

## CLIMATE CHANGE

## Abrupt recent trend changes in atmospheric nitrogen dioxide over the Middle East

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Nitrogen oxides, released from fossil fuel use and other combustion processes, affect air quality and climate. From the mid-1990s onward, nitrogen dioxide (NO<sub>2</sub>) has been monitored from space, and since 2004 with relatively high spatial resolution by the Ozone Monitoring Instrument. Strong upward NO<sub>2</sub> trends have been observed over South and East Asia and the Middle East, in particular over major cities. We show, however, that a combination of air quality control and political factors, including economical crisis and armed conflict, has drastically altered the emission landscape of nitrogen oxides in the Middle East. Large changes, including trend reversals, have occurred since about 2010 that could not have been predicted and therefore are at odds with emission scenarios used in projections of air pollution and climate change in the early 21st century.

## INTRODUCTION

Nitrogen oxides (NO + NO<sub>2</sub> = NO<sub>x</sub>) play a central role in atmospheric chemistry, biogeochemical cycles, and the eutrophication of ecosystems (1–3). Through reaction products such as ozone and aerosol nitrate, anthropogenic NO<sub>x</sub> emissions also contribute to climate change (4). The total global NO<sub>x</sub> source is close to 50 Tg N/year, of which about 23% is natural, 58% is from fossil fuel combustion (predominantly from traffic and power generation) and the remainder is from agriculture, biomass burning, and biofuel use (4, 5). In the Middle East, natural NO<sub>x</sub> sources are minor and the emissions are dominated by fossil fuel combustion. It is well established that NO<sub>x</sub> is a key precursor in the formation of tropospheric ozone (O<sub>3</sub>). However, discrepancies were found between NO<sub>x</sub> trends in emission inventories, used in chemistry transport models to calculate tropospheric O<sub>3</sub>, and observed O<sub>3</sub> trends, because models tend to overestimate O<sub>3</sub> mixing ratios (6). One of the possible causes is that the inventories are based on fuel type and energy consumption reports to estimate both CO<sub>2</sub> and NO<sub>x</sub> sources, but that CO<sub>2</sub>-to-NO<sub>x</sub> emission ratios are not well characterized (7).

In recent years, spectrometric observations from satellites have been used to derive trends in tropospheric NO<sub>2</sub> (7–13). Strong upward NO<sub>2</sub> trends have been reported for the Middle East, including the Arabian Gulf and Eastern Mediterranean, coincident with very high levels of O<sub>3</sub>, especially in summer (14–20). Because anthropogenic NO<sub>x</sub> emissions are largely proportional to fossil energy consumption and traffic intensity, they have been linked to economic and industrial activities (12, 13). It was also shown that NO<sub>2</sub> trends are often related to a combination of economic development and emission controls aimed at improving air quality (7, 13, 21). Here, we analyze the causes of NO<sub>2</sub> trends observed over the Middle East and demonstrate the additional importance of other geopolitical factors.

We present NO<sub>2</sub> (and ancillary SO<sub>2</sub>) measurements performed with the Ozone Monitoring Instrument (OMI) (22), which is a Dutch-Finnish contribution to the Aura satellite program of the National Aeronautics and Space Administration (NASA). It observes backscattered solar radiation in the visible and ultraviolet spectral range, from which tropospheric vertical column densities (TVCDs) of NO<sub>2</sub> are derived at

about 13 × 24 km<sup>2</sup> spatial resolution (in the center of the swath). OMI views the Earth with a relatively wide swath to achieve global coverage in 14 orbits, flown within 1 day. Scientific operations started in October 2004, and we focus on the period 2005–2014. We investigate annual mean TVCDs and trends of NO<sub>2</sub> based on the DOMINO v2 satellite product (23, 24) (Materials and Methods). To help interpret observed NO<sub>2</sub> trends in the Middle East, we made use of the annual world development indicators of the World Bank and data from the U.S. Energy Information Administration (25, 26).

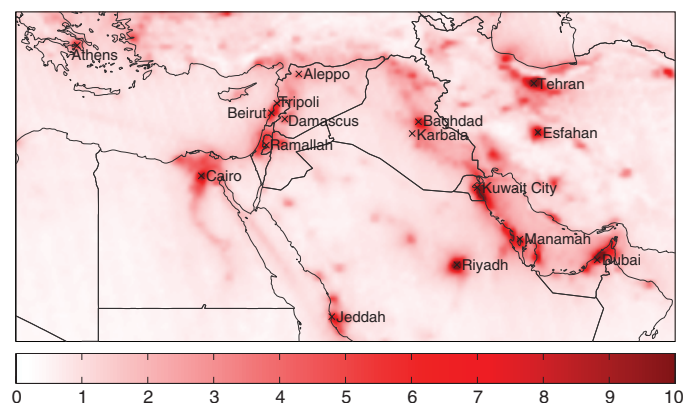
## RESULTS AND DISCUSSION

Figure 1 gives an overview of tropospheric NO<sub>2</sub> column densities over the region considered, between 20° and 40°N latitude and between 20° and 60°E longitude, home to about 350 million people at the crossroads between Europe, Asia, and Africa. The highest NO<sub>2</sub> is observed over Riyadh, Tehran and its surroundings, along the Arabian Gulf, and in Cairo and northern Egypt. Previous studies have derived significant upward NO<sub>2</sub> trends until about 2010 or 2011 over several cities in this region, for example, about 5 to 7%/year in Cairo, 2 to 8%/year in Tehran, 7 to 10%/year in Damascus, 10 to 20%/year in Baghdad, 4 to 5%/year in Jeddah, and 6 to 7%/year in Riyadh (17, 18).

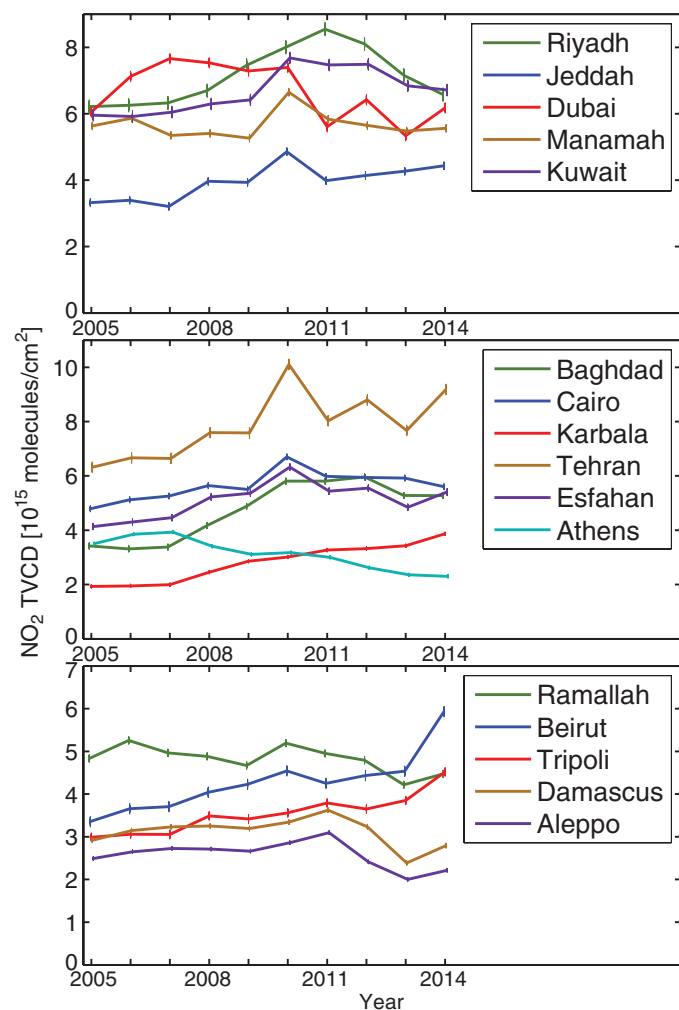
Over Greece and particularly in Athens, however, an overall negative trend of about –4%/year between 1996 and 2011 has been observed (18), which accelerated to –10%/year between 2008 and 2012 (12). In Fig. 2, we show the evolution of annual mean NO<sub>2</sub> column densities over Athens and 15 additional cities in the region between 2005 and 2014, averaging over areas of 100 × 100 km<sup>2</sup>, hence including the city surroundings. Figure S1 shows similar results over smaller areas of 30 × 30 km<sup>2</sup>, focusing more on the central parts of these cities. Both figures demonstrate that the trends and interannual variability are much larger than the standard error of the mean (SEM), indicating that they are significant, most distinct for the 100 × 100 km<sup>2</sup> footprint in Fig. 2. The NO<sub>2</sub> decline over Athens of about 40% since 2008 corroborates previous estimates. The latter likewise applies to the other cities mentioned above, that is, for the period up to 2010, for which strong upward trends have been reported. Figure 2 also reveals that since 2010, trend reversals have occurred over these particular cities. Because the atmospheric lifetime of NO<sub>x</sub> is less than a day in urban plumes, typically several hours in the Middle East, NO<sub>2</sub> over cities is closely related to local NO<sub>x</sub> emissions (27).

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**Fig. 1. Tropospheric NO<sub>2</sub> over the Middle East.** NO<sub>2</sub> column densities in 10<sup>15</sup> molecules/cm<sup>2</sup> observed by OMI, averaged over the period 2005–2014.



**Fig. 2. NO<sub>2</sub> column densities over 16 cities in the Middle East.** Annual mean NO<sub>2</sub> during 2005–2014, indicating NO<sub>x</sub> emission trend changes around 2010. Vertical bars represent the SEM.

On the basis of economic indicators and energy data, the gross domestic product (GDP) in Greece dropped by about 5%/year since 2008, whereas the total primary energy consumption and CO<sub>2</sub> emissions declined by 3 to 4%/year. The correspondence between the negative trend in NO<sub>2</sub> and the World Bank indicators supports the conclusion by Vrekoussis *et al.* (12) that the economic and financial crisis in Greece resulted in substantial reduction of pollution emissions. In the Arabian Gulf area, these relationships appear rather differently. In Saudi Arabia, Bahrain, Kuwait, and the United Arab Emirates (UAE), GDP, energy consumption, and CO<sub>2</sub> emissions increased approximately in parallel, on average 5 to 6%/year between 2005 and 2014. Between 2005 and 2010, NO<sub>2</sub> increased about 5%/year, whereas afterward, it decreased at approximately the same rate. Since about 2010, the positive correlations between economic indicators and NO<sub>x</sub> emissions have vanished. Although shifts of NO<sub>x</sub> relative to CO<sub>2</sub> emissions toward lower ratios have been inferred for East China recently and for North America and Europe in the past decades (7), the marked trend reversals in the Middle East are unique. Figure 3 illustrates the sign changes in NO<sub>2</sub> trends in the Arabian Gulf region and Saudi Arabia, in Iran and central Iraq, and the Eastern Mediterranean with Syria and Egypt.

In Saudi Arabia, positive trends of pollutants including NO<sub>2</sub> have been previously reported (28, 29). In 2008, the country committed to reduce its environmental footprint (30), leading to new legislation (31). In Kuwait, air quality standards have been implemented, for example, to reduce emissions from the petrochemical industry south of Kuwait City (32). In the UAE, a Cabinet Decree on air pollution was issued in 2006 (33), leading to a reduction of NO<sub>2</sub> in Dubai since 2008 (Fig. 2). Hence, our results provide a first indication that air quality control in the Arab Gulf States has become effectual. Nevertheless, along the Red Sea, for example, in Jeddah, the NO<sub>2</sub> tendency is less distinct. By considering NO<sub>2</sub> changes over Riyadh between 2005 and 2014 (fig. S2), we find relatively strong reductions in the wider city area, including the suburbs (Fig. 2), whereas in a smaller area around the city center, NO<sub>2</sub> has decreased less (fig. S1). Continued monitoring of NO<sub>2</sub> will show whether the recent downward trend persists and makes a sustainable contribution to the improvement of air quality.

An NO<sub>2</sub> trend reversal since 2010 is also observed over Cairo (Fig. 2), coincident with the government overthrow in early 2011. In Egypt, the GDP increased by about 6%/year between 2005 and 2010, which declined to 2%/year afterward. Energy consumption and CO<sub>2</sub> emissions increased by about 4%/year before and also after 2010. Hence, NO<sub>x</sub> emission reductions follow a decrease in GDP but seem unrelated to changes in the total energy use. Because no specific air quality control measures were implemented in 2011, we expect that vehicular emission reductions have played a role, for example, related to the decreased availability and increasing prices of petrol relative to income development. Hence, also in Egypt, CO<sub>2</sub>-to-NO<sub>x</sub> emission ratios changed suddenly after 2010, which was driven by political events.

Likewise, in Iran, NO<sub>2</sub> changes appear to be related to political developments, in this case on an international level, because the United Nations Security Council imposed sanctions in 2006, which were extended in 2010. The NO<sub>2</sub> data over the major cities Tehran and Esfahan indicate a rapid increase between 2005 and 2010 by more than 10%/year, unaffected by the sanctions in 2006, whereas this has turned around to about –4%/year since 2010. This development holds pace with changes in GDP, which increased by about 5%/year up to 2010, reduced to 3%/year in the following 2 years, and turned negative afterward (–6%/year). In

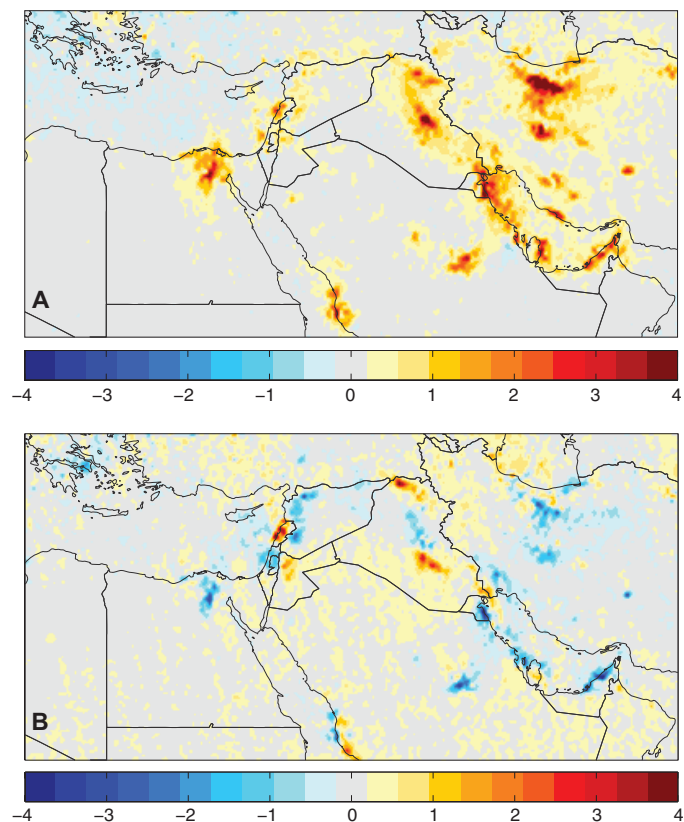
terms of energy use and CO<sub>2</sub> emissions, a growth of 5 to 6%/year occurred until 2010, which has dropped to 1 to 2%/year since then. This development also fits with a strong recent SO<sub>2</sub> emission reduction over the Gulf, likely from shipping, in particular near the main Iranian oil tanker terminal Jazireh Ye at Kharg Island (Fig. 4). This is explained by a large drop in oil export of about 50% since 2010. Figure 4 also in-

dicates an SO<sub>2</sub> trend reversal in emissions from the major Saudi Arabian oil processing facilities at Abqaiq. It will be interesting to keep track of changes after the sanctions against Iran are lifted in the near future.

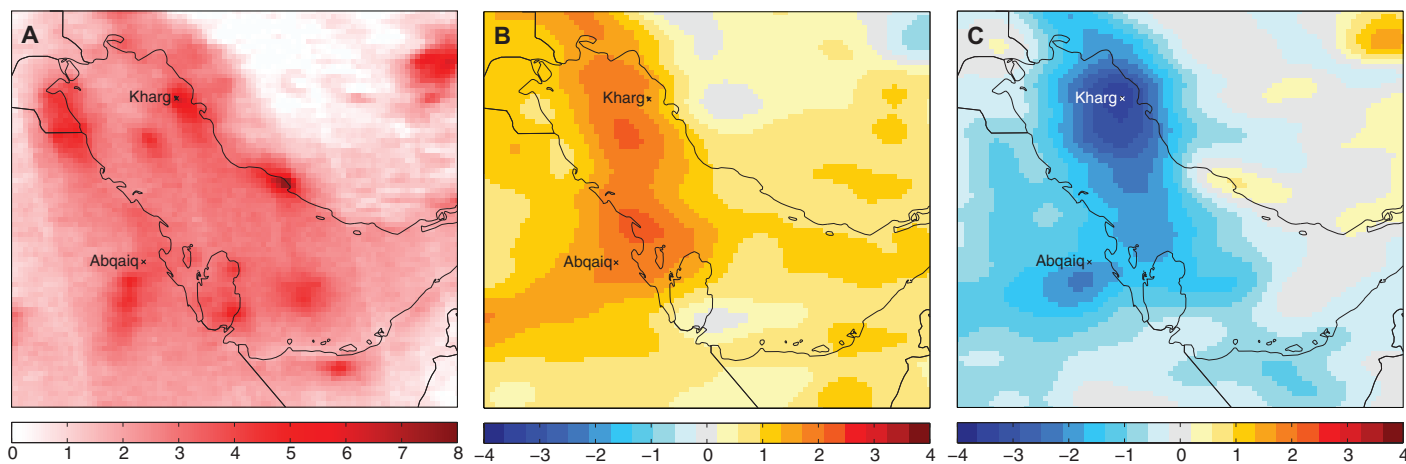
The invasion of Iraq in 2003 has caused dramatic changes in the country. However, since 2005, the GDP has increased again by 6 to 7%/year, accompanied by energy consumption and CO<sub>2</sub> emission increases of 4 to 5%/year. NO<sub>2</sub> over Baghdad increased more than 10%/year until 2011, after which a decline started at nearly the same rate (Fig. 2). This change was accompanied by a particularly rapid GDP increase in 2010 and 2011, after which it dropped by more than half. In Karbala, south of Baghdad, NO<sub>2</sub> increased more gradually by about 10%/year between 2005 and 2014. Figure 3 presents an overview of NO<sub>2</sub> changes in the periods 2005–2010 and 2010–2014, which illustrates that, overall, NO<sub>2</sub> increased substantially in Iraq, especially in Baghdad and the urban areas to the south, and in the north and northeast where the cities of Mosul and Kirkuk are located (see fig. S2 for the period 2005–2014). Since 2013, however, in Baghdad and central Iraq, NO<sub>2</sub> has decreased substantially, for example, including the cities Tikrit and Samarra that have been occupied by the so-called Islamic State. The armed conflict in this area has left marks, including a decrease in NO<sub>x</sub> emissions.

Large NO<sub>2</sub> changes are also found over the Levant. Over Israel, especially near Tel Aviv and Haifa (fig. S1), NO<sub>2</sub> decreased substantially in recent years, following the Clean Air Law of 2008 that entered into force in the early 2011, and consistent with a recent decrease in NO<sub>x</sub> emissions (34). GDP and energy consumption in Israel steadily increased over the 2005–2014 period by about 4%/year, whereas CO<sub>2</sub> emissions grew less rapidly by 2.5%/year. Hence, CO<sub>2</sub>-to-NO<sub>x</sub> emission ratios have also changed markedly in this area since 2010. In the Palestinian territories, developments have been more capricious, with average GDP increases of 4%/year, although as high as 15 to 20% in 2009 and 2011, then turning negative by more than -4%/year. Energy consumption has also been fluctuating, increasing by 25%/year from 2003 to 2007, then dropping to 6%/year and increasing again in recent years. CO<sub>2</sub> emissions have been similarly variable. NO<sub>2</sub> decreased by more than 20% since 2005, for example, in Ramallah (Fig. 2), and the largest changes occurred in recent years, coincident with decreasing GDP. The latter has been associated with economic restrictions in Area C of the West Bank (35).

Over large parts of Lebanon, for example, near Beirut and Tripoli, NO<sub>2</sub> increased by 3 to 4%/year between 2005 and 2013 (Fig. 2). GDP



**Fig. 3. NO<sub>2</sub> trend reversal in many locations in the Middle East. (A and B)** Tropospheric NO<sub>2</sub> column density changes in 10<sup>15</sup> molecules/cm<sup>2</sup> (A) between 2005 and 2010 and (B) between 2010 and 2014.



**Fig. 4. SO<sub>2</sub> trend reversal over the Arabian Gulf. (A)** SO<sub>2</sub> column densities in 10<sup>15</sup> molecules/cm<sup>2</sup> observed by OMI, averaged over the period 2005–2014. **(B and C)** SO<sub>2</sub> changes (B) between 2005 and 2010 and (C) between 2010 and 2014.



increased from 2005 to 2010 by more than 5%/year, after which it fell to 1 to 2%/year. Since 2011, energy use and CO<sub>2</sub> emissions have decreased, by 20% in 2012 (no data available for subsequent years). Then in 2014, a drastic NO<sub>2</sub> increase of 20 to 30% took place over Lebanon. In neighboring Syria, NO<sub>2</sub> over Damascus and Aleppo decreased by 40 to 50% since 2011, coincident with the uprising in spring of that year, which escalated into civil war. Although economic indicator data are not available for this period, the 4 million refugees from Syria, with 1.2 million fleeing to Lebanon (350,000 to Beirut) and 625,000 to Jordan (165,000 to Amman) (36), have had a strong impact on society, accompanied by large changes in NO<sub>x</sub> emissions.

## CONCLUSIONS

The overview of NO<sub>2</sub> variations during the past decade in Fig. 3, comparing the changes between 2005–2010 and 2010–2014, corroborates the large changes and trend reversals in the Middle East, a region troubled by crises. Other crisis regions, for example, in eastern Europe and Africa, are typically not associated with comparably large NO<sub>2</sub> changes. In the Middle East, NO<sub>2</sub> increased substantially in the 2005–2010 period, whereas it mostly declined afterward. Figure 3 also illustrates the shifts in NO<sub>x</sub> emissions that accompanied mass displacements of people in the regions of armed conflict in Syria and Iraq.

Evidently such relatively short-term changes cannot be captured by air pollution emission inventories and future projections, including the Representative Concentration Pathways (RCPs), used by the Intergovernmental Panel on Climate Change (4) [see Emissions of atmospheric Compounds and Compilation of Ancillary Data (ECCAD) at <http://eccad.sedoo.fr>]. For example, the RCP4.5 scenario assumes constant NO<sub>x</sub> emissions for the geographical region outlined by Fig. 1, whereas the RCP8.5 scenario assumes continual increases by 2%/year between 2005 and 2030, both deviating from reality.

Because ground-based air quality measurement networks have been established in only a few areas of the globe, atmospheric monitoring from space can help provide information to policy makers. The present analysis shows that it is feasible to link trends of atmospheric parameters to societal change. Unfortunately, the Middle East is not the only region in the world affected by economic recession and upheaval owing to war, although geopolitical changes appear to be more drastic than elsewhere. It is tragic that some of the observed recent negative NO<sub>2</sub> trends are associated with humanitarian catastrophes.

## MATERIALS AND METHODS

We used the Dutch OMI nitrogen oxide (DOMINO v2) product available at the European Space Agency (ESA) Tropospheric Emission Monitoring Internet Service (TEMIS; [www.temis.nl](http://www.temis.nl)) to retrieve tropospheric column densities of NO<sub>2</sub> (24). Annual means for the period 2005–2014 were calculated on the basis of monthly means to avoid biases due to seasonal variations considering the volume of available data. We applied a filter for an effective cloud fraction less than 30%, and we omitted the outermost 10 pixels on each side of the OMI swath because they have a substantially larger footprint compared to nadir (22). Metropolitan area averages have been calculated over 100 × 100 km<sup>2</sup> around the city center. Averages over smaller areas (30 × 30 km<sup>2</sup>) are shown in fig. S1. Error bars in Fig. 2 and fig. S1 are based on the monthly SEM, assuming Gaussian error propagation for the annual mean.

The analysis of SO<sub>2</sub> data is based on the retrieval algorithm described by Hörmann *et al.* (37). Slant column densities were retrieved using differential optical absorption spectroscopy (38) and converted into geometrical vertical column densities accounting for the OMI measurement geometry. Actual TVCDs can be systematically larger (up to a factor of 2) because much of the detected light is scattered above the boundary layer. However, this does not affect interannual variations and trends. Again, a cloud filter of 30% was applied to the daily data. For SO<sub>2</sub>, only summer months (June to October) were considered as the sensitivity increases when SO<sub>2</sub> reaches higher altitudes in the relatively deep boundary layer during these months. For the calculation of annual differences (Fig. 4, B and C), the SO<sub>2</sub> fields were smoothed by convolution with a two-dimensional Gaussian filter ( $\sigma = 0.5^\circ/0.2^\circ$  in longitude/latitude) to reduce noise and artefacts caused by the OMI “row anomaly” (24).

## SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/1/7/e1500498/DC1>

Fig. S1. NO<sub>2</sub> column densities over 18 cities in the Middle East.

Fig. S2. Tropospheric NO<sub>2</sub> column density changes in 10<sup>15</sup> molecules/cm<sup>2</sup> between 2005 and 2014.

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