Global sensitivities of reactive N and S gas and particle concentrations and deposition to precursor emissions reductions

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Abstract. The reduction of fine particles (PM\textsubscript{2.5}) and reactive N (N\textsubscript{r}) and S (S\textsubscript{r}) species is a key objective for air pollution control policies because of their major adverse effects on human health, ecosystem diversity, and climate. The sensitivity of global and regional N\textsubscript{r}, S\textsubscript{r}, and PM\textsubscript{2.5} to 20\% and 40\% individual and collective reductions in anthropogenic emissions of NH\textsubscript{3}, NO\textsubscript{x}, and SO\textsubscript{2}, (with respect to a 2015 baseline) is investigated using the EMEP MSC-W atmospheric chemistry transport model with WRF meteorology. Regional comparisons reveal that the individual emissions reduction has multiple co-benefits and small disbenefits on different species, and those effects are highly geographically variable. Reductions in NH\textsubscript{3} emissions are effective at decreasing NH\textsubscript{3} concentrations and deposition but much less so for NH\textsubscript{4}\textsuperscript{+}. A 40\% NH\textsubscript{3} emissions reduction decreases regional average NH\textsubscript{3} concentrations by 47-49\%, while sensitivities of NH\textsubscript{4}\textsuperscript{+} concentrations decrease in the order Euro_Medi (Europe and Mediterranean, 18\%), East Asia (15\%), North America (12\%), and South Asia (4\%), reflecting the increasing regional ammonia-richness. A disbenefit is the increased SO\textsubscript{2} concentrations in these regions (10-16\% for 40\% NH\textsubscript{3} emissions reductions) because reduced NH\textsubscript{3} levels decrease SO\textsubscript{2} deposition by altering atmospheric acidity. The 40\% NO\textsubscript{x} emissions reductions decrease NO\textsubscript{x} concentrations in East Asia by 45\%, Euro_Medi and North America by ~38\%, and South Asia by 22\%, whilst decreases in fine NO\textsubscript{3}\textsuperscript{-} are regionally reversed, which is related to enhanced O\textsubscript{3} levels in East Asia (and also, but by less, in Euro_Medi), and decreased O\textsubscript{3} levels in South Asia (and also, but by less, in North America). Consequently, the oxidation of NO\textsubscript{2} to NO\textsubscript{3}\textsuperscript{-} and of SO\textsubscript{2} to SO\textsubscript{4}\textsubscript{2-} is enhanced in East Asia but decreased in South Asia, which in East Asia causes a more effective decrease in NO\textsubscript{x} and SO\textsubscript{2} but a less effective decrease in NO\textsubscript{3}\textsuperscript{-} and even an increase in SO\textsubscript{4}\textsubscript{2-}; in South Asia it causes a less effective decrease in NO\textsubscript{x} and an increase in SO\textsubscript{2} but a more effective decrease in NO\textsubscript{3}\textsuperscript{-} and SO\textsubscript{4}\textsubscript{2-}. For regional policy making, it is thus important to reduce NH\textsubscript{3}, NO\textsubscript{x} and SO\textsubscript{2} emissions together and/or go for stronger reductions to minimise such adverse effects in East Asia and Euro_Medi. Reductions in SO\textsubscript{2} emissions are slightly more effective for SO\textsubscript{2} than SO\textsubscript{4}\textsubscript{2-}. A disbenefit is that SO\textsubscript{2} emissions reductions increase NH\textsubscript{3} total deposition and ecosystem eutrophication (~12\% increase for 40\% emissions reduction). PM\textsubscript{2.5} mitigation in South Asia is most sensitive to 40\% SO\textsubscript{2} reduction (3.10 μg m\textsuperscript{-3}, 10\%) and least sensitive to NH\textsubscript{3} reduction (0.29 μg m\textsuperscript{-3}, 1\%), which is because South Asia is so ammonia-rich that reducing NH\textsubscript{3} has little impact. The most effective measure for North America is reducing NO\textsubscript{x} emissions with an 8\% (0.63 μg m\textsuperscript{-3}) decrease in PM\textsubscript{2.5} in response to a 40\% reduction. In Euro_Medi, the sensitivities of PM\textsubscript{2.5} to 40\% individual emissions reductions range 5-8\% (0.55-0.82 μg m\textsuperscript{-3}). In the UK and Scandinavia PM\textsubscript{2.5} is more sensitive to NH\textsubscript{3} in central Europe it is more sensitive to NO\textsubscript{x} while in the Mediterranean it is more sensitive to SO\textsubscript{2}. In East Asia, reductions in SO\textsubscript{2}, NO\textsubscript{x}, and NH\textsubscript{3} emissions are almost equally effective with PM\textsubscript{2.5} sensitivities to 40\% reductions of 7-8\% (1.89-2.33 μg m\textsuperscript{-3}). Due to the varying contributions of SIA, PM\textsubscript{2.5} sensitivities to 40\% collective reductions in all 3 precursors decrease in the order East Asia (20\%), Euro_Medi and North America (17\%), South Asia (13\%). The geographically-varying non-linear chemical responses of N\textsubscript{r}, S\textsubscript{r}, and PM\textsubscript{2.5} to emissions reductions revealed by this work show the importance of both prioritising emissions strategies in different regions and combining several precursor reductions together to maximise the policy effectiveness.
1 Introduction

Reactive N (N_r) and S (S_r) species are critical determinants of air quality. A substantial proportion of ambient PM_{2.5} (particulate matter with aerodynamic diameter ≤2.5 μm) is secondary inorganic aerosol (SIA) formed from chemical reactions of emissions of the precursor gases NH_3, NO_x (NO and NO_2) and SO_x (sulfur oxides, mainly SO_2) (Behera et al., 2013; Weber et al., 2016; Vasilikos et al., 2018; Nenes et al., 2020). PM_{2.5} is consistently associated with elevated risk of all-cause mortality and other adverse health impacts (Hart et al., 2015; Chen et al., 2017; Chen et al., 2018a; Karimi et al., 2019; Stieb et al., 2020). The gases NO_2 and SO_2 are also direct health pollutants. In addition, oxidized N (e.g., NO_3, HNO_3, and NO_3^−, collectively abbreviated as OXN) and reduced N (e.g., NH_3 and NH_4^+, collectively abbreviated as RDN) species are powerful nutrients for plants and microorganisms, whose deposition leads to eutrophication and loss of ecosystem biodiversity (Erisman et al., 2005; Bergström and Jansson, 2006; Sun et al., 2017; Kharol et al., 2018). The severity of the adverse effects of N deposition is determined not only by the total quantity but also its form. Many studies show different N deposition components having toxicity to different plants, dry deposition of NH_3 is particularly deleterious for example (Van Herk et al., 2003; Sheppard et al., 2011; Sutton et al., 2014, 2020; Pescott et al., 2015) Deposition of oxidized S (i.e., SO_2 and SO_4^{2−}, collectively abbreviated as OXS) also greatly influences precipitation acidity (Lu et al., 2010; Aas et al., 2019; McHale et al., 2021). These observations put greater emphasis on mitigation of certain deposition components.

The East Asia, South Asia, Euro_Medi (Europe and Mediterranean) and North America regions have high population density and high N, and S, pollution. Historically, Europe and North America were the dominant emissions regions, suffering severe air pollution until the late 20th century. However, the combination of emissions controls for SO_x and NO_x in these two regions and rapid industrialisation elsewhere means that emissions in East and South Asia now dominate globally (Weber et al., 2016; NEC, 2019; Fowler et al., 2020), although China, in particular, is now implementing effective SO_x and NO_x emissions controls (Liu et al., 2016; Hoesly et al., 2018; Meng et al., 2022). In contrast, a lack of action on NH_3 emissions in most countries, coupled with the growth in agriculture to feed a rising global population, means that global NH_3 emissions continue to grow (Heald et al., 2012; Fowler et al., 2015; Aksoyoglu et al., 2020). As a result, ambient N, and S, pollution remains a major health and environmental concern in most regions. The European Environmental Agency reported that 97% of the urban population in the European Union in 2019 was exposed to annual mean concentrations of PM_{2.5} above the latest World Health Organization (WHO) air quality guideline of 5 μg m\(^{-3}\) (EEA, 2021; WHO, 2021), whilst the United States Environmental Protection Agency reported that for the period 2014-2016 only 10% of its 429 monitoring sites had PM_{2.5} concentrations <6.0 μg m\(^{-3}\) (USEPA, 2017). Combining satellite retrievals, chemistry model simulations, and ground level measurements, Ma et al. (2014) and Brauer et al. (2016) showed that in 2013 the majority of the East and South Asia population lived in areas where annual mean PM_{2.5} concentrations exceeded the WHO Interim Target 1 of 35 μg m\(^{-3}\) (WHO, 2021).

Furthermore, as mitigation of NO_x emissions in recent years has been more effective than for NH_3, deposition of RDN is now increasingly responsible for the exceedances of N critical loads for eutrophication in many regions (Jovan et al., 2012; Chen et al., 2018b; Simpson et al., 2020; Yi et al., 2021; Jonson et al., 2022).

Understanding the sensitivities of PM_{2.5}, N, and S, pollution to emissions reductions is complicated not only by the substantial regional heterogeneity in relative emissions but also by the substantial meteorological heterogeneity influencing the chemistry and deposition. This necessitates use of atmospheric chemistry transport models (ACTMs) designed to simulate the underlying physical–chemical processes linking emissions, dispersion, chemical reactions, and deposition of atmospheric components. Previous ACTM studies have provided insight into the complexities of sensitivities of PM_{2.5} and its SIA components to changes in emissions in different regions that measurements cannot reveal. For example, using the GEOS-Chem model, Wang et al. (2013) showed that SIA concentrations in 2015 decreased in South China and Sichuan Basin but increased in North China compared to their 2006 levels in response to −16% SO_x and +16% NO_x emissions changes (no change in NH_3 emissions) from 2006 to 2015 according to China’s 12th Five-Year Plan, but if NH_3 emissions increase by +16% (based on their growth rate from 2006 to 2015), the SIA reduction due to SO_x reduction will be totally offset in all regions because of...
the elevated NH$_3$NO$_3$ formation, demonstrating the importance of NH$_3$ control on China’s SIA mitigation. Pommier et al. (2018) reported substantial projected growth in emissions in India between 2011 and 2050, amounting to 304% for SO$_x$, 287% for NMVOC, 162% for NO$_x$, 100% for primary PM$_{2.5}$, and 60% for CO and NH$_3$, leading to increases in annual mean PM$_{2.5}$ and O$_3$ concentrations of 67% and 13% respectively. In the UK, results from EMEP4UK model simulations for 2010 emissions and meteorology indicated that NH$_3$ emissions reductions are the most effective single-component control (compared to individual reductions in NO$_x$, SO$_x$, and primary PM$_{2.5}$) on area-weighted PM$_{2.5}$, whilst weighting by population placed greater emphasis on reductions in emissions of primary PM$_{2.5}$ (Vieno et al., 2016). Holt et al. (2015) used GEOS-Chem to investigate PM$_{2.5}$ sensitivities in the United States to NO$_x$, SO$_x$, and NH$_3$ emissions reductions between two sets of scenarios representing a 2005 baseline (high emissions) and a 2012 analogue (low emissions) for both summer and winter, with the national total emissions exhibiting a 62% decrease in SO$_2$, a 42% decrease in NO$_x$, and a 1% increase in NH$_3$ from 2005 to 2012. They found larger sensitivities of PM$_{2.5}$ to NO$_x$ reductions in winter than in summer in parts of the northern Midwest because PM$_{2.5}$ is more nitrate-limited in the low emissions case, and that lower NO$_x$ and SO$_2$ emissions in 2012 led to larger PM$_{2.5}$ sensitivity to SO$_2$ compared to 2005 since lower NO$_x$ emissions enhance the relative importance of aqueous-phase SO$_2$ oxidation.

These studies analysed N, and S, responses to precursor emissions reductions in the early 2000s and in specific regions, but do not provide a global view of sensitivities to the same reductions everywhere. Given the considerable emissions changes in global and regional NH$_3$, NO$_x$, and SO$_x$ in recent years (Hoesly et al., 2018; Kurokawa and Ohara, 2020), our understanding of the current chemical climate for N, and S, reactions on global and regional scale and how it affects responses of PM$_{2.5}$, N, and S, species to various emissions reductions should be updated. This is the motivation for the work presented here, which provides a global picture of the effectiveness of NH$_3$, NO$_x$, and SO$_x$ emissions reductions for mitigating both concentrations and deposition of N, and S, pollutants. We used the EMEP MSC-W ACTM to simulate the global domain based on global emissions and meteorology in 2015, which enables a regional comparison to be conducted with inherently consistent simulations. The focus here is on annual means, as these are the long-term metric within global and regional air quality standards. We first describe the model set-up and performance and the sensitivity experiments used to simulate responses of PM$_{2.5}$, N, and S, species to 20% and 40% reductions in gaseous precursor emissions (Sect. 2). Section 3 details the global and regional concentration and deposition changes in components of RDN, OXN, OXS, and PM$_{2.5}$ between baseline and emissions reduction scenarios. Section 4 discusses key processes that determine the benefits and disbenefits of emissions reductions and how they vary geographically, and the implications of our findings for policy making.

2 Methods

2.1 Model set-up and performance

The EMEP MSC-W (European Monitoring and Evaluation Programme Meteorological Synthesizing Centre – West) open-source atmospheric chemistry transport model (https://www.emep.int, last access: 8 August 2022) is a three-dimensional Eulerian model widely used for both scientific research and policy development (Bergström et al., 2014; Jonson et al., 2017; Pommier et al., 2018; McFiggans et al., 2019; Karl et al., 2019; Pommier et al., 2020; Jonson et al., 2022). Version rv4.34 was used here. A detailed technical description of EMEP MSC-W rv4.0 is documented in Simpson et al. (2012). A series of overviews of model updates from version rv4.0 to rv4.34 is documented in annual EMEP status reports (Simpson et al., 2013; Tsyro et al., 2014; Simpson et al., 2015, 2016, 2017, 2018, 2019, 2020). Meteorology for 2015 was derived from the Weather Research and Forecasting model (WRF; https://www.wrf-model.org; https://github.com/wrf-model/WRF/releases/tag/v4.2.2, last access: 8 August 2022) version 4.2.2. The coupled EMEP-WRF system has been tested and applied to many regional and global studies (Vieno et al., 2010, 2014, 2016; Werner et al., 2018; Chang et al., 2019; Gu et al., 2021).

Detailed global EMEP-WRF configurations used in this work are presented in Ge et al. (2021b, 2022). In brief, the global domain has a horizontal resolution of 1° × 1° and 21 terrain-following vertical layers from the surface up to 100 hPa. The
height of the lowest model layer is around 45 m. The model outputs of surface concentrations are adjusted to correspond to 3 m above ground level in order to provide concentrations at heights more typical of ambient measurements and human exposure (Simpson et al., 2012). The aerosol module is the Equilibrium Simplified Aerosol Model V4 (EQSAM4clim), which parameterizes a full gas–liquid–solid partitioning scheme for semi-volatile and non-volatile mixtures. Details are described in Metzger et al. (2016, 2018). Dry deposition of gaseous species and aerosol components to the ground surface is simulated utilizing deposition velocity as described in Simpson et al. (2012, 2020). The parameterisation of wet deposition incorporates both in-cloud and below-cloud scavenging of gases and particles (Berge and Jakobsen, 1998; Simpson et al., 2012).

The global model evaluation of N\textsubscript{r} and S\textsubscript{r} concentrations and wet deposition from this model configuration for 2010 and 2015 against measurements from 10 ambient monitoring networks is documented in Ge et al. (2021b) and demonstrates the model’s capability for capturing the spatial and seasonal variations of NH\textsubscript{3}, NH\textsubscript{4}\textsuperscript{+}, NO\textsubscript{2}, HNO\textsubscript{3}, NO\textsubscript{3}\textsuperscript{-}, SO\textsubscript{2}, and SO\textsubscript{4}\textsuperscript{2-} in East Asia, Southeast Asia, Europe, and North America. As an example, the correlation coefficients between model and measurement annual mean concentrations for most species in 2015 are $\geq 0.78$, and for annual wet deposition of RDN and OXN are 0.78 and 0.63 respectively. This is in spite of inherent uncertainty in both model and measurements and differences in their spatial representativeness.

### 2.2 Emissions and model experiments

Baseline emissions for 2015 were from the ECLIPSE V6 (Evaluating the CLimate and Air Quality ImPacts of Short-livEd Pollutant) inventory, available at https://previous.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv6b.html (last access: 8 August 2022). Monthly emissions profiles derived from EDGAR (Emission Database for Global Atmospheric Research, v4.3.2 datasets, available at https://edgar.jrc.ec.europa.eu/dataset_temp_profile) time series (Crippa et al., 2020) were applied to the ECLIPSE annual emissions of SO\textsubscript{2}, NO\textsubscript{2}, NH\textsubscript{3}, CO, CH\textsubscript{4}, NMVOC, primary PM\textsubscript{2.5} and coarse particles. Detailed implementation procedures including the re-assignment of ECLIPSE emissions sectors to EMEP sectors and the calculation of temporal profiles at a given country for a given pollutant are described in Ge et al. (2021b).

A baseline simulation and a set of 8 sensitivity experiments were conducted for emissions and meteorology for 2015. The model experiments applied 20\% and 40\% reductions to global anthropogenic emissions of NH\textsubscript{3}, NO\textsubscript{x}, SO\textsubscript{2} from all sectors both individually and collectively (i.e., reductions applied to all 3 species simultaneously). All other emissions, including natural emissions such as dimethyl sulfide from oceans, lightning NO\textsubscript{x}, and soil NO\textsubscript{x}, were left unchanged.

### 2.3 Definition of world regions

We compared the sensitivities to the emissions reductions of PM\textsubscript{2.5}, N\textsubscript{r}, and S\textsubscript{r} species concentrations and depositions in the four world regions of East Asia, South Asia, Euro_Medi, and North America defined in Fig. 1 (and listed in Table S1). These are based on regions used by the Intergovernmental Panel on Climate Change and as rationalised in Iturbide et al. (2020). All four regions are densely populated and have high N\textsubscript{r} and S\textsubscript{r} pollution as reported by Ge et al. (2022).
Figure 1: The boundaries of the 4 world regions used in this study, which are based on the IPCC reference regions described in Iturbide et al. (2020).

3 Results

3.1 Sensitivities of $N_r$ and $S_r$ gas and aerosol concentrations

The simulated global baseline 2015 annual mean surface concentrations of $N_r$ and $S_r$ have been discussed in detail in Ge et al. (2022). Here we analyse the sensitivities of the modelled surface concentrations to SIA precursor emissions reductions for RDN, OXN and OXS components. Sensitivities differ according to consideration of primary or secondary components and show great geographical variation.

3.1.1 RDN

Figure 2 shows the spatial variations in the sensitivities of NH$_3$ and NH$_4^+$ annual mean surface concentrations to 20% and 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually, and collectively. Regional average sensitivities in East Asia, South Asia, Euro_Medi, and North America are summarised in Fig. 3 and Table S2. Steeper gradients in Fig. 3 correspond to greater concentration changes (sensitivities).

From these figures, it is clear that whilst reducing emissions of NH$_3$ (and all 3 precursors together) decrease NH$_3$ concentrations efficiently, reducing emissions of NO$_x$ and SO$_x$ lead to increases in NH$_3$ concentrations, particularly over densely populated areas. The maximum reduction in model grid NH$_3$ concentration across all scenarios reaches 16.6 µg m$^{-3}$ (44% change relative to baseline; expressed similarly hereafter) and occurs over East Asia in response to a 40% reduction in NH$_3$ emissions. The largest increase in NH$_3$ concentration (1.51 µg m$^{-3}$, 22%) arises in Southeast Asia under the 40% SO$_x$ emissions reduction scenario. Lower NO$_x$ and SO$_x$ emissions decrease the concentration of acidic species available to react with NH$_3$ to form NH$_4^+$ aerosol, so more of the emitted NH$_3$ stays in the gas phase. Other studies have likewise shown that reductions in SO$_2$ or NO$_x$ emissions are an important contributor to the growth in tropospheric NH$_3$ concentrations globally and regionally (Saylor et al., 2015; Warner et al., 2017; Liu et al., 2018; Yu et al., 2018).

In East Asia and North America, NH$_3$ concentrations increase similarly for either NO$_x$ or SO$_x$ reductions (Fig. 3), but in South Asia and Euro_Medi, NH$_3$ concentrations increase more with SO$_x$ reductions than with NO$_x$ reductions, which reflects the larger contribution of (NH$_4$)$_2$SO$_4$ than NH$_3$NO$_3$ to SIA in the latter two regions. However, the increase in NH$_3$ concentrations is relatively small compared to the extent of NO$_x$ and SO$_x$ emissions reductions; 40% reductions in emissions...
of NOx or SOx only increases NH3 concentrations in the 4 regions by 2-6% or 6-9%, respectively (Fig. 3, Table S2). The globally averaged increases in NH3 concentrations for 40% reductions in NOx or SOx emissions are 3% and 9%, respectively. Nevertheless, NH3 concentration decrease resulting from reductions in NH3 emissions is offset by simultaneous effects of NOx and SOx emissions reductions when all 3 precursors are reduced together, as the sensitivities of regional average NH3 concentrations to 40% reductions in all 3 precursor emissions (38-39% across the four regions) are smaller than their sensitivities to 40% reductions in NH3 emissions on its own (47-49%). It is also noteworthy that the sensitivities of regional average NH3 concentrations are essentially linear through 20% and 40% emissions reductions, irrespective of precursor, although the sensitivities are different between regions.

In contrast to NH3, concentrations of NH4+ always decrease when an SIA precursor emission is reduced. Figure 2 shows that NH4+ concentrations in the most densely populated continents (e.g., eastern China, India, Europe, eastern America) respond strongly to emissions reductions in each SIA precursor, whilst they only respond to SOx emissions reductions over oceans. This is related to the production of marine sulfate aerosol from dimethyl sulfide (DMS) and the lack of significant oceanic NOx emissions sources, which means only (NH4)2SO4 formation is important in marine SIA chemistry.

In addition, the impacts of NH3 and SOx emissions reductions on NH4+ concentrations over North Africa are significantly greater than from NOx emissions reductions, indicating a dominance of (NH4)2SO4 within SIA in this region. This is consistent with the results reported by Ge et al. (2022). They showed that large areas in North Africa are characterised by the SO42−-rich chemical domain for SIA formation, which means that NH3 is predominantly taken up by SO42−, leaving no free NH3 to react with HNO3 to form NH4NO3. Given that emissions reductions in all three precursors individually lead to reductions in NH4+ concentrations, it is not surprising that the greatest simulated NH4+ reduction (5.87 µg m⁻³ (43%) in East Asia) arises for the scenario with 40% reductions in all 3 precursors collectively.

Figure 3 also shows that NH4+ sensitivities are essentially linear for emissions reductions to 40%, although responses again vary slightly with region. Among individual precursor reduction scenarios, regional average NH4+ concentrations in East Asia and Euro_Medi are most sensitive to SOx emissions reductions and least sensitive to NOx reductions, while NH4+ concentrations in North America are most sensitive to SOx reductions and least sensitive to NH3 reductions. In South Asia, NH4+ is characterised by strong sensitivity to SOx emissions reductions but only relatively small sensitivities to NOx and NH3 emissions reductions. In the scenario of all 3 species reductions, all regions show relative sensitivities close to the one-to-one line. Another important observation from Fig. 3 is that reductions in NH4+ (4-18%) in response to a 40% NH3 emissions reduction are much smaller than reductions of NH3 concentrations (47-49%) in these regions, which reflects the fact that these regions are so ammonia-rich that reducing NH3 emissions only has limited effects on NH4+ concentrations.
Figure 2: Changes in NH$_3$ and NH$_4^+$ annual surface concentrations for 20% and 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.

Figure 3: The absolute and relative sensitivities of regionally-averaged annual mean surface concentrations of NH$_3$ (upper row) and NH$_4^+$ (lower row) to 20% and 40% emissions reductions in NH$_3$ (blue), NO$_x$ (orange) and SO$_x$ (green) individually, and collectively (red), for the four regions defined in Fig. 1. The solid grey line in each panel illustrates the one-to-one relative response to emissions reductions, whilst the coloured dashed lines are the linear regressions through each set of three model simulations and illustrate the actual responses to emissions reductions of a given precursor. The numbers show the corresponding relative responses to each emissions reduction (with respect to baseline).
3.1.2 OXN

Figure 4 shows the spatial variations in the sensitivities of NOx and fine nitrate annual mean surface concentrations to 20\% and 40\% emissions reductions in NH3, NOx, and SOx, individually, and collectively. Regional average sensitivities in East Asia, South Asia, Euro_Medi, and North America are summarised in Fig. 5 and Table S3. Equivalent global maps and regional sensitivity plots for the responses of HNO3 and coarse nitrate to the same emissions reductions are presented in Figs. S1 and S2 and Table S3.

In contrast to surface NH3, whose concentrations are sensitive to reductions in emissions of each of NH3, NOx and SOx, surface NOx concentrations only respond to NOx emissions reductions and have negligible sensitivity to NH3 and SOx emissions reductions. The 20\% and 40\% reductions in NOx emissions yield a global maximum of 25.6 \(\mu g\) m\(^{-3}\) (22\%) and 51.3 \(\mu g\) m\(^{-3}\) (44\%) reductions in surface NOx concentrations over East Asia. The globally averaged reductions in NOx concentrations for 20\% and 40\% reductions in NOx emissions are 15\% and 30\% respectively (same values for simultaneous reductions in all 3 precursors), whereas the sensitivities for NH3 and SOx emissions reduction are all 0\% (Table S3).

Regionally (Fig. 5), East Asia shows the largest decreases in NOx concentrations (45\%) in response to 40\% NOx emissions reductions, followed by Euro_Medi and North America (36-38\%), and South Asia (23\%). These regional differences in sensitivities to NOx emissions are due to regional differences in oxidation chemistry climate. Figure S3 shows the changes in annual mean surface concentrations of O3 for the 8 emissions reduction scenarios. Concentrations of O3 in eastern China, western and central Europe, and north-eastern US increase as NOx emissions reduce, while O3 in the rest of the world decreases as NOx emissions reduce. In East Asia, the increased oxidant levels enhance NOx chemical removal and results in a greater than one-to-one relative decrease in NOx concentrations with NOx emissions reductions. Consequently, the decrease in fine nitrate in East Asia is offset by enhanced chemical production, which leads to a lower than one-to-one sensitivity (e.g., a 40\% reduction in NOx emissions gives a 33\% decrease in fine NO3\(^{-}\)). In contrast, decreased oxidant levels in South Asia decrease the oxidation of NOx, which partially offsets the decrease in NOx concentrations induced by emissions reductions, causing a more efficient reduction in fine NO3\(^{-}\) concentrations than in NOx, in this region. The variation in regional atmospheric oxidising capacity also alters the SO4\(^{2-}\) formation processes; discussions of this are presented in Sect. 3.1.3 and Sect. 4. The situation is more complex for Euro_Medi and North America as these regions include both positive and negative changes in O3 concentrations with NOx emissions reductions (Fig. S3) and they are not as NOx-rich as East Asia and South Asia. The effects of changes in oxidant levels on NOx and fine NO3\(^{-}\) concentrations are therefore more localised and less apparent in regional averages. Clappier et al. (2021) reported this effect to be most distinct in the Po basin (Italy), western Germany, and Netherlands in Europe, whilst for the United States, Tsimpidi et al. (2008) showed it only becomes pronounced in the north east, both of which are consistent with our results.

For secondary OXN species, the sensitivities of HNO3 and fine and coarse NO3\(^{-}\) to individual reductions in emissions of NH3, NOx and SOx are closely associated with SIA formation chemistry. The principal observation from Fig. 4 is that concentrations of fine NO3\(^{-}\) in all four regions decrease with reduced NH3 and NOx emissions but increase with reduced SOx emissions. This is because H2SO4 and HNO3 compete in their reactions with NH3, and (NH4)2SO4 is formed preferably over NH4NO3. Reductions in NH3 emissions cause the equilibrium between HNO3 and NH3 to shift away from NH4NO3 and therefore to a decrease in fine NO3\(^{-}\) concentrations but to an increase in HNO3 and coarse NO3\(^{-}\) concentrations (Fig. S1).

Reductions in NOx emissions decrease HNO3 and fine and coarse NO3\(^{-}\) concentrations globally. Although the increased oxidant levels that arise in some regions following NOx emissions reductions enhance the chemical formation of these secondary species (as discussed above), NOx emissions reductions of 20\% and 40\% are substantial enough to mean that the lower availability of NOx to form NO3\(^{-}\) dominates the impact on NO3\(^{-}\) concentrations compared with the enhancement in oxidising capacity. Reduced SOx emissions leave more NH3 to equilibrate with HNO3 to form NH4NO3, leading to an increase in fine NO3\(^{-}\) concentrations but to a decrease in coarse NO3\(^{-}\) concentrations as the former takes more HNO3. It is notable that the increase in fine NO3\(^{-}\) concentrations is relatively small compared to the extent of SOx emissions reductions. For example, the...
maximum increase in fine NO$_3^-$ resulting from 40% reductions in SO$_x$ emissions is 1.71 µg m$^{-3}$ (16%), in East Asia. The regional average increases in fine NO$_3^-$ concentrations for 40% SO$_x$ emissions reductions are 8% in East Asia, South Asia, and Euro_Medi, and 4% in North America.

The differences in regional average sensitivities of HNO$_3$ and fine and coarse NO$_3^-$ are highlighted more clearly in Fig. 5 and Fig. S2. Fine NO$_3^-$ in East Asia is equally sensitive to NO$_x$ and NH$_3$ emissions reductions (33% and 32% decreases for 40% NO$_x$ and NH$_3$ emissions reductions respectively), while it is more sensitive to NO$_x$ emissions reductions than to NH$_3$ emissions reductions in South Asia (45% and 39%), Euro_Medi (41% and 33%), and North America (42% and 26%). In terms of absolute concentration changes, the reductions in fine NO$_3^-$ over East Asia in response to 40% NH$_3$ and NO$_x$ emissions reductions (1.62 -1.65 µg m$^{-3}$) are more than 3 times larger than reductions in other regions (0.23 - 0.47 µg m$^{-3}$). On the other hand, if NH$_3$ emissions are reduced then the increases in HNO$_3$ and coarse NO$_3^-$ concentrations in East Asia (15% increases for 40% NH$_3$ reductions) are much larger than the increases in the other three regions (2-6%). All these differences between East Asia and the other three regions reflect the larger abundance of NH$_4$NO$_3$ in SIA over East Asia. This is demonstrated in Fig. S6 which shows that the contribution of fine NO$_3^-$ to PM$_{2.5}$ in the baseline is greatest in East Asia (19%, 5.21 µg m$^{-3}$), followed by Euro_Medi (12%, 1.22 µg m$^{-3}$), North America (11%, 0.86 µg m$^{-3}$), and South Asia (3%, 0.93 µg m$^{-3}$). Detailed discussion on regional SIA composition is presented in Sect. 3.2.

Figure 4: Changes in NO$_x$ and fine NO$_3^-$ annual surface concentrations for 20% and 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.
Figure 5: The absolute and relative sensitivities of regionally-averaged annual mean surface concentrations of NOx (upper row) and fine NO3- (lower row) to 20% and 40% emissions reductions in NH3 (blue), NOx (orange) and SOx (green) individually, and collectively (red), for the four regions defined in Fig. 1. The solid grey line in each panel illustrates the one-to-one relative response to emissions reductions, whilst the coloured dashed lines are the linear regressions through each set of three model simulations and illustrate the actual responses to emissions reductions of a given precursor. The numbers show the corresponding relative responses to each emissions reduction (with respect to baseline).

3.1.3 OXS

Figure 6 shows the global variation in the sensitivities of SO2 and SO42- annual mean surface concentrations to 20% and 40% emissions reductions in NH3, NOx, and SOx individually, and collectively. Figure 7 and Table S4 summarise the sensitivities of the regionally averaged SO2 and SO42- concentrations to the emissions reductions for the four regional domains. Concentrations of SO2 increase in response to reduced NH3 emissions particularly over East Asia, South Asia, Europe, and North America. The largest increases in SO2 resulting from 20% and 40% NH3 emissions reductions are respectively in Southeast Asia (3.62 µg m⁻³, 9%) and East Asia (8.11 µg m⁻³, 20%) (Fig. 6). The response of secondary SO42- concentrations to NH3 emissions reductions varies substantially across the world. In north-eastern China and Europe, SO42- concentrations increase, whilst in southern China, India, and United States they decrease. However, the magnitudes of SO42- concentration changes are much smaller than for SO2. The maximum increases in SO42- concentrations (located in southern China) are only 0.37 µg m⁻³ (2%) and 0.89 µg m⁻³ (5%) for 20% and 40% NH3 emissions reductions, respectively. The maximum decreases in SO42- concentrations, in north-eastern China, are 0.62 µg m⁻³ (6%) and 1.30 µg m⁻³ (12%), respectively. Regionally averaged, East Asia exhibits the largest increase in SO2 for 40% NH3 emissions reductions (16%) (Fig. 7) followed by North America (14%), South Asia (14%), and Euro_Medi (10%), whereas increases in regional average SO42- concentrations are only in the range 0-2%.

Figures S3 and 13 show the global variation in the sensitivities to precursor emissions reductions of total deposition of SO2, and of the wet and dry deposition of all OXS components, respectively. When NH3 emissions are reduced, SO2 total deposition decreases over all of East Asia, South Asia, Europe, and North America (Fig. S3); Fig. 13 shows the decrease is driven by reduced SO2 dry deposition. Several studies have shown that the non-stomatal canopy uptake resistance of SO2 (the inverse of the SO2 dry deposition velocity) is positively correlated to the molar ‘acidity ratio’ $\alpha_{SO} = \frac{[SO_2]}{[NH_3]}$ (Smith et al., 2000; Erisman et al., 2001; Fowler et al., 2009; Massad et al., 2010), a process that is included in the EMEP MSC-W model (Simpson et al., 2010).
et al., 2012). Reduced NH₃ concentrations therefore increase the acidity ratio and hence decrease the rate of SO₂ dry deposition and increase the SO₂ surface concentrations in those regions where this effect is significant. The SO₄²⁻ responses to NH₃ emissions reductions are related to changes in atmospheric acidity as well. The aqueous-phase oxidation of SO₂ by O₃, which is one of the major pathways for SO₄²⁻ production, is significantly pH dependent. In general, the oxidation rate decreases with decreased pH (Penkett et al., 1979; Maahs, 1983; Liang and Jacobson, 1999; Hattori et al., 2021), a process that is incorporated into the EMEP MSC-W model (Simpson et al., 2012). As pointed out by Ge et al. (2022), Europe and north-eastern China are much less ammonia-rich than India, which means that 20% and 40% NH₃ emissions reductions are substantial enough to decrease the pH in the former two regions, leading to decreases in SO₄²⁻ production. However, given that SO₂ levels increase and that there are still other effective oxidation pathways (e.g., OH, H₂O₂) which are independent of pH (McArdle and Hoffmann, 1983; Hoffmann, 1986; Seinfeld and Pandis, 2016), the decreases in SO₄²⁻ concentrations in Europe and north-eastern China due to NH₃ emissions reductions are very small anyway. In contrast, since India is so ammonia-rich, even 40% reductions in NH₃ emissions do not significantly alter the pH in this region. As a result, SO₄²⁻ concentrations in India increase slightly due to the higher availability of its precursor SO₂.

The impacts of NOₓ emissions reductions on SO₂ concentrations show inverse trends in different regions (Fig 7), which reflects regional differences in atmospheric oxidation chemistry. The decreased SO₂ concentrations (maximum reduction: 1.72 µg m⁻³, 6%) in eastern China, Europe, and north-eastern United States (Fig. 6) can be explained by the enhanced O₃ concentrations in these regions arising from the reduced NO + O₃ reaction in these high NOₓ regions (Fig. S3). As a result, SO₂ is more readily oxidised to SO₄²⁻, leading to increased SO₄²⁻ concentrations (maximum increase for 40% NOₓ emissions reductions: 1.59 µg m⁻³ (10%)) in these regions. This positive response of SO₄²⁻ to NOₓ emissions reductions is also reported in regional studies (Botha et al., 1994; Li et al., 2006; Sheng et al., 2018; Fang et al., 2019; Ge et al., 2021a). In contrast, India, north-eastern Africa, and southern Africa show increased SO₂ (maximum increase: 0.29 µg m⁻³, 1%) but decreased SO₄²⁻ concentrations (maximum decrease: 0.73 µg m⁻³, 9%) as NOₓ emissions reduce, which can be explained by the parallel decrease in O₃ concentrations in these regions (Fig. S3). However, these concentration changes are very localised and, from a regional average perspective, are relatively small compared to the extent of emissions reductions applied. For example, in East Asia, the region with the largest response, there is only a 5% decrease in regional average SO₂ concentration (Fig. 7), and 3% increase in SO₄²⁻ concentration, for a 40% reduction in NOₓ emissions. For other regions, the SO₂ and SO₄²⁻ regional average concentration changes are even smaller (from -4% to 2%). The global average sensitivities of SO₂ and SO₄²⁻ annual mean concentrations to 20% and 40% NOₓ emissions reductions are only in the range 0-2% (Table S4).

Under reductions of SO₂ emissions (and of all 3 precursors together), both SO₂ and SO₄²⁻ show almost one-to-one reductions, indicating that SO₂ emissions reductions are crucial for reducing both primary and secondary OXS pollutants and, in the case of reductions of all 3 precursors simultaneously, readily sufficient to dominate over any tendency for NH₃ and NOₓ emissions reductions to increase OXS species concentrations. A 40% reduction in SO₂ emissions leads to a maximum SO₂ decrease of 39.8 µg m⁻³ (40%), in northern Russia, and a maximum SO₄²⁻ decrease of 7.70 µg m⁻³ (39%), in south-eastern China (Fig. 6). For the four regions, average SO₂ concentrations decrease by 22-24% and 42-45%, and SO₄²⁻ concentrations decrease by 17-19% and 34-38%, in response to 20% and 40% SO₂ emissions reductions respectively (Fig. 7). For the 20% and 40% reductions in all 3 precursors together, regionally averaged SO₂ decrease by 19-20% and 38-41% respectively, and regionally averaged SO₄²⁻ decrease by 17-20% and 35-40% respectively.
Figure 6: Changes in SO$_2$ and SO$_4^{2-}$ annual surface concentrations for 20% and 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.

Figure 7: The absolute and relative sensitivities of regionally-averaged annual mean surface concentrations of SO$_2$ (upper row) and SO$_4^{2-}$ (lower row) to 20% and 40% emissions reductions in NH$_3$ (blue), NO$_x$ (orange) and SO$_x$ (green) individually, and collectively (red), for the four regions defined in Fig. 1. The solid grey line in each panel illustrates the one-to-one relative response to emissions reductions, whilst the coloured dashed lines are the linear regressions through each set of three model simulations and illustrate the actual responses to emissions reductions of a given precursor. The numbers show the corresponding relative responses to each emissions reduction (with respect to baseline).
3.2 Sensitivity of PM$_{2.5}$ concentrations

Figure 8 shows the global distribution of the dominant sensitivity of PM$_{2.5}$ towards a 40% reduction in NH$_3$, NO$_x$, or SO$_x$ emissions individually. An annual mean PM$_{2.5}$ concentration threshold of 5 µg m$^{-3}$ has been applied in order to focus attention away from the most remote areas where PM$_{2.5}$ concentrations in the baseline simulation are already below the latest WHO PM$_{2.5}$ air quality guideline (AQG) (WHO, 2021).

The principal observation from Fig. 8 is that the sensitivity of PM$_{2.5}$ to reductions in emissions of individual precursors is highly geographically variable. SO$_x$-sensitive regimes are found in Southeast Asia, South Asia, Africa, and Central America. NO$_x$-sensitive regimes are observed in south-eastern China, France, Germany, most eastern European countries, central and eastern United States, and northern and central parts of South America. Only a few small regions are NH$_3$ sensitive: these include eastern coastal areas around China, the UK and its surrounding seas, southern Scandinavia, and western Russia. The difference in PM$_{2.5}$ sensitivity between northern Europe and the rest of Europe demonstrates that NH$_3$ has become the limiting factor for SIA formation in northern Europe. This greater leverage of NH$_3$ emissions on PM$_{2.5}$ mitigation in this region is due to the effective emissions controls on all SIA precursor emissions here (see also Sect. 4 discussion) (Tørseth et al., 2012; AQEG, 2015; Vieno et al., 2016; Ciarelli et al., 2019; Theobald et al., 2019). In contrast, South Asia is so ammonia-rich that reducing NH$_3$ concentrations has little impact on PM$_{2.5}$ (Ge et al., 2022). The situation in northern Europe exemplifies what to expect in terms of future policy making for the rest of the world.

Many marine areas are characterised as SO$_x$ sensitive but for a different reason than the SO$_x$ sensitivity in South Asia. In the marine areas, sulfate aerosol derived from oceanic emissions of DMS rather than from anthropogenic emissions is the major contributor to PM$_{2.5}$ (Quinn and Bates, 2011; Hoffmann et al., 2016; Novak et al., 2022).

Detailed examination of the magnitudes of the PM$_{2.5}$ sensitivities in each location to each precursor (rather than just their ranking) reveals more complicated regional characteristics. Figure 9 shows the spatial variabilities in PM$_{2.5}$ sensitivities to 40% reductions in emissions of NH$_3$, NO$_x$, and SO$_x$ individually, and collectively, for the four world regions. The regionally averaged PM$_{2.5}$ sensitivities are summarised in Fig. 10 and Table S5. PM$_{2.5}$ concentrations in East Asia show comparable sensitivities to individual emissions reductions in NH$_3$, NO$_x$, and SO$_x$, with the impacts of NH$_3$ and NO$_x$ emissions reductions being more concentrated in continental areas than those for SO$_x$ emissions reductions (Fig. 9). A 40% reduction in NO$_x$ emissions yields a maximum decrease in PM$_{2.5}$ of 11.5 µg m$^{-3}$ (12%) over southern China, while 40% reductions in NH$_3$ and
SO\(_x\) yield slightly smaller maximum PM\(_{2.5}\) decreases of 8.51 \(\mu\)g m\(^{-3}\) (10%) and 8.78 \(\mu\)g m\(^{-3}\) (9%), respectively. The regional average sensitivities of PM\(_{2.5}\) concentrations in East Asia to 40% reductions in individual precursors are ~8% (Fig. 10).

In contrast, PM\(_{2.5}\) concentrations in South Asia only show significant responses to SO\(_x\) emissions reductions, whilst NH\(_3\) emissions reductions have little effect, which is consistent with our previous finding that South Asia has the most ammonia-rich chemical climate for SIA formation (Ge et al., 2022). The dominant proportion of (NH\(_4\))\(_2\)SO\(_4\) in SIA compared to NH\(_4\)NO\(_3\) in South Asia (Fig. S6) also explains the small sensitivity of PM\(_{2.5}\) in this region to NO\(_x\) emissions reductions. The maximum PM\(_{2.5}\) decrease (7.02 \(\mu\)g m\(^{-3}\), 9%) for 40% SO\(_x\) emissions reductions is more than three times larger than the maximum PM\(_{2.5}\) decrease (2.20 \(\mu\)g m\(^{-3}\), 4%) for 40% NH\(_3\) emissions reductions. The decreases in regionally averaged PM\(_{2.5}\) concentrations in South Asia in response to 40% reductions in emissions of individual precursors are in the order 10% for SO\(_x\), 3% for NO\(_x\), and 1% for NH\(_3\).

In the Euro_Medi region, PM\(_{2.5}\) sensitivities vary from north to south. Northern and central Europe is most sensitive to NH\(_3\) and NO\(_x\) emissions reductions, for which the maximum decrease in PM\(_{2.5}\) is ~2.6 \(\mu\)g m\(^{-3}\) (16%) for 40% reductions, while the Mediterranean is more sensitive to SO\(_x\) emissions reductions, for which the maximum decrease in PM\(_{2.5}\) is 2.98 \(\mu\)g m\(^{-3}\) (12%) for 40% reductions. Regionally averaged, however, the PM\(_{2.5}\) concentrations in Euro_Medi show comparable sensitivities to the three precursors with decreases in the range 5-8% for 40% emissions reductions in individual precursors.

Over North America, the eastern US shows larger sensitivities of PM\(_{2.5}\) to all emissions reduction scenarios than the western US, and reductions in NO\(_x\) emissions yield larger decreases in PM\(_{2.5}\) than reductions in SO\(_x\) and NH\(_3\) emissions. The maximum decrease in PM\(_{2.5}\) derived from 40% reductions in NO\(_x\) emissions is 3.10 \(\mu\)g m\(^{-3}\) (16%); for 40% reductions in NH\(_3\) and SO\(_x\) emissions the maximum PM\(_{2.5}\) decreases are 2.37 \(\mu\)g m\(^{-3}\) (14%) and 1.35 \(\mu\)g m\(^{-3}\) (10%) respectively. The regional average sensitivities of PM\(_{2.5}\) concentrations in North America to 40% reductions in emissions of individual precursors decreases in a slightly different order: NO\(_x\) (8%), SO\(_x\) (7%), and NH\(_3\) (4%).

Figure 10 shows that 20% emissions reductions in any precursor lead to decreases in regionally averaged PM\(_{2.5}\) concentrations, although the PM\(_{2.5}\) sensitivities vary substantially with precursor and region. Given the non-one-to-one chemical responses of SIA components to reductions in emissions in individual precursors discussed in Sect. 3.1, even 20% reductions appear substantial enough to ensure that decreased SIA formation due to decreased precursor emissions dominates over any disbenefits to SIA formation from, for example, increases in oxidant levels induced by NO\(_x\) emissions reductions.

For instance, 20% reductions in NO\(_x\) emissions still cause a decrease of 0.77 \(\mu\)g m\(^{-3}\) (3%) in regional average PM\(_{2.5}\) in East Asia, despite increasing regional average SO\(_2^2\) by 0.10 \(\mu\)g m\(^{-3}\) (2%) because it decreases regional average NH\(_4^+\) and fine NO\(_3^-\) by greater amounts (0.17 \(\mu\)g m\(^{-3}\), 5%) and 0.72 \(\mu\)g m\(^{-3}\) (14%) respectively). Similarly, 20% reductions in SO\(_x\) emissions decrease regional average PM\(_{2.5}\) in East Asia by 1.15 \(\mu\)g m\(^{-3}\) (4%) because the decreases in SO\(_2^2\) (1.06 \(\mu\)g m\(^{-3}\), 19%) and NH\(_4^+\) (0.32 \(\mu\)g m\(^{-3}\), 9%) caused by reduced (NH\(_4\))\(_2\)SO\(_4\) formation are larger than the increase in fine NO\(_3^-\) (0.20 \(\mu\)g m\(^{-3}\), 4%) due to elevated NH\(_4\)NO\(_3\) formation. On the other hand, the mitigation of PM\(_{2.5}\) by reducing emissions of all 3 precursors together is impacted by these non-one-to-one chemical responses as well, which causes the net decrease in regional average PM\(_{2.5}\) derived from reductions in all 3 precursors to be smaller than the sum of individual PM\(_{2.5}\) decreases derived from reductions in emissions of precursors individually. For example, 40% reductions in NH\(_3\), NO\(_x\), and SO\(_x\) emissions individually decrease regional average PM\(_{2.5}\) in East Asia by 2.03 \(\mu\)g m\(^{-3}\) (7%), 1.89 \(\mu\)g m\(^{-3}\) (7%), and 2.33 \(\mu\)g m\(^{-3}\) (8%) respectively (sum of the three: 6.25 \(\mu\)g m\(^{-3}\)), while the decrease in regional average PM\(_{2.5}\) derived from 40% reduction in all 3 precursors simultaneously is 5.59 \(\mu\)g m\(^{-3}\) (20%).

The 40% reduction in emissions of all 3 species yields a maximum decrease in PM\(_{2.5}\) of 23.9 \(\mu\)g m\(^{-3}\) (25%) over East Asia, followed by 10.4 \(\mu\)g m\(^{-3}\) (17%) in South Asia, 5.57 \(\mu\)g m\(^{-3}\) (22%) in Euro_Medi, and 5.05 \(\mu\)g m\(^{-3}\) (28%) in North America. The regional average sensitivity of PM\(_{2.5}\) concentrations to 20% and 40% reductions in emissions of all 3 species decreases in the order East Asia (10% and 20% for 20% and 40% reductions respectively), Euro_Medi (9% and 17%), North America (8%...
and 17%), and South Asia (7% and 13%). This trend is related to differences in the contribution of SIA to PM$_{2.5}$ in the different regions. Figure S6 shows the mass contributions of individual PM$_{2.5}$ components to the regional average concentration of PM$_{2.5}$ in the baseline and the 40% emissions reductions scenarios. SIA components in the baseline account for over half of PM$_{2.5}$ in East Asia (52%), followed by Euro_Medi (42%), North America (35%), and South Asia (31%), which explains why reductions in emissions of all three SIA precursors are most efficient for the mitigation of PM$_{2.5}$ in East Asia but least efficient in South Asia. In fact, primary PM$_{2.5}$ is the largest contributor to PM$_{2.5}$ in South Asia, so reducing these emissions will be the most efficient way of abating PM$_{2.5}$ pollution in this region. It is noteworthy that in the scenario of 40% reductions in all 3 species, SIA is still the largest contributor to PM$_{2.5}$ in East Asia, while primary PM$_{2.5}$ and Rest (mainly secondary organic aerosol) become the dominant contributors in other regions. Even with 40% reductions in all three SIA precursors, none of the four regions has its regional average PM$_{2.5}$ concentration decreased to below 5 µg m$^{-3}$. Euro_Medi (8.4 µg m$^{-3}$ after 40% reductions) and North America (6.5 µg m$^{-3}$) are the closest, whilst East Asia (21.8 µg m$^{-3}$) and South Asia (27.0 µg m$^{-3}$) are still far away from achieving the latest WHO AQG for PM$_{2.5}$. Therefore, reductions in emissions of primary PM$_{2.5}$ and in VOCs are also required to achieve further PM$_{2.5}$ reductions in all regions, or even greater reductions in SIA precursors than simulated here.

Figure 9: Changes in PM$_{2.5}$ annual mean surface concentrations for 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually and collectively. The pink star in each map locates the minimum difference within each region.
3.3 Sensitivity of N, and S, deposition

The impacts of reductions in emissions of NH$_3$, NO, and SO$_x$ on total amounts of N and S deposition are straightforward because these must match the emissions mass changes in N and S. However, the relative amounts and spatial pattern of the individual components of N and S deposition are impacted.

Figure 11 shows the spatial variations in the sensitivities of the wet and dry deposition of the RDN components NH$_3$ and NH$_4^+$ to 40% reductions in global emissions of NH$_3$, NO, SO$_x$, individually, and collectively. Figure 12 shows similar for deposition of the OXN components NO$_x$, HNO$_3$, TNO$_3^-$ (total NO$_3^-$, the sum of fine and coarse NO$_3^-$) and Rest (the sum of other oxidized N species). Both wet and dry deposition of NH$_3$ show negative responses to emissions reductions in NH$_3$ and all 3 precursors together, but positive responses to emissions reductions in NO$_x$ and SO$_x$, which is consistent with the responses of surface NH$_3$ concentrations to these emissions reductions (Sect. 3.1.1). This is because reduced NO$_x$ and SO$_x$ emissions lead to decreased concentrations of acidic species in the atmosphere, resulting in more NH$_3$ remaining in the gas phase and
greater NH$_3$ deposition over continents. In contrast, global NH$_4^+$ wet and dry deposition decreases in all emissions reduction scenarios, which is also in line with the decreased NH$_4^+$ concentrations in all scenarios.

The changes in deposition of the OXN components to emissions reductions are more complicated (Fig. 12). The two species with the largest variation in deposition across the emissions reduction scenarios are HNO$_3$ and TNO$_3^-$, which is due to their large contributions to total OXN deposition in most world regions (Ge et al., 2022). For reductions in emissions of NO$_x$ and all 3 species, all OXN deposition components show clear decreasing trends due to the strong reduction in their precursor emissions.

In response to NH$_3$ emissions reductions, Fig. 12 shows that HNO$_3$ wet and dry deposition increases in eastern China, northern India, Europe, and eastern North America, whereas the wet and dry deposition of TNO$_3^-$ decreases in these regions. Further examination of fine and coarse NO$_3^-$ deposition differences in Fig. S7 shows that the decrease in TNO$_3^-$ deposition is driven by the decrease in fine NO$_3^-$ wet and dry deposition, while coarse NO$_3^-$ wet and dry deposition in the four regions actually increases. The reduction in NH$_3$ emissions decreases NH$_4$NO$_3$ formation and therefore liberates more HNO$_3$; as a result, more OXN deposits in the form of HNO$_3$ rather than NO$_3^-$ in these regions. In contrast, impacts of SO$_x$ emissions reductions on HNO$_3$ and TNO$_3^-$ deposition are the opposite of impacts of NH$_3$ emissions reductions. As discussed in Sect. 3.1.2, decreased SO$_4^{2-}$ concentrations promote the formation of NH$_4$NO$_3$, which results in decreased wet and dry deposition of HNO$_3$ and consequently increased TNO$_3^-$ deposition (driven by increased fine NO$_3^-$ deposition, Fig. S7) in eastern China, northern India, Europe, and eastern North America.

Compared to HNO$_3$ and TNO$_3^-$, the responses of NO$_x$ and Rest OXN deposition to reductions in NH$_3$ and SO$_x$ emissions are considerably smaller. For instance, the maximum increases in HNO$_3$ wet and dry deposition in response to 40% NH$_3$ emissions reductions are 328 mgN m$^{-2}$ (85%) and 248 mgN m$^{-2}$ (48%) respectively, whereas the maximum increases in Rest OXN wet and dry deposition are only 1.35 mgN m$^{-2}$ (4%) and 1.64 mgN m$^{-2}$ (4%) respectively. Also, in the 40% NH$_3$ emissions reduction scenario, the maximum decreases in TNO$_3^-$ wet and dry deposition are 359 mgN m$^{-2}$ (25%) and 175 mgN m$^{-2}$ (28%) respectively, whereas the maximum decreases in NO$_x$ dry deposition are only 4.22 mgN m$^{-2}$ (1%).
Figure 11: Changes in wet (WDEP) and dry deposition (DDEP) of NH$_3$ and NH$_4^+$ for 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.

Figure 11: Changes in wet (WDEP) and dry deposition (DDEP) of NH$_3$ and NH$_4^+$ for 40% emissions reductions in NH$_3$, NO$_x$, and SO$_x$ individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.
Figure 12: Changes in wet (WDEP) and dry deposition (DDEP) of NO\textsubscript{x}, HNO\textsubscript{3}, TNO\textsubscript{3}{-} (fine + coarse NO\textsubscript{3}{-}), and Rest OXN species for 40% emissions reductions in NH\textsubscript{3}, NO\textsubscript{x}, and SO\textsubscript{x} individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.

The spatial variations in the sensitivities of SO\textsubscript{2} and SO\textsubscript{4}{2-} wet and dry deposition to the precursor emissions reductions are shown in Fig. 13. Increased SO\textsubscript{2} wet deposition is observed globally in response to NH\textsubscript{3} emissions reductions, whilst SO\textsubscript{2} dry deposition decreases over the continents but increases over the oceans. The non-stomatal canopy resistance of SO\textsubscript{2} is positively correlated to the molar acidity ratio $\alpha_{SO}$ (Smith et al., 2000; Erisman et al., 2001; Fowler et al., 2009; Massad et al., 2010; Simpson et al., 2012). Reductions in NH\textsubscript{3} emissions increase the $\alpha_{SO}$, which therefore increases the canopy resistance...
of SO\textsubscript{2} and decreases SO\textsubscript{2} dry deposition over the continents. The 40% reduction in NH\textsubscript{3} emissions yields a global maximum increase in SO\textsubscript{2} wet deposition of 131 mgS m\textsuperscript{-2} (44%), and a maximum decrease in SO\textsubscript{2} dry deposition of 600 mgS m\textsuperscript{-2} (24%).

The overall effect of increased SO\textsubscript{2} wet deposition and decreased SO\textsubscript{2} dry deposition is a decreased SO\textsubscript{2} total deposition over populated continents (where NH\textsubscript{3} emissions are high) and increased SO\textsubscript{2} total deposition over oceans (Fig. S4). The maximum decrease of SO\textsubscript{2} total deposition located in southern China is 586 mgS m\textsuperscript{-2} (18%) for 40% NH\textsubscript{3} reduction.

The sensitivity of total deposition of SO\textsubscript{4}\textsuperscript{2-} to NH\textsubscript{3} emissions reductions (Fig. S5) follows the trend in the sensitivity of SO\textsubscript{4}\textsuperscript{2-} concentrations (Sect. 3.1.3). The responses of wet and dry deposition of SO\textsubscript{4}\textsuperscript{2-} to NH\textsubscript{3} emissions reductions are similar (Fig. 13). In general, decreased wet and dry SO\textsubscript{4}\textsuperscript{2-} deposition appears over Europe, north-eastern China, and north-eastern US, while increased wet and dry deposition occurs in the rest of the world. As also for their concentration sensitivities, the magnitudes of the SO\textsubscript{4}\textsuperscript{2-} deposition responses are much smaller than for SO\textsubscript{2}. For 40% NH\textsubscript{3} emissions reduction, the maximum decrease of SO\textsubscript{4}\textsuperscript{2-} total deposition is 42 mgS m\textsuperscript{-2} (9%), which is an order of magnitude smaller than that of SO\textsubscript{2} total deposition.

For NO\textsubscript{x} emissions reductions, both wet and dry deposition of SO\textsubscript{2} generally show decreases in eastern China, Europe, and north-eastern US but increases in the rest of the world, which is contrary to the responses of SO\textsubscript{4}\textsuperscript{2-} wet and dry deposition.

This is related to enhanced chemical conversion of SO\textsubscript{2} to SO\textsubscript{4}\textsuperscript{2-} due to increased atmospheric oxidizing capacity over eastern China, Europe, and north-eastern US (details in Sect. 3.1.3). The maximum decrease in SO\textsubscript{2} wet and dry deposition in response to 40% reductions in NO\textsubscript{x} emissions is \textasciitilde 65 mgS m\textsuperscript{-2}, while the maximum increase in SO\textsubscript{4}\textsuperscript{2-} wet and dry deposition is \textasciitilde 50 mgS m\textsuperscript{-2}.

For reductions in SO\textsubscript{x} and in all 3 precursors collectively, decreased wet and dry deposition of SO\textsubscript{2} and SO\textsubscript{4}\textsuperscript{2-} is observed globally and the 40% reductions in these two scenarios yield similar global maximum decreases in SO\textsubscript{2} deposition (wet: \textasciitilde 937 mgS m\textsuperscript{-2}, 41%; dry: \textasciitilde 1338 mgS m\textsuperscript{-2}, 43%) and SO\textsubscript{4}\textsuperscript{2-} deposition (wet: \textasciitilde 828 mgS m\textsuperscript{-2}, 38%; dry: \textasciitilde 150 mgS m\textsuperscript{-2}, 39%).

Figure 13: Changes in wet (WDEP) and dry deposition (DDEP) of SO\textsubscript{2} and SO\textsubscript{4}\textsuperscript{2-} for 40% emissions reductions in NH\textsubscript{3}, NO\textsubscript{x}, and SO\textsubscript{x} individually and collectively. Red and green dots in each map locate the minimum and maximum difference, respectively.
Figure 14 shows the differences of regional total deposition of individual species between baseline and the 40% emissions reduction scenarios. The regional total RDN, OXN and OXS deposition and the contributions of individual components are listed in Table S6 and S7. As expected, the responses of regional total RDN, OXN and OXS deposition are essentially linear through 20% and 40% reductions in their corresponding precursor, and largely insensitive to reductions in the other precursors, although there are slight differences between regions. For example, 20% and 40% reductions in NH$_3$ emissions respectively yield 20-21% and 39-41% decreases in total deposition of RDN in the four regions, whilst having no effect on regional total OXN and OXS deposition. The contributions of different deposition components do, however, vary with emission changes and the different lifetimes of the deposition components contribute to the small variabilities in responses of total RDN, OXN and OXS deposition to emissions reductions via differences in the transport through the regional boundaries.

A 40% reduction in NH$_3$ emissions produces a decrease of 3.58 TgN yr$^{-1}$ (51% of regional total deposition of the same species in baseline; similarly, hereinafter) in NH$_3$ total deposition and 1.16 TgN yr$^{-1}$ (24%) in NH$_4^+$ total deposition over East Asia (Fig. 14), which causes the contribution of NH$_3$ to RDN deposition to decrease from 59% (baseline) to 47% (after 40% NH$_3$ reduction). In other regions, reductions in NH$_3$ emissions also decrease NH$_3$ deposition more effectively than NH$_4^+$ deposition. Also in East Asia, a 40% NH$_3$ emissions reduction increases HNO$_3$ deposition by 0.61 TgN yr$^{-1}$ (20%) and SO$_2^{2-}$ deposition by 0.06 TgS yr$^{-1}$ (2%) but decreases TNO$_3^-$ deposition by 0.55 TgN yr$^{-1}$ (14%) and SO$_2$ deposition by 0.12 TgS yr$^{-1}$ (3%). As a result, the contribution of HNO$_3$ deposition to total OXN deposition increases from 30% to 37%, corresponding to a decrease in TNO$_3^-$ contribution from 54% to 47%, whereas changes in contributions of SO$_2$ and SO$_2^{2-}$ to OXS deposition are very small (Table S7). In other world regions, such changes in total deposition of HNO$_3$, TNO$_3^-$, SO$_2$, and SO$_2^{2-}$ derived from NH$_3$ emissions reductions are similar but of smaller magnitude.

For 40% NO$_x$ emissions reductions, TNO$_3^-$ deposition shows the largest decrease in East Asia (1.57 TgN yr$^{-1}$, 35%), South Asia (0.44 TgN yr$^{-1}$, 26%), and Euro_Medi (0.71 TgN yr$^{-1}$, 37%), whilst HNO$_3$ deposition shows the largest decrease in North America (0.56 TgN yr$^{-1}$, 40%). In contrast, the sensitivities of NO$_x$ dry deposition to NO$_x$ emissions reductions are very small. The contributions of individual OXN deposition components remain fairly constant in all regions. Furthermore, East Asia, Euro_Medi, and North America show a 5% increase in the contribution of NH$_3$ deposition to total RDN deposition and a corresponding 5% decrease in NH$_x^+$ contribution for 40% NO$_x$ emissions reductions, which reflects a small shifting of gas-aerosol partitioning for RDN as well. This kind of contribution change in RDN deposition is 2% for South Asia. The impacts of NO$_x$ emissions reductions on OXS deposition compositions are very small.

The 40% reductions in SO$_x$ emissions yield 3.24 TgS yr$^{-1}$ (39%), 1.07 TgS yr$^{-1}$ (38%), 1.17 TgS yr$^{-1}$ (33%), and 0.78 (37%) TgS yr$^{-1}$ decreases in OXS deposition over East Asia, South Asia, Euro_Medi, and North America respectively. SO$_x$ emissions reductions cause larger decreases in SO$_2$ deposition than in SO$_2^{2-}$ deposition in East Asia and Euro_Medi, while SO$_2$ and SO$_2^{2-}$ deposition in South Asia and North America show similar sensitivities. This is associated with slightly greater proportions of SO$_2$ (56-58%) to OXS deposition in the former regions than in the latter regions, and that these proportions are not affected by SO$_x$ emissions reductions (Table S7). NH$_3$ and NH$_4^+$ deposition is moderately sensitive to SO$_x$ emissions reductions in the four regions. An increase of 0.75 TgN yr$^{-1}$ (11%) in NH$_3$ total deposition and a decrease of 0.71 TgN yr$^{-1}$ (14%) in NH$_4^+$ total deposition for 40% reductions in SO$_x$ emissions is observed over East Asia. For South Asia, Euro_Medi, and North America, the increases in NH$_3$ total deposition due to 40% SO$_x$ reductions are 0.51 TgN yr$^{-1}$ (12%), 0.25 TgN yr$^{-1}$ (11%), and 0.23 TgN yr$^{-1}$ (12%) respectively. This is because reduced SO$_x$ emissions lead to reductions in (NH$_4$)$_2$SO$_4$ formation which then cause increased NH$_3$ but decreased NH$_4^+$ concentrations. Another side effect of SO$_x$ emissions reductions in East Asia is a slight decrease in HNO$_3$ deposition (0.25 TgN yr$^{-1}$, 10%) and an equivalent increase in TNO$_3^-$ deposition (0.21 TgN yr$^{-1}$, 5%). The equivalent deposition changes are considerably smaller in the other three regions, which again indicates a larger amount of NH$_3$NO$_3$ pollution in East Asia than other regions.

For collective reductions in emissions of all precursors, the changes in deposition of each species reflect net effects of individual reductions in emissions of NH$_3$, NO$_x$, and SO$_x$. For instance, Fig. 14 shows that the decrease in NH$_3$ deposition in
East Asia derived from 40% reduction in emissions of all 3 species (2.58 TgN yr\(^{-1}\), 36%) is smaller than that from individual
NH\(_3\) emissions reduction (3.58 TgN yr\(^{-1}\), 51%) due to the compensating effects of simultaneous NO\(_x\) and SO\(_x\) emissions reductions in the former scenario. In contrast, the decrease in NH\(_4^+\) deposition in East Asia for 40% emissions reductions in all 3 species (2.11 TgN yr\(^{-1}\), 43%) is almost double that from individual NH\(_3\) emissions reduction scenario (1.16 TgN yr\(^{-1}\), 24%). The variations in chemical forms of RDN, OXN, and OXS deposition affect where they deposit as well since N, and S, species have different lifetimes and a shorter lifetime causes a more localised deposition. Many studies show that NH\(_3\) and HNO\(_3\) have shorter lifetimes than NH\(_4^+\) and NO\(_3^-\) (Xu and Penner, 2012; Hauglustaine et al., 2014; Bian et al., 2017; Ge et al., 2022). The abatement of total N (TN = RDN + OXN) and S deposition within a certain region is partially offset by this more localised deposition pattern especially in South Asia and Euro_Medi. The 40% reductions in all 3 species emissions yield a 34% (2.93 TgN yr\(^{-1}\)) decrease in regional TN deposition in South Asia, and a 34% (1.18 TgS yr\(^{-1}\)) decrease in regional OXS deposition in Euro_Medi, which means that less N, and S, pollution is transported outside these regions, and more is deposited locally.

Figure 14: The absolute sensitivities (Emission Reduction - Baseline) of regional total deposition (wet + dry) of NH\(_3\), NH\(_4^+\), NO\(_x\), HNO\(_3\), TNO\(_3^-\) (fine + coarse NO\(_3^-\)), SO\(_2\) and SO\(_4^{2-}\) to 40% emissions reductions in NH\(_3\) (top row), NO\(_x\) (2nd row) and SO\(_x\) (3rd row) individually, and collectively (bottom row), for the four regions defined in Fig. 1. The left-hand y axis in each panel is for RDN and OXN species, while the right-hand y axis is for OXS species.
4 Discussion

We use global model simulations for 8 emissions reduction scenarios to investigate the geographical variation in the effectiveness of mitigation of \( \text{N}_x \), \( \text{S}_x \), and PM2.5 pollution to SIA precursor gas emissions reductions. Although the EMEP MSC-W ACTM is state-of-the-art and widely used in scientific research and policy development, the analyses presented in this study are based on data from a single model. The accurate representation of relevant chemical and physical processes in the model is crucial for simulations of sensitivities of \( \text{N}_x \), \( \text{S}_x \), and PM2.5 to emissions reductions in inorganic precursor gaseous. It is possible that variations in aerosol water content under different relative humidity conditions may impact on the exact mass sensitivities of PM2.5. However, the evaluation of surface concentrations and wet deposition of \( \text{N}_x \) and \( \text{S}_x \) species from this model configuration for the same year against global measurements from 10 monitoring networks (Ge et al., 2021b) has demonstrated the model’s capability to capture the overall spatial variations in annual concentrations of NH\(_3\), NH\(_4^+\), NO\(_2\), HNO\(_3\), fine NO\(_3^-\), SO\(_2\), SO\(_4^{2-}\) and their wet deposition in East Asia, Southeast Asia, Europe, and North America. The uncertainties in both model and measurement constrains the extent to which the agreement between model and measurement can be used to evaluate a model’s performance. However, the sensitivities of global and regional \( \text{N}_x \), \( \text{S}_x \), and PM2.5 to various emissions reductions can only be investigated through modelling experiments, and since most model uncertainties will be similar across a set of simulations with the same model configuration, the modelled changes in concentrations between baseline and an emissions reduction scenario should be robust. Nevertheless, considering the fundamental uncertainties in emissions and model parameterizations, all numbers reported in this work should be considered as having uncertainty, albeit that the latest available global emissions inventory and model version were used to minimize the impacts of these uncertainties.

The simulations of emissions reduction scenarios show that the reduction in emissions of one individual precursor has multiple co-benefits and sometimes small disbenefits on mitigating \( \text{N}_x \), \( \text{S}_x \), and PM2.5 pollution, and these effects are geographically variable. In this work, our discussion focuses on East Asia, South Asia, Euro_Medi, and North America because of the high population density and high \( \text{N}_x \) and \( \text{S}_x \) pollution in those regions. The comparison of regional responses to emissions reductions reveals differences in regional oxidation regime, SIA chemistry and deposition pattern which are important processes to consider when designing emissions control policies since transitory increases in PM2.5 and some \( \text{N}_x \) and \( \text{S}_x \) pollutants could occur as emissions reduction measures are gradually applied.

Globally, reductions in NH\(_3\) emissions are effective for reducing NH\(_3\) concentrations and its wet and dry deposition but considerably less effective at reducing NH\(_4^+\). This is because most world regions are in an ammonia-rich chemical domain in which reducing NH\(_3\) emissions only has limited effects on mitigating SIA formation (Ge et al. 2022). Other co-benefits of NH\(_3\) emissions reductions include reductions in fine NO\(_3^-\) surface concentrations and deposition in East Asia, South Asia, Euro_Medi, and North America because of reduced NH\(_3\)NO\(_3\) formation. A notable disbenefit is the increased SO\(_2\) surface concentration and human exposure in these regions which is caused by reduced SO\(_2\) dry deposition. The dry deposition velocity of SO\(_2\) is negatively correlated with the molar acidity ratio \( a_{\text{EN}} \) which is a model parameterisation derived from long-term deposition measurements (Erisman et al., 2001; Simpson et al., 2012). Reduced NH\(_3\) emissions therefore lead to decreased SO\(_2\) dry deposition.

Similarly, whilst reducing NO\(_x\) emissions is of course an effective way of decreasing global concentrations and deposition of OXN species, the degree to which different OXN species are decreased varies across regions. A 40% reduction in NO\(_x\) emissions decreases NO\(_x\) and fine NO\(_3^-\) surface concentrations in East Asia by 45% and 33% respectively, whereas in South Asia this measure has a greater effect on fine NO\(_3^-\) (45% decrease) than on NO\(_x\) (22% decrease). In Euro_Medi and North America, the 40% NO\(_x\) emissions reductions produce similar decreases in regional average NO\(_x\) (36-38%) and fine NO\(_3^-\) (41-42%) concentrations. These trends are consequent on different regional NO\(_x\) oxidation regime and SIA chemistry. The NO\(_x\) emissions reductions decrease NO\(_x\) surface concentrations, which increases \( \text{O}_3 \) concentrations in the high NO\(_x\) areas of eastern China and western and central Europe and therefore increases the atmospheric oxidizing capacity in these regions. As a result, more SO\(_2\) is oxidized to SO\(_4^{2-}\) which leads to decreased SO\(_2\) concentrations and deposition and consequently increased SO\(_4^{2-}\).
concentrations and deposition in these areas. The enhanced SO$_4^{2-}$ production can partially (or even totally) offset the mass reduction in PM$_{2.5}$ caused by reduced NH$_4$NO$_3$ formation when reductions in NO$_x$ emissions are not sufficiently high. The increased oxidant levels will also enhance HNO$_3$ and NO$_3^-$ production in these regions, but this effect does not compensate for the reduction in HNO$_3$ and NO$_3^-$ concentrations due to the reductions in NO$_x$ emissions (at least for 20% and 40% NO$_x$ reductions), so the net effect is globally decreased HNO$_3$ and NO$_3^-$. Consequently, reduced HNO$_3$ and NO$_3^-$ levels caused by NO$_x$ emissions reductions lead to less NH$_4$NO$_3$ formation, which then results in globally increased NH$_3$ concentration and deposition and decreased NH$_4^+$ concentration and deposition. In contrast, decreased O$_3$ concentrations in South Asia and North America in response to NO$_x$ emissions reductions result in less chemical conversion of SO$_2$ to SO$_4^{2-}$, which then causes increased SO$_2$ and decreased SO$_4^{2-}$ concentrations and deposition. Clappier et al. (2021) and Thunis et al. (2021) showed that the increased atmospheric oxidizing capacity induced by reductions in NO$_x$ emissions is the reason for increased PM$_{2.5}$ levels in the Po basin (Italy) especially during wintertime, with increased nitrate, sulfate and SOA concentrations all being closely related to increased O$_3$ levels. Balamurugan et al. (2022) reported that reductions in SIA were much smaller than NO$_x$ emissions reductions during COVID lockdown in Germany, which is because the increased oxidant levels (OH, NO$_3$ and O$_3$) enhanced the formation of sulfate and night-time nitrate which then partially offset the lockdown-induced PM$_{2.5}$ decreases. Fu et al. (2020) noted that the increased oxidation of NO$_x$ to HNO$_3$ due to increased O$_3$ levels makes winter haze NO$_3^-$ in the North China Plain (NCP) almost insensitive to 30% reductions in emissions of NO$_x$, while Le et al. (2020) also revealed an unexpected PM exacerbation caused by unfavourable meteorological conditions and intensified SIA formation due to elevated O$_3$ levels induced by NO$_x$ emissions reductions during COVID lockdown in China.

The greatest effect of SO$_x$ emissions reductions is the direct decrease in global concentrations and deposition of SO$_2$ and SO$_4^{2-}$, which then induces changes in gas-aerosol partitioning of NH$_3$-NH$_4^+$ and HNO$_3$-NO$_3^-$. As discussed above, the reduction in (NH$_4$)$_2$SO$_4$ formation frees more gaseous NH$_3$ and promotes NH$_4$NO$_3$ formation, leading to increased concentrations and deposition of NH$_3$ and fine NO$_3^-$ in all world regions. Considering that one SO$_4^{2-}$ takes up two NH$_3$ molecules under ammonia-rich conditions, but NO$_3^-$ only takes one, the net effect of SO$_x$ emissions reductions still causes globally decreased NH$_4^+$ concentrations and deposition. Liu et al. (2018) noted a significant increase in annual NH$_3$ concentrations caused by rapid SO$_x$ emissions reductions in the NCP. In addition, the shifting of RDN from aerosol-phase NH$_4^+$ to gaseous NH$_3$ in response to reduced SO$_x$ emissions also means that RDN pollution becomes more localized because NH$_3$ has a much shorter lifetime (1.6 days) than NH$_4^+$ (8.9 days) as it deposits more quickly to land rather than being transported to other regions (Ge et al. 2022). Utilizing combined measurements and modelling, Leung et al. (2020) found that the reduction in wintertime PM$_{2.5}$ in the NCP is buffered by enhanced NH$_2$NO$_3$ formation due to decreased SO$_4^{2-}$ concentration liberating free NH$_3$, and increased oxidant levels promoting HNO$_3$ production, despite SO$_2$ and NO$_x$ emissions reductions in China. However, it is important to note that (NH$_4$)$_2$SO$_4$ has greater molecular mass than NH$_4$NO$_3$ and hence has larger leverage on PM$_{2.5}$ mass concentration, which ensures that the reductions in PM$_{2.5}$ mass concentrations derived from reduced (NH$_4$)$_2$SO$_4$ are less readily compensated by increases in NH$_2$NO$_3$ concentrations. This also means that relative changes in the SO$_4^{2-}$ component cause greater mass changes with respect to PM$_{2.5}$ air quality objectives (which must be expressed as mass concentration) than do the same relative changes in the NO$_3^-$ component. The relative changes in the SIA components of PM$_{2.5}$ expressed as molar concentrations would be different.

The simulations for both 20% and 40% emissions reductions scenarios show that the most effective emissions control for decreasing regional average PM$_{2.5}$ concentrations, via an individual component, differs between world regions. PM$_{2.5}$ in South Asia is most sensitive to SO$_x$ emissions reductions, and least sensitive to NH$_3$ emissions reductions, which is because South Asia is extremely ammonia rich (Ge et al., 2022) so reducing NH$_3$ has little impact on mitigating SIA. In this study, 40% reductions in SO$_x$ and NH$_3$ emissions respectively produce 10% and 1% decreases in South Asia regional average PM$_{2.5}$. Pozzer et al. (2017) also showed that NH$_3$ emissions control has negligible impacts on PM$_{2.5}$ levels in South Asia with a 50% reduction in NH$_3$ emissions only reducing annual mean PM$_{2.5}$ by 2%. The most effective measure to reduce annual mean PM$_{2.5}$
in North America is reducing NO\textsubscript{x} emissions, followed by SO\textsubscript{x} and then NH\textsubscript{3}. Liao et al. (2008) reported that the reduction in NO\textsubscript{x} emissions was most effective for decreasing 24-h mean PM\textsubscript{2.5} levels for five cities in the US, while Kelly et al. (2021) reported that reducing NO\textsubscript{x} emissions was more effective for reducing PM\textsubscript{2.5} concentrations in the eastern US than SO\textsubscript{x}, NH\textsubscript{3}, and VOC emissions reductions in both January and July. Tsipmipid et al. (2007, 2008) showed that NO\textsubscript{x} emissions reductions were the most effective measure for controlling PM\textsubscript{2.5} in the eastern US in summer due to the combined effects of lower atmospheric oxidant levels and smaller precursor emissions. Unfortunately, no conclusion on annual mean PM\textsubscript{2.5} sensitivities was drawn from these studies.

In Euro\textsubscript{Medi}, PM\textsubscript{2.5} sensitivities are complex and vary from the north to south. The UK and Scandinavia are more sensitive to NH\textsubscript{3} emissions reductions, central Europe is more sensitive to NO\textsubscript{x} reductions, while the Mediterranean region is more sensitive to SO\textsubscript{x} reductions, which is consistent with conclusions from other European studies (Megaritis et al., 2013; Vieno et al., 2016; Aksoyoglu et al., 2020; Jiang et al., 2020; Clappier et al., 2021). From a perspective of European policy making, it is important to reduce NH\textsubscript{3}, NO\textsubscript{x} and SO\textsubscript{x} emissions together and/or go for stronger reductions to minimise adverse effects caused by enhanced oxidation efficiency. East Asia is another example of mixed sensitivity regimes where individual reductions in SO\textsubscript{x}, NO\textsubscript{x} and NH\textsubscript{3} emissions are almost equally effective for mitigating PM\textsubscript{2.5} at a country level. Studies focusing on East Asia highlight the importance of NH\textsubscript{3} emissions control for reducing annual PM\textsubscript{2.5} pollution especially in central and eastern China (Wang et al., 2011; Pozzer et al., 2017; Cheng et al., 2021). In addition, due to the high proportion of SIA in PM\textsubscript{2.5} in East Asia, reducing all 3 precursors also produces the greatest reduction in regional average PM\textsubscript{2.5} compared to other regions. It is possible that the above conclusions may change when considering population-weighted concentration, but we deliberately selected the four most densely populated regions for analysis in order to focus on reductions relevant to human exposure to SIA, so it is anticipated that findings using population weighting would be broadly similar.

Analyses of both 20% and 40% emissions reductions reveal different kinds of linearity and non-linearity. There is considerable non-linearity in the sense of a lack of one-to-one proportionality between an emission reduction and a species concentration or deposition change. Precursor emissions reduction sometimes even increases other pollutant concentrations. This is consequent on interactions in SIA formation, atmospheric oxidizing capacity, and N and S deposition as discussed earlier, and is highly geographically variable. Such non-linearity may be more significant in a certain area during a certain season due to the inter-annual variability in local meteorology and emissions profiles. However, whilst the sensitivity of an annual quantity of a pollutant to emissions reductions is subject to seasonality, we are confident that the broad conclusions of this study will hold, and from a global and regional policymaking perspective, it is more practical to develop policies of reductions in emissions from different sectors and countries on an annual level. Studies focusing on non-proportional responses of European PM\textsubscript{2.5} to emissions reductions showed that significant seasonality only occurs in a few specific areas (Thunis et al., 2015, 2021; Clappier et al., 2021).

On the other hand, a linearity in response to emissions reductions is apparent via the observation that the responses of PM\textsubscript{2.5}, N\textsubscript{a}, and S annual concentrations and deposition components remain essentially proportional to the precursor emissions reductions (20% and 40%) for a given precursor in a given region, albeit that the magnitude of the slope varies substantially with different precursors and regions. Even if the net concentration changes of one species (e.g., PM\textsubscript{2.5}) induced by reductions in emissions of all 3 precursors are smaller (or greater) than the sum of changes from reductions in individual precursors due to non-linear chemical interactions as discussed earlier, these net changes still follow a very similar gradient when emissions reductions in all 3 precursors change from 20% to 40%. However, where the gradient in the response is not one-to-one, the linearity in the response that is observed up to the 40% emissions reductions simulated here clearly cannot continue to extrapolate linearly all the way to 100% emissions reductions. The gradient of the response must be flatter or steeper (depending on atmospheric component) at the beginning or end of the span from zero to 100% emissions reduction. For instance, 20% and 40% reductions in NH\textsubscript{3} emissions give 24% and 48% decreases in global annual mean NH\textsubscript{3} concentrations respectively (i.e., gradients exceeding one-to-one), whereas these emissions reductions only produce 6% and 14% decreases...
in global NH\textsubscript{4}\textsuperscript{+} concentrations, respectively (i.e., gradients less than one-to-one). When NH\textsubscript{3} emissions are completely switched off, both NH\textsubscript{3} and NH\textsubscript{4}\textsuperscript{+} concentrations will be zero, so the NH\textsubscript{3} concentration sensitivity must become smaller, and the NH\textsubscript{4}\textsuperscript{+} concentration sensitivity must become larger as NH\textsubscript{3} emissions reductions approach 100%. Additional serial sensitivity experiments are required to acquire the full spectrum of NH\textsubscript{3} and NH\textsubscript{4}\textsuperscript{+} (and all other species) sensitivities. It is therefore important for policymakers in different regions to know the emissions reductions required to obtain the mitigation responses needed for specific air quality targets.

Finally, it is also important to remember that reductions in anthropogenic emissions of SIA precursors will have many co-benefits on human health, forests, ecosystem biodiversity, and climate, not just in populated areas but elsewhere.

5 Conclusions

The sensitivities of global and regional annual mean surface concentrations and deposition of gaseous and particle N\textsubscript{2}O\textsubscript{5} and S\textsubscript{O}\textsubscript{2} to 20% and 40% reductions in anthropogenic emissions of NH\textsubscript{3}, NO\textsubscript{x}, and SO\textsubscript{2} both individually and collectively has been investigated using the EMEP MSC-W model coupled with WRF meteorology for 2015. East Asia, South Asia, Euro_Medi, and North America are selected for regional discussions because of their high population densities and N\textsubscript{2}O\textsubscript{5} and S\textsubscript{O}\textsubscript{2} pollution. The comparison in regional responses reveals that the individual emissions reduction in one precursor has multiple co-benefits and sometimes small disbenefits on mitigating N\textsubscript{2}O\textsubscript{5}, S\textsubscript{O}\textsubscript{2}, and PM\textsubscript{2.5} pollution, and these effects are highly geographically variable.

From a global policy-making perspective, whilst reductions in NH\textsubscript{3} emissions are effective for decreasing annual NH\textsubscript{3} concentrations and deposition they are considerably less effective at decreasing NH\textsubscript{4}\textsuperscript{+}. This is because all densely populated continents are ammonia rich so reducing NH\textsubscript{3} emissions only has limited effects on mitigating SIA formation. A 40% reduction in NH\textsubscript{3} emissions decreases regional average NH\textsubscript{3} concentrations in the four regions by 47-49%, while NH\textsubscript{4}\textsuperscript{+} concentrations decrease in the order Euro_Medi (18%), East Asia (15%), North America (12%), and South Asia (4%), the order of increasing regional ammonia-richness. Other side effects of NH\textsubscript{3} emissions reductions include decreases in annual fine NO\textsubscript{3}\textsuperscript{-} surface concentrations and deposition in the four regions due to reduced NH\textsubscript{4}NO\textsubscript{3} formation, and consequently to increased HNO\textsubscript{3} and coarse NO\textsubscript{3}\textsuperscript{-} concentrations and deposition in these regions. Another disbenefit is increased SO\textsubscript{2} surface concentrations in these regions because reduced NH\textsubscript{3} levels affect the pH-dependent SO\textsubscript{2} dry deposition sink. A 40% reduction in NH\textsubscript{3} emissions increases SO\textsubscript{2} concentrations in East Asia by 16%, in South Asia and North America by 14%, and in Euro_Medi by 10%.

Large regional differences are observed in NO\textsubscript{x} emissions reduction scenarios. In East Asia, NO\textsubscript{x} concentrations are very effectively decreased (by 45%) with 40% NO\textsubscript{x} emissions reductions, but they are less effectively decreased in Euro_Medi (38%) and North America (36%) and substantially less effectively decreased in South Asia (22%). By contrast, the regional sensitivities of fine NO\textsubscript{3}\textsuperscript{-} are reversed: South Asia shows the largest decrease (45%), whilst East Asia shows the smallest decrease (33%). This phenomenon is related to different regional oxidation regime and SIA chemistry. NO\textsubscript{x} emissions reductions increase O\textsubscript{3} levels in East Asia (and also, but by less, in Euro_Medi), but decrease O\textsubscript{3} levels in South Asia (and also, but by less, in North America). In East Asia, the increased oxidant levels enhance NO\textsubscript{x} oxidation efficiency, which results in a larger chemical removal of NO\textsubscript{x}. Consequently, the decrease in fine nitrate in East Asia is buffered by its enhanced chemical production. In contrast, decreased oxidant levels in South Asia lessen the oxidation of NO\textsubscript{x} and therefore partially offset the decrease in NO\textsubscript{3}\textsuperscript{-} concentrations due to emissions reductions, and cause a more efficient decrease in fine NO\textsubscript{3}\textsuperscript{-} concentrations. In addition, the reductions in fine NO\textsubscript{3}\textsuperscript{-} levels also cause decreases in NH\textsubscript{4}\textsuperscript{+} concentrations and deposition and corresponding increases in NH\textsubscript{3} concentrations and deposition, albeit to varying degrees in the different regions. The variations in regional oxidant levels also impact the regional oxidation of SO\textsubscript{2} to SO\textsubscript{4}\textsuperscript{2-}. A 40% NO\textsubscript{x} emissions reduction causes a 5% decrease in SO\textsubscript{2} and consequently a 3% increase in SO\textsubscript{4}\textsuperscript{2-} concentrations in East Asia, but a 2% increase in SO\textsubscript{2} and consequently a 4% decrease in SO\textsubscript{4}\textsuperscript{2-} concentrations in South Asia.
Reductions in SOx emissions have globally consistent impacts on SO2 and SO42- concentrations. A 40% reduction in SOx emissions decreases SO2 and SO42- concentrations in the four regions by 42-45% and 34-38% respectively. Reduced SO42- concentrations decrease (NH4)2SO4 formation but promote NH4NO3 formation, which then causes increased NH3 and fine NO3- but decreased NH4+ concentrations and deposition. A 40% reduction in SOx emissions yields ~12% growth in NH3 total deposition in four regions.

PM2.5 sensitivities in South Asia to 40% emissions reductions in precursors individually decrease in the order SOx (3.10 μg m⁻³, 10%), NOx (0.97 μg m⁻³, 3%), and NH3 (0.29 μg m⁻³, 1%), which is consistent with the fact that South Asia is so ammonia-rich that reducing NH3 hardly has any impacts on mitigating PM2.5. The most effective individual measure for North America is reducing NOx emissions with an 8% (0.63 μg m⁻³) decrease in regional average PM2.5 in response to a 40% reduction. In Euro_Medi, PM2.5 sensitivities vary from the north to south with regional average sensitivities to 40% reductions in individual precursors in the range 5-8% (0.55-0.82 μg m⁻³). The UK and Scandinavia are more sensitive to NH3 emissions reductions, central Europe is more sensitive to NOx emissions reductions, while the Mediterranean is more sensitive to SOx emissions reductions. East Asia is another example of mixed sensitivity regimes where individual reductions in SOx, NOx and NH3 emissions are almost equally effective for mitigating PM2.5, with regional sensitivities to 40% reductions in the range 7-8% (1.89-2.33 μg m⁻³). Because of the varying contributions of SIA to PM2.5, the relative sensitivities of PM2.5 to 40% reductions in all 3 precursors simultaneously decrease in the order East Asia (5.59 μg m⁻³, 20%), Euro_Medi (1.78 μg m⁻³, 17%), North America (1.31 μg m⁻³, 17%), and South Asia (4.13 μg m⁻³, 13%).

The simulations conducted in this work reveals geographically-varying non-linear physicochemical responses of N, S, and PM2.5 to emissions reductions. It is important not only to prioritise different precursor reductions in different regions, but also reduce emissions of precursors together in order to minimise various disbenefits.

Code and data availability

As described and referenced in Sect. 2 of this paper, this study used two open-source global models: the European Monitoring and Evaluation Programme Meteorological Synthesizing Centre – West atmospheric chemistry transport model (EMEP MSC-W, 2020, version 4.34, source code available at https://doi.org/10.5281/zenodo.3647990, last access: 8 Aug 2022) and the Weather Research and Forecasting meteorological model (WRF, version 3.9.1.1, https://www.wrf-model.org, last access: 8 Aug 2022; Skamarock et al., 2008). The model outputs presented in figures and tables in this paper and the corresponding Python scripts are available at https://doi.org/10.5281/zenodo.7082661, last access: 16 Sep 2022 (Ge, 2022).

Author contribution

MH, DS and MV conceptualised and supervised the study. MV and PW contributed to model development and set-up and provided modelling support. MV provided computing resource. YG contributed to study design, undertook all model simulations, formal data analyses, visualisation of the results and data curation, with discussion and refinement by all authors. The original draft of the paper was written by YG with contributions and editing by MH. All authors provided review comments and approval of the final version.

Competing interests

The authors declare that they have no conflict of interest.
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