Answers to 4 Policy-Relevant Questions about Hemispheric Transport Based on HTAP2, AQMEII3, MICS3, and TOAR

Prepared by Terry Keating, Frank Dentener,[1] Heather Morrison, Tim Butler, and Jacek Kaminski,
Co-Chairs and Vice-Chairs of the Task Force on Hemispheric Transport of Air Pollution

Executive Summary

Starting in 2012, the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) began an ambitious effort to conduct a nested global and regional-scale multi-model intercomparison using a new global emissions dataset compiled for this purpose. The main objectives of the intercomparison were:

- to conduct base simulations that could be compared between models and with observations, providing a basis for assessing the uncertainty in model results;
- to conduct a large suite of sensitivity analyses that would allow the development and testing of a parameterized set of source/receptor relationships that would form the basis of a global model emulator that could be used to estimate the effect of emissions changes in a source region on air quality and its impacts in a receptor region.

One goal of the effort was to produce an emulator, with some quantitative characterization of uncertainties, that could be used to assess a wide-variety of policy-relevant questions or emissions scenarios at the global scale.

The HTAP2 (and associated AQMEII-3 and MICS-Asia 3) exercises constitute one of the largest air quality model intercomparisons ever undertaken. A special issue of the journal *Atmospheric Chemistry and Physics* collected 48 articles from 2015 to 2018 reporting results from the effort so far, and additional scientific publications are still being produced. The multi-model ensemble of simulations provides a useful data base and set of source/receptor relationships that the TF HTAP can continue to use to address policy-relevant questions in the future. However, some policy-relevant messages are clear from the ensemble of base and sensitivity simulations and are highlighted below.

- The HTAP2 modeling results are consistent with the conclusions of TF HTAP's 2010 report:
  - The impact of extra-regional anthropogenic emissions is more important for ground-level ozone than for particulate matter or sulfur or nitrogen deposition.
    - Extra-regional emissions are more important for column ozone burdens and ozone direct radiative forcing than for surface ozone concentrations.
    - For surface ozone, the impact of extra-regional emissions sources is highest in Spring and lowest during Summer.
    - For surface ozone, the impact of extra-regional emission sources is larger for longer-term average concentrations than for peak concentrations that tend to be driven by local emissions.
  - Methane, in combination with nitrogen oxides, is an important ozone precursor on a global basis, and projected increases in methane emissions could, over time, offset the ozone decreases associated with decreases of emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs).
- For Europe, ground-level ozone concentrations are more sensitive to anthropogenic emissions outside of Europe than to anthropogenic emissions inside of Europe. This is the case for all

[1] Frank Dentener was Co-Chair of the TF HTAP until the end of 2018.

European sub-regions, all seasons, and for a range of ozone metrics including annual and seasonal averages, SOMO35, and POD1.[2] The relative influence of these extra-regional emissions on ozone in Europe varies by location, season, and ozone metric. Extra-regional anthropogenic emissions of NO_x and VOCs outside of Europe are estimated to contribute between 2-12 ppb of ozone depending on the season, with an annual average contribution of 4-8 ppb of ozone depending on the model used. The contribution of anthropogenic methane emissions to ozone in Europe is estimated to be about 5-8 ppb (on the basis of 6mDMA1).[3]

- For North America, region-wide annual average ozone concentrations appear equally sensitive to anthropogenic emissions outside of North America and emissions inside of North America. Extra-regional anthropogenic emissions of NO_x and VOCs outside of North America and changes in global methane, together, are estimated to contribute between 8-13 ppb of ozone in the western United States, 2-12 ppb of ozone in the central United States, and 2-10 ppb of ozone in the eastern United States, depending on the season.
- Maritime shipping is well understood to have significant impacts on air quality in coastal regions. In the HTAP2 experiments, all maritime shipping was considered to be extra-regional, leading to an increase in the estimated extra-regional contribution to air pollution levels in all regions compared to HTAP1. Further analysis is needed to understand the source-receptor relationships associated with shipping in different coastal regions subject to ECAs (Emission Control Areas) compared to shipping emissions in the open ocean.
- In HTAP2, the differences in predictions of particulate matter concentrations from different models decreased relative to the HTAP1 but this was not the case for ozone concentrations. The lack of improvement in model agreement for ozone was surprising given that all HTAP2 models used the same emissions inputs, whereas in HTAP1 emissions inputs were not harmonized. More work is needed to understand the processes contributing to the spread in model predictions, but analysis to date suggests that differences in the vertical transport and deposition processes between models may be resulting in this spread. Higher resolution regional models generally performed better in comparison to observations than did the coarser resolution global models. However, the best performing global models compared better to observations than did the worst performing regional models.

[2] SOMO35 = the annual sum of the positive differences between the daily maximum 8-hour average ozone value and the cutoff value set at 35 ppb; POD1 = phytotoxic ozone dose (accumulated stomatal flux above a threshold of 1 nmol/m^2/s) accumulated over a growing season

[3] 6mDMA1 = the running mean of the 6-month average of the daily maximum 1-hour ozone concentration

Introduction

The Task Force on Hemispheric Transport of Air Pollution (TF HTAP) was created by the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) in December 2004 to improve the understanding of the intercontinental transport of air pollutants across the Northern Hemisphere. Under the leadership of the European Union and the United States of America, the Task Force organized a series of projects and collaborative experiments (hereafter referred to as HTAP1) that culminated in 2010 with the publication of the first comprehensive assessment (hereafter referred to as the 2010 Assessment) of the intercontinental transport of ozone (O_3), particulate matter (PM), mercury (Hg) and persistent organic pollutants (POPs)(Dentener 2010; Dutchak 2010; Keating 2010a; Keating 2010b; Keating 2010c; Pirrone 2010).

The TF HTAP's mandate was further elaborated in 2010 (Executive Body for the Convention on Long-range Transboundary Air Pollution 2010) and the TF HTAP began a second series of projects and collaborative experiments in 2012 (hereafter referred to as HTAP2). The HTAP2 experiments included a series of nested simulations at the global and regional scale that were made possible through a collaboration with two ongoing regional-scale model intercomparison activities: the Air Quality Model Evaluation International Initiative (AQMEII), focused on Europe and North America,(Galmarini; Rao 2011) and the Model Inter-Comparison Study – Asia (MICS), encompassing South and East Asia (Carmichael 2002; Carmichael 2008; Fu In press). Regional modelers participating in AQMEII (Phase 3) and MICS (Phase 3) were provided boundary conditions for base case and sensitivity simulations from a selected group of global models participating in HTAP2 to enable nested base case and sensitivity simulations at higher spatial and temporal resolutions, while maintaining consistency with the ensemble of global models. This body of work culminated in the publication of 48 articles in a special issue of the journal Atmospheric Chemistry and Physics entitled "Global and regional assessment of intercontinental transport of air pollution: results from HTAP, AQMEII, and MICS (Dentener 2019)."

Building in part on efforts under TF HTAP to support global model evaluation and in parallel with the HTAP2 modeling experiments, development of the first Tropospheric Ozone Assessment Report (TOAR) began in 2014 under the auspices of the International Global Atmospheric Chemistry Project (IGAC) Project and FutureEarth. The goals of TOAR were to compile a global database of tropospheric ozone observations and to generate a series of consistent metrics that would enable trend analysis of tropospheric ozone's health, ecosystem, and climate impacts. The resulting database and initial analyses of trends have been published as a series of articles in a 2018 special issue of the journal Elementa (Cooper 2020).

Based on these recent contributions to the scientific literature, the co-chairs of TF HTAP have summarized the key lessons learned in the form of answers to four questions relevant to the LRTAP Convention:

- Based on current models, what is the contribution of and sensitivity to anthropogenic emissions sources outside Europe and North America to O_3 and PM concentrations and S and N deposition inside Europe and North America, respectively?
- How have these contributions or sensitivities changed?
- What confidence do we have in the estimates of these contributions and sensitivities?
- How are these contributions and sensitivities expected to change and what emissions sectors contribute significantly?

Before discussing what we have learned with respect to each of these questions, it is useful to summarize how the HTAP2 experiments differed from the previous HTAP1 analyses.

HTAP2 Experimental Design

The HTAP1 experiments were designed to assess the relative performance of global models for predicting the impacts of changes in anthropogenic emissions in North America (NA), Europe (EU), East Asia (EA), and South Asia (SA) on air quality. The results painted a picture of intercontinental

transport with broad strokes based on annual average, region-wide impacts. The HTAP2 experiments were designed to refine this picture and to begin to understand the source of differences between the models. The main differences between the HTAP1 and HTAP2 experimental designs are shown in Table 1.

Table 1. Comparison of the HTAP1 and HTAP2 experimental designs.

Design Element	**HTAP1**	**HTAP2**
Meteorology	2001	2008-2010
Emissions	Each model chose data representative of 2001.	Global and regional models were provided consistent emissions data for 2008 and 2010.
Source Regions	4 rectangular regions (in latitude-longitude coordinates) over North America, Europe, East Asia, and South Asia. Rest of World	16 regions covering the globe defined primarily by national boundaries and oceans. 6 regions identified as "priority regions":": North America, Europe, East Asia, South Asia, Russia/Belarus/Ukraine, and Middle East.
Spatial Resolution	Global models (resolutions typically 3°x2°)	Nested global and regional simulations (global model resolutions between 0.5°x0.5° and 3.x2; regional models 10-50 km)
Pollutants	O_3, PM, Hg, POPs	O_3, PM
Sensitivity simulations	20% decreases in anthropogenic emissions from each source region separately for NO_X, VOC, CO, and all anthropogenic emissions. 20% decrease in global methane concentration	20% decreases in anthropogenic emissions from each source region by pollutant and sector. Priority given to separate simulations for NO_X, CO, all emissions from transportation, all power and industry, all residential, all fire, and all mineral dust. Priority given to an 18% increase in global methane (2121 vs 1798 ppb) concentration, representative of 2030 in RCP 8.5.

Given multiple years of base meteorology, a larger number of source regions, and the potential to examine the sensitivity to changes in specific sectors, the number of sensitivity simulations exploded from 19 requested under HTAP1 to a possible 600 under the HTAP2 design. Twenty-one of the sensitivity simulations were identified as high priority and participants chose where to focus their effort. To ensure some comparability, and for evaluation against observations, all participants were requested to conduct a 2010 base case simulation. The result is a sparse matrix of simulations which provides the opportunity to explore a variety of questions, albeit with varying numbers of models and, thus, varying levels of confidence.

In this document, we refer to the difference in concentration (or deposition) between the base simulation and an emission perturbation simulation as the estimated "response" to the 20% emissions change. The "contribution" of a source region to a receptor region can be estimated by multiplying the 20% response by 5, as an approximation of the response to a 100% emission decrease. The contribution may be expressed in absolute (ppb) or relative (%) terms.

Comparing model results in HTAP1, we found that there was a larger variation between models in the absolute concentrations predicted than in the relative responses to the emissions perturbations. To compare the results of different simulations across different models, we defined a metric to characterize the relative Response to Extra-Regional Emissions Reductions (RERER). This metric is defined as the change in concentration or deposition within a region *i* in response to a 20% change in

emissions from anthropogenic sources outside the region ($\sum R_{foreign}$) divided by the change in concentration or deposition within the region in response to a 20% change in all anthropogenic emissions (inside and outside the region)($\sum R_{all}$). In HTAP1, we determined that the contribution of pollution from different regions was relatively additive, allowing us to calculate the RERER metric from a global perturbation simulation and a regional perturbation simulation as in eq.1:

$$RERER_i = \frac{\Sigma R_{foreign}}{\Sigma R_{all}} = \frac{R_{global} - R_{region,i}}{R_{global}} \qquad \text{(eq 1)}$$

The resulting ratio is equal to 0 if pollutant concentrations or deposition are sensitive only to changes of emissions within the region. The ratio is equal to 1 if pollutant concentrations or deposition are only sensitive to changes in emissions outside the region. If ozone formation was linear, the value of RERER would always fall between 0 and 1 and the greater the RERER value, the greater the importance of intercontinental or hemispheric transport. However, ozone chemistry is non-linear, and the value of RERER can be greater than 1 or less than 0. Such values occur in areas where the chemistry is dominated by the titration of ozone by NO_x emissions, usually in winter months. Under these conditions, the response to changes within the region can be the opposite sign of the responses to changes outside the region. Depending on the relative magnitude of these responses, the RERER can be greater than 1 or less than 0.

Question 1: Estimates of Current Hemispheric Transport

Based on current models, what is the contribution of and sensitivity to anthropogenic emissions sources outside Europe and North America to O_3 and PM concentrations and S and N deposition inside Europe and North America, respectively?

The HTAP2 experiments generally confirm the patterns of intercontinental source/receptor relationships identified in HTAP1 and the 2010 Assessment. For O_3, a large fraction of the observed concentrations is due to non-anthropogenic emission sources, including precursor emissions from vegetation, fires, lightning, and soils, and intrusion from the stratosphere. The complex and varying background sources of ozone precursor emissions are augmented by and interact with varying anthropogenic sources of precursor emissions within a region and outside a region. The contribution of each of the natural and anthropogenic components varies on different time scales from annual to hourly and by location and elevation. Thus, the contribution of and sensitivity to extra-regional emission sources depends on the metric, or policy objective, of interest. The highest O_3 concentrations are typically associated with local or regional emissions sources, stagnant meteorology, and high temperatures. However, natural and extra-regional anthropogenic sources alone can produce O_3 concentrations that exceed health-protective levels.

Within Europe, models predict a wide range of sensitivities to extra-regional emissions reductions for area-wide annual average O_3 concentrations, with RERER values of 1.2 ensemble average (model range 0.5-2.16) from global models and 0.9 (0.62-1.03) from regional models. Using global models, but subdividing Europe into smaller regions, the models show a decrease in the impact of extra-regional emissions from west to east with RERER values of 1.10 (global model range 0.63-1.12) for Northwest Europe, 0.81 (0.56-1.04) for Southwest Europe, 0.93 (0.57-1.1) for Eastern Europe, and 0.71 (0.63-0.83) for Greece and Turkey. However, there is a large seasonal and strongly model-dependent variation in the computed O_3 response to European emission reductions, illustrated by results from two models in Figure 1. In winter, to a varying degree, models find positive O_3 responses to (mainly NO_x) emission reductions, which can explain RERER values greater than 1. The relative importance of regional or extra-regional emission sources is also dependent on what O_3 metric is used. The contribution of European anthropogenic emissions to European ozone was estimated by the EMEP model to be only 16% on the basis of annual average concentration, but 41% for the summer seasonal average, 31% for annual SOMO35 (a health based metric) and 37% for annual POD1 (a metric for vegetation effects.(Jonson 2018) A recent analysis using the TM5 model for 2010 estimates that anthropogenic methane emissions contribute 5-8 ppb (or 9-16%) of ozone in Europe, measured on the basis of a the highest 6-monthly mean of daily maximum 1 hourly ozone value (6mDMA1)(vanDingenen 2018).

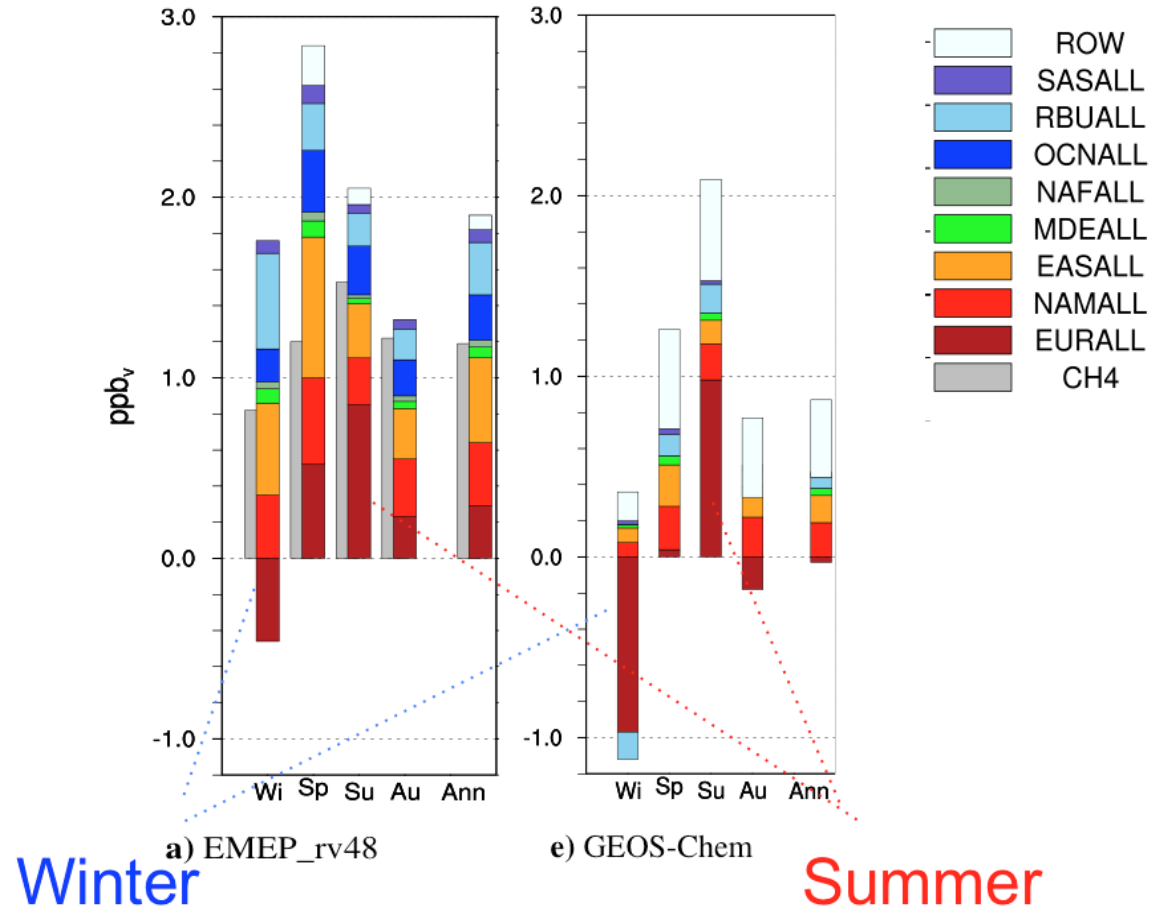

Figure 1. Responses of seasonal European surface ozone (ppb) to 20% changes in emissions of ozone precursors in different regions of the world. WI (DJF), SP(MAM), SU(JJA), AU(SON) for two models. Note that the separate responses to emissions changes in northern Africa (NAFALL) and for ocean shipping (OCNALL) are only included in the EMEP_rv48 model calculations (a). Model dependent contributions from remaining regions are included as ROW (rest of the world). The effect of a 20 % increase in global concentrations of CH_4 is shown as a separate bar for the EMEP_r48 model. Adapted from Figure 5 in (Jonson 2018).

Within North America, annual area-average O_3 concentrations show a RERER of 0.69 (0.48-0.75) in global models and 0.54 (0.46-0.65) in regional models. The extra-regional contributions peak in spring and decrease strongly from west to east, as illustrated in Figure 2, which displays RERER values calculated from monthly mean O_3 concentrations in each model grid cell. The global model GEOS-CHEM was used to estimate the various natural and anthropogenic contributions to O_3 levels in the United States from 2004 to 2012 (Guo 2018). Daily maximum 8-hour average (MDA8) O_3 concentrations in the absence of U.S. anthropogenic emissions (i.e., background O_3) was estimated to be approximately 35 ppb in spring and autumn, and approximately 40 ppb in summer. Anthropogenic emissions outside North America and changes in global methane were estimated to contribute between 8- 13 ppb in the western United States, 2-12 ppb in the central United States, and 2-10 ppb in the eastern United States, depending on the season. On high ozone days the contributions were typically 0-3 ppb lower than the seasonal average contributions. The largest difference between average and high ozone days was shown to be due to changes in U.S. anthropogenic emissions followed by changes in biogenic emissions. Between 2004-2006 and 2010-2012, observed and simulated summertime surface MDA8 O3 levels averaged over the entire United States declined by 3 ppb, estimated by the model as the net impact of rising contributions to U.S. background (+2 ppb) and declining US anthropogenic O3 precursor emissions (-6 ppb). The model estimated large interannual variability in background ozone, driven mostly by variations of natural emissions.

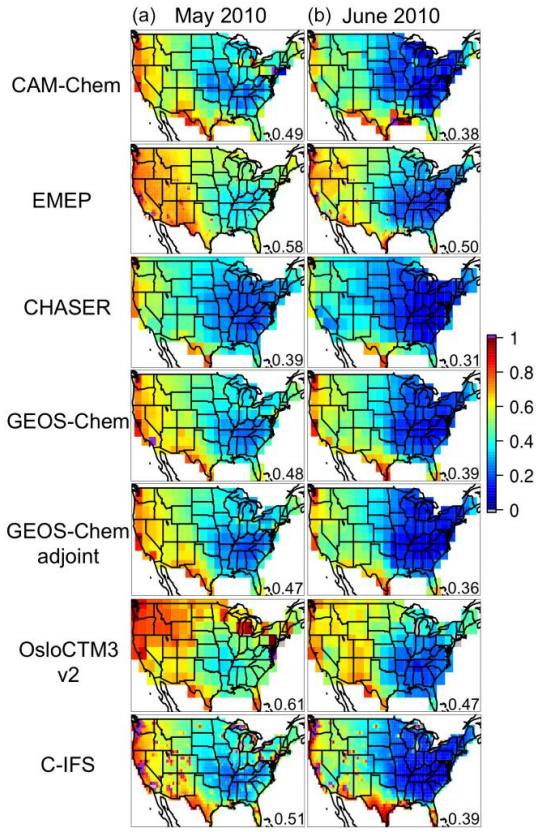

Figure 2. Relative influence of foreign compared to North American emission reductions through the RERER metric.

A value of 1 indicates mainly foreign influence, while 0 suggests local emission reductions of higher importance. The mean values for the continental US and Hawaii are indicated for each panel at the lower right corner. Adapted from (Huang 2017).

As with O_3, observed $PM_{2.5}$ concentrations are the result of contributions from non-anthropogenic sources, including emissions from vegetation, natural fires, wind-blown dust, sea-salt, and volcanos. Anthropogenic sources from near and far add to these natural sources. The contribution of each of the natural and anthropogenic components varies on different time scales from annual to hourly and by location and elevation. Both natural and anthropogenic sources can affect $PM_{2.5}$ concentrations on intercontinental scales.

The relative impact of extra-regional anthropogenic sources on $PM_{2.5}$ concentrations is smaller than on O_3 concentrations. In Europe, the sensitivity of $PM_{2.5}$ concentrations to anthropogenic emissions outside the region (including maritime shipping) is reflected in RERER values of 0.29 ensemble average (model range 0.19-0.36) for global models and 0.17 (0.01-0.34) for regional models. In North America, the relative contributions of anthropogenic emissions outside the region are lower with RERER values for $PM_{2.5}$ of 0.16 (0.09-0.21) from global models and 0.02 (-0.03-0.07) from regional models. Note that the exceptionally low regional model RERER value here is based on results from only 3 models.

The sensitivity of the deposition of sulfur (S), oxidized nitrogen (NO_y), and reduced nitrogen (NH_x) to changes in extra-regional anthropogenic emissions is similar to the sensitivity of $PM_{2.5}$ concentrations. Using the results of 11 global models, the RERER for S deposition in Europe was estimated to be 0.36 averaged regionally, with values of 0.53 for coastal areas and 0.27 for non-coastal areas. The RERER for NOy deposition in Europe is 0.34 (0.48 for coastal areas, 0.27 for non-coastal areas). For NH_x deposition, the RERER across Europe in 0.12 (0.22 in coastal areas, 0.09 for non-coastal areas). In North America, the RERERs are smaller and show a similar pattern: for S deposition, 0.17 overall (0.40 in coastal areas, 0.12 in non-coastal areas); for NOy deposition, 0.17 overall (0.43 in coastal

areas, 0.12 in non-coastal areas); and for NH_x deposition, 0.07 overall (0.31 in coastal areas, 0.05 in non-coastal areas). The larger sensitivity in coastal areas may be due to the location of coastal areas in proximity to upwind sources, including maritime shipping, and differences in deposition processes in coastal regions.

In North America, 83% of S deposition, 83 % of NOy deposition, and 93% of NH_3 deposition are due to sources within North America. In Europe, the fractions are 64%, 66%, and 88% for S, NOy, and NH_X deposition, respectively. However, in the region defined by Russia, Ukraine, and Belarus, only 39%, 41%, and 45% of S, NOy, and NH_X deposition, respectively, are due to emissions within the region. The percent of emissions transported and deposited outside each of the priority source regions is shown in Table 2.

Table 2. Percent of emissions in a source region transported out of the region and deposited elsewhere (Tan 2018).

Source Region	S	NOy	NHx
North America	31%	29%	12%
Europe	40%	34%	17%
Russia/Belarus/Ukraine	38%	39%	23%
East Asia	27%	26%	
South Asia	34%	34%	15%
Middle East	58%	46%	51%

Question 2: Evidence of Trends

How have the contribution of and sensitivity to anthropogenic emissions sources outside Europe and North America changed over time?

O_3 observations through 2014 collected for TOAR show peak O_3 values strongly decreasing in North America and Europe, and strongly increasing in parts of East Asia. However, the trends are more mixed for summer daytime average O_3 concentrations in North America and Western Europe, with some sites showing significant increases (Chang 2017; Schultz 2017). Methane concentrations have been increasing at a rate of about 6 ppb per year in 2007-2013 and accelerating to 10 ppb per year during 2014-2018 (Dlugokencky 2020). For fine particles, similar regional trends have been seen in analyses of satellite observations over the period 1992-2012, with decreasing trends in North America and Europe and strong increasing trends in South and East Asia (Boys 2014).

Emissions trends are generally consistent with the spatial patterns in the observed trends. Between 1990 and 2010, anthropogenic emissions of PM increased little globally, but shifted geographically, with a 30% decline in Europe and North America and a 50% increase in Asia (Klimont 2017). Similarly, between 2000 and 2010, global anthropogenic emissions of ozone precursors (NO_X, CO, and NMVOCs) grew modestly. However, emissions in Europe and North America decreased by 10% to 50% while emissions in South Asia and East Asia and other regions of the world increased by 10% to 50% (Turnock 2018). Globally, anthropogenic CH_4 emissions increased by 17% between 1990 and 2012, with decreases in Europe, little change in North America, and strong increases in East Asia, South Asia, and other regions of the world. The changing spatial patterns of emissions have shifted ozone precursors into the tropics where ozone production is more efficient. Zhang et al.(2016) suggest that this equatorward shift of emissions has increased the total global ozone burden more than

the combined effect of the increase in global methane emissions and the increase in the total mass of non-methane precursor emissions.

Table 2.1 Percentage change in global and regional emissions of NOx, CO, and NMVOC between 2000-2010 and annual average ozone concentrations between 2001 and 2010 as estimated by the mean of the HTAP1 and HTAP2 ensembles. Adapted from (Turnock 2018).

		Global	Europe	North America	South Asia	East Asia
% change in annual emissions 2000-2010	**NOx**	9.5	-8.4	-25.0	49.8	42.1
	CO	-1.2	-27.1	-47.1	18.8	15.6
	NMVOC	5.2	-9.7	-32.1	32.1	24.8
% change in annual concentration 2001-2010	**O_3**	-3.6	-4.3	-2.0	1.5	0.0

Analyses of recent satellite observations have shown decreasing emissions of NO_X and SO_2 and decreasing PM over East Asia, which have been attributed to the implementation of emissions control policies in China (Karplus 2018; Liu 2017; Zheng 2018).

Between the HTAP1 simulations based on 2001 and the HTAP2 simulations based on 2008-2010, there were no significant changes in the sensitivities of O_3 and $PM_{2.5}$ concentrations in Europe and North America to 20% perturbations in anthropogenic emissions in other regions. However, changes in emissions patterns have changed the absolute and relative contributions of regional and extra-regional sources. For example, Jonson et al.(2018) found that for European annual average surface O_3 levels, the contribution of European emissions had decreased significantly between 2001 and 2010, while the contribution of North American emissions had declined far less and the contribution of East Asian emissions had increased. Thus, the relative contribution of extra-regional sources increased. Note that comparisons of the RERER values calculated in HTAP2 and the similar RAIR metric defined for HTAP1 are complicated by the difference in the boundaries of the source and receptor regions for HTAP1 and HTAP2 and the treatment of marine shipping.

Question 3: Global and Regional Model Evaluation

What confidence do we have in estimates of the contribution of and sensitivity to anthropogenic emissions sources outside Europe and North America?

Confidence in model (or emissions) estimates is based on judgments about how well important physical and chemical processes (or emissions sources) are represented in the model and how well the model reproduces observed concentrations. HTAP2 involved a more theoretically advanced set of models with higher spatial resolution than used in HTAP1, but the ability to reproduce observations did not change significantly for all pollutants. Models are able to reproduce broad spatial and seasonal patterns of observations, but biases can be as large as the estimates of extra-regional contributions. Thus, we have some confidence in the ability of models to qualitatively describe the role of regional and extra-regional sources and processes. However, uncertainty in quantitative estimates of extra-regional contributions generally are the same order of magnitude as the estimates themselves.

Our estimates of emissions sources globally have improved as previously unaccounted sources have been included (e.g. sources of black carbon). However, uncertainties in emissions inventories in some parts of the world remain high. For example, HTAP2 emissions for India were greater by a factor of

1.5 to 2 for NOx, NMVOC, and SO2 as compared to an inventory created at the national scale (Venkataraman 2018). Satellite observations have helped improve estimates of emissions trends in many parts of the world.

Our confidence in model estimates of the sensitivity of O_3 in Europe and North America to anthropogenic emissions in other regions has not changed since HTAP1. The range of estimates of surface O_3 concentrations in the HTAP2 ensemble is large and of similar magnitude to that in HTAP1, despite having used the same emissions in all HTAP2 models. Regional models are less biased than global models for surface O_3 when compared to observations. The larger errors in global models are particularly important for threshold-based metrics, such as AOT40. Nevertheless, the best performing global models have less error than the worst performing regional models. The largest sources of error are found in temporal processes acting on longer time scales (weeks or months), including emissions, their interaction with chemistry, and long-range transport processes.

The range of estimates for $PM_{2.5}$ narrowed between HTAP1 and HTAP2. Global and regional models perform similarly for $PM_{2.5}$, with a tendency to underestimate $PM_{2.5}$ concentrations in Europe and North America. Removing this low bias may significantly increase estimates of $PM_{2.5}$ exposure and health impacts.

Table 3. Model evaluation of annual average O_3 and $PM_{2.5}$ in the AQMEII3 ensemble of regional models over Europe and North America (Im 2018) and HTAP2 global models (Liang 2018). Im et al. compared 12 models over Europe and 3 models over North America to the ENSEMBLE database. Liang et al. used 11 models for O_3 compared to the TOAR database and 8 models for $PM_{2.5}$ compared to the Global Burden of Disease PM estimates for 2010.

Pollutant	Annual Average Hourly Ozone					Annual Average Daily $PM_{2.5}$				
Region	Europe		North America		World	Europe		North America		World
Model ensemble	regional	global	regional	global	global	regional	global	regional	global	global
Regression Coefficient (r)	0.84	0.55	0.82	0.52	0.53	0.68	0.73	0.71	0.50	0.77
Normalized Mean Bias (NMB) (%)	-4.0	4.3	8.6	14.1	13.2	-38.0	-36.0	-19.9	-10.5	-23.1
Normalized Mean Gross Error (NMGE) (%)	5.9	9.8	15.5	17.6	13.2	52.1	39.7	36.2	27.6	35.4

Compared to HTAP1, there is increased understanding that purpose-built model ensembles (based on quality criteria or common features of ensemble members) may further reduce errors compared to using simple mean or median results under baseline conditions. However, it is not yet clear whether these ensembles are more reliable under emissions scenario conditions, or for the evaluation of long-range transport phenomena.

The nested design of the HTAP2 and AQMEII3 simulations allow for an analysis of how the boundary conditions defined by global models impact the estimates and model performance of higher-resolution regional models. Different global models simulating the same scenario were shown to produce significantly varying boundary conditions for the North American and European regional domains. These different boundary conditions were shown to have substantial impacts on the base case performance of a regional model, even affecting the directionality of the overall bias. As discussed above, regional models are generally less sensitive to extra-regional emissions changes than are global models. However, different boundary conditions can increase or decrease the sensitivity of

a regional model in comparison to the global model. The largest contributor to the sensitivity to boundary conditions was shown to be related to differences in mid-tropospheric ozone (Hogrefe 2018), which affects ground-level ozone in the regional model through horizontal advection and, in particular, vertical mixing (Liu 2018). Comparison of regional models to observed O_3 vertical profiles broadly confirmed that meteorological drivers and vertical mixing processes were among the key-factors explaining differences between models and observations (Astitha 2018).

Question 4: Future Scenarios and Mitigation Potential

How are the contribution of and sensitivity to anthropogenic emissions sources outside Europe and North America expected to change and what emissions sectors contribute significantly?

In Europe, changes in emissions outside Europe and global methane concentrations will largely drive future annual average O_3 levels. Without additional controls, global methane emissions are expected to grow, increasing O_3 mortality in Europe in 2050 by up to 8,000 additional premature deaths compared to 2010 levels. Implementation of mitigation policies, largely outside of Europe, can decrease methane emissions overall and decrease O_3 mortality in Europe by up to 2000 premature deaths per year compared to 2010 levels, a difference of 10,000 deaths per year between the highest and lowest global CH_4 emissions scenarios. In North America, the difference between the highest and lowest global CH_4 emissions scenarios corresponds to a difference of up to 5,000 deaths per year in 2050. The sectors with substantial mitigation potential are fossil fuel production, waste and wastewater management, and agriculture, with the largest emissions in China, followed by Latin America, Africa, India, and North America (vanDingenen 2018).

Shipping makes a significant contribution to both O_3 and $PM_{2.5}$ levels in Europe and North America, particularly in coastal regions. The impact of planned emissions control policies under the International Maritime Organization have not been examined here but should be explored.

Decreases in emissions in North America over the last decade are likely to have contributed to flattening or declining trends observed recently at remote sites in Europe. Likewise, controls on the electricity and industrial sectors in China have led to decreasing emissions of some pollutants in the last 3-5 years. The implications of these recent emissions decreases and their influence on observed trends at remote sites in North America and Europe should be explored.

Air pollution in South Asia is expected to continue to increase largely because of changes in emissions within the region. Implementation of clean technologies and climate change mitigation policies could substantially decrease pollution levels with large benefits for human health, crops, and ecosystems. The benefits of emissions decreases in South Asia would mostly accrue to the region itself, and then downwind regions in East Asia and the Middle East. However, the global implications of emissions reductions in South Asia should be further explored.

Since 2012, TF HTAP has organized a series of workshops with TFIAM and CIAM to explore future global scenarios and the potential for mitigation. IIASA's work under the ECLIPSE project has contributed significantly to these discussions. Sessions have focused on improving our understanding of emissions and mitigation potential for marine shipping; residential heating, cooking, and lighting; agricultural burning; transportation; electricity production; and various sources of methane.

Next Steps

The HTAP2 analyses have provided confirmation of previous findings and some new insights, and have provided new data and tools with which to explore a variety of questions. Looking forward, several important questions stand out with respect to the science and policy interests of the LRTAP Convention:

- How are emissions continuing to change globally?
- What information is needed to decrease the differences and biases in current global and regional model estimates of the intercontinental transport of ozone or ozone precursors?

- What specific mitigation measures are available that will most likely decrease the impact of intercontinental transport on ground-level ozone in Europe and North America, and what are their costs and benefits?

Given the review of the revised Gothenburg Protocol and the potential for future negotiations, the Task Force will focus its efforts in the near term (within two years) on addressing these questions as they pertain to ground-level ozone; ozone precursors, including methane; and ozone impacts.

To address the first question, TF HTAP must continue to update the global emissions mosaic produced for the HTAP experiments with more recent emissions estimates. The 2001 (HTAPv1) and 2010 (HTAPv2) emissions mosaics are compiled from national, regional, and global emissions estimates that are accepted by governments and expert communities (Janssens-Maenhout 2015; Janssens-Maenhout 2012). These inventories have been used widely by the atmospheric modeling community. Ideally, an update will include estimates for the most recent year possible and consistent trends back to 2000. Trend information is useful for understanding how extra-regional emissions have contributed to changes in air quality and deposition observed within the LRTAP Convention and the effectiveness of the Gothenburg Protocol. Producing such an update will be a high priority for the Task Force over the next few years.

Over the longer term, TF HTAP's emissions analyses should explore emerging techniques to link emissions estimates to trade relationships, separating emissions associated with consumption within a region from emissions associated with exported goods and services (e.g., Zhang 2017). Such analyses add additional dimensions to transboundary pollution management, enabling estimates of the impacts that the consumption of goods and services in one region have on air quality, health, ecosystems, and climate in other regions of the world.

To address the second question (improving model performance), a more detailed examination of the individual processes represented in global and regional models needs to be conducted and best practices and benchmark datasets for model evaluation should be identified. Model evaluation efforts in HTAP2 and AQMEII3 have pointed to the importance of differences in deposition schemes for S, N, and O_3. Deposition processes are important sinks, affecting estimates of concentrations and transport, as well as important pathways for vegetation and ecosystem damage. Deposition (or more broadly, surface exchange) processes are also important for Hg and POPs, which are part of the TF HTAP's mandate, but have not been a focus in HTAP2. For estimating O_3 impacts, it is important to go beyond deposition to understand uptake by vegetation (i.e., stomatal flux). Methane has a relatively long atmospheric lifetime, thus for annual simulations included in HTAP1 and HTAP2 methane emissions were not explicitly modelled and ozone sensitivity to methane controls was approximated by changing methane concentrations. Ongoing assessments under the CCAC and AerChemMIP are generating multi-model ensembles of the dynamic response of ozone to changes in methane emissions, which may be incorporated into future HTAP source/receptor estimates. With these challenges in mind, TF HTAP organized a meeting in April 2020 to focus on identifying near term research activities that may improve the tools available to the Convention to assess the benefits of measures to decrease ozone precursors, including methane, both inside and outside the Convention. The workshop took stock of work going on within the Convention as well as efforts outside the Convention, including TOAR, AQMEII, CCAC, WMO GAW (including the Measurement-Model Fusion for Global Total Atmospheric Deposition (MMF GTAD), AeroCom, CCMI, and AerChemMIP.

To address the third question (identifying sector specific costs and benefits), the TF HTAP must work cooperatively with TFIAM, TFTEI, and other experts with knowledge of sector specific technology options and costs. TF HTAP would also benefit from input from WGSR concerning sectors of potential interest. To facilitate cooperation and information exchange with other bodies, TF HTAP will strive to increase participation in meetings of other subsidiary bodies of the Convention and schedule joint or adjacent meetings where appropriate. To improve the communication of policy relevant information, TF HTAP proposed at its April 2020 meeting to establish an online document to present answers to policy-relevant questions that could be collaboratively developed with the participation of scientists and policy audiences. This online collaboration can build upon the content

in this current report to further clarify messages and to add new information as new results are available and new questions arise.

Two efforts that will contribute to TF HTAP's ability to address all three questions are continued work on global model evaluation and the development of tools, such as openFASST, to visualize and explore the implications of different global models and scenarios.

The compilation of best practices and benchmark datasets used for global model evaluation is a long-term goal of TF HTAP that is important for ensuring the quality of information relied upon by the Convention. Based on recommendations from the April 2020 meeting, TF HTAP will explore collaboration with TOAR-II to develop metrics specifically for evaluation of global and regional ozone models. Such an effort contributing to TOAR-II could build upon work of FAIRMODE and other efforts at the regional scale as well as CCMVal and other past efforts at the global scale.

Further development of the functionality of the openFASST screening tool will allow exploration of the implications of multi-model ensemble estimates of source/receptor relationships and future global emissions scenarios, such as those provided by IIASA's GAINS model. Building upon JRC's TM5-FASST tool, TF HTAP is working to enable FASST to use and compare source/receptor relationships from a variety of models and/or source attribution methods, from the HTAP experiments and other exercises. At the same time, TF HTAP is working to make the FASST codes available in an open-source environment so that interested experts can collaborate in further development.

Over the last 15 years, TF HTAP has been able to tap into the interests of the international scientific community and facilitate a large body of scientific work, leveraging a relatively small investment by the lead parties. With a renewed mandate (Executive Body, 2019), new members of the leadership team, and strong interest from the policy side of the Convention, TF HTAP will continue to work to both improve the scientific understanding of the international transport of air pollutants and to communicate policy-relevant findings and insights to the Convention and other relevant audiences.

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HTAP2/AQMEII3/MICS3 ACP Special Issue

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Appendix 1: Overview of publications in the HTAP2 special issue, and attribution to coordinated activity, regional coverage, component, and science topic

		Activity				Regional Focus					Pollutant		Topic					
Authors	**Title**	HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Curci et al., 2018	Modelling black carbon absorption of solar radiation: combining external and internal mixing assumptions		x				x	x				x						x
Dong et al., 2018	Long-range Transport Impacts on Surface Aerosol Concentrations and the Contributions to Haze Events in China: an HTAP2 Multi-Model Study	x							x									
Liu et al., 2018	Multi-Model Comparison in the Impact of Lateral Boundary Conditions on Simulated Surface Ozone across the United States Using Chemically Inert Tracers		x					x				x			x			
Palacios-Peña et al., 2018	Aerosol optical properties over Europe: an evaluation of the AQMEII Phase 3 simulations against satellite observations		x				x					x						x
Van Dingenen et al., 2018	TM5-FASST: a global atmospheric source-receptor model for rapid impact analysis of emission changes on air quality and short-lived climate pollutants				x	x					x	x				x	x	x
Fiore et al., 2018	Regional and intercontinental pollution signatures on modeled and measured PAN at northern mid-latitude mountain sites	x				x	x				x			x				

Authors	Title	Activity				Regional Focus					Pollutant		Topic					
		HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Sobhani et al., 2018	Source Sector and Region Contributions to Black Carbon and PM2.5 in the Arctic				x					x		x	x					
Baró et al., 2018	Comparison of regional meteorology-chemistry models with satellite cloud products over Europe				x		x					x						x
Crippa et al., 2017	Sectorial and regional uncertainty analysis of the contribution of anthropogenic emissions to regional and global PM2.5 health impacts				x	x						x	x			x		
Astitha et al., 2018	Seasonal ozone vertical profiles over North America using the AQMEII3 group of air quality models: model inter-comparison and stratospheric intrusions		x					x				x			x			
Jonson et al., 2018	The effects of intercontinental emission sources on European air pollution levels	x				x	x					x		x				
Tan et al., 2018	Source contributions to sulfur and nitrogen deposition – an HTAP II multi-model study on hemispheric transport	x				x											x	
Guo et al., 2018	Average versus high surface ozone levels over the continental USA: model bias, background influences, and interannual variability				x			x							x			

Authors	Title	Activity				Regional Focus					Pollutant		Topic					
		HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Liang et al., 2018	HTAP2 multi-model estimates of premature human mortality due to intercontinental transport of air pollution and emission sectors	x				x					x	x				x		
Vivanco et al., 2018	Modeled deposition of nitrogen and sulfur in Europe estimated by 14 air quality model systems: evaluation, effects of changes in emissions and implications for habitat protection		x				x					x					x	
Turnock et al., 2018	The impact of future emission policies on tropospheric ozone using a parameterised approach	x				x					x			x	x			
Im et al., 2018b	Influence of anthropogenic emissions and boundary conditions on multi-model simulations of major air pollutants over Europe and North America in the framework of AQMEII3		x				x	x			x	x			x	x		
Galmarini et al., 2018	Two-scale multi-model ensemble: is a hybrid ensemble of opportunity telling us more?	x	x				x	x			x	x		x				
Venkataraman et al., 2018	Source influence on emission pathways and ambient PM2.5 pollution over India 2015–2050				x				x			x				x		
Tan et al., 2018b	Multi-model study of HTAP II on sulfur and nitrogen deposition	x				x												

Authors	Title	Activity				Regional Focus					Pollutant		Topic					
		HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Im et al., 2018a	Assessment and economic valuation of air pollution impacts on human health over Europe and the United States as calculated by a multi-model ensemble in the framework of AQMEII3		x				x	x			x	x				x		
Gao et al., 2018	Air quality and climate change, Topic 3 of the Model Inter-Comparison Study for Asia Phase III MICS-Asia III – Part 1: Overview and model evaluation			x							x	x						x
Han et al., 2018	Characteristics of intercontinental transport of tropospheric ozone from Africa to Asia				x				x		x			x	x			
Hogrefe et al., 2018	Impacts of different characterizations of large-scale background on simulated regional-scale ozone over the continental United States		x					x			x			x				
Li et al., 2018	Comparison and evaluation of anthropogenic emissions of SO2 and NOx over China	x		x					x			x	x					
Itahashi et al., 2018	A 15-year record 2001–2015 of the ratio of nitrate to non-sea-salt sulfate in precipitation over East Asia				x				x			x		x				
Doherty et al., 2017	Multi-model impacts of climate change on pollution transport from global emission source regions				x	x	x								x			

Authors	Title	Activity				Regional Focus					Pollutant		Topic					
		HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Uno et al., 2017	Seasonal variation of fine- and coarse-mode nitrates and related aerosols over East Asia: synergetic observations and chemical transport model analysis				x				x			x		x				
Mathur et al., 2017	Extending the Community Multiscale Air Quality CMAQ modeling system to hemispheric scales: overview of process considerations and initial applications		x			x						x	x					
Knowland et al., 2017	The influence of mid-latitude cyclones on European background surface ozone					x	x				x							x
Xu et al., 2017	Source attribution of Arctic black carbon constrained by aircraft and surface measurements					x				x		x			x			
Ikeda et al., 2017	Tagged tracer simulations of black carbon in the Arctic: transport, source contributions, and budget					x				x		x			x			
Solazzo et al., 2017b	Advanced error diagnostics of the CMAQ and Chimere modelling systems within the AQMEII3 model evaluation framework		x				x	x			x			x				
Liu et al., 2018	Modeling intercontinental transport of ozone in North America with CAMx for the Air Quality Model Evaluation International Initiative AQMEII Phase 3		x					x			x			x				

Authors	Title	Activity				Regional Focus					Pollutant		Topic					
		HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Nopmongcol et al., 2017	Response of the global surface ozone distribution to Northern Hemisphere sea surface temperature changes: implications for long-range transport				x	x					x				x			x
Klimont et al., 2017	Global anthropogenic emissions of particulate matter including black carbon					x	x					x	x					
Huang et al., 2017	Impact of intercontinental pollution transport on North American ozone air pollution: an HTAP phase 2 multi-model study	x						x			x				x			
Karamchandani et al., 2017	Source-sector contributions to European ozone and fine PM in 2010 using AQMEII modeling data		x				x				x	x			x			
Itahashi et al., 2017	Nitrate transboundary heavy pollution over East Asia in winter				x				x			x		x	x			
Solazzo et al., 2017a	Evaluation and error apportionment of an ensemble of atmospheric chemistry transport modeling systems: multivariable temporal and spatial breakdown		x				x				x				x			
Galmarini et al., 2017	Technical note: Coordination and harmonization of the multi-scale, multi-model activities HTAP2, AQMEII3, and MICS-Asia3: simulations, emission inventories, boundary conditions, and model output formats	x	x	x	x	x												

Authors	Title	Activity				Regional Focus					Pollutant		Topic					
		HTAP	AQMEII	MICS	Other	Global	Europe	North America	Asia	Arctic	Ozone	Aerosol	Emissions	Model Evaluation, observation	Boundary Condition	Health Impacts	Deposition, Crop Impacts	Climate Impacts
Li et al., 2017	MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP	x		x									x					
Kioutsioukis et al., 2016	Insights into the deterministic skill of air quality ensembles from the analysis of AQMEII data		x					x			x			x	x			
Stjern et al., 2016	Global and regional radiative forcing from 20 % reductions in BC, OC and SO4 – an HTAP2 multi-model study	x				x						x						x
Gao et al., 2016	Response of winter fine particulate matter concentrations to emission and meteorology changes in North China				x				x			x						x
Kasoar et al., 2016	Regional and global temperature response to anthropogenic SO2 emissions from China in three climate models				x				x			x						x
Solazzo and Galmarini, 2016	Error apportionment for atmospheric chemistry-transport models – a new approach to model evaluation		x				x	x						x	x			
Janssens-Maenhout et al., 2015	HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution	x				x							x					