.



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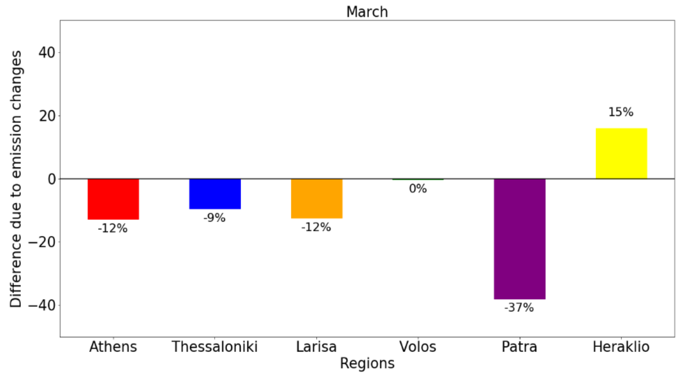
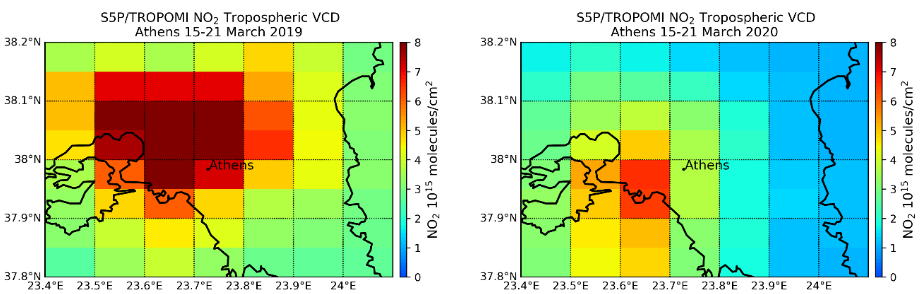


Figure 5. Upper. Monthly mean absolute differences in tropospheric NO_2 columns (10^{15} molecules/cm^2) between 2020 and 2019 are shown for TROPOMI [orange] and LOTOS-EUROS [blue] for the five major cities in Greece, from left to right, Athens, Thessaloniki, Larisa, Volos, Patra and Heraklio. Lower. The percentage differences attributed to emission changes.

Apart from the reduction in NOx emissions due to land transport restrictions, some decline is also expected in shipping emissions over the Aegean Sea routes. In Figure S2 the land is masked so as to reveal the shipping tracks between the Bosporus Strait and the port of Piraeus in Athens for March 2019 [left] and March 2020 [right.] Excluding the background values over the open sea, we calculate a monthly mean of 1.50x10^{15} molecules/cm^2 for 2019 compared to 1.32x10^{15} molecules/cm^2 for 2020, a -12% [-5% for April] reduction, well in line with the reductions found over land.

3.2. Lockdown effects on weekly NO_2 levels over Athens



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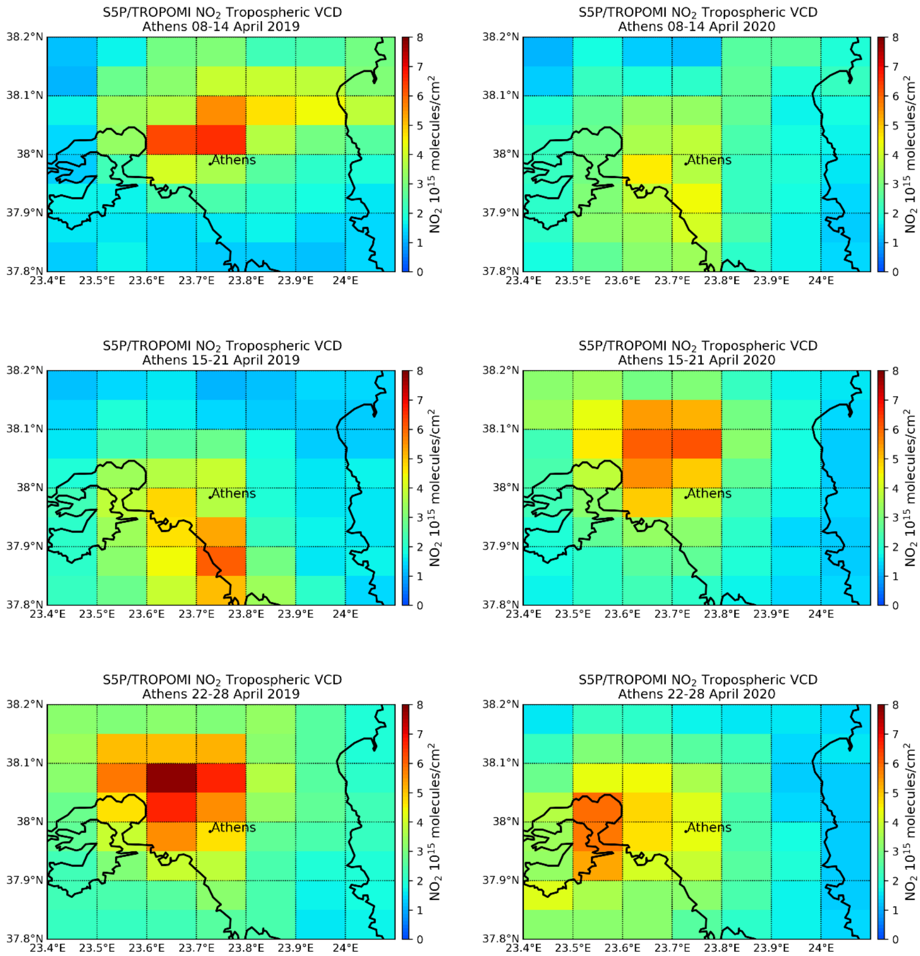

Figure 6. Weekly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, over Athens for 2019 [left] and 2020 [right]. First row, 15-21 March 2019; second, 8-14 April, third, 15-21 April and fourth, 22-28 April.

https://doi.org/10.5194/acp-2020-600
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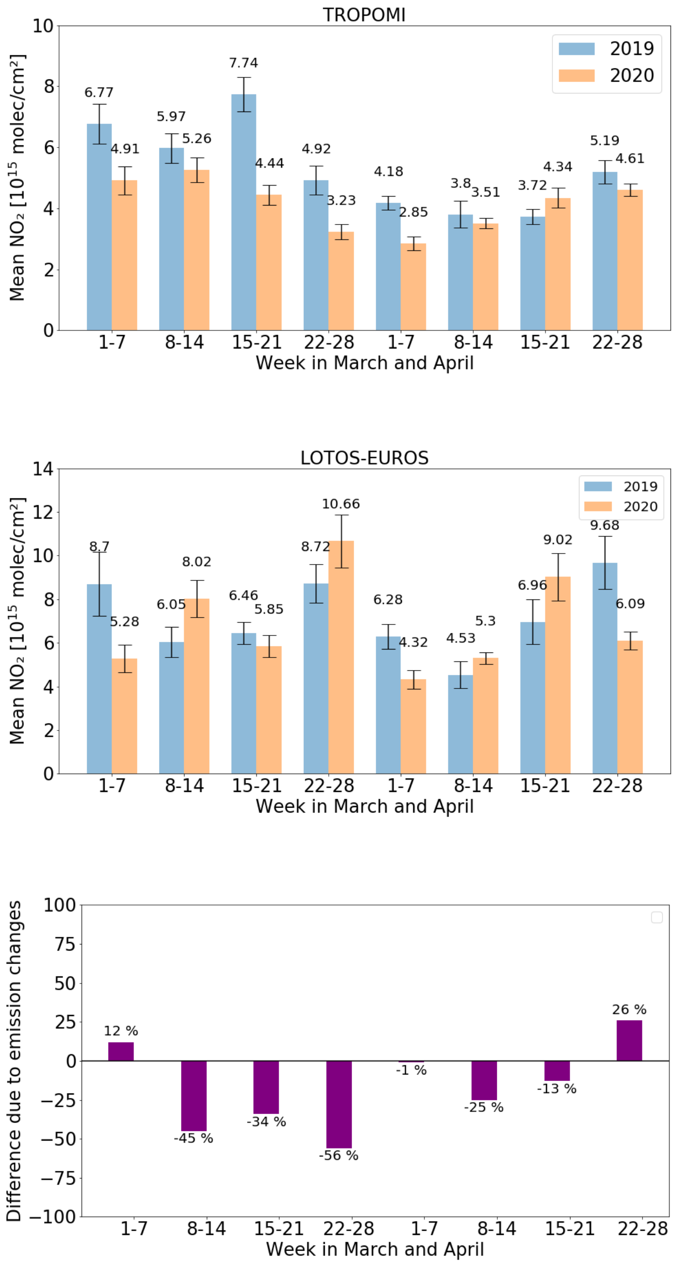

Figure 7. Upper. Weekly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, for weeks in the 2019 [blue] and 2020 [orange] for Athens. Middle. Weekly mean LOTOS-EUROS tropospheric NO_2 columns, in 10^{15} molecules/cm^2, for weeks in the 2019 [blue] and 2020 [orange] for Athens. Lower Lower. The percentage differences attributed to emission changes, revealing the actual magnitude of the NOx emissions decrease.

https://doi.org/10.5194/acp-2020-600
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Without disregarding the possible contribution of central heating to total NOx emissions, the largest decrease due to the COVID-19 lockdown is indeed observed over the main Greek hotspot, the city of Athens and its surrounding. In Figure 6, weekly mean TROPOMI tropospheric NO_2 columns, in 10^{15} molecules/cm^2, over Athens for 2019 [left] and 2020 [right] are shown for weeks 15-21 March (first row), 8-14 April (second row), 15-21 April (third row) and 22-28 April (fourth row). Apart from the obvious reduction in magnitude this year, what is most prominent in this composite is the effect of the winds for both the location of the local maximum as well as the spread of the pollution plume, which further strengthens our decision not to perform one-on-one comparisons between the different NO_2 fields. In numbers, the average weekly NO_2 load over Athens sensed by TROPOMI is presented in the upper panel of Figure 7 where the 2019 averages are shown in blue and the 2020 ones in orange for weeks of March and April. Out of the 12 pixels considered for this sub-domain, which may give up to 84 measurements for each week, for year 2019 an average of 53±16 [median of 52] S5P/TROPOMI observations where found whereas for year 2020 an average of 52±25 [median of 56]. Even though the representativeness of the weekly levels can by no means be considered equal between the years, apart from the penultimate week, TROPOMI reports lower NO_2 columns ranging between -8% to -43%. The contribution of the meteorological factors to these estimates can be assessed by the equivalent LOTOS-EUROS weekly averages, shown in the middle panel of Figure 7. As for Figure 5, bottom, the percentage difference of the LOTOS-EUROS simulations between 2019 and 2020 are calculated, as those for TROPOMI. The difference between those two relative differences is given in the lower panel of Figure 7. The fact that the CTM predicted an increase in NO_2 production for most weeks, under the assumption that the primary emissions remained stable between the two years, results in higher reduction levels ranging between -1% to -56%.

4. Conclusions

In this work, Sentinel-5P/TROPOMI tropospheric NO_2 observations were studied in order to examine the possible positive effect on Greek air quality caused the recent COVID-19 pandemic lockdown. The country enforced severe movement restrictions and entire economic sectors gradually were shut down, starting from the last weekend of February and gradually, activity per activity, reaching a total lockdown in effect from Monday 23rd up to May 4th. The time period between March and April 2020, and the equivalent weeks in 2019, were analyzed and compared for six, largest in population, cities in Greece, as well as the shipping lanes of the Aegean Sea. TROPOMI monthly mean tropospheric nitrogen dioxide, NO_2, observations showed a decrease of between -3% and -26% [-1% to -27%] with an average of -22% [-11%] for March and April 2020 respectively, compared to the previous year, for the urban areas and approximately -12% [-5%] for the shipping sector. For the capital city of Athens, weekly reductions, between -8% and -43%, for the seven of the eight weeks studied were found. Similar reductions were reported by six in situ air quality stations in Athens that reported measurements to the European Environmental Agency Air Quality database, with monthly decreases ranging between 0% to -47% and an average of -23%. In order to eliminate the expected meteorological effects on the observed NO_2 levels, Chemical Transport Modelling simulations, provided by the LOTOS-EUROS CTM, show that the magnitude of these satellite-sensed reductions cannot solely be attributed to the difference in meteorological factors affecting NO_2 levels during March and April 2020 and the equivalent time periods of the previous year. Taking this factor into account, the resulting decline due to the COVID-19 related measures was estimated to range between 0% and -37% for the five largest Greek cities, with an average of ~ -10%.

Author Contributions: The data analysis was performed by A.K., I.S. and M.E.K.; methodology and conceptualization by D.S. and I.P.; software development by I.S. and A.K.; writing—original draft preparation by M.E.K.; review and editing by D.S., A.S., A.M., J.v.G. and H.E. All authors have read and agreed to the published version of the manuscript.

Funding: This research has been co-financed by the European Union (European Regional Development Fund) and Greek national funds through the Operational Program "Competitiveness, Entrepreneurship and Innovation" (NSRF 2014-2020) by the "Panhellenic Infrastructure for Atmospheric Composition and Climate Change" project (MIS 5021516) and well as the

"Innovative system for Air Quality Monitoring and Forecasting" project [code T1EDK-01697, MIS 5031298), implemented under the Action "Reinforcement of the Research and Innovation" Infrastructure.

Acknowledgments: We acknowledge the usage of modified Copernicus Sentinel data 2019-2020. Results presented in this work have been produced using the Aristotle University of Thessaloniki (AUTh) High Performance Computing Infrastructure and Resources. M.E.K., I.S. and D.S. would like to acknowledge the support provided by the IT Center of the AUTh throughout the progress of this research work. M.E.K. and A.K. would also like to acknowledge the support provided by the Atmospheric Toolbox®.

Conflicts of Interest: The authors declare no conflict of interest.

Data availability: The S5P/TROPOMI data are publicly available via the Copernicus Open Data Access Hub, https://scihub.copernicus.eu/. The LOTOS-EUROS simulations are available upon request. The air quality monitoring station data are publicly available via the European Environmental Agency Air Quality monitoring service, https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm.

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