Supplement of

Examining the competing effects of contemporary land management vs. land cover changes on global air quality

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Text S1. Here we provide our reasoning for considering changes in displacement height \( (d) \) as negligibly small compared to changes in roughness length \( (z_o) \). Equation S1 demonstrates how aerodynamic resistance \( (R_a) \) is typically calculated in land surface exchange scheme:

\[
R_a = \frac{1}{\kappa u^*} \left( \frac{z - d}{z_0} - \Psi \left( \frac{z - d}{L} \right) + \Psi \left( \frac{z_0}{L} \right) \right) \tag{S1}
\]

where \( \kappa \) is von Kármán constant, \( u^* \) is friction velocity \( (m \ s^{-1}) \), \( L \) is Obukhov Length \( (L \) ), \( z \) is altitude, \( z_o \) is roughness length, and \( d \) is displacement height \( (m) \) (Foken, 2006). Since the middle of the first vertical grid of GEOS-Chem \( (z) \) is around 60 – 70 meters (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_vertical_grids), which is significantly larger than \( d \) such that \( z - d \approx z \) under most conditions, the changes in \( d \) are expected to be less important than the changes in \( z_0 \).

Text S2. The population-weighted averaged changes surface \( \text{O}_3 \) (ppb) or \( \text{PM}_{2.5} \) (\( \mu \text{g} \ \text{m}^{-3} \)) \( (\Delta [X]_{\text{pop_weighted}}, Y) \) for region \( Y \) is calculated as follow:

\[
\Delta [X]_{\text{pop_weighted}, Y} = \frac{\sum_{\text{gridcells in } Y} \Delta [X]_{i, \text{gridcells in } Y} \cdot \text{Pop}_i}{\sum_{\text{gridcells in } Y} \text{Pop}_i} \tag{S2}
\]

where \( \Delta [X]_i \) is changes in surface concentration of concerned chemical species, and \( \text{Pop}_i \) is the population count for individual gridcell \( i \). The global gridded population is from the fourth version of The Gridded Population of the World (GPWv4) (CIESIN, 2018), and remapped to match the resolution of GEOS-Chem output.

**Fig. S1** Modelled annual surface concentration of \( \text{SO}_4^{2-} \), \( \text{NO}_3^- \) and \( \text{NH}_4^+ \) aerosol for 2014. Filled circles indicate measured annual means for 2014 compiled from Air Quality System (AQS) of Environmental Protection Department (EPD) for United States (US), National Atmospheric Chemistry (NAtChem) database for Canada, European Monitoring and Evaluation Programme (EMEP) for Europe, Acid Deposition Monitoring Network in East Asia (EANET) for eastern Asia, and Geng et al. (2017) for China. The data from AQS, NAtChem, EMEP and EANET.
**Fig. S2** Comparison between mean modelled and observed surface ozone compiled by Sofen et al. (2016) at 2014. MAE and MB represents the mean absolute error and mean bias, respectively.

**Figure S3.** Regionally changes (2014 – 1992) in agricultural and non-agricultural NH$_3$ emissions from CEDS.
**Figure S4.** LULCC-induced changes in annual mean dry deposition ($v_d$) velocity (cm s$^{-1}$) and flux ($F_d$) (molec cm$^{-2}$ s$^{-1}$) in of NO$_2$ and SO$_2$.

**Figure S5.** Change in annual mean PM$_{2.5}$ (in μg m$^{-3}$) due to a) LULCC and changes in agricultural emissions at 1992 anthropogenic emissions background (simulation 5 – simulation 1), and b) anthropogenic emission changes (including agricultural emissions) (simulation 4 – simulation 5).
**Figure S6.** Annual mean surface HNO$_3$/H$_2$O$_2$ ratio under 1992 and 2014 anthropogenic emission background.

**Figure S7.** Change in annual mean surface O$_3$ (in ppbv) due to a) LULCC and changes in agricultural emissions at 1992 anthropogenic emissions background (simulation 5 – simulation 1), and b) anthropogenic emissions changes (including agricultural emissions) (simulation 4 – simulation 5).
Figure S8. Contribution of different pathways (wet vs dry, reduced (NH$_x$) vs oxidized (NO$_y$)) to the changes in total nitrogen deposition. NH$_x$ ≡ NH$_3$ + NH$_4$ and NO$_y$ ≡ NO + NO$_2$ + HONO + organic nitrates + aerosol nitrate.

Figure S9. Grid cells (denoted in red) with annual mean NO$_2$ concentration exceeding 0.05 and 0.6 ppb.
### Table S1. Comparison between modelled and observed annual average surface sulphate, nitrate and ammonium aerosol mass (in μg m\(^{-3}\)) over different region/observational network. Measurements are compiled from Air Quality System (AQS) of Environmental Protection Department (EPD) for United States (US), National Atmospheric Chemistry (NAtChem) database for Canada, European Monitoring and Evaluation Programme (EMEP) for Europe, Acid Deposition Monitoring Network in East Asia (EANET) for eastern Asia, and Geng et al. (2017) for China. The data from AQS, NAtChem, EMEP and EANET are collect in 2014.

| Region       | Species | Mod | Obs  | Mod/Obs |
|--------------|---------|-----|------|---------|
| US           | SO\(_4^{2-}\) | 0.88 | 1.27 | 0.70    |
|              | NO\(_3^-\)  | 0.77 | 0.93 | 0.83    |
|              | NH\(_4^+\)  | 0.54 | 0.69 | 0.79    |
| Canada       | SO\(_4^{2-}\) | 0.78 | 0.92 | 0.85    |
|              | NO\(_3^-\)  | 0.84 | 0.44 | 1.92    |
|              | NH\(_4^+\)  | 0.52 | 0.36 | 1.46    |
| Europe       | SO\(_4^{2-}\) | 1.84 | 2.08 | 0.88    |
|              | NO\(_3^-\)  | 1.67 | 1.51 | 1.10    |
|              | NH\(_4^+\)  | 1.16 | 1.11 | 1.05    |
| China        | SO\(_4^{2-}\) | 8.53 | 18.93| 0.45    |
|              | NO\(_3^-\)  | 6.16 | 10.15| 0.61    |
|              | NH\(_4^+\)  | 4.83 | 7.61 | 0.64    |
| EANET        | SO\(_4^{2-}\) | 2.33 | 3.64 | 0.64    |
|              | NO\(_3^-\)  | 1.09 | 1.31 | 0.84    |
|              | NH\(_4^+\)  | 1.15 | 1.01 | 1.14    |

| Region                  | Countries included                                                                 |
|-------------------------|--------------------------------------------------------------------------------------|
| Former Soviet Union (FSU) | Armenia, Azerbaijan, Belarus, Georgia, Kazakhstan, Kyrgyzstan, Moldova, Russia, Tajikistan, Turkmenistan, Ukraine, Uzbekistan |
| Western Europe (WEU)     | Austria, Belgium, Switzerland, Germany, Denmark, Spain, Finland, France, United Kingdom, Greece, Ireland, Iceland, Italy, Luxembourg, Netherlands, Norway, Portugal, Sweden |
| Central and eastern Europe (CEU) | Albania, Bulgaria, Bosnia and Herzegovina, Cyprus, Czechia, Estonia, Croatia, Hungary, Kosovo, Lithuania, Latvia, Montenegro, Poland, Romania, Serbia, Slovakia, Slovenia |
| China                   | China                                                                                 |
| South Asia (SAs)         | Afghanistan, Bangladesh, Bhutan, India, Sri Lanka, Nepal, Pakistan                    |
| Middle East (ME)         | United Arab Emirates, Egypt, Iran, Iraq, Israel, Kuwait, Lebanon, Oman, Qatar, Saudi Arabia, Syria, Turkey, West Bank, Yemen |
| Southeast Asia (SEA)     | Brunei, Indonesia, Cambodia, Laos, Myanmar, Malaysia, Philippines, Thailand, Vietnam  |
Table S2. Definition of regions.

| Region† | ΔPM$_{2.5}$ (LULCC+agr_emis,1992) | ΔO$_3$ (LULCC+agr_emis,1992) |
|---------|----------------------------------|-----------------------------|
| FSU     | -0.69 (-1.95)                   | -                           |
| WEU     | -0.49 (-1.24)                   | -                           |
| CEU     | -2.03 (-2.22)                   | -                           |
| China   | +0.34 (+0.75)                   | +0.09 (-0.21)               |
| SAs     | +0.55 (+0.82)                   | +0.26 (+0.25)               |
| ME      | +0.20 (+0.33)                   | -                           |
| SEA     | +0.21 (+0.17)                   | +0.32 (+0.26)               |
| JK      | -0.08 (-0.23)                   | -                           |
| NAm     | +0.15 (+0.56)                   | -0.05 (-0.21)               |
| CAm     | +0.10 (+0.22)                   | -                           |
| WAf     | -                                | +0.20 (+0.30)               |
| EAf     | -                                | +0.16 (+0.27)               |
| Global  | -0.08 (+0.23)                   | +0.01 (+0.03)               |

Table S3. Changes in area averaged, and population-weighted (in parentheses), annual mean surface PM$_{2.5}$ (ΔPM$_{2.5}$(LULCC+agr_emis,1992), in μg m$^{-3}$) and O$_3$ (ΔO$_3$(LULCC+agr_emis,1992), in ppbv) concentrations due to combined effects of LULCC and agricultural emission evaluated under 1992 anthropogenic emission background. Results only from regions with area- or population-weighted average ΔPM$_{2.5}$(LULCC+agr_emis,1992) > 0.2 μg m$^{-3}$ or ΔO$_3$(LULCC+agr_emis,1992) > 0.2 ppbv are shown.

†The definitions and abbreviations of all regions can be found in Table S2.
Reference

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