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# **MULTIFACETED AEROSOL EFFECTS ON PRECIPITATION**

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#### **ABSTRACT**

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Aerosols have been proposed to influence precipitation rates and spatial patterns from scales of individual clouds to the globe. However, large uncertainty remains regarding the underlying mechanisms and importance of multiple effects across spatial and temporal scales. Here, we review the evidence and scientific consensus behind these effects, categorised into radiative effects via modification of radiative fluxes and the energy balance, and microphysical effects via modification of cloud droplets and ice crystals. Broad consensus and strong theoretical evidence exist that aerosol radiative effects (aerosol-radiation interactions (ARIs) and aerosol-cloud interactions (ACIs)) act as drivers of precipitation changes because global mean precipitation is constrained by energetics and surface evaporation. Likewise, aerosol radiative effects cause well-documented shifts of large-scale precipitation patterns, such as the Inter-Tropical Convergence Zone (ITCZ). The extent of aerosol effects on precipitation (APEs) at smaller scales is less clear. Although there is broad consensus and strong evidence that aerosol perturbations microphysically increase cloud droplet numbers and decrease droplet sizes, thereby slowing precipitation droplet formation, the overall aerosol effect on precipitation across scales remains highly uncertain. Global cloud resolving models (CRMs) provide opportunities to investigate mechanisms that are currently not well-represented in global climate models (GCMs) and to robustly connect local effects with larger scales. This will increase our confidence in predicted impacts of climate change.

#### INTRODUCTION

Less than three percent of water on Earth sustains life. Precipitation is the most important mechanism delivering fresh water from the atmosphere to the surface. Although climate change discussions are commonly framed in terms of global temperature change, precipitation changes significantly drive actual impacts of climate change on the planet<sup>1,2</sup>.

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A substantial body of literature exists describing the impact of greenhouse gas (GHG) induced warming on precipitation, and the concepts are well understood<sup>2,3</sup>. In contrast, the uncertainty regarding aerosol (nano- to micrometre sized particles suspended in air of anthropogenic or natural origin) effects on precipitation (APEs) remains large. Many hypotheses describe APE based on radiative and cloud microphysical arguments. Some are included in current climate models, others are not (cf. Figure 1, Table 1). Large uncertainty remains regarding the underlying mechanisms and relative importance of proposed effects across spatial and temporal scales.

This article builds on the results of an expert workshop held under the auspices of the Global Energy and Water cycle Exchanges (GEWEX) Aerosol Precipitation (GAP) initiative<sup>4</sup>. It critically reviews the current evidence and scientific consensus (in the authors' view) for APEs and their proposed mechanisms. To facilitate this assessment, we categorise mechanisms according to their degree of scientific support: Category A: strong evidence / broad consensus; Category B: some evidence / limited consensus; Category C: hypothesised / no consensus.

#### THE PHYSICAL MECHANISMS OF AEROSOL EFFECTS ON PRECIPITATION

The physical drivers of APE can be categorised into i) radiative effects via modification of radiative fluxes and the energy balance, which occur due to aerosol scattering and absorption, and modification of cloud radiative properties by ii) microphysical effects via modification of cloud droplet and ice crystal number, size and morphology that can affect growth to precipitation-size particles, as well as latent heat from phase changes (enthalpy of vaporisation or fusion). All these effects can induce dynamical feedbacks across scales.

In addition to this mechanistic (bottom up) view, conservation laws provide a complementary (top down) perspective: conservation of energy constrains global mean precipitation<sup>5-7</sup>, as changes in latent heat of condensation (L) associated with precipitation changes (dP) have to be compensated by opposite changes in net column integrated cooling (dQ) through adjustment of net surface or top-of-atmosphere fluxes, and vice versa. At smaller spatial scales, net latent heating associated with precipitation changes can also be balanced through divergence of dry static energy<sup>5,8-10</sup> ( $d(\nabla \cdot \underline{u}s)$ ) (column integrated, with  $\underline{u}$  horizontal velocity, neglecting changes in energy and liquid or solid water storage and kinetic energy transport), as illustrated in Figure 2:

$$L dP = dQ + d(\nabla \cdot us)$$

Conservation of water provides additional constraints. In the global mean and for sufficiently long time-scales, precipitation P must be balanced by evaporation E so P-E=0. On smaller spatial scales, moisture ( $q_v$ ) flux convergence can compensate for imbalances in P-E so that:

$$dP - dE = -d(\nabla \cdot uq_v)$$

This implies the existence of *breakdown scales of budgetary constraints on precipitation* - a scale below which energy and water budget constraints on precipitation do not strictly apply due to efficient horizontal transport<sup>11</sup>. In the extra-tropics, this scale is expected to be related to the first baroclinic Rossby radius of deformation ( $L = \frac{NH}{\pi f_0} \approx 1000 km$ , where N is the Brunt–Väisälä frequency, H is the scale height, and  $f_0$  is the Coriolis parameter). This latitudinally dependent precipitation constraint on aerosol perturbations implies varying effects in the tropics and extra-tropics (Figure 3). Even for regional aerosol perturbations, energetic constraints apply to the global mean. Reductions in surface insolation and atmospheric heating by aerosol absorption decrease global mean precipitation in both simulations, with teleconnections in the tropical simulation.

Evidence from climate models shows that localised aerosol absorption could affect tropical precipitation over thousands of kilometres<sup>12</sup>. Similar scale arguments apply to the moisture budget, with limitations on moisture convergence constraining the susceptibility of regional APEs<sup>13</sup>. The combination of energy and water budget constraints (smallest closure scale) yields a characteristic scale for regional precipitation responses<sup>11</sup> of 3000 km to localized aerosol perturbations, similar to scales of weather systems<sup>14</sup>.

- It is important to note that this budgetary framework does not provide direct constraints on precipitation intensity distributions, despite constraints on its mean. APEs could invoke an additional feedback mechanism through the radiative effects of atmospheric humidity and clouds<sup>15</sup>. Combined, energy and moisture budget constraints can provide physical mechanisms underpinning the "buffering" of APEs<sup>16</sup> in equilibrium conditions, which is also related to radiative convective equilibrium concepts<sup>17-19</sup>.
- APEs can be decomposed into adjustments due to instantaneous atmospheric net diabatic heating, including rapid adjustments of the vertical structure of water vapour, temperature and clouds (hours to days), and a slower response mediated by surface temperature changes <sup>6,20,21</sup> defined as "hydrological sensitivity"<sup>9,22</sup>. Due to difficulties in separating fast surface temperature changes (days to months) from rapid adjustments in climate models, these are commonly considered jointly<sup>20,21</sup>.
- Finally, both radiative and microphysical effects and associated changes to the regional energy balance can lead to dynamical effects and regional circulation changes with concomitant changes in precipitation<sup>23,24</sup>.
- We now discuss each potential mechanism underlying APEs and assess their evidence and scientific consensus.

#### RADIATIVE EFFECTS

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- i. *Surface ENERGY BUDGET* ARIs and ACIs modulate radiative surface fluxes and, consequently, sensible and latent heat fluxes. These effects generally reduce surface insolation, decreasing surface evaporation which has been linked to a "spin down" of the hydrological cycle<sup>25</sup>. This is corroborated by the observed precipitation response to ARIs following major volcanic eruptions, showing substantial decreases in precipitation over land and river discharge into ocean<sup>26,27</sup>. (Near-surface absorbing aerosol can enhance precipitation through diabatic heating, even when surface sensible heat fluxes are reduced<sup>28</sup>.) Energetically, the net-negative total ARIs<sup>29</sup> reduce the global mean temperature, atmospheric water vapour, and associated long-wave emission, which is compensated by reductions in precipitation and associated latent heat: climate models show that negative aerosol radiative forcing masks almost all temperature-driven GHG effects on precipitation over land up to present (with GHG effects dominating the future)<sup>9,30,31</sup>. However, such radiative arguments cannot be decoupled from dynamical feedbacks, as shown below.
- 109 That ARIs reduce global precipitation through changes in surface temperature and surface fluxes 110 builds on our physical understanding of the energy budget, is supported by observational evidence<sup>32</sup> 111 and reproduced by climate models. We assess this effect as Category A, supported by strong evidence 112 and broad scientific consensus, although magnitudinal uncertainties remain.
- 113 The following two mechanisms could be combined as aerosol absorption effects, but we retain the mechanistic separation prevailing in existing literature.
- ii. **Atmospheric diabatic heating** by aerosol absorption creates local energetic imbalances. To ensure energy conservation, this is compensated by reductions in latent heat release through precipitation, by rapid adjustments of net surface or top-of-atmosphere fluxes, or, on smaller scales or in the tropics<sup>11,33</sup>, through divergence of dry static energy<sup>8,34</sup>. The energetic framework provides a useful tool to diagnose APEs<sup>9,21,28,34,35</sup> and can explain the contrasting behaviour of absorbing and non-absorbing aerosols<sup>21,36</sup>.
- That diabatic heating of absorbing aerosol reduces global mean precipitation is consistent with our physical understanding of the energy budget, is reproduced by climate models but builds on limited observational evidence. We therefore assess this effect as Category A, supported by strong evidence and broad scientific consensus but with remaining magnitudinal uncertainties.
- 125 *iii.* **Semi-direct effects**<sup>9,37-40</sup> are rapid adjustments associated with aerosol absorption affecting the vertical temperature and humidity structure, with potential effects on clouds and precipitation.
  127 These effects are generally accompanied by corresponding surface flux changes (cf. ii). Elevated layers of absorbing aerosol can modify lower-tropospheric static stability and sub-tropical inversion strength<sup>39,41</sup>, suppressing boundary layer deepening and concomitant entrainment<sup>42</sup>. Although the focus has been on shallow clouds<sup>43</sup>, the impact on deep convection and associated precipitation has

been demonstrated in CRMs, revealing a complex diurnal cycle<sup>44</sup>, and climate models<sup>28</sup>. However, most prior research focused on semi-direct effects of shallow clouds in the context radiative forcing<sup>43</sup>, not precipitation. Hence, the overall uncertainty remains large.

Semi-direct effects of absorbing aerosol on the thermodynamic structure of the atmosphere are 134 based on a sound physical foundation and have been well documented. However, the sign and 135 136 magnitude of the effect on clouds and subsequently precipitation are sensitive to the vertical 137 collocation of clouds and aerosols as well as the cloud regime. Some consistency exists across CRM studies, however, the observational evidence remains limited. We therefore assess this effect as 138 Category B, backed up by physical conceptual models, modelling studies and limited observational 139 140 evidence and some scientific consensus, even if the magnitude and sign of the impact on 141 precipitation remain unclear.

The following mechanisms iv) - vi) could be combined as aerosol effects on regional precipitation patterns but we retain the mechanistic separation prevailing in existing literature.

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iv. *Changes in regional-scale precipitation and monsoon dynamics* have been attributed to regional patterns in ARI-induced surface cooling and atmospheric heating, both locally and remotely <sup>12,34,45-146</sup>. The precipitation response can be attributed to a combination of the modulation of surface fluxes over land, hence of the thermal gradient between land and sea<sup>50,51</sup>, as well as aerosol absorption effects, driving thermally direct circulations<sup>12,52</sup> and moisture convergence<sup>52</sup> (linked to extreme precipitation <sup>53,54</sup>), the sea breeze circulation<sup>55</sup>, and teleconnections<sup>56</sup>.

Aerosol effects on regional scale precipitation and monsoon dynamics have been shown to affect precipitation patterns. This builds on climate model and CRM simulations and general physical understanding, with some observational evidence. However, uncertainties remain regarding the attribution of observed precipitation to aerosol effects and overall strength of the effects. We therefore assess this effect as Category B, backed by some evidence and limited scientific consensus.

v. Aerosol radiative effects on sea surface temperature patterns (SSTs) have been linked to observed climatological trends<sup>57,58</sup>. Associated changes in multi-decadal SST variability<sup>59</sup> have previously been linked to the Sahel drought<sup>60-63</sup>. In addition to the local effects on the SST distribution, aerosols may also affect ocean dynamics and thereby SSTs. For example, aerosol forcing was shown to strengthen the Atlantic Meridional Overturning Circulation (AMOC) thereby modulating SST patterns in the Atlantic Ocean<sup>64-67</sup>, and affecting the Northern Hemisphere climate and precipitation patterns<sup>63,68</sup>. SSTs also control hurricane activity<sup>61,69-71</sup>, providing a mechanism for potential aerosol effects on hurricanes<sup>72,73</sup>. Forcing trends associated with European sulfuremissions as aerosol precursor, have been linked to a pronounced North Atlantic "hurricane drought" from the 1960s through early 1990s<sup>74</sup> during which hurricane power dissipation, a measure of storm damage<sup>75</sup>, was strongly inversely correlated with European sulfur emissions. Much of the direct SST forcing was from Saharan mineral dust, which in turn was associated with reduced monsoonal flow resulting from high sulfate aerosol concentrations<sup>76</sup>.

The SST mediated effect of aerosol on regional precipitation patterns and hurricane activity builds on climate model simulations and general physical understanding, with limited observational evidence. We therefore assess this effect as Category B, backed up by some evidence and limited scientific consensus.

172 vi. Hemispheric asymmetry in aerosol radiative effects<sup>77</sup> shifts the energy flux equator to where the column-integrated meridional energy flux vanishes<sup>78,79</sup>. The position of the energy flux equator is 173 174 closely linked to the ITCZ position and associated precipitation. With anthropogenic aerosol predominantly located in the northern hemisphere, associated negative/positive aerosol radiative 175 effects, e.g. from sulfate/black carbon, lead to a southward/northward ITCZ shift<sup>62,78-87</sup>. For sulfate, 176 177 this is a slow (SST mediated) response, whereas for black carbon adjustments in response to absorption contribute88. Dynamical cloud feedbacks can further amplify the hemispheric 178 asymmetry<sup>89</sup> and ITCZ shifts can interact with local monsoon regimes<sup>90</sup>. 179

The effect of hemispherically asymmetric aerosol radiative effects on the energy flux equator and ITCZ position builds on a robust theoretical foundation<sup>79</sup>, agrees with observational evidence<sup>83,91</sup> and

is reliably reproduced by GCMs. We therefore assess this effect as Category A, backed up by strong 182 evidence and broad scientific consensus. 183

#### MICROPHYSICAL EFFECTS

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185 vii. CCN mediated effects on stratiform liquid clouds, including stratocumulus: enhanced loading of CCN (hygroscopic or wettable aerosols of sufficient size to facilitate droplet growth) can increase cloud droplet numbers and, at constant liquid water content, lead to smaller droplets. This effect saturates for high aerosol concentrations<sup>92</sup> and/or low updraft velocities due to the depletion of supersaturation by condensation. This pathway can slow droplet growth to the threshold size for precipitation<sup>93-96</sup>, thereby supressing precipitation efficiency; this mechanism can also apply to warm phase of stratiform mixed-phase clouds<sup>97</sup>. The reduced removal of cloud water by precipitation has been hypothesized to increase cloud liquid water path (LWP) and lifetime95. There is clear observational evidence of an increase in cloud droplet numbers and associated decrease in droplet radii due to aerosol perturbations from aircraft data98, ship-track observations99-103 and satellite remote sensing<sup>104-106</sup>. This is reproduced in CRMs and qualitatively in climate models<sup>105,107</sup>. Analysis of satellite-retrieved CloudSat<sup>108</sup> radar reflectivity and MODIS<sup>109</sup> effective radius data provides observational evidence for droplet size dependence of precipitation onset, with enhanced (low) drizzle rates above effective radii of 15 (10) µm. Combined with the documented impact of CCN on effective radii, this indicates warm rain susceptibility to CCN perturbations<sup>110</sup>. These observations are limited to liquid-top shallow clouds, which represent a small fraction of global mean precipitation<sup>111</sup>. The observational evidence for an increase in liquid water paths via precipitation suppression due to increased aerosol concentrations is still disputed and cloud-regime dependent<sup>101,112-114</sup>. Many climate models simulatestrong LWP responses to aerosol perturbations<sup>112,115</sup>, likely because their simplified representations of warm rain formation ("autoconversion") have built-in power-law dependences on cloud droplet number but lack smallscale feedbacks, such droplet size effects on evaporation and associated cloud entrainment feedbacks<sup>16,116,117</sup>. This uncertainty propagates into climate model assessments of APEs.

CCN mediated effects on stratiform liquid cloud, including stratocumulus, have been shown to increase droplet numbers and suppress warm rain formation. This is consistent with warm rain formation theory, supported by observational evidence from space-born cloud radars and reproduced by high-resolution CRMs. The expected effect is reduced light rain occurrence, possibly compensated by increasing occurrence of stronger rain events. However, the overall impact on largescale precipitation remains unclear. We therefore assess this effect as Category B, backed up by some evidence and limited scientific consensus.

The following mechanisms viii) and ix) could be combined as aerosol effects on convection but we 215 retain the mechanistic separation by cloud phase prevailing in existing literature. 216

217 viii. **CCN mediated effects on shallow convection:** for shallow (liquid) convective clouds, an aerosol mediated increase in cloud droplet numbers has several effects: associated smaller droplet radii enhance evaporation that increases the buoyancy gradient at the cloud edge, creating vorticity and increasing associated entrainment/detrainment<sup>116</sup>, which results in a reduction of cloud size, liquid water path, buoyancy and precipitation. At the same time, suppression of rain production via the droplet number effect on autoconversion can produce enhanced condensation and latent heat release due to larger numbers of remaining cloud droplets and associated increase in surface area, often referred to as "warm phase or condensational invigoration" 118-120. It can also enhance cloudtop detrainment; subsequent evaporative cooling can destabilize the environment<sup>121</sup>. Both mechanisms could generate deeper clouds<sup>122</sup> with potentially enhanced precipitation. The net effect on mean precipitation could therefore be small 16,17 or even positive, depending on environmental conditions: high-resolution large-eddy simulations demonstrate a non-monotonic precipitation response with increases at low aerosol concentrations up to an optimal aerosol concentration, followed by a precipitation decrease<sup>118-120,123-125</sup>. For larger spatio-temporal scales, idealised simulations of shallow convection approach a radiative-convective equilibrium state<sup>17</sup>. Although the transient behaviour approaching equilibrium responds to increasing cloud droplet number

concentrations through deepening and delays precipitation onset<sup>126</sup>, in the equilibrium state associated decreases in relative humidity and faster evaporation of small clouds compensates for much of the radiative effects with broader intensity precipitation distributions<sup>19</sup>. The overall effect depends on the relative importance of transient and equilibrium states<sup>17,93,127</sup> with recent evidence highlighting limitations of idealised simulations that unrealistically favour equilibrium states<sup>128</sup>. However, contrasting environmental factors, such as boundary layer development or humidity, can influence the overall effects<sup>123,129</sup>.

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CCN mediated effects on shallow convection have been shown to increase droplet numbers and slow warm-phase precipitation formation. This is based on high-resolution CRMs and observational evidence. It is important to note that convection parameterisations in most GCMs do not represent any microphysical aerosol effects on convection. The overall effect on precipitation is less certain. We assess this effect as Category B, backed up by some evidence and limited scientific consensus.

CCN mediated effects on deep convection: for deep (liquid & ice phase) convective clouds, ix. "convective invigoration" is widely discussed, generally referring to enhanced aerosol levels causing stronger updrafts or higher clouds and an associated increase in precipitation<sup>93,98,130-136</sup>. Several hypotheses about underlying mechanisms exist. Often overlooked, these share a common starting point with shallow convection in the liquid base of clouds: the suppression of warm rain formation from reduced autoconversion with enhanced CCN in the lower, liquid part of the cloud<sup>137,138</sup>, with an associated reduction in droplet size and resulting entrainment/detrainment feedbacks. Subsequent invigoration hypotheses include: enhanced condensation and associated latent heat release ("warm phase invigoration", c.f. viii)118,119,139,140; enhanced evaporation and downdraft formation affecting cold pool strength and surface convergence 141,142; delay of warm-phase precipitation increasing the amount of cloud water reaching the freezing level, enhancing the release of latent heat of freezing<sup>93,98,132</sup> although the importance of this ("cold phase invigoration") is disputed<sup>143</sup>; the hypothesis that depletion of cloud water through precipitation in low aerosol environments could generate high supersaturations and subsequent activation of small aerosol particles into cloud droplets, enhancing condensation and (warm phase) latent heat release 144 – a hypothesis shown to be inconsistent with a limited set of observations<sup>145</sup>; and that enhanced CCN levels increase environmental humidity through clouds mixing more condensed water into the surrounding air, preconditioning the environment for invigorated convection<sup>146</sup>. The latter result is likely a consequence of idealised equilibrium simulations as it is not observed in realistic simulations across a wide range of environmental conditions<sup>147</sup>. Feedbacks between convective clouds and their thermodynamic environment may modulate or buffer APEs. Overall, the strength and relative importance of mechanisms underlying convective invigoration are disputed 143 – it is sensitive to uncertain microphysical effects<sup>148,149</sup> and strongly dependent on environmental regimes<sup>49,130,141,150</sup>-<sup>152</sup>. In addition, the excess buoyancy associated with the respective mechanisms can be partially offset by negative buoyancy associated with condensate loading 153,154, with the net effect dependent on condensate offloading through precipitation. The role of condensate loading has been explored through theoretical calculations that show the potential of aerosol-induced invigoration is significantly limited for cold-based storms, and that aerosol-induced cold-phase processes weaken, rather than strengthen the updrafts in warm-based storms (referred to as aerosol enervation)<sup>155</sup>. The first systematic multi-model assessment of these competing aerosol effects on deep convective updrafts<sup>154</sup> has been performed as part of a deep convection case study<sup>137</sup> over Houston, USA, under the umbrella of the Aerosol, Cloud, Precipitation, and Climate initiative (Figure 4). This intercomparison revealed updraft increases by 5%-15% in the mid-storm regions (4-7 km above ground) with increased CCN, primarily driven by enhanced condensation, with waning and mixed difference in levels above. Condensate loading contributions are generally limited. Despite this apparent invigoration, 6 of 7 models produce precipitation decreases (of -10% to -80%), highlighting the complexity of precipitation responses to aerosol perturbations. There are indications that microphysical effects strengthen deep and weaken shallow clouds in convective cloud fields, thereby broadening the precipitation intensity distribution 18,44. Observations and modelling suggest a nonmonotonic effect, with precipitation peaking at an optimal aerosol concentration<sup>156,157</sup>. It should be re-iterated that even high-resolution CRM simulations of aerosol effects on deep convection remain subject to large uncertainty, particularly with mixed-phase and ice-cloud microphysics, affecting the simulated base states as well as their response to aerosol perturbations<sup>137,148,158</sup> (Figure 4). Few current climate models include aerosol aware convection parameterisations and their early results indicate limited aerosol effects on convective precipitation on the global scale<sup>159,160</sup>. However, the associated uncertainties remain large, providing challenges for the next generation of cloud resolving climate models.

CCN mediated effects on deep convection consistently show increased droplet numbers and reduced warm rain formation in the lower parts of the cloud. This builds on a robust theoretical foundation, is supported by limited observations and is consistently reproduced by CRMs. The propagation of these perturbations through the mixed- and ice-phase microphysics of clouds remains uncertain across models, with limited observational constraints. Severally potheses exist on associated changes in buoyancies leading to invigoration, with models consistently simulating an increase in latent heating of condensation due to the increased surface area of enhanced droplet numbers. However, their importance remains highly uncertain. The overall effect on aggregated precipitation remains highly uncertain. We therefore assess this effect as Category C, backed up by plausible hypotheses, but with limited evidence and limited scientific consensus.

x. *INP mediated effects* on clouds are likely to be significant, but still highly uncertain, given the unknown proportion of cloud ice between -38°C and 0°C that forms by INP-induced heterogeneous freezing or remains supercooled. Clouds glaciate below approximately -38°C, where droplets freeze homogeneously. Increased concentrations of INPs (generally solid or crystalline aerosols which provide a surface onto which water molecules are likely to adsorb, bond and form ice-like aggregates) have been proposed to enhance the glaciation of clouds<sup>97,161,162</sup> with an associated increase in precipitation efficiency and reduction of cloud lifetime<sup>163</sup>. Low INP concentrations in remote marine environments consistently inhibit precipitation<sup>164</sup>. However, the complexity of microphysical pathways in mixed- and ice-phase clouds is significant<sup>149</sup> with potential compensating pathways buffering the response, leading to low precipitation susceptibility<sup>165</sup>. Modification of precipitation through controlled INP emissions ("cloud seeding") has been extensively attempted in the weather modification community, with demonstrated impact on cloud microphysical processes<sup>166</sup>; however, limited evidence exists for its effectiveness in terms of large-scale precipitation modulation<sup>167,168</sup>. The role of INPs is further complicated by secondary ice production processes that are ill-constrained but can lead to rapid cloud glaciation<sup>169</sup>.

INP mediated effects have been shown to affect cloud phase and microphysics. A number of hypotheses exist on subsequent effects on precipitation. However, there is no complete theoretical framework, and evidence from modelling and observations is limited. We therefore assess this effect as Category C, backed up by plausible hypotheses, but only limited evidence and limited scientific consensus.

322 It is important to re-iterate that occurrence and strength, and spatiotemporal extent, of radiative and 323 microphysical APEs are modulated by environmental conditions<sup>49,142,150,170,171</sup> as well as energy/water 324 budget constraints<sup>11,33,36</sup>, which complicates their detectability. Also, the potential exists for 325 compensation between individual mechanisms, buffering the overall precipitation response<sup>16</sup>.

#### **DETECTABILITY AND ATTRIBUTION OF PRECIPITATION CHANGES**

In-situ observations provide the most detailed insights into processes underlying APEs and are invaluable for the development and evaluation of theories and models. However, due to the inhomogeneous and intermittent nature of precipitation it is generally impossible to measure areal average precipitation reliably. Representation errors<sup>172</sup> are likely to exceed the expected magnitude of aerosol effects.

Statistical analysis of satellite-retrieved aerosol radiative properties and precipitation shows higher precipitation rates with higher aerosol optical depth<sup>134</sup> with potentially non-monotonic behaviour<sup>173</sup>.

Confounding factors (as aerosol extinction, cloud and precipitation are controlled by common factors, such as relative humidity<sup>174</sup>, and precipitation is the predominant aerosol sink<sup>175</sup>) complicate the interpretation. More fundamentally, remotely sensed aerosol properties are not always representative of the relevant aerosol perturbations<sup>176</sup> and statistical analyses rely on assumptions of spatial representativeness of not co-located retrievals<sup>177,178</sup>. However, satellites provide the only source for global observational constraints and the abundance of data permits robust statistical relationships. When environmental conditions are controlled for<sup>179</sup>, the apparent increase in precipitation with aerosol extinction is significantly reduced, although a positive relationship remains for cloud regimes<sup>179-181</sup> with tops colder than 0°C, suggesting a role of ice processes<sup>180</sup>. Furthermore, satellite data provide constraints on microphysical processes: TRMM and CloudSat observations show a systematic shift in the relationship between rain drop size distribution and liquid water path with enhanced aerosol concentrations off the coast of Asia<sup>182</sup>.

Situations with well-characterised aerosol perturbations can serve as analogues for APEs<sup>183</sup>. Aerosols emitted from point sources, such as ships, volcanoes, industrial sites, or cities, can cause distinct tracks in clouds that can be analysed from satellite data<sup>101,184,185</sup>, even when invisible<sup>186</sup>. The analysis of cloud droplet size in ship-track data shows a consistent effective radius reduction in the track<sup>99,113</sup>, consistent with observed effective radii reductions in response to SO<sub>2</sub> emissions from a degassing volcano<sup>187</sup>. In general, cloud droplet effective radius is expected to be positively correlated with precipitation formation through warm rain formation<sup>188</sup>. However, the precipitation in ship-tracks reveals a differentiated response across cloud regimes<sup>113</sup>. Satellite observations of lightning enhancement over shipping lanes<sup>189</sup> also provide strong indications of aerosol effects on convective microphysics and potential aerosol-driven mesoscale circulations, although APEs itself remain more elusive<sup>190</sup> and contributions from dynamical factors cannot be ruled out.

The difficulty remains to consistently reconcile observations with modelling data: any shift in the precipitation intensity distribution also implies a shift in the fraction of rain detectable from radar or microwave data<sup>191</sup>. Also, the formation of detectable perturbations in clouds is limited to a sub-set of environmental conditions<sup>102,186</sup> with overall limited precipitation amounts, thereby limiting the global representativeness of such observations.

On larger scales, observational uncertainty and low signal-to-noise ratios complicate the attribution of observed changes of regional APEs<sup>192</sup>. Detection and attribution techniques<sup>193</sup> use GCMs to estimate spatio-temporal response patterns ("fingerprints") of precipitation to aerosol perturbations, which then can be compared to observed precipitation changes. However, observational and modelling uncertainties still obscure unambiguous evidence of such fingerprints of aerosol on regional scale precipitation<sup>194-196</sup>.

## CONCLUSIONS

This article reviews the evidence and scientific consensus for APEs and the underlying set of physical mechanisms. Broad consensus and strong theoretical evidence exists that because global mean precipitation is constrained by conservation of energy<sup>6</sup> and water<sup>11,13</sup> as well as surface evaporation<sup>25</sup>, aerosol radiative effects act as direct drivers of precipitation changes<sup>8</sup>. Likewise, aerosol radiative effects cause well-documented shifts of large-scale precipitation patterns, such as the ITCZ. The extent to which APEs are i) applicable to smaller scales and ii) driven or buffered by compensating microphysical and dynamical mechanisms and budgetary constraints is less clear. Despite broad consensus and strong evidence that suitable aerosols increase cloud droplet numbers and reduce warm rain formation efficiencies across cloud regimes, the overall aerosol effect on cloud microphysics and dynamics, as well as the subsequent impact on local, regional and global precipitation, is less constrained. Air-pollution control measures will reduce aerosol levels in the future, with an expected reversal of aerosol effects on regional precipitation patterns<sup>197</sup>.

Research on APEs has been limited by the fact that: locally to regionally, precipitation is controlled by complex non-linear interactions with multiple microphysical, radiative and dynamical feedbacks; the expected aerosol-induced change in precipitation is potentially smaller than the internal variability<sup>198</sup>

and uncertainty in current observations; current observations can only constrain some of the processes involved – satellite retrievals are often limited to proxies of the parameters involved and insitu measurements are limited, in particular in convective updrafts; isolating causal effects of aerosol on precipitation in the presence of multiple confounding variables remains challenging – it is easier to identify a strong "effect" than to prove that it is the consequence of confounding; and finally, because the representation of clouds in current climate models is inadequate to represent key microphysical processes and, importantly, the coupling between microphysics and cloud dynamics. Consequently, significant uncertainty remains, limiting our ability to quantify and predict past and future precipitation changes.

It should be emphasised that, in terms of local impacts on humans and ecosystems, absolute precipitation changes are likely to be less important than relative precipitation changes in the mean and in the frequency of occurrence of extremes. To illustrate this point, the absolute precipitation changes over the Sahel region simulated by the CMIP6 multi-model intercomparison seem negligible – but constitute ~ 40% of the local precipitation (Figure 1). Likewise, local impacts may be dominated by regional shifts of precipitation patterns rather than precipitation process changes. These aspects have not been given sufficient attention.

#### **NEW FRONTIERS**

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Out of ten mechanisms reviewed, only three have been assessed to be supported by strong evidence and broad consensus and two primarily based on hypotheses without consensus (Table 1). Future research should define critical tests for numerical models based on observations, in particular of convective updraft microphysics and thermodynamics, including observational simulators for comparability. Active remote sensing and systematic in-situ observations<sup>199,200</sup>, including from uncrewed aerial vehicles, will provide novel constraints on particularly uncertain mixed-phase cloud microphysics and dynamics. Advanced geostationary satellites and cube-sat fleets will allow monitoring the full cloud life cycle. Idealised aqua-planet<sup>33,201</sup> or radiative convective equilibrium simulations<sup>18,202</sup>, such as the GAP Radiative Convective Equilibrium aerosol perturbation model intercomparison<sup>140</sup>, connect evidence from local scale effects to regional and global precipitation. The availability of global CRMs<sup>203</sup> and digital twin Earths<sup>204</sup> provides significant opportunities to overcome our reliance on climate models with parameterised local-scale processes and inadequate microphysics, that currently do not represent three of the ten mechanisms reviewed here (Table 1). However, even CRMs have large uncertainties in cloud microphysical processes that can obscure aerosol effects<sup>148</sup> and remain to be systematically constrained by observations. The shift to global CRMs, which will be a focus of the GAP initiative<sup>4</sup>, will also allow for robust quantification of the connection between local ACIs and large-scale dynamical feedbacks and teleconnections.

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## **COMPETING INTERESTS DECLARATION**

The authors declare no competing interests.

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## **AUTHOR CONTRIBUTION STATEMENTS**

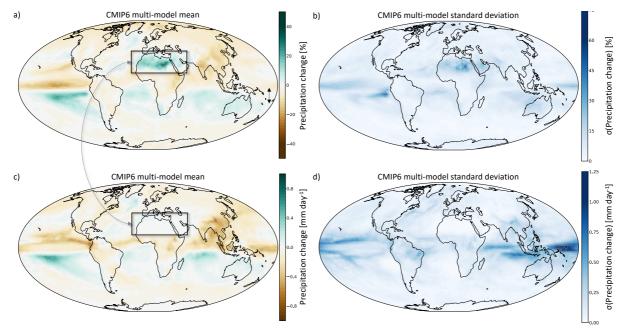
P.S. and S. vd H. developed the structure of the GEWEX Aerosol Precipitation Initiative workshop programme providing the basis for this review paper. M. W.C and E. G. served as raporteurs provided detailed meeting notes. P.S. and S. vd H. drafted the first version of the manuscript that was extended upon with contributions from all authors to the literature review, the synthesis of the results and revising the manuscript. G.D. created the figures in Figure 1 and Figure 3. S.M.S created Figure 4.

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# **TABLES & FIGURES**

**Table 1:** Assessment of the effect of increasing aerosol on precipitation. Microphysical and radiative pathways are distinguished in the second column. Columns 3 and 4 indicate the expected effect on mean precipitation or the intensity distribution; column 5 indicates whether the effect is included in current generation (CMIP6) climate models. The scientific consensus (A strong evidence / broad consensus; B some evidence / limited consensus; C hypothesised / no consensus) is summarized in column 6.

Physical driver of aerosol effect on precipitation	Pathway	Effect on mean	Effect on intensity distribution	Included in CMIP6 climate models	Scientific consensus
(i) Surface energy budget	Radiative	Decrease	Uncertain	Yes	А
(ii) Diabatic heating	Radiative	Decrease	Uncertain	Yes	Α
(iii) Semi-direct effects	Radiative	Uncertain	Uncertain	Yes	В
(iv) Regional scale and monsoon dynamics	Radiative	Regional shifts	Uncertain	Yes	В
(v) Sea surface temperature patterns	Radiative	Regional shifts	Uncertain	Yes	В
(vi) Hemispheric asymmetry	Radiative	Regional shifts	Neutral	Yes	А
(vii) CCN effects on stratiform liquid clouds	Microphysical	Neutral	Uncertain	Yes (significant uncertainties)	В
(viii) CCN effects on shallow convection	Microphysical	Uncertain	Broaden	No	В
(ix) CCN effects on deep convection	Microphysical	Uncertain	Broaden	No	С
(x) INP effects	Microphysical	Uncertain	Uncertain	No (in most models)	С



**Figure 1:** Climate model simulated a) relative and c) absolute precipitation changes [%] due to anthropogenic aerosol from the coupled model intercomparison project phase 6 (CMIP6) Detection and Attribution Model Intercomparison Project (DAMIP<sup>205</sup>, difference between last 30 years of present-day DAMIP *hist-aer* minus pre-industrial *picontrol* control simulations) and the corresponding multi-model standard deviations b), d), respectively. Note the significant differences between relative (a) and absolute (b) precipitation changes, highlighted in the box over northern Africa and the Middle-East.

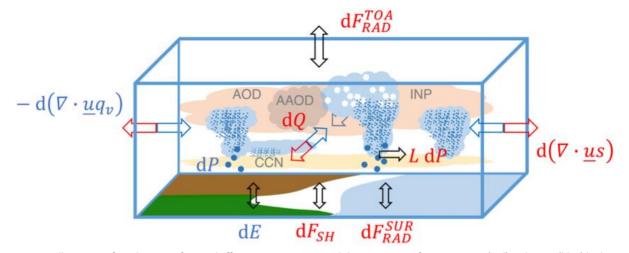


Figure 2: Illustration of mechanisms of aerosol effects on precipitation and their constraints from an energy (red) and water (blue) budget perspective. Radiative and microphysical effects are mediated by variations in Aerosol Optical Depth (AOD), Aerosol Absorption Optical Depth (AAOD) and Cloud Condensation Nuclei (CCN) as well as Ice Nucleating Particles (INP), respectively.

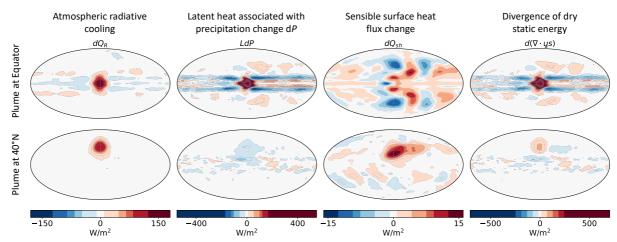
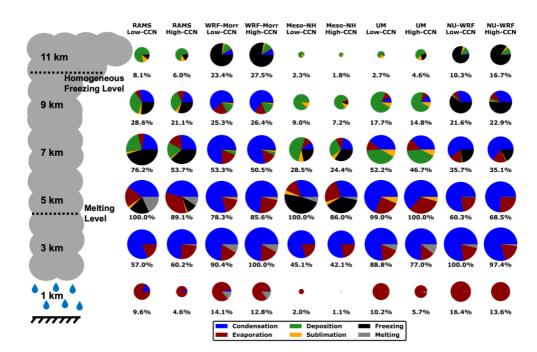


Figure 3: Idealised aqua-planet ICON<sup>206</sup> general circulation model simulations of changes of precipitation and the atmospheric energy balance in response to idealised circular absorbing aerosol radiative plumes (of 10° size and identical aerosol radiative properties with peak aerosol optical depth of 2.4 and single scattering albedo of 0.8)<sup>33</sup>. Top row: plume located on the equator. Bottom row: plume located at 40°N.  $dQ_R$ : atmospheric radiative cooling; LdP: latent heat associated with precipitation change dP;  $dQ_{SH}$ : sensible surface heat flux;  $d(\nabla \cdot \underline{u}_S)$ : divergence of dry static-energy.



**Figure 4:** Cloud-resolving model intercomparison of CCN mediated effects on deep convection from the Aerosol, Clouds, Precipitation and Climate deep convection study<sup>137,154</sup>: fractional mass process rates for tracked deep convective systems for low and high CCN conditions as a function of height. Results for each model, named in the top row, are shown for low and high CCN conditions in individual columns. The size of the pies is scaled logarithmically by the largest mass production rate of the model. Significant differences in the model base state and the response to cloud condensation nuclei perturbations illustrate associated large uncertainties.