

# A two-pollutant strategy for improving ozone and particulate air quality in China

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**Fine particulate matter (PM<sub>2.5</sub>) decreased by 30–40% across China during 2013–2017 in response to the governmental Clean Air Action. However, surface ozone pollution worsened over the same period. Model simulations have suggested that the increase in ozone could be driven by the decrease in PM<sub>2.5</sub>, because PM<sub>2.5</sub> scavenges hydroperoxy (HO<sub>2</sub>) and NO<sub>x</sub> radicals that would otherwise produce ozone. Here we show observational evidence for this effect with 2013–2018 summer data of hourly ozone and PM<sub>2.5</sub> concentrations from 106 sites in the North China Plain. The observations show suppression of ozone pollution at high PM<sub>2.5</sub> concentrations, consistent with a model simulation in which PM<sub>2.5</sub> scavenging of HO<sub>2</sub> and NO<sub>x</sub> depresses ozone concentrations by 25 ppb relative to PM<sub>2.5</sub>-free conditions. PM<sub>2.5</sub> chemistry makes ozone pollution less sensitive to NO<sub>x</sub> emission controls, emphasizing the need for controlling emissions of volatile organic compounds (VOCs), which so far have not decreased in China. The new 2018–2020 Clean Air Action plan calls for a 10% decrease in VOC emissions that should begin to reverse the long-term ozone increase even as PM<sub>2.5</sub> continues to decrease. Aggressive reduction of NO<sub>x</sub> and aromatic VOC emissions should be particularly effective for decreasing both PM<sub>2.5</sub> and ozone.**

China is confronting serious air pollution, with surface air concentrations of fine particulate matter (PM<sub>2.5</sub>) and ozone routinely exceeding air quality standards<sup>1–3</sup>. In 2013, the Chinese government introduced a 5 yr (2013–2017) plan for Clean Air Action<sup>4</sup>. The plan targeted PM<sub>2.5</sub> as the most urgent threat to public health<sup>5</sup>, and it principally focused emission controls on primary particles and sulfur dioxide (SO<sub>2</sub>) from coal combustion. As a result, PM<sub>2.5</sub> concentrations across urban areas of China decreased by 30–40% during 2013–2017 (ref. <sup>6</sup>). At the same time, however, surface ozone concentrations increased<sup>1,7</sup>. Annual mortality attributable to surface ozone pollution in China is at present over 50,000 deaths according to ref. <sup>3</sup> and 154,000–316,000 deaths according to ref. <sup>8</sup>. Ozone pollution is also detrimental to vegetation<sup>9</sup>, crop yields<sup>10</sup> and building materials<sup>11</sup>.

Surface ozone is produced rapidly in polluted air by photochemical oxidation of volatile organic compounds (VOCs) catalysed by nitrogen oxide radicals (NO<sub>x</sub> ≡ NO + NO<sub>2</sub>) and hydrogen oxide radicals (HO<sub>x</sub> ≡ OH + HO<sub>2</sub> + organic peroxy radicals (RO<sub>2</sub>)). VOCs and NO<sub>x</sub> are emitted mainly from fuel combustion and industrial sources<sup>12</sup>, and biogenic sources also contribute to VOC emissions<sup>13</sup>. HO<sub>x</sub> originates mainly from photochemical oxidation of water vapour and photolysis of carbonyls<sup>14</sup>, so ozone pollution is generally worst in summer. Summer ozone pollution is particularly severe over urbanized eastern China, with most sites from the Ministry of Ecology and Environment (MEE) network far exceeding the 8 h daily maximum (MDA8) air quality standard of 82 ppb (refs. <sup>1,13,15</sup>). The chemistry involved in ozone formation is highly nonlinear and may be limited by the supply of either NO<sub>x</sub> or VOCs, which has implications for which sources to control<sup>16,17</sup>. Satellite observations of the ratio of formaldehyde (HCHO) to nitrogen dioxide (NO<sub>2</sub>) in the atmospheric column suggest that urban regions of China are in

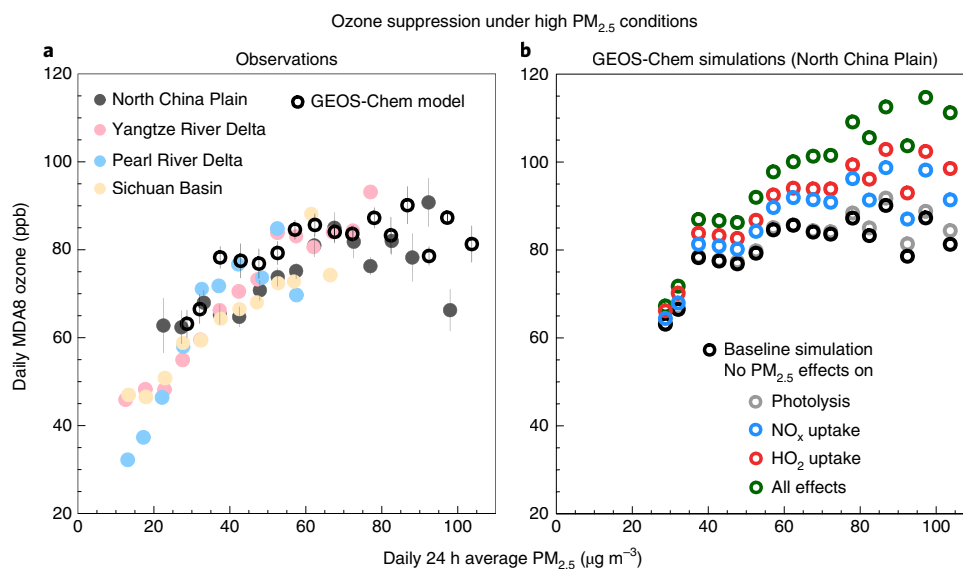
a transition regime between NO<sub>x</sub>-limited and VOC-limited, while rural areas are NO<sub>x</sub>-limited<sup>18</sup>. However, ozone production in urban centres is thought to be VOC-limited<sup>13</sup>.

In previous work<sup>15</sup>, we showed that the 2013–2017 ozone increase in China was of anthropogenic rather than meteorological origin. According to the Multiresolution Emission Inventory for China (MEIC)<sup>12</sup>, anthropogenic NO<sub>x</sub> emissions in China decreased by 20% during 2013–2017, while VOC emissions increased by 2%. These emission trends alone cannot explain the increase in ozone. Using simulations from the Goddard Earth Observing System Chemical Transport Model (GEOS-Chem), we suggested that the ozone increase could instead be driven by the PM<sub>2.5</sub> decrease, due to the role of PM<sub>2.5</sub> as scavenger of hydroperoxy (HO<sub>2</sub>) radicals that would otherwise react with nitric oxide (NO) to produce ozone. Here we demonstrate this ozone–PM<sub>2.5</sub> linkage with MEE network observations during pollution episodes, and we show that it affects whether ozone production is limited by the supply of NO<sub>x</sub> or VOCs. The results have important implications for developing two-pollutant control strategies to decrease both PM<sub>2.5</sub> and ozone in China.

## Observed ozone–PM<sub>2.5</sub> relationship

Figure 1a shows the ozone–PM<sub>2.5</sub> relationship in daily summer observations of MDA8 ozone and 24 h PM<sub>2.5</sub> concentrations at sites operated by the MEE during 2013–2018. We focus on the four megacity clusters targeted by the 2013–2017 Clean Air Action: North China Plain (106 sites), Yangtze River Delta (84 sites), Pearl River Delta (66 sites) and Sichuan Basin (71 sites). We averaged the daily MDA8 ozone and PM<sub>2.5</sub> data over all sites in a given cluster to obtain the daily time series, removed the 2013–2018 trends (Methods) to avoid the influence from the PM<sub>2.5</sub> concentration decrease over the period and binned the data into 5 μg m<sup>-3</sup> PM<sub>2.5</sub> increments.

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**Fig. 1 | Ozone- $PM_{2.5}$  relationship in China in summer.** **a**, Observed 2013–2018 relationships in daily data for MDA8 ozone and 24 h average  $PM_{2.5}$  concentrations. Values are regional averages over  $5 \mu\text{g m}^{-3}$   $PM_{2.5}$  bins (at least three data points required) for the monitoring sites operated by China's MEE in the four megacity clusters targeted by the Clean Air Action (see Methods for geographical definitions): North China Plain, Yangtze River Delta, Pearl River Delta and Sichuan Basin. Multi-year trends (2013–2018) of ozone and  $PM_{2.5}$  have been removed (Methods) to avoid obfuscating influence from the large  $PM_{2.5}$  decrease over the period (see Supplementary Fig. 5 for the relationships in the non-detrended data). GEOS-Chem simulation results for the North China Plain (2016–2017) are also shown. Error bars shown for the North China Plain are standard errors of ozone concentrations for each bin. **b**, GEOS-Chem baseline simulation for the North China Plain (as in **a**) and sensitivity simulations removing the effects of  $PM_{2.5}$  on photolysis rates,  $NO_x$  uptake and  $HO_2$  uptake, first separately and then together.

We see in Fig. 1a a generally positive correlation between daily MDA8 ozone and  $PM_{2.5}$ , which is consistent across the four megacity clusters. This positive correlation is typical of summer observations in polluted regions<sup>19,20</sup>. It can be understood as reflecting common dependences of ozone and  $PM_{2.5}$  on meteorology, manifested in particular by strong correlations of both with temperature and wind speed<sup>6,15</sup>. The North China Plain has particularly high  $PM_{2.5}$ , frequently exceeding  $60 \mu\text{g m}^{-3}$ . There we find that ozone levels off at about 80 ppb even as  $PM_{2.5}$  increases to about  $100 \mu\text{g m}^{-3}$ . As shown in Fig. 1a, a GEOS-Chem model simulation (Methods) conducted for 2016–2017 and sampled in the same way as the observations closely reproduces this behaviour.

The levelling off of ozone under highly polluted conditions might simply reflect the relatively short ozone lifetime against sinks from deposition and chemistry<sup>21</sup> or the self-suppression of ozone production as the  $NO/NO_2$  ratio shifts towards  $NO_2$  with increasing ozone. To isolate the role of  $PM_{2.5}$ , we conducted a GEOS-Chem sensitivity simulation removing  $PM_{2.5}$  effects on chemistry and photolysis (Fig. 1b). We find in that simulation that ozone does not level off until it reaches about 110 ppb. For polluted conditions with  $PM_{2.5}$  exceeding  $80 \mu\text{g m}^{-3}$ , we see from Fig. 1b that  $PM_{2.5}$  effects are responsible for about a 25 ppb suppression of ozone relative to  $PM_{2.5}$ -free conditions. This is a much larger effect than previously reported by ref. 15, which did not focus on pollution episodes.

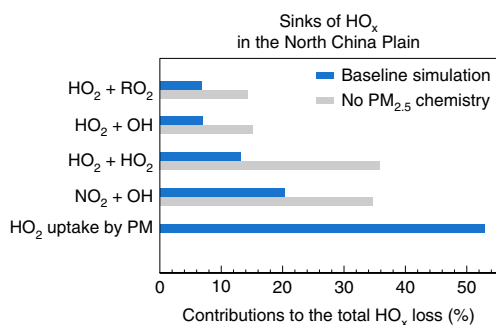
We conducted further sensitivity simulations in the model to isolate the contributions from different  $PM_{2.5}$  effects on ozone (Methods). For the days with  $PM_{2.5}$  exceeding  $80 \mu\text{g m}^{-3}$  in Fig. 1b, we find that not including individual  $PM_{2.5}$  effects on photolysis rates,  $NO_x$  uptake and  $HO_2$  uptake increases ozone on average by 2 ppb, 9 ppb and 15 ppb, respectively. The small effect from photolysis is consistent with previous model simulations<sup>22,23</sup>. Changes in atmospheric dynamics through aerosol–radiation interactions are not considered in our work but increase ozone by less than 2 ppb in the aforementioned model simulations<sup>22</sup>.

The largest effect of  $PM_{2.5}$  on ozone is through  $HO_2$  uptake, as reported in our previous work<sup>15</sup>. This uptake provides a sink for  $HO_x$  radicals and hence suppresses ozone formation. There is considerable evidence in the literature for  $HO_2$  uptake by particles with a reactive uptake coefficient  $\gamma_{HO_2}$  in the range 0.1–1.0 (refs. 24–26). Our GEOS-Chem simulations use  $\gamma_{HO_2} = 0.2$  with hydrogen peroxide ( $H_2O_2$ ) as product, as recommended by ref. 24. Ref. 26 recommends  $\gamma_{HO_2} = 0.24$  based on their ensemble of measurements for aerosol samples collected at two mountain sites in eastern China, showing consistent  $HO_2$  uptake with no apparent dependence on  $PM_{2.5}$  composition. There is uncertainty as to whether the product of  $HO_2$  uptake is  $H_2O_2$  or  $H_2O$  (ref. 27), but this has little effect on our ozone simulation<sup>15</sup>.

The effect of  $NO_x$  uptake by  $PM_{2.5}$  involves aqueous-phase conversion of  $NO_2$ , nitrate radical ( $NO_3$ ) and dinitrogen pentoxide ( $N_2O_5$ ) to nitric acid ( $HNO_3$ ) (ref. 24). Here we updated the reactive uptake coefficients for these reactions in GEOS-Chem following ref. 28 (Methods). The effect of  $NO_x$  uptake on ozone is larger than previously reported by ref. 15 for 2013 conditions, and this is partly due to the 20% decrease in  $NO_x$  emissions since then, which has made ozone production more  $NO_x$  sensitive (Supplementary Fig. 1).

### **$PM_{2.5}$ impact on emission control strategies for ozone**

A critical issue in developing an emission control strategy for ozone pollution is to understand the relative benefits of  $NO_x$  and VOC emission controls. This is generally framed in terms of whether ozone production is  $NO_x$ -limited or VOC-limited, which depends on the dominant  $HO_x$  loss pathways<sup>29</sup>. In standard gas-phase mechanisms, ozone production is  $NO_x$ -limited if the dominant  $HO_x$  sink is the self-reaction of peroxy radicals to produce peroxides, and VOC-limited if the dominant  $HO_x$  sink is the reaction of  $NO_2$  with OH to produce  $HNO_3$ . In urban areas of China, however, we find that the dominant sink is  $HO_2$  uptake by  $PM_{2.5}$ , as illustrated in Fig. 2 for the North China Plain. By analogy with gas-phase formation of peroxides, one might expect this sink to push ozone production



**Fig. 2 | HO<sub>x</sub> radical sinks in the North China Plain.** Relative contributions of different chemical pathways to the total sink of HO<sub>x</sub> radicals in the GEOS-Chem simulation for the boundary layer (surface to 1.3 km) over the North China Plain (114°–118° E, 34°–40° N). Results are summer 2017 averages from the baseline simulation and from a sensitivity simulation with no PM<sub>2.5</sub> chemistry (grey).

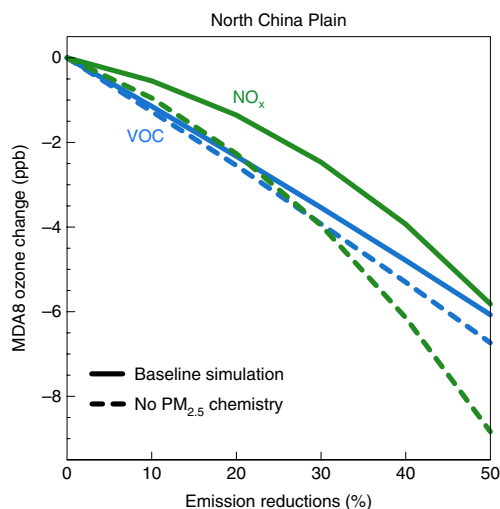
towards NO<sub>x</sub>-limited conditions. NO<sub>x</sub> uptake by PM<sub>2.5</sub> would do the same. However, uptake of HO<sub>2</sub> decreases the HO<sub>2</sub>/OH ratio and a lower supply of HO<sub>x</sub> radicals lengthens the lifetime of NO<sub>x</sub>, which causes the NO<sub>2</sub> + OH reaction to account for a greater contribution to the HO<sub>x</sub> sink<sup>30,31</sup>. Indeed, Fig. 2 shows a greater contribution of the NO<sub>2</sub> + OH reaction relative to gas-phase formation of peroxides (HO<sub>2</sub> + HO<sub>2</sub> and RO<sub>2</sub> + HO<sub>2</sub>) when PM<sub>2.5</sub> chemistry is included.

To clarify this issue, we conducted simulations with uniform 10%, 20%, 30%, 40% and 50% decreases of anthropogenic NO<sub>x</sub> or VOC emissions relative to 2017 values with and without PM<sub>2.5</sub> chemistry. Figure 3 shows the response for mean MDA8 ozone concentrations over the North China Plain. PM<sub>2.5</sub> chemistry weakens the ozone response to decreases of either NO<sub>x</sub> or VOC emissions because of the lower concentrations of HO<sub>x</sub> radicals. The effect is much larger for NO<sub>x</sub> than for VOCs, because HO<sub>2</sub> uptake drives a net increase in NO<sub>x</sub> concentrations as a result of lower OH concentrations and hence slower conversion to HNO<sub>3</sub> by the NO<sub>2</sub> + OH reaction (Supplementary Fig. 2). As a result, ozone production becomes more strongly VOC-limited (Fig. 3). The shift to more VOC-limited conditions because of HO<sub>2</sub> uptake by PM<sub>2.5</sub> also holds for the other megacity clusters, even though their PM<sub>2.5</sub> concentrations are lower (Supplementary Fig. 3).

Chemical indicators are often used in observations and models to diagnose whether ozone production is NO<sub>x</sub>-limited or VOC-limited<sup>32</sup>. A frequently used indicator is the H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> concentration ratio<sup>32,33</sup>, which tracks the prevailing HO<sub>x</sub> sink. But this can be misleading if PM<sub>2.5</sub> is the major HO<sub>x</sub> sink, as is the case here, particularly if the product of PM<sub>2.5</sub> uptake is H<sub>2</sub>O<sub>2</sub>. In that case, PM<sub>2.5</sub> chemistry causes the H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> ratio to increase (Supplementary Fig. 4), whereas ozone production in fact becomes more VOC-limited. The alternative indicators of the HCHO/NO<sub>2</sub> column concentration ratio, derived from satellite data<sup>18,34</sup>, and the ozone/NO<sub>y</sub> concentration ratio<sup>35</sup> (NO<sub>y</sub> referring to the sum of NO<sub>x</sub> and its oxidation products) are more robust, as shown in Supplementary Fig. 4.

### Towards a joint ozone–PM<sub>2.5</sub> air pollution control strategy

The effect of PM<sub>2.5</sub> chemistry on ozone production has important implications for a coordinated emission control strategy to decrease both PM<sub>2.5</sub> and ozone. There is strong impetus for continuing to decrease PM<sub>2.5</sub> levels in China, as present-day levels still greatly exceed the air quality standard<sup>1,6</sup>. PM<sub>2.5</sub> decreases over the 2013–2017 period were driven mostly by emission controls on SO<sub>2</sub> and primary particles from coal combustion<sup>12</sup>. Data for the North China Plain indicate that organic and nitrate components of PM<sub>2.5</sub> are now more important than sulfate<sup>36–38</sup>. As leverage from decreasing SO<sub>2</sub> emissions diminishes, decreasing VOC and NO<sub>x</sub> emissions may

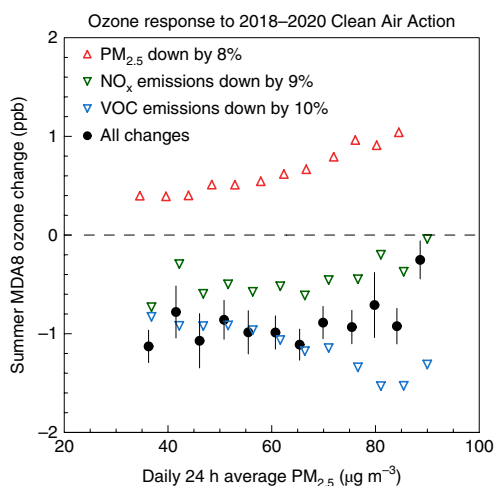


**Fig. 3 | Response of mean summer MDA8 surface ozone concentrations in the North China Plain to reductions in NO<sub>x</sub> and VOC emissions relative to 2017 levels.** Results are from the baseline simulation including PM<sub>2.5</sub> chemistry (solid lines) and from a sensitivity simulation without PM<sub>2.5</sub> chemistry (dashed lines).

become relatively more effective for decreasing PM<sub>2.5</sub>. Decreasing VOC and NO<sub>x</sub> emissions would also have benefits for ozone to offset the ozone penalty from decreasing PM<sub>2.5</sub>.

The recent 2018–2020 Clean Air Action plan calls for an 18% national decrease in PM<sub>2.5</sub> concentrations in 2020 relative to 2015, a 15% decrease in NO<sub>x</sub> and SO<sub>2</sub> emissions and a 10% decrease in VOC emissions<sup>39</sup>. Taking into account the already realized 2015–2017 decreases in NO<sub>x</sub> and SO<sub>2</sub> emissions<sup>12</sup> and in PM<sub>2.5</sub> concentrations<sup>6</sup>, the remaining reduction target for 2018–2020 is 9% for NO<sub>x</sub> emissions, 10% for VOC emissions and 8% for PM<sub>2.5</sub> concentrations. We applied the GEOS-Chem simulation for 2017 to examine the effects of these reductions, first separately and then together, on ozone pollution in the North China Plain. Results in Fig. 4 show that further decreases of PM<sub>2.5</sub> alone cause continuing ozone increases but that this is offset by the planned decreases of NO<sub>x</sub> and VOC emissions, resulting in a small overall net benefit for ozone of about 1 ppb. This ozone benefit is similar for relatively clean (low PM<sub>2.5</sub>) and highly polluted (high PM<sub>2.5</sub>) conditions. Although the ozone penalty from decreasing PM<sub>2.5</sub> is largest under highly polluted conditions, the benefit from decreasing VOC emissions is also largest under those conditions. Larger VOC reductions than the 10% planned under the 2018–2020 Clean Air Action would provide larger ozone benefits, as illustrated in Fig. 3.

NO<sub>x</sub> and VOC emission controls improve not only ozone but also PM<sub>2.5</sub>. Nitrate and organic components contribute 20% and 30%, respectively, of present-day winter PM<sub>2.5</sub> composition in the North China Plain<sup>37,38</sup>. Nitrate PM<sub>2.5</sub> formation in China is generally limited by the supply of NO<sub>x</sub> (refs. 40,41), so a 9% decrease in NO<sub>x</sub> emissions by 2020 would decrease PM<sub>2.5</sub> on average by about 1.8%. VOC reactivity for ozone formation in China is at present dominated by aromatics<sup>42</sup>, whose emissions have increased rapidly over the past decade relative to other VOCs<sup>43</sup>. Aromatics are also a large source of organic PM<sub>2.5</sub> (refs. 44,45). Assuming that 40% of organic PM<sub>2.5</sub> is secondary, that is, produced from the oxidation of VOCs<sup>45,46</sup>, and that this component scales with VOC emissions<sup>47</sup>, a 10% decrease in VOC emissions would decrease PM<sub>2.5</sub> by 1.2%. In this simple calculation, the 2018–2020 decreases of NO<sub>x</sub> and VOC emissions planned under the Clean Air Action would result in a 3% decrease in PM<sub>2.5</sub>, contributing substantially to the 8% PM<sub>2.5</sub> reduction goal while also improving ozone air quality. Further work should go beyond this



**Fig. 4 | Expected ozone concentration change in the North China Plain under the 2018–2020 Clean Air Action plan.** The figure shows mean MDA8 ozone responses in  $5 \mu\text{g m}^{-3}$  bins for the different changes projected by the 2018–2020 Clean Air Action plan relative to the 2017 GEOS-Chem baseline simulation. The changes are applied separately and then together ('All changes'). Values are regional averages for the North China Plain. The model data are sampled in the same way as in Fig. 1. Vertical bars for the 'All changes' simulation are the standard error on mean ozone for each bin.

simple estimate by examining  $\text{PM}_{2.5}$  sensitivity to individual VOCs and the linearity of the response. Future research should also investigate the role of biogenic VOCs, which interact with the formation of anthropogenic  $\text{PM}_{2.5}$  (refs. 48–50), and whose emissions would be sensitive to changes in land use and climate<sup>51</sup>.

In summary, we have shown through analysis of observed ozone– $\text{PM}_{2.5}$  relationships in China and supporting model simulations that ozone production is suppressed under high  $\text{PM}_{2.5}$  conditions ( $\text{PM}_{2.5} > 60 \mu\text{g m}^{-3}$ ) because of  $\text{PM}_{2.5}$  chemistry involving reactive uptake of both  $\text{HO}_x$  and  $\text{NO}_x$  radicals. Ozone concentrations in the North China Plain are reduced by about 25 ppb relative to  $\text{PM}_{2.5}$ -free conditions when  $\text{PM}_{2.5}$  exceeds  $80 \mu\text{g m}^{-3}$ .  $\text{PM}_{2.5}$  chemistry results in ozone production that is more VOC-limited than it otherwise would be, emphasizing the importance of controlling reactive VOC emissions to abate ozone pollution in China. The 2018–2020 Clean Air Action plan of the Chinese government calls for a 10% decrease in VOC emissions; we find that this should offset the effect on ozone of the continued decrease in  $\text{PM}_{2.5}$  concentrations, reversing the trend of increasing ozone. More aggressive VOC emission controls would further decrease ozone concentrations. With organic and nitrate  $\text{PM}_{2.5}$  now contributing a large share of total  $\text{PM}_{2.5}$ , controlling VOC and  $\text{NO}_x$  emissions would substantially improve both  $\text{PM}_{2.5}$  and ozone air quality.

### Online content

Any methods, additional references, Nature Research reporting summaries, source data, statements of code and data availability and associated accession codes are available at <https://doi.org/10.1038/s41561-019-0464-x>.

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## Author contributions

K.L. and D.J.J. conceived the study. K.L. performed the analysis. H.L., J.Z., V.S., L.S. and S.Z. contributed to interpreting the data. V.S. and K.H.B. helped with model simulations. Q.Z. provided emission data. K.L. wrote the draft of the paper with D.J.J. and H.L. All authors contributed to discussing and improving the paper.

## Competing interests

The authors declare no competing interests.

## Additional information

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## Methods

**Observations.** Hourly surface air ozone and PM<sub>2.5</sub> concentrations for summer (June–August) 2013–2018 were obtained from the public website of the MEE. We focused on the four megacity clusters targeted by the Clean Air Action: North China Plain (34°–40°N, 114°–118°E, 106 sites), Yangtze River Delta (30°–33°N, 119°–122°E, 84 sites), Pearl River Delta (21.5°–24°N, 112°–115.5°E, 66 sites) and Sichuan Basin (28.5°–31.5°N, 103.5°–107°E, 71 sites). See Supplementary Fig. 1 for the geographical domains. We calculated daily MDA8 ozone and 24 h PM<sub>2.5</sub> concentrations at individual sites and averaged them over the megacity clusters.

Rigorous analysis of the observed ozone–PM<sub>2.5</sub> relationships in terms of ozone suppression under high PM<sub>2.5</sub> conditions (Fig. 1a) must correct for the effect of the 2013–2018 trends driven by changes in emissions<sup>6,12,15</sup>, because these emission-driven trends (decreases in PM<sub>2.5</sub>, increases in ozone) would otherwise produce a negative relationship between the two. We have argued previously that the 2013–2018 increase in ozone is driven mostly by the decrease in PM<sub>2.5</sub>, but other factors, including trends in NO<sub>x</sub> emissions and meteorology, could also play a role<sup>15</sup>. Here we detrended the time series for ozone and PM<sub>2.5</sub> by removing the ordinary linear regressions of daily concentrations versus time for each megacity cluster over the 2013–2018 period and then adding back the 2013–2018 mean concentrations. The data without detrending show an even stronger suppression of ozone under high-PM<sub>2.5</sub> conditions (Supplementary Fig. 5).

**Chemical transport model simulations.** GEOS-Chem simulations for 2016–2017 over the East Asia domain (including all of China and adjacent regions) were conducted with the nested-grid version of the model (version 12.0.0; <http://www.geos-chem.org/>). The model configuration is the same as in ref. <sup>15</sup> except for minor updates to HO<sub>2</sub> and NO<sub>x</sub> uptake by PM (see the following paragraph). The model includes detailed ozone–NO<sub>x</sub>–VOC–PM–halogen tropospheric chemistry<sup>52,53</sup> and is driven by meteorological data from the NASA Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2) (<https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2>). The model's horizontal resolution is 0.5°×0.625°, following the grid of the MERRA-2 data. Chemical boundary conditions at the edges of the East Asia domain are updated every 3 h from a 4°×5° global simulation. The GEOS-Chem simulation of surface ozone in China has been evaluated in a number of previous studies<sup>15,54–56</sup>. In ref. <sup>15</sup>, we showed that the model can reproduce the observed ozone distribution across China in summer with no notable bias.

PM<sub>2.5</sub> affects ozone in GEOS-Chem by perturbing the radiation field (and hence photolysis rates) and by providing surfaces for reactive uptake. Effects on photolysis rates are computed with the Fast-JX radiative transfer code of ref. <sup>57</sup> as implemented in GEOS-Chem by refs. <sup>58,59</sup>. The HO<sub>2</sub> reactive uptake coefficient ( $\gamma_{\text{HO}_2}$ ) in GEOS-Chem is 0.2, producing either H<sub>2</sub>O or H<sub>2</sub>O<sub>2</sub> (refs. <sup>24,27</sup>). Here we set H<sub>2</sub>O<sub>2</sub> as a product, following refs. <sup>24,33</sup>, unlike ref. <sup>15</sup>, which set H<sub>2</sub>O as a product, but the choice of product has no notable effect on the ozone simulation<sup>15</sup>. Reactive uptake coefficients for N<sub>2</sub>O<sub>5</sub>, NO<sub>2</sub> and NO<sub>3</sub> are updated from GEOS-Chem version 12.0.0 on the basis of recent work by refs. <sup>28,60</sup>. This includes the parameterization of  $\gamma_{\text{N}_2\text{O}_5}$  for converting N<sub>2</sub>O<sub>5</sub> to HNO<sub>3</sub> on sulfate–nitrate–ammonium PM<sub>2.5</sub> (ref. <sup>61</sup>) and organic PM<sub>2.5</sub> (ref. <sup>62</sup>),  $\gamma_{\text{NO}_2} = 1 \times 10^{-5}$  for conversion of NO<sub>2</sub> to HONO and HNO<sub>3</sub> (which yields a good simulation of HONO/NO<sub>2</sub> concentration ratios in China<sup>60</sup>) and  $\gamma_{\text{NO}_3} = 1 \times 10^{-3}$  for conversion of NO<sub>3</sub> to HNO<sub>3</sub> (ref. <sup>24</sup>).

Anthropogenic emissions are from MEIC (<http://www.meicmodel.org/>)<sup>12</sup> and from MIX Asian emission inventory<sup>63</sup> for other Asian countries. Open fire emissions are from the Global Fire Emissions Database version 4 (ref. <sup>64</sup>). Natural emissions include NO<sub>x</sub> from lightning<sup>65</sup> and soil<sup>66</sup>, and biogenic emissions of VOC from vegetation calculated according to the Model of Emissions of Gases and Aerosols from Nature version 2 (ref. <sup>57</sup>).

A number of GEOS-Chem sensitivity simulations were carried out for this work, as listed in Supplementary Table 1 and described below. All reported results are after a one-month initialization.

**Ozone–PM<sub>2.5</sub> relationship.** We conducted sensitivity simulations removing the effects of PM<sub>2.5</sub>-induced changes in photolysis rates, reactive uptake of HO<sub>2</sub> and reactive uptake of NO<sub>x</sub> (as N<sub>2</sub>O<sub>5</sub>, NO<sub>2</sub> and NO<sub>3</sub>). Each was first removed separately, and then all were removed concurrently. The changes in PM<sub>2.5</sub> were applied only to the boundary layer below 1.3 km (ref. <sup>67</sup>) to avoid changes in the ozone background.

**Sensitivity to NO<sub>x</sub> and VOC emissions.** We conducted sensitivity simulations for 2017 with uniform 10%, 20%, 30%, 40% and 50% decreases of anthropogenic NO<sub>x</sub> or VOC emissions relative to the baseline simulation. We repeated the same ensemble of simulations without PM<sub>2.5</sub> effects below 1.3 km altitude.

**Effectiveness of 2018–2020 Clean Air Action plan.** We conducted sensitivity simulations relative to the 2017 baseline simulation by applying a uniform 9% decrease of anthropogenic NO<sub>x</sub> emissions over China, a 10% decrease of anthropogenic VOC emissions and an 8% decrease of PM<sub>2.5</sub> concentrations, first separately and then together.

## Data availability

The surface measurements for PM<sub>2.5</sub> and ozone from China's Ministry of Ecology and Environment can be downloaded from [beijingair.sinaapp.com](http://beijingair.sinaapp.com). The anthropogenic emission inventory is available from [www.meicmodel.org](http://www.meicmodel.org). The MERRA-2 reanalysis data are from <https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2>. The GEOS-Chem simulation results are available from the corresponding authors on request.

## Code availability

The GEOS-Chem model code is open source (<https://doi.org/10.5281/zenodo.2658178>). Code for calculation and data processing is available from the corresponding authors on request.

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