



Enhanced land–sea warming contrast elevates aerosol pollution in a warmer world

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Many climate models simulate an increase in anthropogenic aerosol species in response to warming¹, particularly over the Northern Hemisphere mid-latitudes during June, July and August. Recently, it has been argued that this increase in anthropogenic aerosols can be linked to a decrease in wet removal associated with reduced precipitation², but the mechanisms remain uncertain. Here, using a state-of-the-art climate model (the Community Atmosphere Model version 5), we expand on this notion to demonstrate that the enhanced aerosol burden and hydrological changes are related to a robust climate change phenomenon—the land–sea warming contrast^{3,4}. Enhanced land warming is associated with continental reductions in lower-tropospheric humidity that drive decreases in low clouds—particularly large scale (stratus) clouds—which, in turn, lead to reduced large-scale precipitation and aerosol wet removal. Idealized model simulations further show that muting the land–sea warming contrast weakens these hydrological changes, thereby suppressing the aerosol increase. Moreover, idealized simulations that only feature land warming yield enhanced continental aridity and an increase in aerosol burden. Thus, unless anthropogenic emission reductions occur, our results add confidence that a warmer world will be associated with enhanced aerosol pollution.

Since the pre-industrial era, anthropogenic activities have resulted in a significant increase in anthropogenic aerosol burden⁵, which in turn has affected Earth's radiative balance. According to the Fifth Assessment Report of the IPCC, the total effective aerosol radiative forcing is -0.9 W m^{-2} (90% uncertainty range: -1.9 to -0.1 W m^{-2})⁶, indicating that aerosols cause a net cooling effect, which has probably offset $\sim 40\%$ of GHG warming⁷. Aerosols can also adversely affect air quality and human health, with a recent study attributing 3.3 million premature deaths each year to aggravated aerosol pollution, led by fine particulate matter ($\text{PM}_{2.5}$)⁸, particularly in heavily polluted areas such as India and China. Continued GHG-induced global warming is expected to be associated with changes in the physical, chemical and biological factors that control the lifetime, transport, chemistry and atmospheric burden of aerosols^{9,10}. Considering the climatic and societal importance of aerosol pollution, an improved understanding of how future climate change can influence the amount of aerosol is needed for climate and air pollution policy decisions.

Studies show a mixed aerosol response to GHG-induced warming, with some analyses yielding a decrease in aerosol burden, particularly SO_4 ^{11,12}. However, more recent studies show an increase in aerosols under warming^{12,13}. State-of-the-art Atmospheric Chemistry Climate Model Intercomparison Project (ACCMIP)¹⁴ models yield robust increases in most aerosol species, particularly

over the Northern Hemisphere mid-latitudes during summer, which are largely due to a decrease in wet removal from reductions in large-scale precipitation (LSP)². This results in a negative aerosol climate feedback, ranging from -0.21 to $-0.46 \text{ W m}^{-2} \text{ K}^{-1}$. Similarly, these same ACCMIP models yield a robust increase in $\text{PM}_{2.5}$ and, in turn, an increase in premature mortality¹⁵.

Uncertainty in the aerosol response to future warming is related to several factors, including uncertainty in the simulation of aerosol processes^{16,17}, including transport, removal and chemistry. Furthermore, models must accurately simulate how GHG-induced warming impacts the climate system and, in turn, how these warming perturbations affect the important physical processes controlling aerosol burden. However, some climate warming responses are more robust than others. The land–sea warming contrast (LSWC), where continents warm more than the ocean^{3,4,18,19}, is a robust feature found in both observations and climate model simulations. This phenomenon is caused by contrasts in surface sensible and latent fluxes over land³, land–ocean contrasts in boundary-layer lapse rate changes²⁰, boundary-layer relative humidity and associated low-level cloud cover changes over land²¹, and soil moisture reductions²². Thus, enhanced continental warming is associated with an increase in land aridity, which in turn may affect the burden of anthropogenic aerosols. Here, we demonstrate, using novel simulations, that the LSWC is a dominant driver of the anthropogenic aerosol increase under future warming.

Figure 1a,d,g,j shows that the Community Atmosphere Model version 5 (CAM5)²³ simulates a significant increase in all anthropogenic aerosol species in response to warming. Responses are estimated from the difference between a ten-year control simulation, based on the year 2000 climate and aerosol emissions, and a ten-year warming simulation, based on the year 2100 climate and year 2000 aerosol emissions (Methods). The significance of all of the responses was determined by Student's *t*-test for the difference of means, using the pooled variance. In all cases, the annual mean aerosol burden increases globally, with the maximum increase over the Northern Hemisphere mid-latitudes ($30\text{--}60^\circ \text{ N}$)—more specifically, over the Northern Hemisphere mid-latitude continents during summer (June through August (JJA)). For example, the annual mean sulphate (SO_4) burden increases 3.5% globally, 3.8% over the Northern Hemisphere mid-latitude land, and 9.8% over the Northern Hemisphere mid-latitude continents during JJA.

A similar sequence in decreasing wet deposition (Fig. 1b,e,h,k) is found from global annual to Northern Hemisphere mid-latitude continents during JJA. Wet deposition is the primary removal mechanism for atmospheric aerosols and soluble gases²⁴. Hence, the increase in burden is consistent with a reduction in wet deposition, and moreover, the maximum increase in aerosol burden over Northern Hemisphere mid-latitude continents during JJA is

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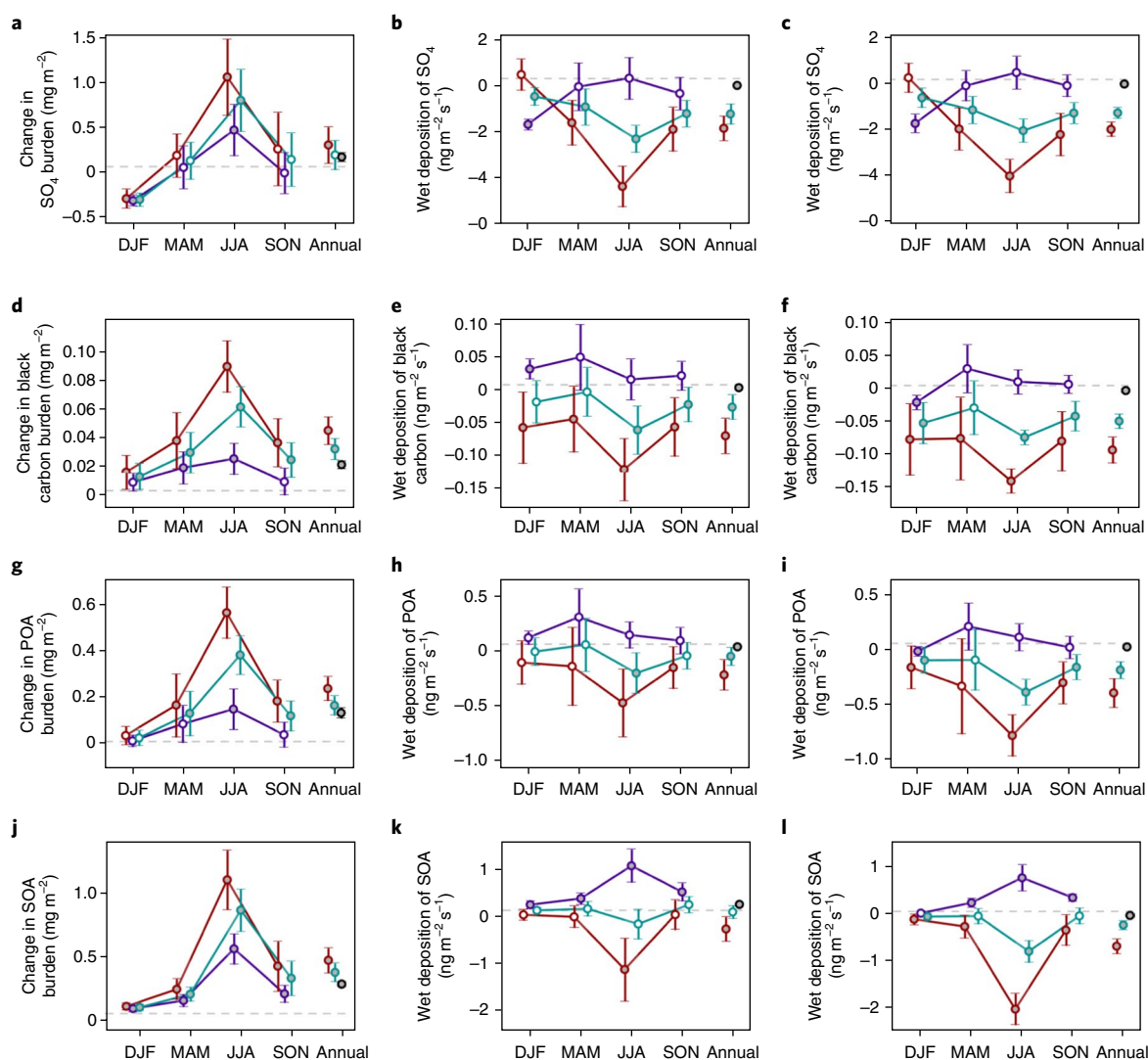


Fig. 1 | CAM5 seasonal and annual mean aerosol burden and wet deposition response to climate change. a–l, Changes in aerosol burden (**a, d, g** and **j**), and wet deposition due to total precipitation (**b, e, h** and **k**) and LSP (**c, f, i** and **l**) over the Northern Hemisphere mid-latitudes for SO_4 (**a–c**), black carbon (**d–f**), POA (**g–i**) and SOA (**j–l**) stratified by all grid points (turquoise), land only (maroon) and ocean only (purple). Annual mean responses for the globe (black), Northern Hemisphere mid-latitudes (turquoise) and Northern Hemisphere mid-latitude continents (maroon) are shown to the right of each panel. Error bars represent the 99% confidence interval based on a Student's *t*-test for the difference of means, using the pooled variance. DJF, December through February; SON, September through November.

consistent with a corresponding maximum reduction in wet removal. Furthermore, most of this reduction in aerosol wet removal is driven by a decrease in wet deposition due to LSP (Fig. 1c,f,i,l). Over the oceans, the weaker decreases and increases in wet deposition, respectively, are consistent with: (1) the increase in aerosol burden over the continents, some of which gets transported over the ocean, leading to an increase in wet removal; and (2) increases in LSP (Supplementary Fig. 1). These results agree with earlier findings based on ACCMIP models².

We now focus on what contributes to the reduction in aerosol wet deposition, most of which occurs over the continents of the Northern Hemisphere mid-latitudes during JJA. A decrease in wet deposition is in contrast with the expected global mean precipitation increase in response to warming¹⁰. CAM5 supports previous findings and simulates a 2.5% increase in global annual mean total precipitation, which becomes larger over the Northern Hemisphere mid-latitude continents (9.4% annual increase). Most of this annual mean increase over the Northern Hemisphere

mid-latitude continents is driven by convective precipitation (a 21% annual increase) as opposed to LSP (a 2% annual increase; Supplementary Fig. 1). During JJA, Northern Hemisphere mid-latitude total precipitation increases slightly, which is decomposed into a 14% increase in convective precipitation, but an 18% decrease in LSP (Fig. 2a). Similar results exist across the CMIP5 models². Despite this increase in convective precipitation, the change in wet deposition due to convective precipitation is negligible over the Northern Hemisphere mid-latitudes during JJA under warming (Supplementary Fig. 2). The decrease in LSP is consistent with a maximum reduction in wet deposition due to large-scale (and total) precipitation over the Northern Hemisphere mid-latitude continents during JJA. Although the dominant role of LSP, and the corresponding wet removal, is consistent with ACCMIP models², we acknowledge relatively large model diversity in terms of the proportion of wet removal due to convective precipitation¹⁶, as well as additional uncertainties in aerosol simulations (Methods).

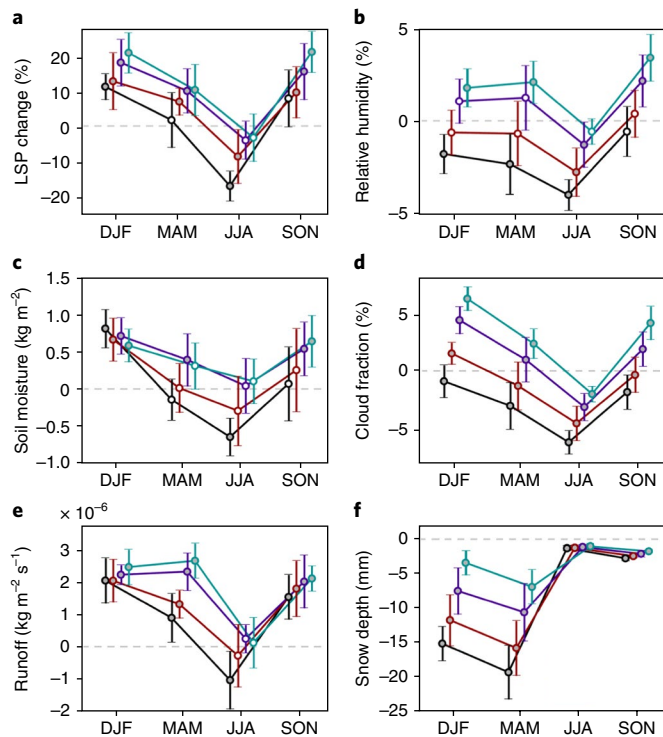


Fig. 2 | CAM5 seasonal mean hydrology response for default warming and muted land warming simulations. a–f, Changes for the Northern Hemisphere mid-latitudes over land for LSP (**a**), lower tropospheric relative humidity (**b**), 10 cm soil moisture (**c**), low cloud cover (**d**), surface runoff (**e**) and snow depth (**f**). Responses are shown for 0% nudging (that is, default warming; black), 1.0% nudging (maroon), 2.5% nudging (purple) and 5.0% nudging simulations (turquoise). Error bars represent the 99% confidence interval based on a Student's *t*-test for the difference of means, using the pooled variance. The low cloud cover response in **d** is nearly identical to the low-level, large-scale (stratus) cloud response.

Reductions in relative humidity and soil moisture are important components of the projected summer drying over the Northern Hemisphere mid-latitudes²⁵. To a large extent, land moisture is dictated by the transport of moisture from the oceans²⁶. When the continental lower tropospheric temperature warms more than that over the ocean, the air can hold more moisture relative to the amount of moisture advected from the oceans²⁷. As a result, the relative humidity over the continents decreases. This relative humidity reduction promotes low-level cloud (CLOW) reductions over the land, causing further land warming, thus constituting a positive feedback that acts to further warm and dry out the land^{21,28}. Soil moisture is also a crucial factor for the positive land-drying feedback during the summer. Less soil moisture has been associated with less precipitation through atmospheric feedbacks²⁹.

CAM5 captures this summertime drying over the Northern Hemisphere mid-latitude continents (Supplementary Fig. 3), including the reduction in relative humidity and CLOW, as well as a reduction in soil moisture, all of which are largest during JJA (Fig. 3). The CLOW reduction is largely due to decreases in low-level large-scale (stratus) cloud (SCLOW; Methods). Furthermore, snow depth exhibits a maximum decrease during March through May (MAM), which implies less snow melt during the late spring and early summer, probably contributing to the decrease in soil moisture. Also consistent with the enhanced continental aridity is a decrease in surface runoff. These CAM5 hydrological changes are generally consistent across CMIP5 models (Supplementary Fig. 4).

However, some exceptions do exist, including the seasonal cycle of runoff and the magnitude of the snow depth response.

To evaluate the importance of enhanced land warming to the aforementioned hydrological responses, and the increase in aerosol burden under warming, a set of idealized simulations were performed to mute the land warming. These were identical to the default warming simulation, but the near-surface land temperature was nudged to the control simulation's near-surface land temperature (Methods). Three separate nudging simulations were performed, with nudging strengths of 1.0, 2.5 and 5.0%. These simulations successfully muted the enhanced land warming, with larger nudging producing a larger LSWC reduction (Supplementary Fig. 5 and Supplementary Table 1). The LSWC amplitude can be measured in terms of the warming ratio, defined as the lower-tropospheric continental warming relative to that over the ocean³⁰. In CAM5 simulations with 5.0% nudging, the global annual warming ratio drops from 1.46 to 1.08 (26% decrease). This warming ratio reduction occurs at all latitudes, but the largest decrease (~41%) occurs over the Northern Hemisphere mid-latitudes during JJA. Hence, these nudging simulations successfully weakened the LSWC.

The importance of muted land warming was explored in two steps: (1) by analysing the hydrology changes; and (2) by investigating the aerosol burden and wet deposition changes. The muted land warming simulations weaken the decrease in all hydrology variables, particularly during JJA, including lower tropospheric relative humidity, soil moisture, surface runoff, low clouds and LSP (Fig. 2). The decrease in MAM snow depth is also weakened. Thus, with a weaker LSWC, less Northern Hemisphere mid-latitude continental drying occurs, and this response generally scales with the magnitude of the nudging. However, we note possible nonlinearities with some aspects of the response, as the changes in LSP, soil moisture and runoff are similar for the 2.5 and 5.0% nudging experiments, despite clear separation of the change in aerosol burdens.

Figure 3 shows that muted land warming results in a weaker increase (or decrease) in anthropogenic aerosol species over the Northern Hemisphere mid-latitude continents, particularly during JJA. Consistently, the decrease in wet deposition due to LSP is also weakened. This weakening is consistent with the changes in hydrology and strength of the nudging—1.0% nudging yields the smallest reduction during JJA, and 5.0% nudging yields the largest reduction.

Climate change may also affect aerosol burden through modification of chemical production pathways, particularly in the context of SO₄ and secondary organic aerosols (SOA). In CAM5, changes in chemical production act to mute the increase in SO₄ and SOA burden over the Northern Hemisphere mid-latitude continents, especially during JJA (Supplementary Figs. 6 and 7). The two primary chemical production pathways of SO₄ are aqueous and gaseous production (Methods). In response to warming, CAM5 exhibits a decrease in aqueous SO₄ production, consistent with the decrease in low clouds (Fig. 2). This, in turn, results in more of the SO₄ gaseous precursor SO₂ (not shown), and a corresponding increase in SO₄ gaseous production. However, the decrease in aqueous production dominates, and the total chemical production of SO₄ decreases in response to warming (Supplementary Fig. 6).

In CAM5, a relatively simple treatment of SOA is assumed (Methods). A gaseous precursor for SOA formation (SOAG) requires oversaturation to condense and form SOA. The SOAG partial pressure increases with enhanced warming, which decreases the condensation of SOAG to SOA under warming, resulting in less SOA (Supplementary Fig. 7). Hence, changes in chemical production are not responsible for the increase in SO₄ or SOA burden under warming—in fact, they act to weaken the increase. Furthermore, these conclusions are consistent across the muted warming simulations (Supplementary Figs. 6 and 7).

Finally, a third set of simulations were performed to investigate the impact of increasing the LSWC. They were identical to the

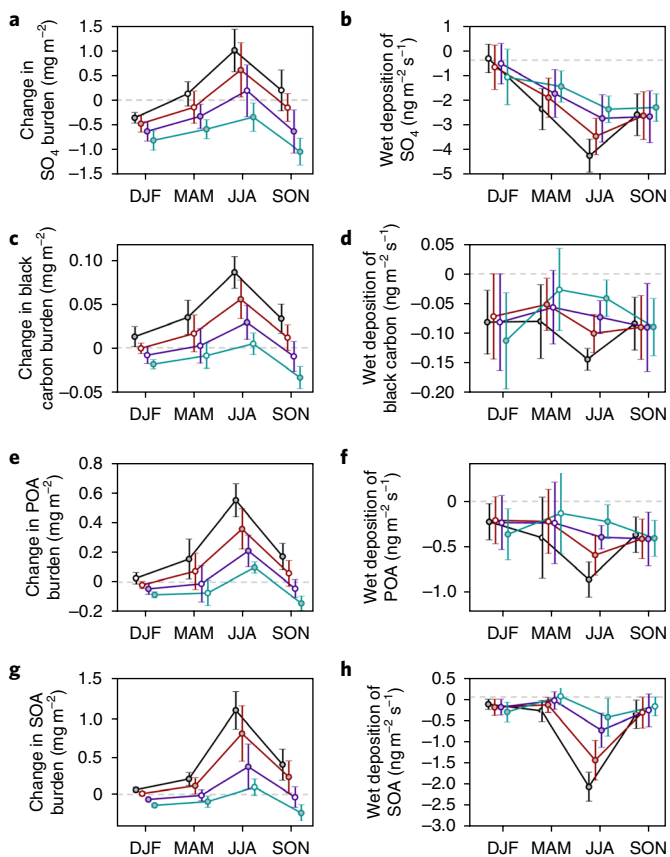


Fig. 3 | CAM5 seasonal mean response of aerosol burden and wet deposition due to LSP for default warming and muted land warming simulations. **a–h**, Changes in aerosol burden (**a**, **c**, **e** and **g**), and wet deposition due to LSP (**b**, **d**, **f** and **h**), for the Northern Hemisphere mid-latitudes over land for SO₄ (**a** and **b**), black carbon (**c** and **d**), POA (**e** and **f**) and SOA (**g** and **h**). Responses are shown for 0% nudging (that is, default warming; black), 1.0% nudging (maroon), 2.5% nudging (purple) and 5.0% nudging simulations (turquoise). Error bars represent the 99% confidence interval based on a Student's *t*-test for the difference of means, using the pooled variance.

control simulation, but the near-surface land temperature was nudged to that based on a future warming simulation, based on the year 2150 Representative Concentration Pathway (RCP) 8.5 climate conditions (Methods). Three different nudging strengths of 1.0, 2.5 and 5.0% were used. These simulations therefore enhance the LSWC (Supplementary Fig. 8), and a larger contrast is obtained with a larger nudging strength. These simulations, which only feature enhanced land warming, yield an increase in all anthropogenic aerosol species, in addition to a decrease in wet removal by LSP, over the Northern Hemisphere mid-latitude continents (Fig. 4). These simulations also show the expected hydrological changes, including decreases in lower-tropospheric relative humidity, soil moisture, low clouds and LSP, particularly during JJA (Supplementary Fig. 9). Thus, land warming alone causes continental drying and an increase in aerosol burden, with more land warming yielding a larger response.

Now, we further elucidate the cause of the JJA LSP decrease over the Northern Hemisphere mid-latitude continents. Extratropical storm tracks play an important role in mid-latitude precipitation, and global warming may lead to a decrease in extratropical storm track activity^{31,32}. CAM5 simulations support this finding, yielding a decrease in JJA Northern Hemisphere mid-latitude storm track

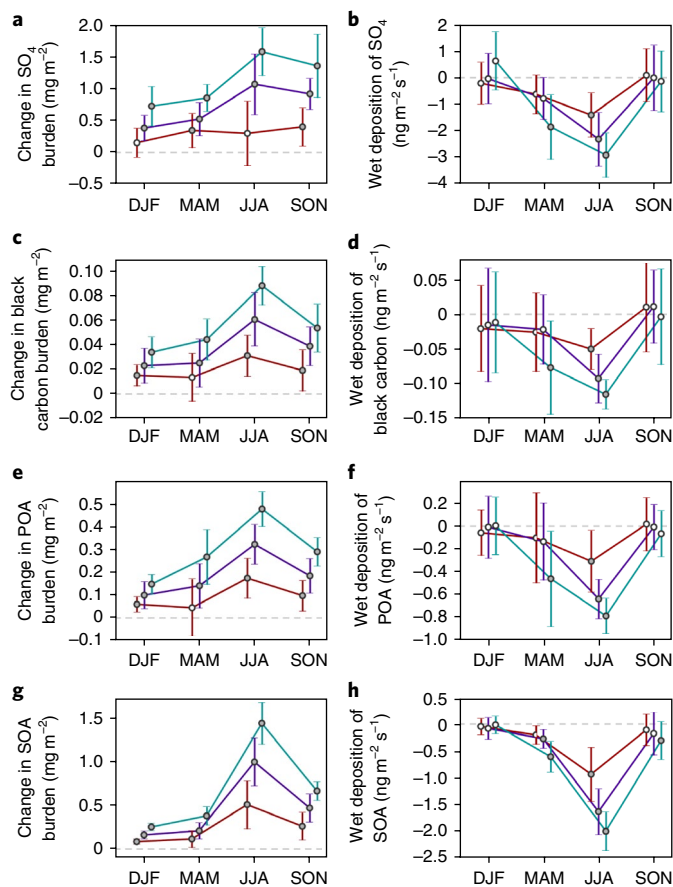


Fig. 4 | CAM5 seasonal mean response of aerosol burden and wet deposition due to LSP for enhanced land warming simulations. **a–h**, Changes in aerosol burden (**a**, **c**, **e** and **g**), and wet deposition due to LSP (**b**, **d**, **f** and **h**), are shown for the Northern Hemisphere mid-latitudes over land for SO₄ (**a** and **b**), black carbon (**c** and **d**), POA (**e** and **f**) and SOA (**g** and **h**). Responses are shown for 1.0% nudging (maroon), 2.5% nudging (purple) and 5.0% nudging simulations (turquoise). Error bars represent the 99% confidence interval based on a Student's *t*-test for the difference of means, using the pooled variance.

activity (Supplementary Fig. 10). However, the decrease in JJA storm track activity occurs over both land and ocean, with similar magnitude, in opposition to the land–sea contrast in LSP, low cloud and other hydrological variables (for example, Supplementary Fig. 3). Additional analyses also suggest that a decrease in storm track activity is not the dominant cause of the LSP decrease (Supplementary Information).

Instead, the LSP decrease is largely consistent with decreases in SCLOW. As with most models, CAM5 parameterizes large-scale cloud cover based on relative humidity²³. The JJA Northern Hemisphere mid-latitude decrease in continental relative humidity under warming is consistent with the decrease in large-scale cloud cover, particularly in the lower troposphere. Moreover, the net condensation rate of water vapour into liquid stratus droplets depends on the stratus cloud cover. This implies that a decrease in large-scale cloud cover should be associated with a decrease in LSP. Thus, we suggest that the decrease in LSP is a direct consequence of the decrease in large-scale cloud cover.

Statistical analyses support this conclusion. The spatial (grid box) JJA Northern Hemisphere mid-latitude land correlation between SCLOW and low-level relative humidity (LSP) is 0.72 (0.55) in the control simulation. The corresponding correlation between the

change in SCLOW and the change in low-level relative humidity (LSP) is 0.52 (0.43). In contrast, the corresponding correlations between LSP and storm track activity are much weaker, at 0.16 in the control simulation and 0.19 based on responses. Similar results are obtained in CMIP5 models (Supplementary Information). Furthermore, a regression model comprising SCLOW versus LSP values from the CAM5 control simulation predicts reasonably well the actual change in JJA Northern Hemisphere mid-latitude continental LSP (Supplementary Fig. 11).

To further test the importance of SCLOW to LSP, we performed additional perturbed parameter experiments with CAM5 that involved reducing the sensitivity of SCLOW to relative humidity (Methods). When decreasing the sensitivity of SCLOW to relative humidity, smaller (relative) decreases in SCLOW under warming are expected. Furthermore, if decreases in SCLOW drive decreases in LSP under warming, smaller (relative) decreases in LSP would also be expected. In the default warming simulation, the Northern Hemisphere mid-latitude JJA continental decrease in SCLOW is -3.7% and the decrease in LSP is $-3.3\text{ mm month}^{-1}$. The corresponding percentage changes are -33.9 and -13.3% , respectively. In our sensitivity experiment, the corresponding decreases in SCLOW and LSP were -2.5% and $-2.6\text{ mm month}^{-1}$, respectively. More importantly, the percentage changes exhibited weaker decreases, at -30.5 and -10.7% , respectively. Thus, as we reduce the sensitivity of SCLOW to relative humidity over land, warming results in a smaller SCLOW decrease, and a correspondingly smaller LSP decrease.

Similar to state-of-the-art ACCMIP models, CAM5 simulates a global annual mean increase in anthropogenic aerosols in response to GHG-induced warming, with a maximum increase over the Northern Hemisphere mid-latitude continents during JJA. Targeted CAM5 simulations show that this response is related to the LSWC and associated increases in continental aridity, which result in less aerosol wet removal. Muting the LSWC weakens the increase in aerosol burden, as well as the decrease in soil moisture, runoff, snow depth, lower tropospheric relative humidity, LSP and associated aerosol wet removal. Furthermore, land warming alone yields an increase in aerosol burden and the opposite hydrological changes. Additional analyses suggest that the reduction in LSP is largely due to decreases in SCLOW, which is consistent with reductions in continental relative humidity. Although aerosol simulations have uncertainty, we have related the increase in aerosol burden under warming to a robust climate change phenomenon—the land warms more than the ocean, which leads to enhanced continental aridity, and less LSP and aerosol wet removal. Furthermore, although our results are based on a single climate model, a larger suite of CMIP5 models yields similar hydrological changes to CAM5. ACCMIP models also support the importance of reduced LSP and aerosol wet removal under warming. Since our default warming responses are (1) based on a business-as-usual warming scenario and (2) assume no reductions in anthropogenic aerosol emissions, they represent an upper bound on future aerosol increases in response to climate change. Unless emission reductions occur, our results add confidence that a warmer world will be associated with enhanced anthropogenic aerosol pollution, or alternatively, that larger emission reductions will be necessary to obtain a desired level of air quality.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, statements of data availability and associated accession codes are available at <https://doi.org/10.1038/s41558-019-0401-4>.

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References

- Westervelt, D. et al. Quantifying $\text{PM}_{2.5}$ -meteorology sensitivities in a global climate model. *Atmos. Environ.* **142**, 43–56 (2016).
- Allen, R. J., Landuyt, W. & Rumbold, S. T. An increase in aerosol burden and radiative effects in a warmer world. *Nat. Clim. Change* **6**, 269–274 (2016).
- Sutton, R. T., Dong, B. & Gregory, J. M. Land/sea warming ratio in response to climate change: IPCC AR4 model results and comparison with observations. *Geophys. Res. Lett.* **34**, L02701 (2007).
- Boer, G. The ratio of land to ocean temperature change under global warming. *Clim. Dynam.* **37**, 2253–2270 (2011).
- Bond, T. C. et al. Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000. *Glob. Biogeochem. Cycles* **21**, GB2018 (2007).
- Boucher, O. et al. in *Climate Change 2013: The Physical Science Basis* (eds Stocker, T. F. et al.) 571–657 (IPCC, Cambridge Univ. Press, 2013).
- Ramanathan, V., Crutzen, P., Kiehl, J. & Rosenfeld, D. Aerosols, climate, and the hydrological cycle. *Science* **294**, 2119–2124 (2001).
- Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D. & Pozzer, A. The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature* **525**, 367–371 (2015).
- Vecchi, G. A. & Soden, B. J. Global warming and the weakening of the tropical circulation. *J. Clim.* **20**, 4316–4340 (2007).
- Held, I. M. & Soden, B. J. Robust responses of the hydrological cycle to global warming. *J. Clim.* **19**, 5686–5699 (2006).
- Pye, H. et al. Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. *J. Geophys. Res. Atmos.* **114**, D01205 (2009).
- Racherla, P. N. & Adams, P. J. Sensitivity of global tropospheric ozone and fine particulate matter concentrations to climate change. *J. Geophys. Res. Atmos.* **111**, D24103 (2006).
- Fang, Y. et al. The impacts of changing transport and precipitation on pollutant distributions in a future climate. *J. Geophys. Res. Lett.* **116**, D18303 (2011).
- Lamarque, J.-F. et al. The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics. *Geosci. Model Dev.* **6**, 179–206 (2013).
- Silva, R. A. et al. Future global mortality from changes in air pollution attributable to climate change. *Nat. Clim. Change* **7**, 647–651 (2017).
- Textor, C. et al. Analysis and quantification of the diversities of aerosol life cycles within AeroCom. *Atmos. Chem. Phys.* **6**, 1777–1813 (2006).
- Allen, R. J. & Landuyt, W. The vertical distribution of black carbon in CMIP5 models: comparison to observations and the importance of convective transport. *J. Geophys. Res. Atmos.* **119**, 4808–4835 (2014).
- Byrne, M. P. & O’Gorman, P. A. Land–ocean warming contrast over a wide range of climates: convective quasi-equilibrium theory and idealized simulations. *J. Clim.* **26**, 4000–4016 (2013).
- Byrne, M. P. & O’Gorman, P. A. Link between land–ocean warming contrast and surface relative humidities in simulations with coupled climate models. *Geophys. Res. Lett.* **40**, 5223–5227 (2013).
- Joshi, M. & Gregory, J. Dependence of the land–sea contrast in surface climate response on the nature of the forcing. *Geophys. Res. Lett.* **35**, L24802 (2008).
- Fasullo, J. T. Robust land–ocean contrasts in energy and water cycle feedbacks. *J. Clim.* **23**, 4677–4693 (2010).
- Clark, R. T., Murphy, J. M. & Brown, S. J. Do global warming targets limit heatwave risk? *Geophys. Res. Lett.* **37**, L17703 (2010).
- Neale, R. B. et al. *Description of the NCAR Community Atmosphere Model (CAM 5.0)* (NCAR, 2012).
- Atlas, E. & Giam, C. Ambient concentration and precipitation scavenging of atmospheric organic pollutants. *Water Air Soil Pollut.* **38**, 19–36 (1988).
- Rowell, D. P. & Jones, R. G. Causes and uncertainty of future summer drying over Europe. *Clim. Dynam.* **27**, 281–299 (2006).
- Simmons, A., Willett, K., Jones, P. & Dee, D. Low-frequency variations in surface atmospheric humidity, temperature, and precipitation: inferences from reanalyses and monthly gridded observational data sets. *J. Geophys. Res. Atmos.* **115**, D01110 (2010).
- Byrne, M. P. & O’Gorman, P. A. Understanding decreases in land relative humidity with global warming: conceptual model and GCM simulations. *J. Clim.* **29**, 9045–9061 (2016).
- Joshi, M., Lambert, F. & Webb, M. An explanation for the difference between twentieth and twenty-first century land–sea warming ratio in climate models. *Clim. Dynam.* **41**, 1853–1869 (2013).
- Allen, R. & Zender, C. The role of eastern Siberian snow and soil moisture anomalies in quasi-biennial persistence of the Arctic and North Atlantic Oscillations. *J. Geophys. Res.* **116**, D16125 (2011).

30. Sejas, S. A., Albert, O. S., Cai, M. & Deng, Y. Feedback attribution of the land–sea warming contrast in a global warming simulation of the NCAR CCSM4. *Environ. Res. Lett.* **9**, 124005 (2014).
31. Chang, E. K., Guo, Y., Xia, X. & Zheng, M. Storm-track activity in IPCC AR4/CMIP3 model simulations. *J. Clim.* **26**, 246–260 (2013).
32. Chang, E. K., Guo, Y. & Xia, X. CMIP5 multimodel ensemble projection of storm track change under global warming. *J. Geophys. Res. Atmos.* **117**, D23118 (2012).

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Author contributions

R.J.A. conceived the project, designed the study and performed the CAM5 simulations. R.J.A. and T.H. led the writing of the paper. T.H. carried out the data analysis and figure construction. C.A.R. and H.S. advised on interpretation of the results. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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Methods

All simulations for this study were performed with the state-of-the-art CAM5 (ref. 23)—the atmospheric component of the Community Earth System Model version 1.2.2, developed primarily at the National Center for Atmospheric Research. CAM5 incorporates the three-mode modal aerosol model (MAM3)³³, which provides internally mixed representations of number concentrations and mass for Aitken, accumulation and coarse aerosol modes. The simulated aerosols were composed of SO₄, black carbon, primary organic aerosols (POA), SOA, sea salt and mineral dust. Aerosol wet removal was based on ref. 34, but with modifications for the consistency with cloud macrophysics and microphysics. The routine treats both in-cloud and below-cloud scavenging. For in-cloud scavenging, cloud water first-order loss rates (based on cloud water mixing ratios and precipitation production rates) were multiplied by ‘solubility factors’ to obtain aerosol first-order loss rates. The solubility factors can be interpreted as the aerosol fraction in cloud drops multiplied by the tuning factor. The stratiform in-cloud scavenging only affects the stratiform-cloud-borne aerosol particles, and these have solubility factors of 1.0 (0 for interstitial aerosols).

In CAM5 (and most models), two types of clouds were diagnosed: large-scale (stratus) and convective (cumulus). The large-scale cloud cover is derived from the assumed triangular distribution of total relative humidity. The cumulus cloud cover is a function of the convective mass flux from both the deep and shallow convection schemes.

The spatial correlation (JJA Northern Hemisphere mid-latitude land) between the change in CLOW and SCLOW is 0.98. Furthermore, the JJA Northern Hemisphere mid-latitude continental decreases in CLOW and SCLOW are -4.1 and -3.7% , respectively (significant at the 99% confidence level). The corresponding decrease in low-level convective cloud cover is much weaker at -0.4% . In terms of percentage changes, CLOW and SCLOW exhibit large responses at -28.8 and -33.9% , respectively (-11.5% for low-level convective cloud cover). Thus, the bulk of the CLOW decrease is due to decreases in SCLOW, and the spatial patterns of the responses are very similar. Using CLOW as a surrogate for SCLOW is a reasonable approximation.

Furthermore, the change in SCLOW, as for most hydrological variables including LSP, exhibits a land–sea contrast (for example, Supplementary Fig. 3). During JJA in the Northern Hemisphere mid-latitudes, SCLOW decreases by -3.7% over land, but increases by 1.5% over sea (both significant at the 99% confidence level). This is consistent with the land–sea contrast in low-level relative humidity, which decreases by -4.5% (significant at the 99% confidence level) over land, but increases by 0.5% (significant at the 95% confidence level) over the sea.

Uncertainty in the aerosol response to future warming is related to several factors, including uncertainty in the simulation of aerosol processes. For example, models parameterize aerosol removal processes differently, including both wet and dry removal. Based on AeroCom models, relatively large diversity exists in terms of the proportion of wet removal due to convective precipitation versus LSP. Across AeroCom models and aerosol species, the percentage of convective wet removal (relative to the total) ranges from ~ 10 – 80% ¹⁶. CAM5 falls in the middle of this relatively large range, with a percentage of removal by convective precipitation ranging from 43–52%, depending on the aerosol species. Over the Northern Hemisphere midlatitude continents during JJA, the numbers are a bit larger than the global annual mean values, with approximately 50–60% of wet removal due to convective precipitation. Thus, CAM5 does not lack a sensitivity of wet removal by convective precipitation.

Another source of uncertainty is how changes in precipitation intensity versus frequency impact aerosol wet removal³⁵. During JJA, the CMIP5 multi-model mean shows a 6–7% decrease in the LSP frequency rate in the Northern Hemisphere mid-latitudes. The corresponding decrease in the intensity of precipitation is 13–14%. This indicates that a decrease in the intensity of LSP, as opposed to the frequency, is probably the most important driver of the decrease in LSP and the associated wet removal³. However, we note that models tend to overestimate the frequency of precipitation (rain almost every day), implying that they may underestimate the role of precipitation frequency in wet scavenging.

Similar results are obtained with CAM5, as the frequency of JJA LSP decreases by -3.5% in the default warming simulation over Northern Hemisphere mid-latitude continents, whereas the intensity decreases by -13.8% . Furthermore, the changes in LSP in the nudged simulations are driven by changes in the intensity, as opposed to the frequency, of LSP. For example, the frequency of JJA LSP decreases by -3.5 , -4.2 , -3.1 and -2.7% over the Northern Hemisphere mid-latitude continents in the default warming and 1.0, 2.5 and 5.0% nudged (muted land warming) simulations, respectively. The corresponding intensity of LSP decreases by -13.8 , -4.6 , -1.0 and -0.6% . Similarly, in the enhanced land-warming simulations, the frequency of LSP decreases by -1.4 , -1.3 and -1.8% in the 1.0, 2.5 and 5.0% nudged (enhanced land warming) simulations, respectively. The corresponding intensity of LSP decreases by -6.5 , -15.5 and -17.3% . Thus, changes in the intensity, as opposed to the frequency, of LSP are driving the bulk of the LSP signal. We note that changes in precipitation frequency are probably more related to dynamics (for example, changes in the frequency of storm tracks), whereas changes in precipitation intensity are probably more related to changes in thermodynamics (for example, enhanced land warming and aridity). Thus, the

dominant role of the LSP intensity decrease in the overall LSP reduction supports the importance of thermodynamics, as opposed to dynamics.

Although we did not use the full chemistry version of CAM5 (CAM5-chem), simple on-line chemistry is included in CAM5. In terms of SOA, the most straightforward representation (which is used in many climate models) is to assume fixed mass yields for precursor volatile organic compounds and then emit this mass as primary aerosol particles. MAM adds one level of sophistication by simulating a single lumped gas phase SOA species (SOAG). MAM then simulates condensation or evaporation of the SOAG to or from several aerosol modes. This provides a realistic method for estimating the distribution of SOA among different modes, and a minimal representation of the temperature dependence of the gas/aerosol partitioning.

Simple gas-phase chemistry is included for SO₄. This includes: dimethyl sulfide oxidation with OH and NO₃ to form SO₂; SO₂ oxidation with OH to form H₂SO₄ (gas); H₂O₂ production; and H₂O₂ loss. Rate coefficients and oxidant concentrations (O₃, OH, HO₂ and NO₃) are provided from the Model for Ozone and Related chemical Tracers³⁶.

CAM5 time-slice simulations were integrated for ten years and were based on climatological sea-surface temperatures and sea-ice concentrations, along with anthropogenic aerosol and precursor gas emissions. Nearly identical results were obtained with a longer, 20-year integration. Sea-surface temperatures and sea-ice concentration anomalies (2090–2099 relative to 2006–2015) from CMIP5 RCP 8.5 models were added to the default warming simulation, along with end-of-the-century RCP 8.5 GHG concentrations. The control and default warming simulations both have identical aerosol and precursor gas emissions, based on the year 2000. We note that our simulations lack additional climate warming–land feedbacks, including prognostic wildfire emissions and changes in vegetation that may be important for changes in aerosol burden under future warming.

Additional idealized ten-year time-slice simulations were performed to investigate the effect of the LSWC on aerosol burden. Muted land warming simulations are identical to the default warming simulation, but near-surface land temperatures are nudged to the control simulation. The simulated meteorological field (near-surface land temperature) T_{nudged} is calculated in the following manner:

$$T_{\text{nudged}} = (1-\alpha)T_{\text{warming}} + \alpha T_{\text{control}} \quad (1)$$

where T_{warming} and T_{control} are the near-surface land temperatures from the default warming simulation and control simulation, respectively. T_{control} is fed every 6 h and the nudging is applied at every model time step (that is, every 30 min). The fraction α denotes the strength of the nudging, which varies between 0.010, 0.025 and 0.050 (that is, 1.0, 2.5 and 5.0% nudging, respectively).

A series of ten-year time-slice simulations with enhanced land warming was also performed. These simulations are identical to the control simulation, but near-surface land temperatures are relaxed to a warming simulation, which is based on RCP 8.5 climate conditions in the year 2150. Thus, these simulations only feature enhanced land warming.

Similar to most climate models, CAM5 diagnoses the liquid stratus cloud cover as a function of the grid box mean relative humidity over water. There is also a critical relative humidity (U_{cl}) that must be exceeded for liquid stratus cloud cover to form (that is, stratus cloud only exists when the grid mean relative humidity exceeds U_{cl}). In CAM5, U_{cl} is an externally specified function of height and surface properties. U_{cl} is specified at 0.8875 in the layers below 700 hPa (SCLOW). However, for SCLOW over land with a water-equivalent snow depth less than 10^{-6} m, $U_{cl} = 0.7875$. Similarly, $U_{cl} = 0.80$ in the layers above 400 hPa (high-level stratus). Between 700 and 400 hPa (mid-level stratus), a linearly interpolated U_{cl} value is used.

Sensitivity experiments were performed by increasing U_{cl} in the layers below 700 hPa over land with a water-equivalent snow depth of less than 10^{-6} m, from 0.7875 to 0.8875 (both a new control and a new warming simulation). By increasing U_{cl} , the amount of SCLOW over land is reduced, particularly during the summer months when the snow depth is low. The JJA Northern Hemisphere mid-latitude climatological SCLOW over land decreases from 10.9% in the default simulation ($U_{cl} = 0.7875$) to 8.4% in the sensitivity simulation ($U_{cl} = 0.8875$). Corresponding values over the sea remain relatively unchanged at 32.3% (32.7%) in the default (sensitivity) simulation.

More importantly, the sensitivity of SCLOW to relative humidity is reduced. This is confirmed by regressing SCLOW versus low-level relative humidity using values from each control simulation. With $U_{cl} = 0.7875$, the regression slope ($\Delta\text{CLOW}/\Delta\text{RH}$ (where RH stands for relative humidity); that is, the sensitivity of SCLOW to relative humidity) is 0.51, implying that a 1% decrease in relative humidity yields a 0.5% decrease in SCLOW (significant at the 99% confidence level). With $U_{cl} = 0.8875$, the regression slope decreases to 0.41, implying that a 1% decrease in relative humidity yields a 0.4% decrease in SCLOW (significant at the 99% confidence level). Thus, this perturbed parameter experiment reduces the sensitivity of SCLOW to relative humidity by 20%. Similar results are obtained using relative humidity at other levels (for example, 700 hPa).

In addition to smaller reductions in JJA Northern Hemisphere mid-latitude continental SCLOW and LSP in our sensitivity simulation, other hydrological

variables change in a consistent way, which supports the idea that feedbacks exist between the hydrological variables. For example, there is less JJA Northern Hemisphere mid-latitude land warming (6.0 to 5.6 K), a smaller decrease in low-level relative humidity (−4.5 to −4.3%) and a smaller decrease in soil moisture (−0.85 to −0.46 kg m^{−2}). Similar conclusions exist based on percentage changes. Thus, with reduced sensitivity of SLOW to relative humidity, there is reduced land warming, as well as smaller decreases in low-level relative humidity and soil moisture. In other words, there is a smaller increase in continental aridity.

Consistent with our default warming and nudged simulations, the decrease in LSP (as U_{ci} is increased) is entirely dominated by decreases in LSP intensity. The change in the frequency of LSP remains essentially unchanged at −3.5%. However, the intensity of LSP changes from −13.8 to −6.8% as U_{ci} is increased from 0.7875 to 0.8875.

Code availability

The codes used to process the CAM5 simulations are available from R.J.A.

Data availability

CAM5 data and simulations are available from R.J.A.

References

33. Liu, X. et al. Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5. *Geosci. Model Dev.* **5**, 709–739 (2012).
34. Rasch, P. et al. A comparison of scavenging and deposition processes in global models: results from the WCRP Cambridge Workshop of 1995. *Tellus B* **52**, 1025–1056 (2000).
35. Hou, P., Wu, S., McCarty, J. L. & Gao, Y. Sensitivity of atmospheric aerosol scavenging to precipitation intensity and frequency in the context of global climate change. *Atmos. Chem. Phys.* **18**, 8173–8182 (2018).
36. Emmons, L. K. et al. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). *Geosci. Model Dev.* **3**, 43–67 (2010).

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