

An increase in aerosol burden and radiative effects in a warmer world

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Atmospheric aerosols are of significant environmental importance, due to their effects on air quality, as well as their ability to alter the planet's radiative balance. Recent studies characterizing the effects of climate change on air quality and the broader distribution of aerosols in the atmosphere show significant, but inconsistent results, including the sign of the effect^{1–3}. Using a suite of state-of-the-art climate models, we show that climate change is associated with a negative aerosol–climate feedback of -0.02 to $-0.09 \text{ W m}^{-2} \text{ K}^{-1}$ for direct radiative effects, with much larger values likely for indirect radiative effects. This is related to an increase in most aerosol species, particularly over the tropics and Northern Hemisphere midlatitudes, largely due to a decrease in wet deposition associated with less large-scale precipitation over land. Although simulation of aerosol processes in global climate models possesses uncertainty, we conclude that climate change may increase aerosol burden and surface concentration, which may have implications for future air quality.

The burden of atmospheric aerosols depends on several factors, including emissions, chemistry and weather patterns. Although emissions are the dominant factor determining ambient aerosol concentrations, multiple links between global climate change and aerosol concentrations exist. Future responses of the climate system to greenhouse gas (GHG) warming will lead to changes in the hydrologic cycle⁴ and atmospheric circulation^{5–9} that will subsequently affect air quality and the distribution of aerosols irrespective of changes in emissions.

Projections of future climate change yield a global increase in precipitation, and local increases in precipitation are expected to simultaneously decrease aerosol burdens^{10–12}. However, changes in the frequency/intensity of precipitation and storm tracks may offset any potential increases in wet removal associated with a global increase in precipitation¹². Recent studies suggest that the warming associated with increasing GHGs may lead to an overall increase in the burden of soluble aerosols (for example, sulphate, black carbon)^{13–15}, and this ‘climate penalty’ may impact a region’s ability to attain a specified air quality standard.

In addition to its association with air quality, aerosols impact the radiative balance of the planet. Most aerosol species, such as sulphate, reflect solar radiation and alter cloud microphysical properties, which enhances cloud albedo and lifetime. Changes in aerosols due to future climate warming will therefore affect the planetary energy balance, constituting a feedback loop^{16,17}. With respect to both anthropogenic and natural (sea salt, dust) aerosols, significant positive and negative feedbacks have been found^{16,18–20}.

Table 1 shows the response of several aerosol species to climate change, as simulated by climate models from the Atmospheric

Chemistry and Climate Model Intercomparison Project (ACCMIP; refs 21,22). Responses are based on the difference between two time-slice simulations representing climate in the year 2000 and 2100 (based on Representative Concentration Pathway 8.5, RCP8.5), both with identical emissions from year 2000. Also included are similar experiments with the Community Atmosphere Model versions 4 and 5 (CAM4/5; Supplementary Methods). Models show that climate change associated with GHG-induced global warming will result in significantly elevated surface concentrations of all primary anthropogenic aerosol species (SO_4 , BC and POM). For example, the average increase in surface sulphate concentration is 11.4%, with a range of 1.7%–17.7%. Similarly, the average increase in black carbon (primary organic matter) surface concentration is 10.5% (6.8%), with a range of 0.3%–30.2% (0.1%–15.3%). Models also show significant increases in primary anthropogenic aerosol burden, particularly for sulphate, where the mean increase is 12.5%, with a range of 0.6%–33.1%. Increases in BC and POM burden also exist (except in GISS-E2-R-p3), with several models yielding more than a 20% increase. Fine particulate matter ($\text{PM}_{2.5}$) also shows increases. Thus, climate change associated with GHG warming may result in enhanced anthropogenic aerosol burden and surface aerosol concentration.

Natural aerosols, including dust and sea salt, are also generally projected to increase. Table 1 shows that the average increase in sea salt burden is 2.9%, with a range of 1.6–4.2%. An increase in dust burden also exists, except in HadGEM2. A larger number of models from the Coupled Model Intercomparison Project version 5 (ref. 23) (CMIP5; Supplementary Table 1) also suggest natural aerosols may increase in a warmer world. The CMIP5 ensemble mean yields a 0.4% increase in sea salt burden and a corresponding 5.2% increase in dust burden (both significant at the 99% confidence level). More importantly, 74% (78%) of the models yield a positive change in sea salt (dust) burden (Supplementary Fig. 1). An increase in GHGs may therefore be associated with an increase in natural aerosols.

Consistent with the increase in aerosol burden under global warming, there is a corresponding increase in the magnitude (that is, a larger negative number) of both the aerosol direct radiative effects (DRE) and the aerosol cloud radiative effects (CRE; Supplementary Methods). The average change in DRE across ACCMIP models is -0.21 W m^{-2} , with a range of -0.03 to -0.43 W m^{-2} (Table 1). CAM5, GISS-E2-R-p3 and CESM-CAM-superfast also yield a stronger CRE, ranging from -1.14 to -5.43 W m^{-2} (the relevant diagnostics were not available for the other models). Thus, climate change associated with GHG warming may result in both enhanced aerosol burden and a negative aerosol–climate feedback.

ACCMIP models show that climate change is associated with a decrease in aerosol wet deposition (the primary removal

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Table 1 | Global average changes in surface aerosol concentration, burden and radiative effects due to climate change.

Model	SO ₄	BC	POM	PM _{2.5}	SOA	DUST	SS	DRE	CRE
CAM4	3.4/6.2	1.5/8.1	0.7/8.9	n/a	n/a	0.30/2.2	4.7/1.5	n/a	n/a
CAM5	6.6/4.6	6.7/12.1	5.7/11.5	n/a	−7.2/13.3	2.7/6.6	11.1/4.2	−0.19	−1.14
CESM-CAM-superfast	14.0/10.0	n/a	n/a	n/a	n/a	n/a	n/a	n/a	−5.43
GFDL-AM3	16.9/33.1	9.7/20.9	12.1/24.5	9.1/21.7	9.6/22.0	3.0/10.3	2.3/3.5	−0.43*	n/a
GISS-E2-R-p3	1.7/0.6	0.3/−2.6	0.1/−5.2	1.5/2.1	−6.6/−10.9	1.7/5.2	2.6/2.6	−0.03	−1.30
HadGEM2	17.5/25.0	30.2/19.7	6.4/18.4	n/a	0.0/−1.3	−11.2/−10.8	2.9/1.6	−0.25*	n/a
MIROC-CHEM	17.7/8.7	14.9/23.4	15.3/25.6	18.9/24.2	12.1/25.0	21.7/25.5	9.4/2.1	−0.14	n/a
STOC-HadAM3	12.3/14.8	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a

Surface concentrations (first number) and burden (second number) are percentage changes, and include sulphate (SO₄), black carbon (BC), primary organic matter (POM), fine particulate matter (PM_{2.5}), secondary organic aerosol (SOA), dust (DUST) and sea salt (SS). Radiative effect changes include direct radiative effects (DRE) and cloud radiative effects (CRE), and are in W m^{−2}. Radiative effects include both shortwave and longwave radiation, unless otherwise denoted by *. n/a is not available. All changes are significant at the 99% confidence level, unless denoted by bold.

mechanism), whereas dry deposition increases (Supplementary Tables 2–4). Climate change is also associated with a decrease in the removal rate of an aerosol species, k , which is dominated by a decrease in the wet removal rate, k_{wet} (Supplementary Methods). In the case of sulphate, for example, k decreases by -1.5 to -23.6% , with a mean decrease of -10.5% . This is driven by a decrease in k_{wet} , which ranges from -2.0 to -24.8% , with a mean decrease of -11.5% . Similar results exist for black carbon and primary organic matter. Moreover, nearly all of the decrease in SO₄, BC and POM wet deposition is due to removal by large-scale precipitation (Supplementary Table 5).

The decrease in wet deposition and wet removal rate, however, is inconsistent with the change in global mean precipitation, which increases by 1.4 – 9.8% , with a mean increase of 6.7% (or 1.6% per degree of surface warming). This is consistent with other studies showing a 1 – 3% increase in global mean precipitation per degree of surface warming⁴. This result shows that the efficiency with which aerosols are removed from the atmosphere by precipitation decreases in a warmer world.

The importance of precipitation—particularly large-scale precipitation—in driving changes in aerosol burden in a warmer world is also supported by analysing the corresponding spatial correlations, all of which are negative and range from -0.27 to -0.45 for sulphate burden (Supplementary Table 6), with a multi-model average of -0.36 (-0.48 in the Northern Hemisphere only). The corresponding model mean correlation based on zonal means increases to -0.52 . Correlations based on the frequency of large-scale precipitation are generally weaker, as are correlations based on convective precipitation. The model mean spatial correlations between the change in BC and POM burden and large-scale precipitation are -0.33 and -0.30 , respectively, which are also larger than those based on convective precipitation. Based on zonal means, these correlations increase to -0.51 and -0.44 . Most, but not all, models yield larger corresponding correlations when percentage changes are used, as opposed to raw differences.

Similarly, the spatial correlations between the change in sulphate wet deposition and precipitation are largest with large-scale precipitation, ranging from 0.20 to 0.46 , with a multi-model mean of 0.30 (Supplementary Table 7). The corresponding model mean correlation based on zonal means increases to 0.60 . Based on convective precipitation, this correlation is much weaker at 0.13 . Positive, but weaker, model mean zonal mean correlations also exist between the change in large-scale precipitation and BC and POM wet deposition at 0.40 and 0.30 , respectively. The corresponding model mean zonal mean correlations between the percentage change in both large-scale precipitation and wet deposition are much larger, at 0.72 for SO₄, 0.67 for BC and 0.58 for POM. These results suggest that areas with a decrease in large-scale precipitation also tend to experience a decrease in wet removal and an increase in burden (and vice versa).

We now focus more specifically on what drives the increase in aerosol burden, most of which occurs over the tropics and Northern Hemisphere (NH) midlatitudes (30° – 60° N; Supplementary Figs 2–4). Figure 1 shows that the increase in sulphate burden is largest during June, July and August (JJA) over the NH midlatitudes (Supplementary Fig. 5 shows the other models). This is associated with a maximum reduction in JJA wet deposition over NH midlatitude land. Most of this decrease is due to a wet removal by large-scale precipitation (based on HadGEM2 and CAM5; Supplementary Figs 6 and 7). Many models also show a corresponding increase in wet deposition over NH midlatitude ocean. Furthermore, the NH midlatitude land experiences a maximum decrease in JJA sulphate-burden weighted large-scale precipitation (weighted by the control experiment's burden). Similar, but weaker results exist over the tropical land masses throughout most of the year. Sulphate-burden weighted convective precipitation shows weaker, if not opposite, changes. The increase in sulphate burden is not caused by an increase in chemical production (Supplementary Fig. 8). Similar results generally exist for other aerosol species, including BC and POM (Supplementary Figs 9 and 10). Further CAM4/5 simulations with more extreme global warming continue to support an increase in aerosol burden in response to warming, and the importance of a decrease in large-scale precipitation and wet removal (particularly due to large-scale precipitation) in driving this increase (Supplementary Figs 11 and 12).

Similar to total precipitation, global warming is associated with a global mean increase in large-scale (and convective) precipitation. However, Fig. 2 shows that the NH midlatitude and, to a lesser extent, tropical land masses feature a significant decrease in large-scale precipitation, particularly during JJA. GFDL, HadGEM2 and MIROC-CHEM yield a decrease in JJA NH midlatitude large-scale precipitation of $\sim 40\%$; CAM4/5 yield a 20 – 30% decrease (Supplementary Fig. 13). GISS-E2-R-p3 yields a weak decrease, consistent with its weaker increase in burden (Table 1). This is a robust response to global warming across the ACCMIP models, as well as CMIP5 models, where the multi-model mean shows a $\sim 20\%$ decrease in JJA NH midlatitude large-scale precipitation (Supplementary Fig. 14). Models also yield significant, but smaller, decreases in the frequency of precipitation (based on the number of days per year with more than zero precipitation), particularly over land (Supplementary Fig. 15 and Supplementary Table 1). The CMIP5 JJA multi-model mean shows a 6 – 7% decrease in the large-scale precipitation frequency in the NH midlatitudes. This implies the corresponding decrease in the intensity of precipitation is ~ 13 – 14% (Supplementary Methods), indicating a decrease in the intensity of large-scale precipitation, as opposed to the frequency, is the most important driver. We note, however, that models tend to overestimate the frequency of precipitation (they rain almost every day), implying that they may underestimate the role of precipitation frequency in wet scavenging.

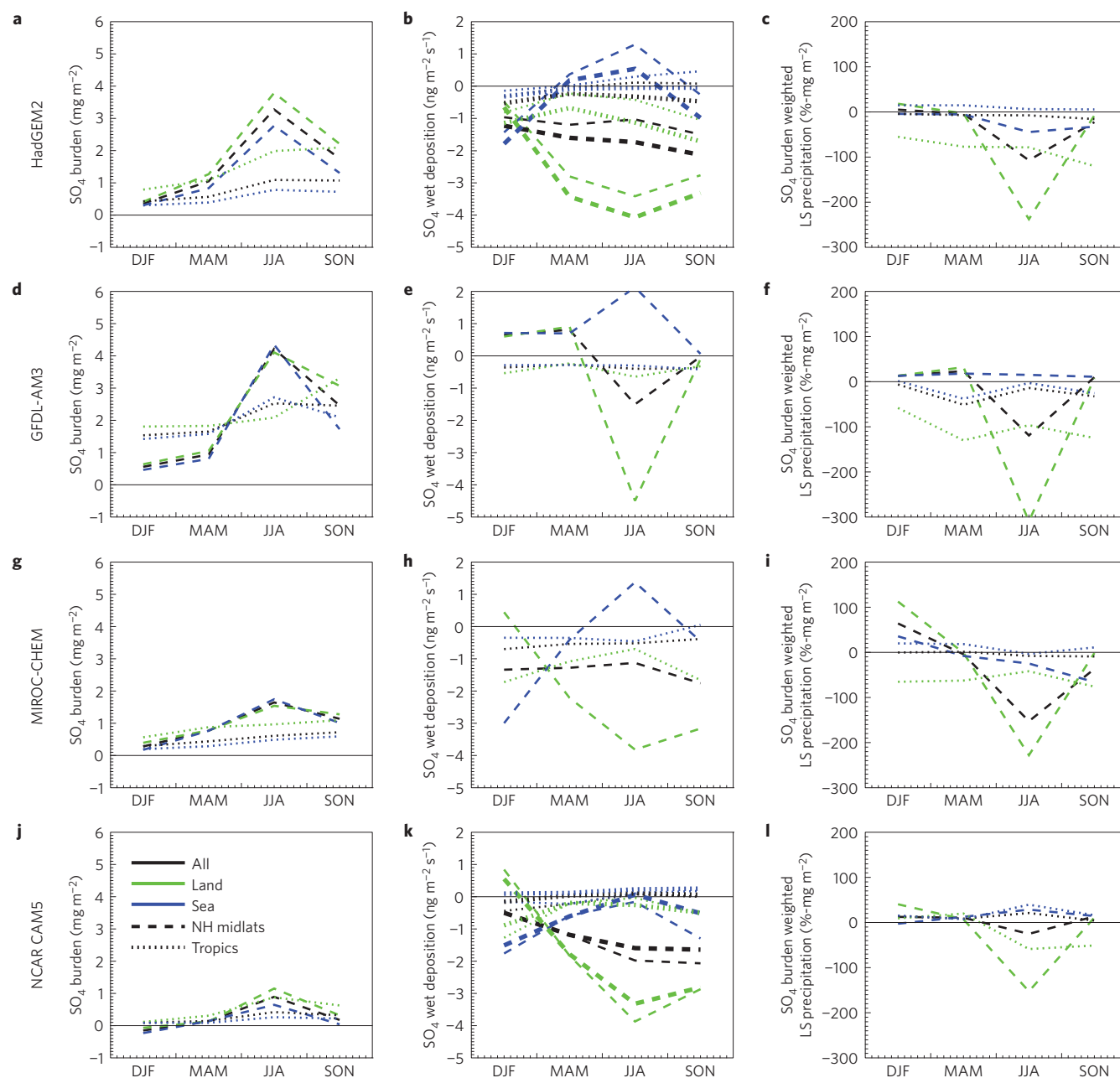


Figure 1 | ACCMIP seasonal sulphate-burden response to climate change. **a–l**, Sulphate burden (mg m^{-2}) (**a,d,g,j**), sulphate wet deposition ($\text{ng m}^{-2} \text{s}^{-1}$) (**b,e,h,k**) and sulphate-burden weighted large-scale (LS) precipitation (\%-mg m^{-2}) (**c,f,i,l**) for HadGEM2 (**a–c**), GFDL-AM3 (**d–f**), MIROC-CHEM (**g–i**) and NCAR CAM5 (**j–l**). Seasonal responses are shown for the NH midlatitudes (30° – 60° N) and the tropics (30° S– 30° N) over land, sea and all grid points. Extra sets of thick lines for HadGEM2 and CAM5 SO_4 wet deposition (**b,k**) represent wet deposition by large-scale precipitation only. DJF: December, January, February; MAM: March, April, May; JJA: June, July, August; SON: September, October, November.

These results can be summarized as follows: the increase in sulphate burden, most of which occurs during JJA, is associated with a decrease in JJA large-scale precipitation (particularly its intensity) over NH midlatitude and tropical land areas where burden is already relatively large. This drives a corresponding decrease in wet deposition, and an increase in sulphate burden, some of which gets transported over the ocean. This leads to an increase in wet deposition over the ocean, but this increase is not large enough to offset the enhanced transport, resulting in a net increase in burden over the oceans as well. We note that, unlike the primary anthropogenic aerosols, the increase in sea salt is largely driven by an increase in surface winds, particularly at high-latitudes (Supplementary Fig. 16).

Figure 3 shows the response of several aerosol species to more extreme global warming experiments in CAM5 (Supplementary Methods). All aerosol species yield a significant, positive slope with respect to surface temperature, indicating an increase in aerosol burden is a robust response to warming. The correlation coefficient between surface temperature and burden is ≥ 0.91 for all aerosol species. Similar results exist for surface concentration (except for secondary organic aerosol) and for the primary anthropogenic aerosol species in CAM4 (Supplementary Figs 17 and 18). Furthermore, global mean wet deposition of SO_4 , BC and POM in CAM5 decreases linearly as the surface warms, all with correlations of -0.99 (Supplementary Fig. 19). This further supports the importance of a decrease in wet removal

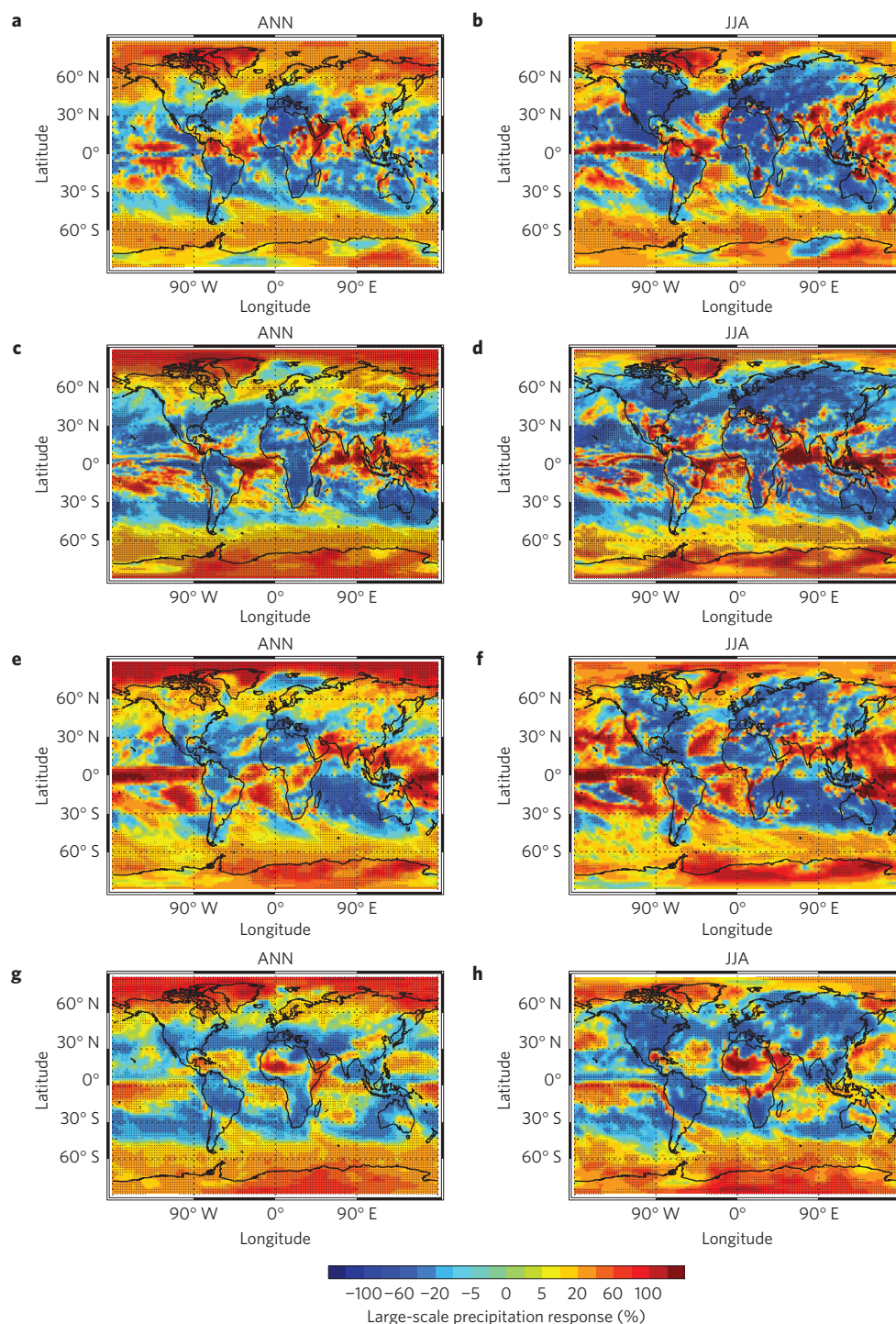


Figure 2 | Percentage change in large-scale precipitation in response to climate change. a–h, Annual (a,c,e,g) and JJA (b,d,f,h) mean for GDFL (a,b), HadGEM2 (c,d), CAM5 (e,f) and MIROC-CHEM (g,h). Symbols represent significance at $\geq 95\%$ confidence level based on a t-test.

as the dominant driver of the increase in aerosol burden as climate warms.

The change in the direct radiative effect of all aerosols due to more extreme warming in CAM5 is also illustrated in Fig. 3. Global warming increases the (negative) direct radiative effect of aerosols (Fig. 3g), which acts as a negative feedback to the imposed warming at $-0.05 \text{ W m}^{-2} \text{ K}^{-1}$. Similar results are obtained with the ACCMIP models (using available 1850, 2000, 2030 and 2100 climate simulations with constant emissions), which yield an average DRE-climate feedback of -0.05 , with a range from

-0.02 to $-0.09 \text{ W m}^{-2} \text{ K}^{-1}$ (Supplementary Fig. 20). More detailed analysis of CAM5 shows that all aerosol species lead to a stronger, more negative DRE. The lone exception is black carbon, which also yields a stronger, but more positive DRE. Anthropogenic aerosols account for $\sim 30\%$ of the total DRE change and natural aerosol account for $\sim 70\%$, with most of this due to sea salt (at 60%). Although the majority of the aerosol-climate DRE feedback in CAM5 is caused by sea salt, ACCMIP models generally yield weaker increases in sea salt, and stronger increases in anthropogenic aerosols (Table 1). Thus, ACCMIP models probably yield a stronger

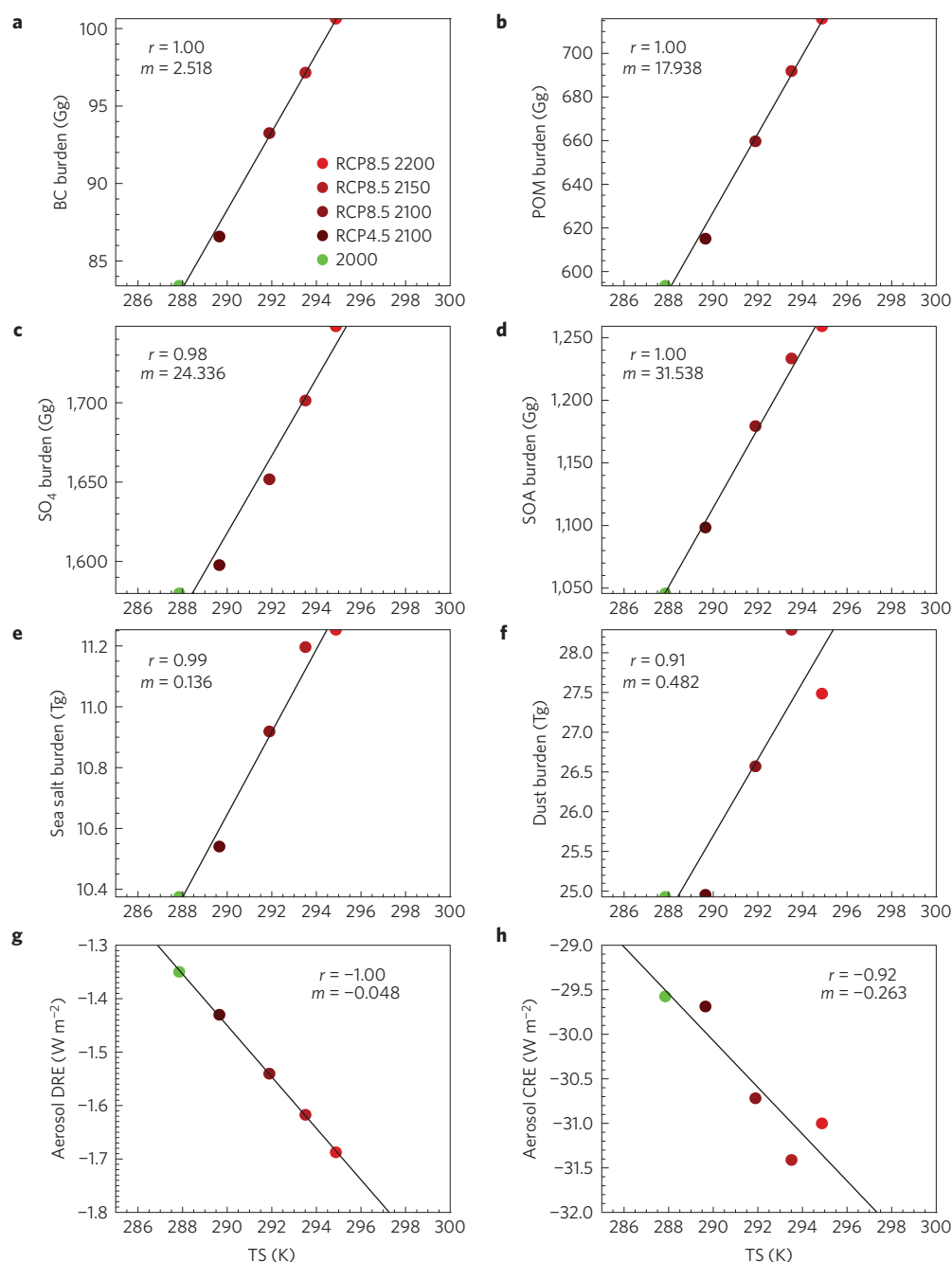


Figure 3 | CAM5 aerosol burden and radiative effects in response to warming. a–f, Global annual mean surface temperature (K) versus burden of black carbon (a), primary organic matter (b), sulphate (c), secondary organic aerosols (d), sea salt (e) and dust (f). **g,** Global annual mean surface temperature (K) versus aerosol DRE. **h,** Global annual mean surface temperature (K) versus aerosol CRE. Experiments are driven with identical emissions, but different climates based on RCP4.5 and 8.5. Also included is the corresponding slope (m) and correlation coefficient (r).

(weaker) anthropogenic (natural) aerosol contribution to the DRE feedback.

Models also yield a significant negative aerosol cloud radiative feedback, which is much larger than the aerosol feedback due to direct effects. CAM5 yields a CRE feedback of -0.26 W m^{-2} per K of warming (Fig. 3h). Similarly, the two ACCMIP models that archived the relevant diagnostics yield a CRE feedback of $-0.43 \text{ W m}^{-2} \text{ K}^{-1}$ for GISS-E2-R-p3 and $-1.90 \text{ W m}^{-2} \text{ K}^{-1}$ for CESM-CAM-superfast (Supplementary Fig. 20e,f). Thus, the total aerosol feedback (DRE+CRE) based on CAM5 and GISS-E2-R-p3 ranges from -0.31 to $-0.46 \text{ W m}^{-2} \text{ K}^{-1}$, most of which is due to aerosol cloud interactions. We note that future anthropogenic

aerosol emissions are projected to decrease, implying the magnitude of this aerosol–climate feedback probably represents an upper limit.

Simulation of aerosol processes and radiative effects—particularly indirect effects—remain uncertain^{20,24}. Nonetheless, we have shown that state-of-the-art climate models simulate a consistent increase in most aerosol species under climate change associated with GHG-induced warming. The bulk of the response is driven by a decrease in large-scale precipitation over land, particularly over the NH midlatitudes during JJA. We conclude that future global warming may exacerbate air quality and drive a negative aerosol–climate feedback.

Received 13 April 2015; accepted 17 September 2015;
published online 9 November 2015

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Acknowledgements

We acknowledge the climate modelling groups participating in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) for producing and making available their model output, and the World Climate Research Programme's Working Group on Coupled Modelling, which is responsible for CMIP. For CMIP the US Department of Energy's Program for Climate Model Diagnosis and Intercomparison provides coordinating support and led development of software infrastructure in partnership with the Global Organization for Earth System Science Portals.

Author contributions

R.J.A. conceived the project, designed the study, carried out the data analysis and led the writing of the paper. W.L. assisted in writing of the paper. S.T.R. provided un-archived HadGEM2 data. All authors discussed the results and commented on the manuscript.

Additional information

Supplementary information is available in the [online version of the paper](#). Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to R.J.A.

Competing financial interests

The authors declare no competing financial interests.