IGAC*tivities* Newsletter

of the International Global Atmospheric Chemistry Project
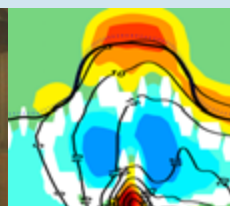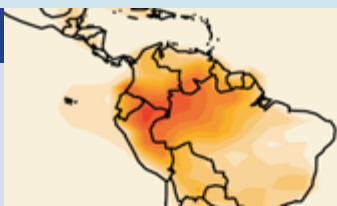
A Note from the IGAC Co-chairs: Paul Monks and Tong Zhu

There is no doubt the world is changing and one of our parent organization's IGBP is also evolving with a greater focus on science for a global sustainable future. The nature of the interaction between humankind and nature is at the centre of sustainable life on earth. Through the great shifts in Earth's geochemical and biological systems over time, life has generated ever-more-sophisticated responses to varying planetary conditions and there is a need to explore these interactions within a global multi-disciplinary research program. IGBP is moving to provide the essential scientific leadership and knowledge of the Earth system to help guide society onto a sustainable pathway during rapid global change. The IGAC community is working on many issues of direct consequence to global sustainability but we also must begin to think how our research fits into the evolving framework of the IGBP.

Since much of the global sustainable future depends on the next generation of scientist, IGAC is always excited when it has the opportunity to recognize and highlight the work being done by young scientists in atmospheric chemistry. For this reason, this issue of the IGAC Newsletter is very excellent as we feature articles from three of the winners of the Young Scientist Best Presentation Prize from the 12th Symposium of the International Commission on Atmospheric Chemistry and Global Pollution (iCACGP) and 11th Science Conference of the International Global Atmospheric Chemistry (IGAC) Project held jointly in Halifax, Nova Scotia, Canada July 2010.

Alexander T. Archibald, a current post doctorate at the University of Cambridge UK, discusses the importance of HO_x oxidation on isoprene levels. Ian R. Burling, a post doctorate at the University of Montana US, looks into emission from biomass burning in the southern United States. Finally, Yuayuan Fang, a post doctorate at Princeton University US, investigates the impact of changes in precipitation frequency and intensity on particulate matter concentrations under a changing climate. These three young scientists highlight all the hard work and excellent research young scientists across the world are doing. IGAC looks forward to continually highlighting and supporting young scientist activities.

IGAC recently has launched some very exciting new initiatives including the IGBP Air Pollution & Climate Initiative and the Atmospheric Chemistry & Health Initiative. In addition, IGAC now sponsors National/Regional Working Groups. Recently, the IGAC Scientific Steering Committee overwhelming approved the IGAC China Working Group being led by IGAC co-chair Tong Zhu. We are looking forward to sponsoring other National/Regional Working Groups in order to facilitate coordination of research both within nations/regions and between the nation/region and the international atmospheric chemistry community. Finally, we had to sadly say goodbye to Sarah Doherty, who as the IGAC Executive Officer for 7 years did a tremendous job. However, we are also happy to welcome Megan L. Melamed as the new IGAC Executive Officer. At the time of writing, Megan has her feet under the table and is keeping us on a steady course.

We look forward to implementing the new and moving forward current IGAC Activities. Please feel free to contact IGAC at igac@uw.edu if you would like to become more involved or have any suggestions/comments.

Sincerely,

Paul Monks (IGAC SSC co-chair) ■ Tong Zhu (IGAC SSC co-chair)

GLOBAL IGBP CHANGE
International Geosphere-Biosphere Programme

iCACGP
In Cooperation with IAMAS Commission on Atmospheric Chemistry and Global Pollution

Introducing the New Executive Officer

Sarah Doherty

As of February 2011 Sarah Doherty stepped down as IGAC Executive Officer in order to focus on her research at the Joint Institute for the Study of the Atmosphere and Ocean/University of Washington. Sarah served as the IGAC Executive officer from 2003 to 2010. She will be greatly missed by the IGAC community.

Megan L. Melamed replaced Sarah Doherty as the new IGAC Executive Officer. Megan has a unique history that makes her an ideal candidate to be the IGAC Executive Officer. In 2000, Megan received two Bachelor of Arts degrees, in Chemistry and in Spanish Literature, from Colby College in Waterville, Maine. She continued her education at the University of Colorado in Boulder earning a Master of Arts and a Ph.D. in environmental engineering in 2002 and 2006, respectively. Megan conducted her Ph.D. research at the National Oceanic and Atmospheric Administration (NOAA) with The Chemistry and Climate Process group in the Earth System Research Laboratory (ESRL) under the guidance of Susan Solomon. Her graduate research focused on airborne differential optical absorption spectroscopy (DOAS) observations of nitrogen dioxide and sulfur dioxide emissions from power plants and urban areas as part of the Texas Air Quality Study (TexAQS) 2000 and the New England Air Quality Study (NEAQS) 2004.

Megan Melamed

Upon completion of her doctorate work, Megan received the National Science Foundation International Research Fellowship Program award to conduct research at the Universidad Nacional Autónoma de México (UNAM) in Mexico City, Mexico. Her research in Mexico City focused on identifying transportation events to the south of Mexico City using nitrogen dioxide DOAS observations as part of the Megacity Initiative: Local and Global Research Observations (MILAGRO) 2006 field campaign. Following her post-doctoral work in Mexico, Megan was awarded an AAAS Science and Technology Policy Fellowship. As an AAAS fellow, Megan worked at the U.S. Environmental Protection Agency on a wide breadth of projects on global change research. Megan is excited to be using her scientific research background coupled with her international and policy experience to coordinate activities as the Executive Officer of IGAC.

Investigating the impacts of HO_X recycling in the oxidation of isoprene: Sensitivity studies of past, present and future atmospheres using the UKCA model

Contributed by **Alexander T. Archibald** (alex.archibald@atm.ch.cam.ac.uk)
National Centre for Atmospheric Science, UK and Department of Chemistry, University of Cambridge, Cambridge, CB2 1EW, UK.

▪ Introduction

Isoprene (C_5H_8, 2-methyl-1,3-butadiene) represents a major fraction of the total volatile organic compounds (VOC) that are emitted to the atmosphere (vegetative isoprene emissions are estimated to be in the range of ~ 400-500 Tg C/yr, a mass similar to that of the entire human population!). Owing to the high reactivity of isoprene towards the hydroxyl radical (OH), model studies invariably implicate isoprene as a large sink of OH. However, recent field studies in major isoprene emitting regions such as Amazonia (Lelieveld et al., 2008) and South East Asia (Hewitt et al., 2010) have found large discrepancies between modelled and measured HO_X ($OH+HO_2$) concentrations. In particular Lelieveld et al. (2008) have shown that current models for atmospheric chemistry predict OH up to a factor of 10 lower than measurements in the Amazon. However, issues with representing HO_X in models are not limited to the tropics. Rather it appears that models struggle to represent HO_X when isoprene levels are high (> 1 ppbv) and NO_X ($NO+NO_2$) levels are low ($\leq$ 1 ppbv). This under-simulation of HO_X has potentially far reaching implications due to the role of OH in "cleansing" the atmosphere of pollutants and greenhouse gases (e.g. CO and CH_4). Suggestions

Alex Archibald

Alex studied for a BSc in Chemistry at the University of Bristol from 2003-2006. In his final year he started a research project on modeling atmospheric chemistry which spurred him on to stay at Bristol to undertake studies towards a PhD. Alex's PhD research was jointly funded by the UK Met Office and Great Western Research Council and focused on using models to try and understand the degradation and dispersion of atmospheric trace gases. Alex graduated from Bristol in June 2010 and now works as an NCAS postdoctoral associate at the University of Cambridge, Department of Chemistry.

for the cause of the discrepancies are (i) the presence of unmeasured (NO-like) species, capable of propagating the interconversion of HO_X (red shaded species in Figure 1) (ii) a fundamental lack of understanding of the mechanism for low-NO_X isoprene oxidation.

Owing to the importance of isoprene to atmospheric chemistry there have been an extensive number of laboratory investigations probing the pathways involved in its atmospheric degradation. However, many of these experiments have been limited because they have been performed in conditions that do not represent the background atmosphere (i.e. under high concentrations of NO_X and or HO_2/RO_2). Recently, several theoretical (Peeters et al., 2009; Peeters and Müller 2010) and experimental (Paulot et al., 2009) studies have provided new insights into the oxidation mechanism of isoprene under low NO_X conditions. The work of Peeters et al. (2009) concerning unimolecular reactions of the isoprene hydroxy-peroxy radicals (ISOPOHO2) provides a new framework with which to re-assess the low NO_X oxidation mechanism of isoprene. Here we summarise our latest work looking at assessing these impacts based on performing a set of chemistry-climate model integrations (Archibald et al., 2011).

▪ Modelling method

A number of experiments were carried out using the UKCA model (http://www.ukca.ac.uk) run at a horizontal resolution of 3.75° in longitude × 2.5° in latitude on 60 hybrid height levels that stretch from the surface to ~84 km (for more details of the model setup see Archibald et al. (2011)). The UKCA model is a global chemistry-climate model that has been developed jointly by NCAS and the UK Met Office
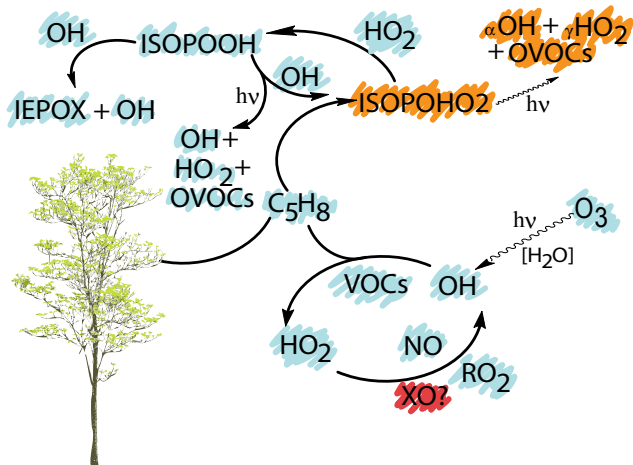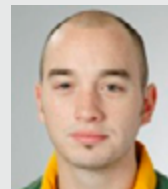

Figure 1. Schematic of isoprene photochemistry. The modification made to isoprene chemistry is shown in the orange shading where we include competitive unimolecualr reactions of the ISOPOHO2 that lead to the production of OH, HO_2, and other VOCs. The traditional HO_X chemistry is shown in blue shading. In red is the postulated reaction of an as yet un-measured species that is able to convert HO_2 to OH.

to provide community capability for modelling chemistry-climate interactions. Our experiments involved performing base simulations (**B**) using our existing chemical mechanism and then performing modified runs (**M**) where the isoprene chemistry was modified to include isomerisation reactions of the ISOPOHO2. Essentially the modified chemistry allows for the regeneration of HO_x through the isomerisation's and subsequent degradation of the ISOPOHO2 (this is shown in Figure 1 as the reactions shaded orange). Further experiments were then performed by changing the climate and emissions so that the effects of the modified chemistry on the composition of the atmosphere could be calculated in the present-day (**PD**) (*ca.* 2000), pre-industrial (**PI**) (*ca.* 1850), and future (**FC**) (*ca.* 2100).

■ Present-day impacts

Including the isomerisations of ISOPOHO2 leads to large increases in OH and HO_2 relative to the base model run.

These increases are largest near the surface in regions where isoprene emissions are high but extend throughout the tropical troposphere (see Figure 2 of Archiabald et al. (2011)). Figure 2 shows that including the isomerisation of ISOPOHO2 can increase OH by as much as 400%. Such large increases in OH lead to changes in the distribution of other species important for tropospheric chemistry. Not shown are the increases in HO_2 which although large (up to ~ 200% near the surface) are relatively smaller compared to the increases in OH. In Figure 2, the modelled OH Base run (red lines in all plots) and Modified run (blue lines in all plots) are compared against measured OH during the OP3 campaign in Borneo (Hewitt et al., 2010). The measurements are plotted as campaign mean values (black crosses) and corresponding standard deviations (black envelope). Clearly the Modified run leads to improved agreement with the OH measurements. However, Figure 2 shows that the Modified run worsens the agreement
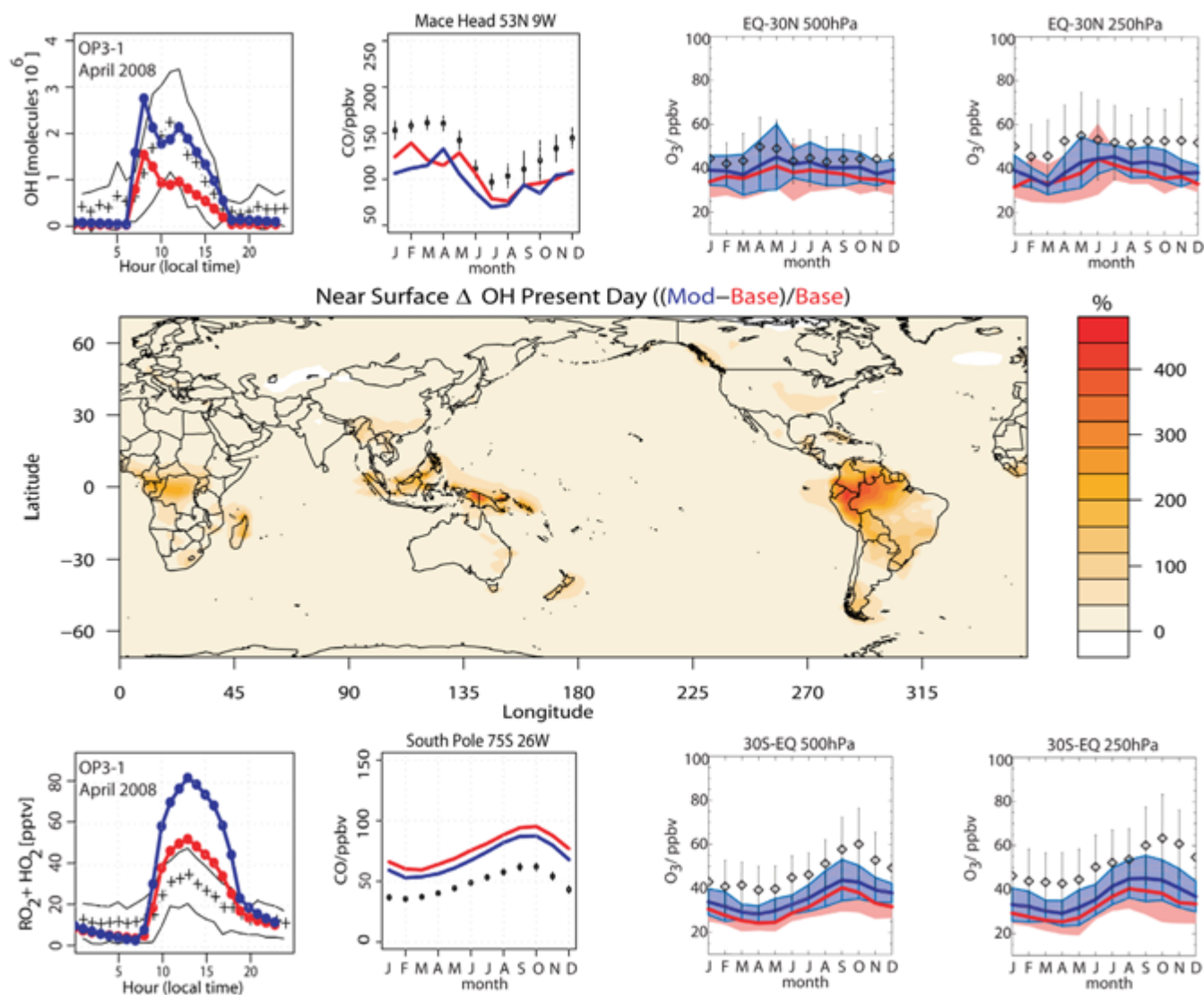

Figure 2. Results from PD runs. The central plot shows the near surface change in OH between the modified and base runs. Also shown are the results of model measurement comparisons against OH and ΣRO_2+HO_2 made during the OP3 campaign in Borneo, CO from long term surface measurement sites, and ozone compiled from a climatology of ozone sonde data (see Archibald et al. (2011) for references to data sources). In all cases the run with modified chemistry is shown in blue, whilst the base run is shown in red.

between the modelled and measured peroxy radicals (RO_2+HO_2). The impact of the Modified chemistry is also shown for CO (whose major sink is via reaction with OH) where measurements from surface stations in the Northern Hemisphere and Southern Hemisphere are compared with model results. The increases in OH lead to global reductions in CO that bring the model more in line with observations in the Southern Hemisphere. One of the notable effects of the increase in HO_X is that ozone production is increased throughout the troposphere in our Modified run. This can be seen in Figure 2 where modelled ozone is plotted against an ozone sonde climatology for different latitude bands and pressure regions (see Archibald et al. (2011) for more details). The increase in ozone production as a consequence of the modified isoprene chemistry tends to improve the model agreement with these measurements over the regions shown but slightly worsens the agreement near the surface at the equator (not shown).

Pre-industrial impacts

The chemical environment of the pre-industrial (PI) era is markedly different than present day (PD). During the pre-industrial era, the minimal anthropogenic emissions lead to a much greater ratio of VOC:NO_X emissions (thus an environment potentially more sensitive to changes in low NO_X chemistry). Including the modifications to our models isoprene chemistry leads to very large changes in OH and HO_2 in the pre-industrial era (enhancements of OH greater than 1000% near the surface with zonal mean increases of up to 170% in places). As with the present day simulation, the pre-industrial simulations show that the increased HO_X leads to decreases in CO and increases in ozone. The increases in ozone represent somewhat of a paradox in reconciling the model with observations as, in accord with a range of other chemical transport models, we tend to simulate ozone concentrations under pre-industrial conditions that are higher than those inferred from measurements. This clearly is an issue that will require further work to reconcile.

Future climate impacts

There are many possible trajectories for the future chemical composition of the atmosphere. In this study we focused on a set of simulations where we specified a relatively optimistic emissions scenario (IPCC B2+CLE) where VOCs increase (partly driven by the effect of warmer conditions on increasing isoprene emissions) but NO_X is slightly lower than in our present day simulations.

As with both the present day and pre-industrial runs the modification made to isoprene oxidation lead to increases in HO_x and ozone and decreases in CO. The increases in OH are smaller than those of the PD and PI simulations but still significant with near surface increase in OH > 400% in places. As with both the PD and PI simulations this leads to increased ozone production and an increase in the tropospheric ozone burden of ~ 10%.

Impacts on methane

In all simulations the methane mixing ratio was prescribed to levels appropriate for the time being simulated (see Table 1 of Archibald et al. (2011) for details). Methane

is a powerful greenhouse gas in its own right, but this is further complicated by the fact that methane oxidation (via its reaction with OH) can lead to the production of ozone (also a greenhouse gas). In all the simulations we performed we have calculated a significant increase in the tropospheric burden of OH when we include the modified isoprene chemistry. These increases in OH can then be used to calculate what the effect would be on the tropospheric chemical lifetime of methane (τCH_4). Figure 3 shows the relative change in τCH_4 for the different simulations. Clearly the greatest effect is seen in the PI simulation where the τCH_4 decreases by ~ 35%. However, the effects on the PD and FC simulations are also significant with decreases of τCH_4 calculated to be around 15 and 10% respectively.
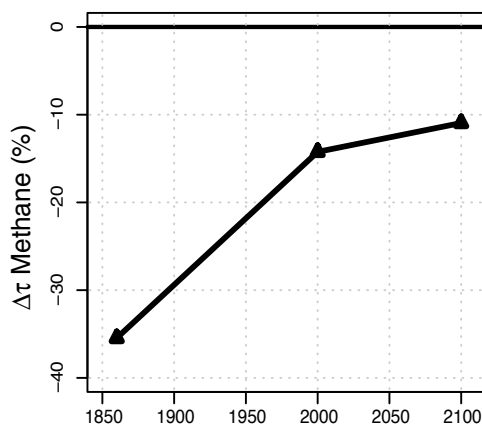

Figure 3. Relative (%) change in methane lifetime calculated between different pairs of simulations. The abscissa shows the approximate year the simulations are representative of.

Summary

Observations of HO_X radicals in the pristine environments of Amazonia and South East Asia suggest that HO_X levels are maintained much higher than models suggest (Lelieveld et al., 2008). Several ideas have been put forward for reconciling this issue including the presence of un-identified species that may interconvert the HO_X radicals. In this study we have assessed the impact of including unimolecular isomerisations and subsequent degradation of the isoprene hydroxy-peroxy radicals as postulated by Peeters et al. (2009) and Peeters and Müller (2010). Our results from present-day simulations suggest that this chemistry can help reconcile issues with OH but at a determent to ΣRO_2+HO_2. However, other species (CO and ozone) show that in general adoption of the new chemistry leads to improved agreement between model and measurements for the present-day.

In order to investigate if this chemistry could have wider implications we then looked at its impacts under pre-industrial and future climate and emission scenarios. By far the greatest impact is seen in the pre-industrial where adoption of the new chemistry leads to a reduction in

τCH_4 of ~ 35%. No doubt more studies will be required to unravel the intricacies of the oxidation of isoprene, including targeted laboratory experiments performed under low NO_x and low HO_2/RO_2 conditions. However, issues with our understanding of the chemical composition of the contemporary atmosphere can clearly not be ignored in our simulations of the pre-industrial atmosphere.

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Results from a large multi-platform study of trace gas and particle emissions at southern US DoD bases: A comparison of laboratory and field study results

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■ Introduction

Biomass burning is the largest source of primary, fine carbonaceous particles and the second largest source of trace gases in the global atmosphere (*Bond et al., 2004*; *Crutzen and Andreae, 1990*) and impacts both the chemical composition and radiative balance of the atmosphere. The majority of biomass burning occurs unregulated in the tropics (*Crutzen and Andreae, 1990*). In the United States, formal land management programs use prescribed burning to reduce wildfire hazards and improve wildlife habitats, and to improve access (*Biswell, 1999*; *Wade and Lunsford, 1989*). Many fire-adapted ecosystems depend on the regular occurrence of fire for survival.

The gas-phase emissions from biomass burning are dominated by water vapor (H_2O) and carbon dioxide (CO_2), but also include significant amounts of carbon monoxide (CO), nitrogen oxides (NO_x = NO + NO_2), methane (CH_4), ammonia (NH_3), sulfur dioxide (SO_2) and a multitude of non-methane organic compounds (NMOC) of which oxygenated volatile organic compounds (OVOC) comprise a large fraction (*Christian et al., 2003,*

Ian Burling

Ian received a Bachelor of Science in Chemistry at Brock University, Canada. He received his PhD in Chemistry from York University in Toronto, Canada. He is currently a post-doctoral scholar at the University of Montana Department of Chemistry. He is currently studying the emissions from biomass burning and their contributions and transformations in the atmosphere.

2004). These NMOC may contribute to photochemical ozone (O_3) production and secondary organic aerosol (SOA) formation.

Laboratory studies of the emissions from biomass burning offer many advantages (*Yokelson et al., 2008*). More extensive instrumentation can be utilized in lab fire studies, and since smoke concentrations tend to be higher, more chemical species can be quantified. Also, in a lab experiment, all the smoke from an entire

fire may be sampled, so that emission factors per fuel for simulated fires can be determined accurately, whereas field measurements sample a small fraction of the total smoke. Characterization and measurement of the composition of fuels is much easier in the laboratory.

For field measurements of smoke emissions an airborne-based measurement platform is usually required for sampling of flaming combustion emissions due to the lofting of smoke from convection created by high flame temperatures. Airborne measurements also allow downwind sampling of a smoke plume to probe plume aging processes. Ground-based measurements are necessary for sampling the smoke of unlofted residual smoldering combustion which often stays near the ground, impacting local visibility and air quality. The emissions from residual smoldering combustion (RSC) are often quite different from those observed during flaming combustion as RSC emissions are dominated by incomplete combustion products (*Bertschi et al.*, *2003*; *Christian et al.*, *2007*). RSC can continue for weeks after initial ignition.

We report results from a large, multi-component study focused on North American biomass burning that measured both initial emissions and post-emission processing. In a laboratory component, vegetation commonly managed by prescribed fires was collected from the southeastern and southwestern US and burned under "realistic" conditions at the USDA Forest Service Fire Sciences Laboratory fire simulation facility in

Figure 1. USDA Forest Service Fire Sciences Laboratory combustion facility in Missoula, MT.

Missoula, MT. The smoke emissions were measured with a comprehensive suite of state-of-the-art trace gas and particle instruments. The University of Montana open-path FTIR system measured many trace gases and additional instruments (some deployed in smoke for the first time) allowed measurements of over 150 additional volatile organic compounds (VOC). A subsequent, extensive field campaign featured ground-based sampling of smoldering emissions in North Carolina with ground-based FTIR and airborne measurements of initial emissions and plume aging in California, Arizona, and North Carolina on a Twin Otter aircraft from November 2009 through March 2010.

▪ Laboratory component

For the laboratory component, we collected samples of vegetation from military bases in the southeastern (Camp Lejeune, NC and Fort Benning, GA) and southwestern US (Fort Hunter-Liggett, CA, Vandenberg AFB, CA and Fort Huachuca, AZ). The samples were shipped to and burned at the US Forest Service Fire Sciences Laboratory combustion facility in Missoula, MT (Figure 1). The

smoke emissions were sampled at a single point for the duration of the fire to obtain data on the initial emissions from the fire. We employed an extensive suite of gas- and particle-phase instrumentation, including an open-path Fourier transform infrared spectrometer (OP-FTIR), negative ion proton transfer chemical ionization mass spectrometer (NI-PT-CIMS), proton transfer reaction mass spectrometer (PTR-MS), proton transfer ion trap mass spectrometer (PIT-MS) and gas chromatography (GC), gas chromatography/mass spectrometry (GC-MS), aerosol mass spectrometer (AMS), filter sampler, nephelometer, particle into liquid sampler (PILS), and various particle number and sizing instruments.

Figure 2 shows an example of the temporal profile of gas-phase emissions as measured by OP-FTIR (*Burling et al.*, *2010*). We observed emissions associated with high efficiency flaming combustion (e.g. CO_2, SO_2, NO_X, HCl, HONO) and those associated with lower efficiency smoldering combustion (e.g. CO, CH_4, NH_3, OVOC). As is usually the case with real fires there was often no clear distinction between the two processes, as the smoke from smoldering combustion was entrained with the flaming
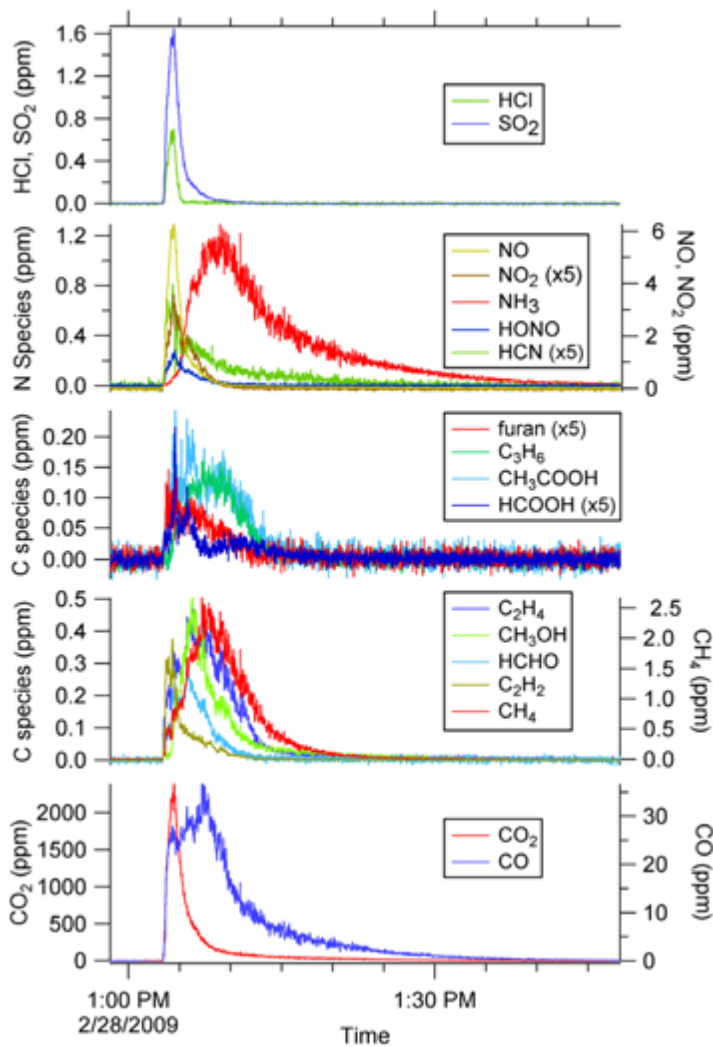
Figure 2. Temporal profile of gas-phase emissions for a typical laboratory burn as measured by OP-FTIR (*Burling et al. 2010*).
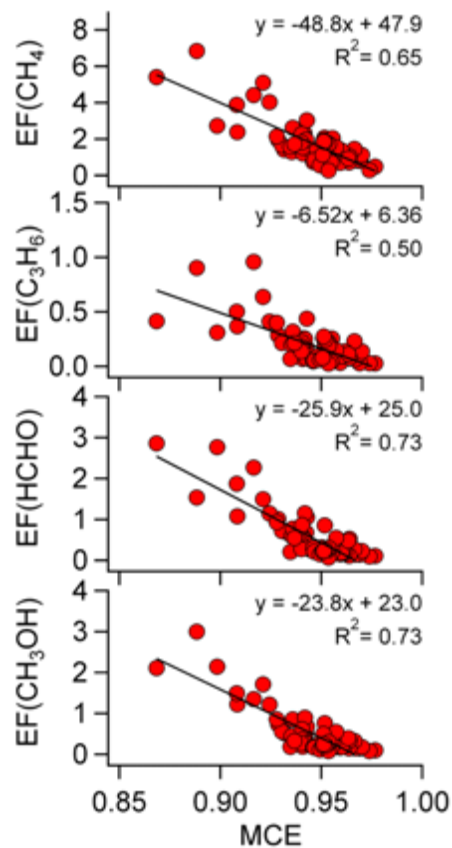

Figure 3. Emission factors (g kg[1]) for selected hydrocarbon and oxygenated organic species as a function of MCE for all burns measured in the laboratory. Investigation of the dependence of EF on fuel type and other factors, synthesis with field-measured EF, as well as incorporation of additional species from mass spectrometric techniques are all subjects of on-going work.

combustion emissions as the flame front travelled across the fuel bed. We integrated these emissions over the course of the fire and determined emission ratios with respect to CO or CO_2. We determined fire-integrated emission factors (EF: g species emitted per kg dry fuel burned) for all gas-phase species as measured by the OP-FTIR using the carbon mass-balance method. We also compared the emission factors to the modified combustion efficiency (MCE), an indicator of the relative amount of flaming and smoldering combustion, where MCE = $\Delta CO_2/(\Delta CO_2 + \Delta CO)$ and Δ refers to the mixing ratio enhancement above background. Higher MCE indicates relatively more flaming combustion while lower MCE more smoldering. Figure 3 shows the emission factors for selected hydrocarbons and oxygenated organic species as a function of MCE (*Burling et al. 2010*). The anti-correlation of EF with MCE indicates that these compounds are products of smoldering combustion. Emissions of species containing elements other than carbon, hydrogen or oxygen (e.g. NO_X, NH_3, SO_2, HCl) were observed to be dependent on the elemental composition of the fuels as well as MCE. As shown

in Figure 2, we also observed gas-phase nitrous acid (HONO) in the emissions of all fires during flaming combustion. HONO is a source of the hydroxyl radical (OH) in the atmosphere and thus can promote rapid photo-oxidation as the smoke plume ages. The HONO/NO_x ratios ranged from approximately 0.03 to 0.20.

Figure 4 shows the mass spectra as recorded by the PIT-MS for two burns, with mass spectra separated by flaming and smoldering combustion (*Warneke et al. 2011*). Since PIT-MS measures full mass spectra and the sensitivity increases linearly with mass it is a useful tool to investigate the importance of emissions of higher mass compounds from fires. The identified and unidentified mass fractions of the total PIT-MS signal were determined for the smoldering and the flaming phase of the fires and it was found that in some cases, only half of the mass was identified. Most of the unidentified mass was from compounds with molecular weights above 100 atomic mass units. The large unidentified mass fraction, which is not included in current models, is likely important for the production of secondary organic aerosol (SOA).
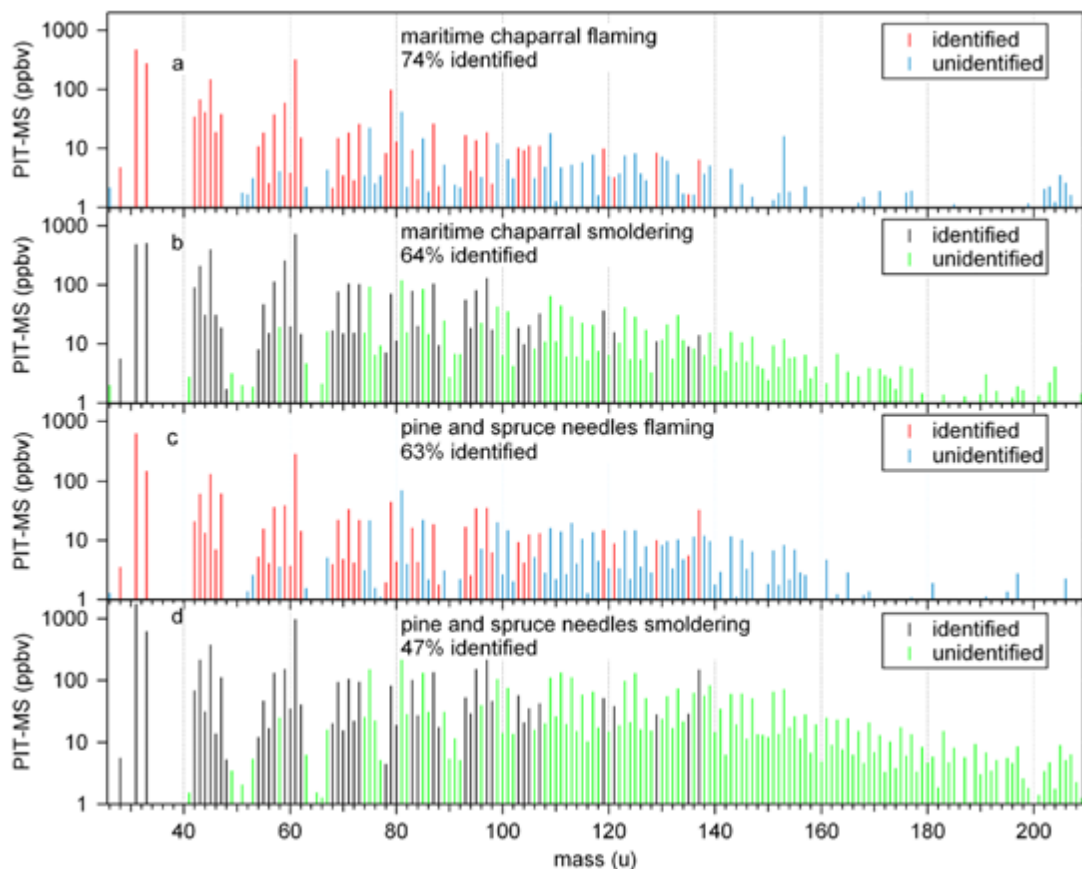
Figure 4. PIT-MS mass spectra: (a) maritime chaparral during flaming, (b) maritime chaparral during smoldering, (c) pine and spruce needles during flaming, (d) pine and spruce needles during smoldering (*Warneke et al. 2011*).

These unidentified, high molecular weight compounds are likely OVOC or aromatics and therefore often have a low volatility. Due to this low volatility, a large fraction of the mass could end up in the aerosol phase after only a few oxidation steps (e.g. *Robinson et al., 2007*). The high molecular weight unidentified NMOC coupled with the high direct emissions of HONO could lead to significant and rapid O_3 and SOA formation as the smoke ages.

We are currently working on calculating emission factors based on the other instrumentation. Please see the following for more information on the recent results from the various instruments: *Burling et al. (2010)*; *Roberts et al. (2010)*; *Veres et al. (2010)*; *Warneke et al. (2011)*.

■ Field measurements

We sampled 14 prescribed fires at or near the locations where the fuels for the lab experiment were collected from a Twin Otter aircraft in the air and also from the ground with our ground-based FTIR system. Due to space and weight restrictions, the instrumentation onboard the Twin Otter was less extensive than in the lab. Instrumentation included our closed-cell airborne FTIR (AFTIR), nephelometer, whole air sampling canisters, and for eight fires a single particle soot photometer (SP2) and AMS. Unlike in the laboratory experiments, where we sampled the smoke from a fixed point over the course of the fire,

we collected grab samples in the FTIR cell both in the plume and in adjacent clean air to provide background information. We collected samples close to the plume several hundred meters above the flame front for initial emissions, and in one case (Williams Fire, CA) we were also able to also sample the aged plume up to 60 km downwind from the source. Figure 5 shows a picture of the

Figure 5. View from Twin Otter of March 1 burn of pine understory (ME unit) at Camp Lejeune, NC.

ME unit prescribed fire at Camp Lejeune, NC as taken from the Twin Otter.

We obtained emission ratios (and emission factors) for all gas-phase species by plotting the excess mixing ratio of the species vs. ΔCO or ΔCO_2. The slope of this plot, with the intercept forced through zero, yields the emission ratio. Example plots of emission ratios for several smoldering compounds are shown in Figure 6 for the oak woodland burn at Fort Huachuca, Arizona. We observed excellent correlation for most species on most fires.

For one of the pine understory burns in North Carolina we were able to simultaneously sample the lofted smoke with our airborne FTIR and the smoke from residual smoldering of stumps on the ground. Although the relative contribution of the residual smoldering combustion to the total smoke emissions is often small for prescribed fires, this unlofted residual smoldering combustion smoke can impact local air quality and visibility. We observed significantly different emissions between the airborne and ground-based platforms with the emission factors shown in Figure 7. The ground-based emissions were products of low efficiency combustion (MCE ranged from 0.759 to 0.850) characterized by much higher hydrocarbon and oxygenated organic species than in the airborne samples (MCE = 0.945).
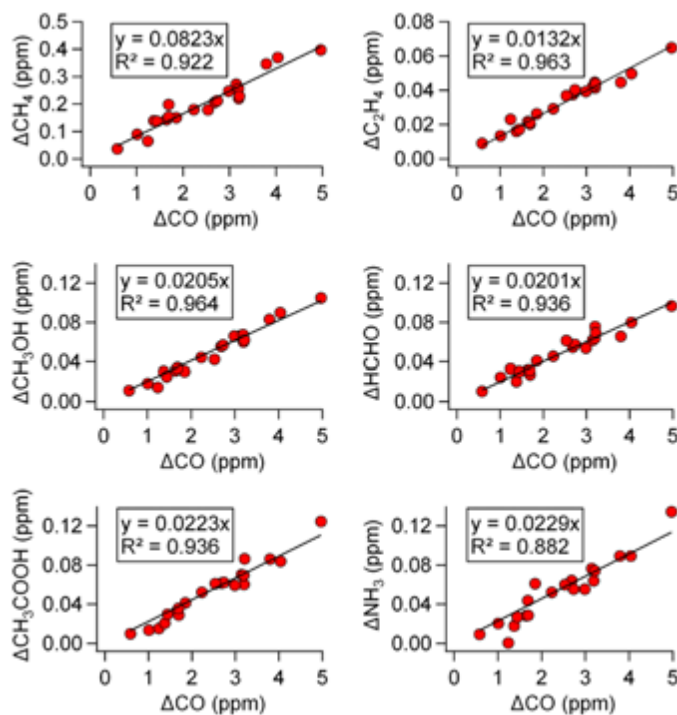

Figure 6. Emission ratios to CO for a few of the gas-phase species measured in airborne samples of oak savanna smoke in Arizona. Each point represents an individual smoke sample.

Figure 7. Comparison of emission factors as observed in the lofted smoke by the airborne FTIR (red) and of residual smoldering combustion (dark and light blue) measured by ground-based FTIR for the same pine understory fire. Ground samples 1 through 4 (s1-s4) were of smoldering dead stumps while sample 5 (s5) was from the burning base of a living tree. Note the log scale, e.g. the EF(HCHO) for samples s1-s4 is ~3 times higher than the airborne EF.
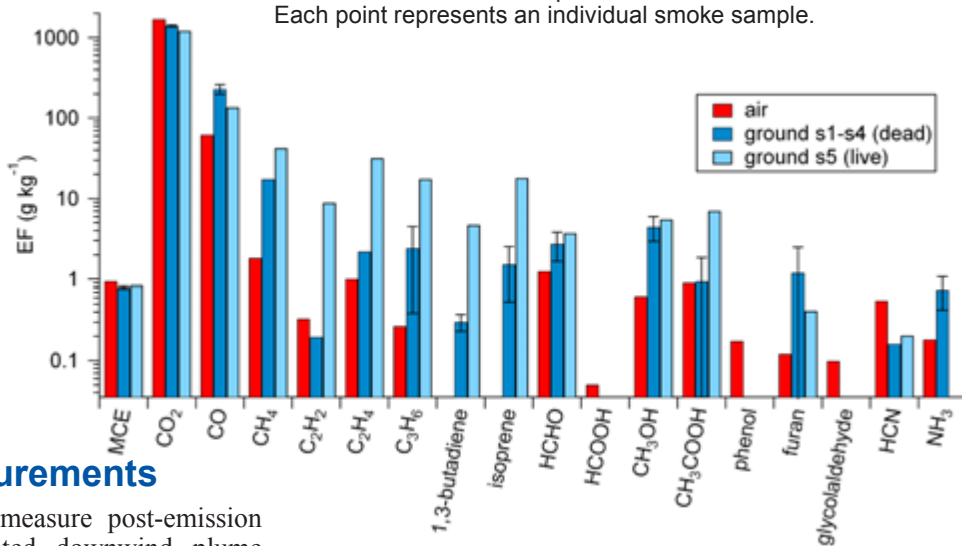
▪ Smoke aging measurements

On one fire we were able to measure post-emission chemical changes in the isolated downwind plume for ~4.5 hours of smoke aging time (*Akagi et al., in preparation*). We measured significant changes in several gas-phase constituents with our airborne FTIR. The molar $\Delta O_3/\Delta CO$ ratio increased from below background levels to 0.102 in 4.5 h (Figure 8). Biomass burning is a major source of global tropospheric ozone (*Sudo and Akimoto, 2007*) and this provides a rare measurement of the rate of O_3 formation in an isolated plume, which is ideal for testing the photochemical mechanism of a smoke plume model. Preliminary comparisons show good agreement when the direct emissions and a secondary source of HONO are included in the model.

Acetic acid (Figure 8) and formic acid ratios to CO increased by factors of 1.73 ± 0.43 and 7.33 ± 3.02 (respectively) over the same aging period. These species can be major carbon reservoirs in aged biomass burning plumes (*Yokelson et al., 2003*) and also impact the pH of rainfall in rural tropical regions (*Crutzen and Andreae, 1990*). A rapid decay in C_2H_4 (Figure 8) was consistent with an in-plume average OH of $5.16 \times 10^6 \pm 6.0 \times 10^4$ molecules cm^{-3}, providing further evidence that significant OH concentrations can occur in biomass burning plumes.

The aerosol light scattering to CO_2 ratio (Figure 8) increased by a factor of 2.3 ± 0.7 over 4 h. The organic and inorganic aerosol (NH_4^+, NO_3^-, and SO_4^{2-}) composition as measured by the AMS also changed as the plume aged. The single-particle soot photometer (SP2) observed changes in the fraction of thickly coated refractory black carbon (rBC) particles. The results from these instruments are currently under investigation and will be published in the near future.
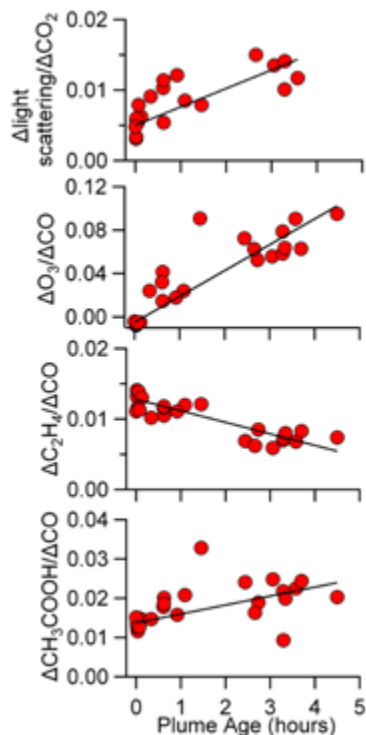
Figure 8. Evolution of selected normalized excess mixing ratios and light scattering during smoke plume aging.

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Warmer wetter climate, more soluble pollutants

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▪ Introduction

Air quality is strongly influenced by meteorological conditions and thus is sensitive to climate change (e.g., *Denman et al.*, 2007). Understanding the impact of climate change on air quality is essential for managing future air quality and evaluating the societal consequences of climate change. Recent modeling studies have examined implications of climate change for air pollutants, especially for surface ozone (O_3) and particulate matter (PM) since they are of most concern for public health (e.g., *Jacob and Winner*, 2009). These studies agree that as a result of 21st-century climate change, background O_3 in the lower troposphere (where O_3 loss by reacting with OH via water vapor is dominant) will decrease while surface O_3 over polluted regions at northern mid-latitudes will increase (+1-10 ppbv) (*Wu et al.*, 2008a, b; *Lin et al., 2008*; *Nolte et al*., 2008; *Weaver et al.,* 2009). For PM, while significant changes (±0.1-1 µg m^{-3}) were found, neither the sign nor the magnitude was consistent across models (*Jacob and Winner*, 2009). The uncertainty regarding the impact of climate on PM may reflect the complexity of the dependence of its components on meteorological variables, and the key role of precipitation in modulating PM sinks (*Racherla and Adams*, 2006; *Tagaris et al.*, 2007; *Avise et al*., 2009; *Pye et al*., 2009; *Dawson et al*., 2007; *Tai et al.*, 2010). Although *Racherla and Adams* (2006) showed that a lower PM burden corresponds to an increase in global precipitation in a future climate, *Pye et al*. (2009) find that changes in precipitation are not always the governing factor for PM concentrations. *Jacob and Winner* (2009) argued that precipitation frequency is likely the dominant factor determining PM concentration changes. Such disparate results in the literature motivate an investigation into the impact of changing precipitation on soluble pollutants in a warmer climate. We apply idealized tracers, with both insoluble and soluble versions, in a global chemistry-climate model (Atmospheric Model version 3, AM3, developed by the Geophysical Fluid Dynamics Laboratory (GFDL), a prototype of that described in *Donner et al.,* 2010) in order to isolate the role of changing transport and precipitation on future air pollutants. We present below highlights from a study that is described in detail in a submitted manuscript (*Fang et al., 2011*).

▪ Methods

We conduct a pair of idealized simulations using GFDL AM3 to magnify climate change signals relative to model internal variability: for the present-day climate (denoted as "1981-2000"), we use a monthly 20-year mean annually invariant climatology of observed sea surface temperature and sea ice from the Hadley Center to drive our AM3 simulation; for the future climate (referred to as "2081-2100"), we add the 20-year mean monthly anomalies of sea surface temperature and sea ice extent (calculated from a 19-model IPCC AR4 A1B scenario ensemble mean) to the present-day observed climatological values. Both simulations are run for 21 years with the first year as

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spin-up. We keep emissions of aerosols and trace gases in both simulations at 1990s level. The concentrations of the long-lived greenhouse gases (CO_2, N_2O and CFCs) follow the A1B scenario. These settings allow us to study the effect of climate change on pollutants, without considering the additional complexity of changes in anthropogenic emission changes. The AM3 simulated global surface temperature increase from 1981-2000 to 2081-2100 is 2.7 K, and precipitation increases globally by 5%, consistent with the IPCC AR4 (2.8 K and 6% increases of surface temperature and precipitation, respectively in the AR4 model ensemble mean) (*Meehl et al*., 2007).

To diagnose the impact of circulation changes in 2081-2100 on pollutant distributions, we use a passive COt tracer that decays exponentially with a 25-day lifetime. The emissions of COt tracer mimic CO emissions in 2001, which include anthropogenic emissions from the RETRO project (*Schultz et al*., 2007; http://www.retro.enes.org) and biomass burning emissions from GFED version2 (*van der Werf et al*., 2006; http://www.geo.vu.nl/~gwerf/GFED.htm). Another tracer, SAt, follows the same emission and decays as COt, but is subjected to additional removal by wet deposition as for sulfate aerosols. The application of these tracers is adapted from the diagnostic tracer experiments (TP) of the Task Force on Hemispheric Transport of Atmospheric Pollutants (*HTAP*, 2007; http://www.htap.org). The total CO emissions (including anthropogenic and biomass burning) are about 1000 Tg CO yr^{-1}.

▪ The impact of changing transport on insoluble pollutants

Figures 1a show the zonal mean distribution in 1981-2000 and its change from 1981-2000 to 2081-2100 for the COt tracers. During 1981-2000, COt has maximum concentrations at the surface near the tropics and over the northern hemispheric mid-latitudes, which reflects the emissions distribution. In 2081-2100, there is a distinctive vertical redistribution of COt. At the surface,
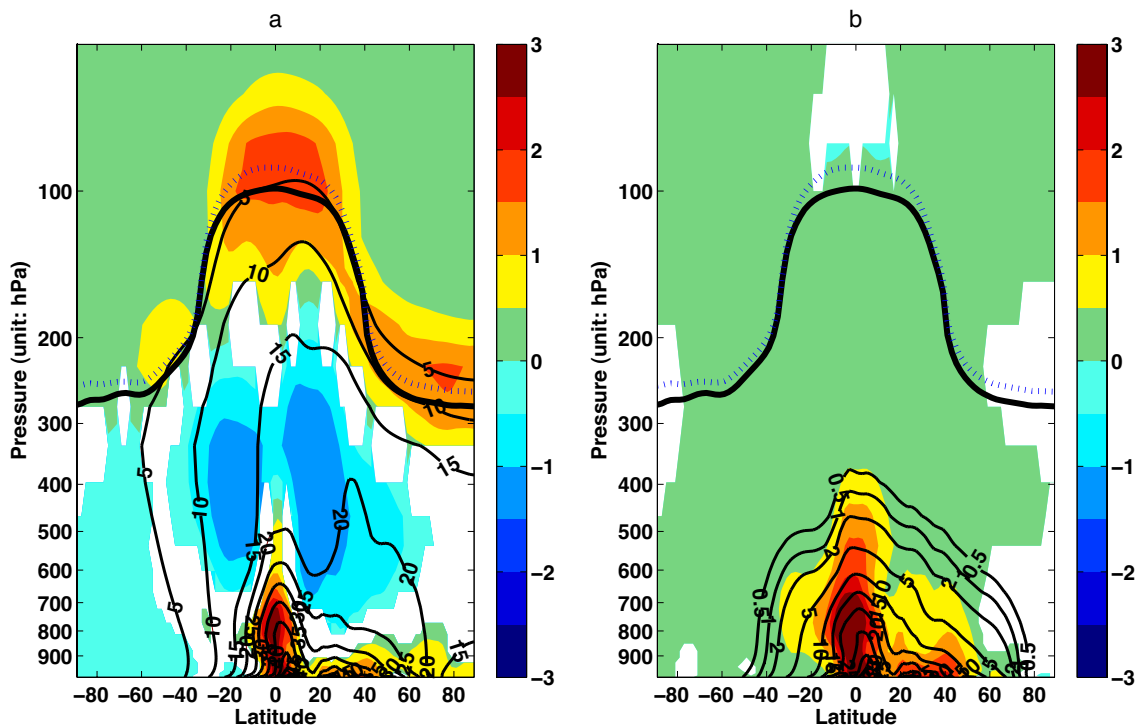
Figure 1. Zonal mean distributions of two idealized tracers plotted at each model pressure level, averaged over the years 1981 to 2000 (black contour; ppb) and the changes in the 20-year mean tracer abundance from 1981-2000 to 2081-2100 (color; ppb). Shown are (a) COt tracer, with a 25-day lifetime and emissions of carbon monoxide for the year 2001 and (b) SAt, equivalent to COt except for wet deposition, which mimics that for sulfate aerosol. White areas indicate regions where changes are not significant at the 95% confidence level. Thick black dashed and dotted lines show the tropopause location during 1981-2000 and 2081-2100 respectively (identified as in Reichler et al. [2003], based on the World Meteorological Organization (WMO) lapse-rate criterion). Adapted from *Fang et al.,* 2011, in revision at *J. Geophys. Res.*

COt concentrations increase by up to 6 ppbv in the tropics and up to 2 ppbv at northern hemispheric mid-latitudes, with the relative change ranging from 2-7%. Meanwhile, in the free troposphere, COt concentrations decrease with a maximum reduction of up to 2 ppbv (-2 to -12% at 400hPa). To help understand the vertical redistribution of tropospheric COt (i.e., increases in the lower troposphere and decreases in the free troposphere), we apply a two-box model between the lower troposphere and the free troposphere (the boundary between these two boxes defined as 500 hPa level following *Held and Soden* (2006)), except that we use the mass conservation of COt in the free troposphere box. We find that the relative change of the mass flux exchange between these two boxes to be -13%. The reduced mass flux exchange between lower troposphere and free troposphere accounts for the vertical redistribution of COt in a future climate. Besides the redistribution in the troposphere, we also see a strong enhancement in COt concentraion near the tropopause, which is mostly due to a higher tropopause in the future (*Fang et al.*, 2011, in revision at J. Geophys. Res.)

■ The impact of changing precipitation on soluble pollutants

Figure 1b shows the zonal mean distribution in 1981-2000 and the changes from 1981-2000 to 2081-2100 for the SAt tracers. In 1981-2000, the SAt zonal distribution in the northern hemisphere is close to that of COt, reflecting the use of the same emission inventories. However, because of

the wet removal, SAt decreases much more rapidly from its source region than COt (mean SAt concentrations in the lower and free troposphere are 7.4 and 0.3 ppbv, compared to 19 and 13 ppbv for COt).

From the present to the future simulations, the SAt distribution changes differently from that of COt. SAt surface concentrations increase both in the tropics and at the northern hemispheric mid-latitudes, similar to COt, but with a greater relative change (above 10%). In the free troposphere and the southern hemisphere (where COt concentration decreases), SAt concentrations increase. The tropospheric SAt burden increases from 17 to 19 Gg (+12%) in the future, indicating a 12% increase of the SAt lifetime from the 1981-2000 to 2081-2100 (as emissions are identical). Since wet deposition via precipitation is the only difference between these two tracers, we are thus able to isolate the impact of future precipitation changes on soluble pollutants by comparing the changes of COt and SAt.

The increase of SAt throughout the whole troposphere and the longer lifetime of SAt are consistent with the decrease of wet deposition (-5%) and the resulting increase (18%) in the lifetime against wet deposition from present-day to the future. As the total global precipitation increases in the future (+6%), the increasing SAt burden and decreasing wet deposition imply that, globally, precipitation is less effective at removing soluble tracers (SAt) in the future. In the following sections, the causes of this apparent disparity between increased global precipitation and decreased wet
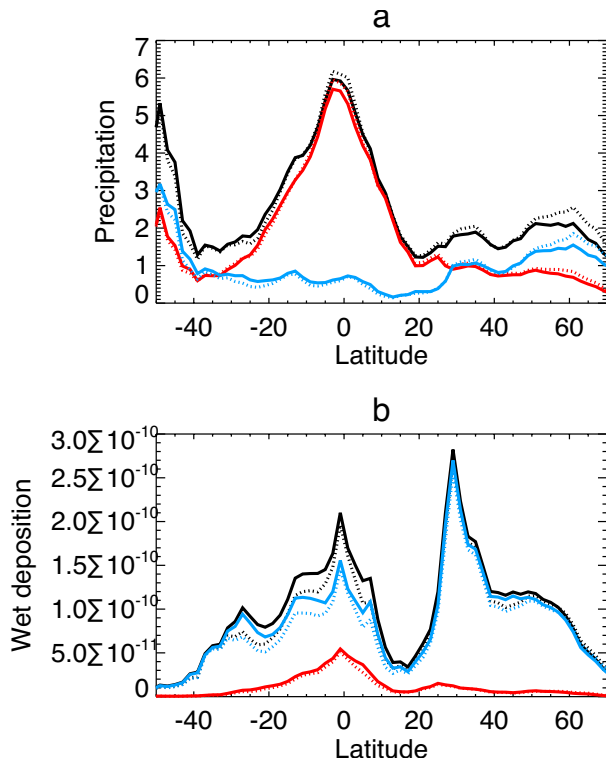
a

b

Figure 2. The 20-year annual zonal mean latitudinal distribution of total (black), large-scale (blue) and convective (red) precipitation (a; mm/day) and wet deposition (b; kg/m²/day) over land. Solid lines and dashed lines represent 1981-2000 and 2081-2100 cases respectively. Adapted from *Fang et al.,* 2011, in revision at *J. Geophys. Res.*
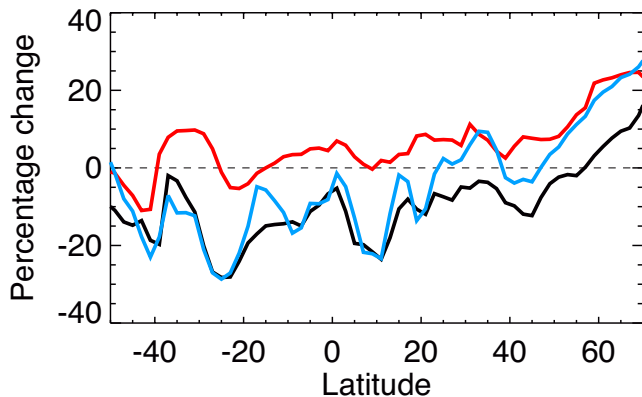

Figure 3. The percentage change by 2081-2100 of the 20-year averaged zonal mean large-scale precipitation (blue), convective precipitation (red) and SAt wet deposition (black). The correlation coefficient between the large-scale precipitation percentage change and that of wet deposition is above 0.9 and the root mean square difference between them is around 8%. Adapted from *Fang et al.*, 2011, in revision at *J. Geophys. Res.*

$\longrightarrow$

Figure 4. The 20-year mean seasonal cycle of SAt burden (unit: 10⁻¹⁰kg/m²/day) over land within North America during 1981-2000 (black solid) and 2081-2100 (blue dashed). Adapted from *Fang et al.*, 2011, in revision at *J. Geophys. Res.*

deposition are investigated.

The higher surface concentration of both SAt and COt suggest that air pollution will worsen in a warmer and wetter global climate.

■ The role of large-scale vs. convective precipitation

Globally, total precipitation is highest in the tropics, where it is dominated by convective precipitation; large-scale precipitation is comparable to convective precipitation in the mid-latitudes (Figure 2a). In contrast to the total precipitation, SAt wet deposition has two latitudinal maxima (in the tropics and the extra-tropics), both of which are driven by large-scale precipitation (Figure 2b), suggesting that wet deposition is dominated by large-scale precipitation in the AM3 model. In the scavenging parameterization for sulfate, convective precipitation is a more efficient scavenging process than large-scale precipitation. If convective and large-scale scavenging were applied to the same area, convective scavenging would be the dominant removal pathway for our SAt tracer. In the model, however, convective precipitation (and the subsequent tracer scavenging) occurs only within sub-grid updraft plumes. In contrast, large-scale precipitation allows for more widespread scavenging of pollutants over the entire cloud-covered region. As a result, on a regional scale, wet deposition by large-scale precipitation overwhelms that by convective precipitation, at least in our model.

Textor et al. (2006) examined 9 models within the AeroCom initiative (http://dataipsl.ipsl.jussieu.fr/AEROCOM/) and they found that the fractional contribution of simulated large-scale wet deposition to total wet deposition varies widely across individual models (range of 0.1 to 0.9). The lack of model agreement results from differences in many aspects of the models, such as emissions, resolution, and wet deposition parameterizations and indicates a need for more research on wet deposition pathways and how they are represented in global models.

Since large-scale precipitation dominates total wet deposition, we then compare the changes in large-scale precipitation and in wet deposition (Figure 3). The relative change in latitudinal wet deposition follows that of large-scale wet deposition in most latitudinal bands. This implies that the latitudinal variability of the change in the large-scale precipitation can largely explain that of SAt wet deposition in the future climate simulation.
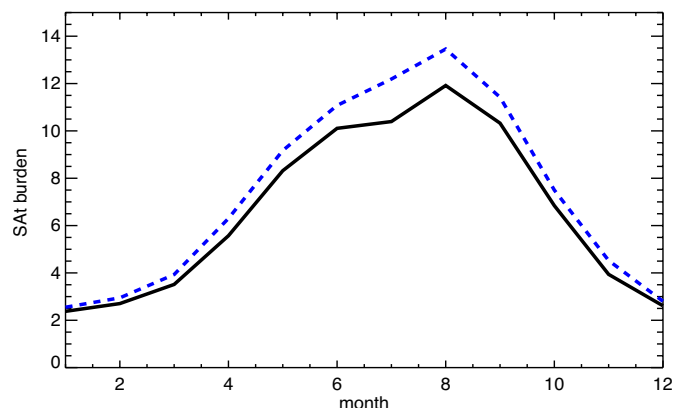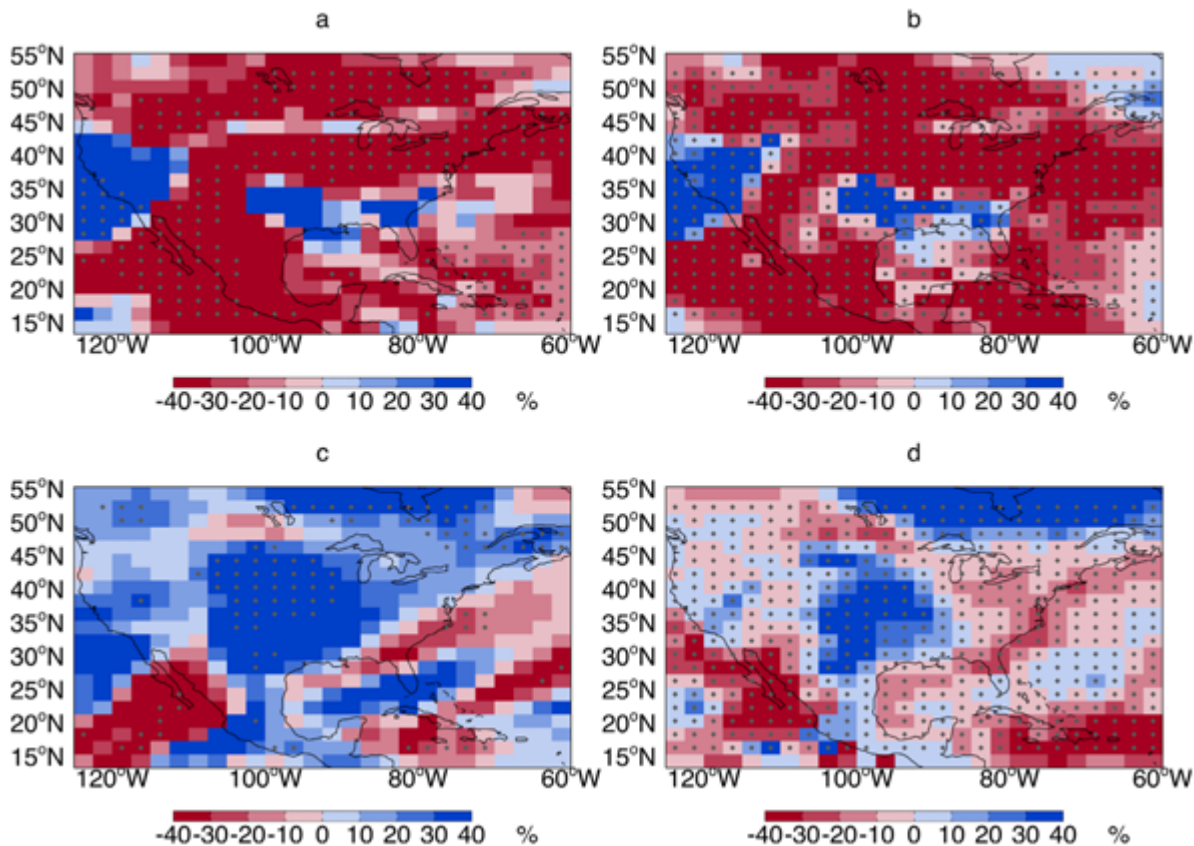
Figure 5. 20-year mean (a) July large-scale precipitation percentage change; (b) July large-scale wet deposition percentage change; (c) January large-scale precipitation percentage change; (d) January large-scale wet deposition change. Dotted area indicates changes satisfying 95% significance level. The maximum reduction of large-scale precipitation and wet deposition in July are about 100% and 90% while in January are both about 70%. Adapted from *Fang et al.*, 2011, in revision at *J. Geophys. Res.*

▪ Seasonal changes over North America

Despite the general consistency between zonal mean changes in large-scale precipitation and the SAt wet deposition (Figure 3), opposite-signed changes occur in the northern mid-latitude regions (25-40°N), where annual wet deposition decreases while large-scale precipitation increases. North America stands out as a good region for examining this inconsistency, and we begin with a discussion of the seasonality of changes in precipitation versus wet deposition.

The SAt burden over North America maximizes during summer, almost 6 times that during winter (Figure 4). Clearly, the precipitation change occurring during summer will be most important in driving the annual change in the SAt tracer. In July, large-scale precipitation decreases in the northern part of United States and over Mexico, and wet deposition follows accordingly over these regions (Figures 5a, b). During winter, large-scale precipitation increases almost everywhere over the inland eastern and central United States, and the wet deposition increases consistently in general (Figures 5c,d). Our analysis indicates that differences in seasonal variations of the SAt burden and the changes in precipitation lead to the apparent discrepancy between annual mean changes in precipitation and wet deposition that emerges in Figure 3 at northern mid-latitudes.

▪ Changes in precipitation frequency over North America

During winter, the agreement between the spatial patterns of changes in precipitation and wet deposition is weaker (Figures 5c, d). *Dawson et al.* (2007) also find a weaker sensitivity in the concentration of PM in January than in July over the eastern United States when perturbing precipitation rates in their chemical transport model. *Jacob and Winner* (2009) argue that this sensitivity difference implies that precipitation frequency, rather than the precipitation intensity, is a dominant factor determining the wet scavenging of the soluble tracers. In this section, we evaluate the impact of precipitation frequency on wet deposition. The mean precipitation rate $P_r = P_i \times P_f$, where P_i represents the intensity of precipitation in an average precipitation event and P_f represents the frequency of such precipitation events. Changes in precipitation rate in a future climate can be explained by changes in average precipitation intensity or in precipitation frequency: $\frac{\delta P_r}{P_r} = \frac{\delta P_i}{P_i} + \frac{\delta P_f}{P_f}$. The soluble

tracer burden is much lower in January than in July (Figure 3). Meanwhile, the large-scale precipitation, which is more effective in washing out the soluble tracers, is much stronger than convective precipitation in January. It is likely that in January, wet deposition over some regions is already
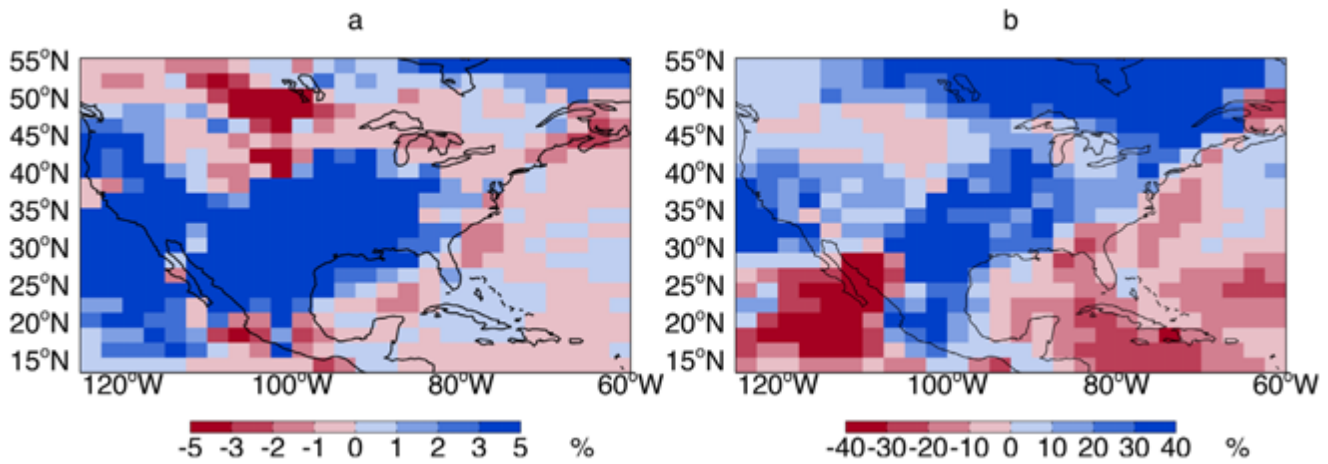
Figure 6. (a) Percentage change of the number of days with large-scale precipitation in all Januarys and (b) percentage change of large-scale precipitation intensity (calculated as the difference between the relative changes in large-scale precipitation (Figure 5d) and precipitation frequency (Figure 6a)) in January from 1981-2000 to 2081-2100 (unit: %). Adapted from *Fang et al.,* 2011, in revision at *J. Geophys. Res.*

saturated with precipitation intensity during a single event (i.e., increasing precipitation intensity cannot increase wet scavenging because all of the soluble pollutant has already been washed out). In this case, wet deposition is not sensitive to $\dfrac{\delta P_i}{P_i}$ (and as a result, it is not consistent with $\dfrac{\delta P_r}{P_r}$). Instead, changes in precipitation frequency ($\dfrac{\delta P_f}{P_f}$) play a more important role. We count the number of January days with precipitation in the 1981-2000 and in the 2081-2100 periods to estimate daily precipitation frequency. We find that both precipitation frequency and precipitation intensity (calculated as $\dfrac{\delta P_i}{P_i} = \dfrac{\delta P_r}{P_r} - \dfrac{\delta P_f}{P_f}$) change from the present to the future climate during January over the United States. The decrease in precipitation frequency over the Great Lakes, Maine, and New Brunswick, Canada appears to drive the wet deposition decrease (compare Figure 5d and Figure 6a). The spatial correlation coefficient between the relative change in precipitation frequency and wet deposition in January is 0.6 while between that of precipitation intensity and the wet deposition is only 0.2 over the United States. These results suggest that during winter, precipitation frequency, rather than precipitation intensity, plays a more important role in controlling the wet deposition of soluble pollutants.

■ The Diagnosed Precipitation Impact index

We test here whether we can infer future changes in global burdens and the spatial wet deposition patterns of soluble pollutants without needing to use a fully coupled chemistry-climate model. Specifically, we assess whether we can infer these changes directly from the present day tracer and precipitation distributions combined with two-dimensional patterns of changes in precipitation, fields routinely available from climate models. In the model,

the wet deposition flux is directly proportional to the local concentration, and depends non-linearly on the local precipitation production, particularly large-scale precipitation. This calculation assumes that the main climate change influence on soluble pollutants occurs primarily through precipitation that requires that changes in the spatial distribution of the pollutant burden from other processes (most notably, transport) are small. These assumptions are reasonable for our COt and SAt tracers.

We first examine the possibility of using the large-scale precipitation change weighted by the present-day SAt tracer burden to directly infer the changes in the spatial patterns of SAt wet deposition by 2081-2100. Although the wet deposition change over each grid box is affected by many processes other than local wet deposition, the spatial pattern of changes in the simulated wet deposition is well correlated with that of the burden-weighted large-scale precipitation change: global spatial pattern correlation coefficients of 0.6, 0.5 and 0.8 for annual, January and July (not shown). Consistent with our findings in Section 5.3, January has the lowest correlation coefficient.

The considerable correlation, particularly for July, motivates us to define a diagnosed precipitation impact index (DPI) based on the present-day large-scale precipitation, its change in a future climate and the present-day SAt burden: $DPI = \dfrac{\overline{B \cdot \Delta P_{ls}}}{\overline{B \cdot P_{ls}}}$, where B represents the present-day SAt burden, ΔP_{ls} and P_{ls} represents the change in large-scale precipitation and the present-day large-scale precipitation and the overbar represents a spatial or spatiotemporal average. The DPIs are consistent in sign with the relative changes in global wet deposition simulated annually and in July (annual and July DPIs are -6% and -11%, within a factor of 0.5-2 of the corresponding relative changes in wet deposition, -9% and -6%). The agreement between the DPI and the wet deposition change annually and in July confirms that the large-scale precipitation change determines the global wet

deposition change from the 1981-2000 to 2081-2100 in our model. Thus this DPI index may be useful to estimate the sign of future changes in the global burdens and wet deposition of soluble pollutants directly from a present-day burden distribution and large-scale precipitation alone. We emphasize that this DPI index can be used only under several assumptions relevant to our simulations with the AM3 model: 1) large-scale precipitation dominates wet deposition; 2) precipitation frequency is not the major driver of changes in wet deposition; 3) changes in the burden are predominantly driven by wet deposition rather than transport or chemistry.

This DPI index provides a simple way to examine the precipitation impact on soluble pollutant distributions by using only precipitation fields from different climate models. However, the relative importance of large-scale versus convective wet deposition is highly inconsistent across models (e.g., *Textor et al.*, 2006). Thus the DPI calculated using large-scale precipitation change would not be relevant for a model in which convective precipitation dominates wet deposition. Improved understanding and parameterizations of wet deposition processes would build confidence in projecting impacts of precipitation changes on wet deposition of soluble pollutants.

▪ Conclusions

We have used the Atmospheric Model version 3 (AM3) (*Donner et al.,* 2010) developed by the Geophysical Fluid Dynamics Laboratory (GFDL) to investigate how changes in climate may drive pollutant distributions to change in a warmer climate. In order to isolate climate-induced transport and precipitation changes and their implications for atmospheric pollutant distributions, we incorporate a simple carbon monoxide (CO)-like tracer (COt) with CO emissions and a fixed 25-day lifetime and a soluble version of that tracer (SAt) in the model for 1981-2000 and 2081-2100 meteorological conditions.

From a global and hemispheric perspective, a warming climate causes the mass exchange between the lower troposphere and the free troposphere to decrease by 13%. This leads to higher surface COt concentration (by less than 5%) near the source region (Figure 1a). Meanwhile, surface SAt concentration also increases (by about 10%; Figure 1b), due to the combined impact of reduced ventilation from lower troposphere to the free troposphere and reduced wet deposition in a warming climate. These results support prior emphasis on the need for tighter emission regulations to achieve a desired level of air quality as climate warms (e.g., *Holzer and Boer*, 2001; *Wu et al.*, 2008b).

The fact that wet deposition decreases despite the increased global precipitation in our model indicates that it may be misleading to use changes in the total precipitation as a simple predictor for the sign of soluble tracer changes in a warmer climate. We find that in AM3 model, large-scale precipitation dominates the wet scavenging of soluble pollutants, even in the tropics where convective precipitation exceeds large-scale precipitation (Figure 2). The zonal mean changes in wet deposition largely follow the regional pattern of large-scale precipitation (correlation

coefficient for zonal annual mean large-scale precipitation and tracer wet deposition change is above 0.9; Figure 3). We conclude that as climate warms, the longer lifetime of soluble pollutants with respect to wet deposition is mostly due to the simulated decrease in the large-scale precipitation over land. We note that this conclusion depends on the emission distribution of the tracer, which in our case is restricted to land. The seasonality of precipitation changes also modulates their impact. For example, over North America, precipitation and wet deposition both increase in January, and decrease in July (Figure 5). The absolute magnitude of the precipitation change is greater in January, so annual precipitation increases; the absolute magnitude of the wet deposition change is greater in July, so annual mean wet deposition decreases. The agreement between the precipitation and wet deposition changes over North America is weaker in January than in July, implying a possible role for changes in precipitation frequency on tracer wet removal, as shown to occur over the Great Lakes (Figure 6).

As the changes in wet deposition are dominated by the changes in soluble pollutant distribution as opposed to a transport-driven redistribution, we then explore if SAt burden-weighted large-scale precipitation changes offers a good estimate of the precipitation impact on soluble tracer burdens in a future climate. We find that the SAt burden-weighted large-scale precipitation correlates fairly well (spatially) with the SAt wet deposition changes in a future climate, especially for July and annually (r = 0.8 and 0.6 respectively). We thus develop a diagnosed precipitation impact (DPI) index: the global mean of present-day pollutant burden weighted large-scale precipitation change divided by the global mean of present-day pollutant weighted large-scale precipitation. This DPI could be used to directly infer soluble pollutant wet deposition responses from changes in precipitation as simulated by a climate model. This index captures the sign and magnitude (to a factor of 0.5-2) of the relative changes in the global wet deposition of the soluble pollutant tracer. If our findings that LS precipitation dominates wet deposition and that the horizontal transport pattern changes little in a future climate are broadly applicable, the DPI could be applied to LS precipitation fields in other climate models to obtain an estimate of the distribution of soluble pollutants under future scenarios.

The robustness of any predictions of future soluble pollutant tendencies should be evaluated with an ensemble of models. Climate models, however, are notoriously inconsistent in their simulated seasonal and regional precipitation changes (*Christensen et al.*, 2007). Applying our diagnosed precipitation impact index to other models that have precipitation change patterns available provides us a simple yet quantitative way to estimate the impact of precipitation change in those models on soluble tracers in a future climate. Such an approach, however, requires our finding that large-scale precipitation dominates wet deposition to be broadly applicable. Given the discrepancy in large-scale versus convective precipitation simulations across climate models and their relative importance in determining wet depositions (*Textor et al.*, 2006), there is a critical need for observational studies to advance

our understanding of these processes and improve their representation in models.

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Air Pollution & CLIMATE
a science-policy dialogue

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As part of its second phase synthesis activities, the International Geosphere Biosphere Programme (IGBP) has identified several key areas that cut across research in its own core projects and that also reach out beyond IGBP with the aim of exploring future cross disciplinary research needs. As such the IGBP Air Pollution & Climate initiative led by IGAC aims to engage scientist and policy makers to assess the status of knowledge with regard to current understanding about air pollution and climate and their interactions in particular with relation to current and proposed mitigation options and policy discussions. The idea is centered around setting up a "conversation" between scientist and policy-makers across the globe. Former IGAC co-chair Kathy Law (LATMOS, France) and current IGAC co-chair Paul Monks (University of Leicester, U.K.) are leading this effort. In addition to the co-chairs, the Air Pollution & Climate initiative steering group includes Nadine Unger (Yale University, USA), Denise Mauzeral (Princeton University, USA), and Terry Keating (USA EPA).

Motivation

Air pollution has negative impacts on human health and ecosystems and control strategies are a priority for reducing disease and mortality. In addition, to its impacts on human health and ecosystems, air pollution also has impacts on the climate. For example, tropospheric ozone and black carbon can lead to warming while sulphur containing compounds can lead to cooling. The indirect effects of aerosols on cloud properties and precipitation patterns, although still not well understood, could cause both warming and cooling effects. Deposition of black carbon on snow and ice surfaces leads to reduced surface albedo, enhancing warming in sensitive regions like the Arctic or the Himalayas. Owing to the short lifetime of air pollution (hours to weeks), its impacts on climate are observed at the local to regional scale. In addition, the long-term climate change resulting from a build up of CO_2 in the atmosphere could impact air pollution concentrations by modifying circulation and precipitation patterns. In the developing world, where air pollution reductions are pressing, there is also international pressure to address climate change, which could result in policies that further degrade air quality. Clearly, air pollution and climate change can no longer be considered as separate issues in the scientific and policy communities.

Whilst controls of CO_2 emissions should remain a priority for mitigation of climate change, policy makers have begun to consider reductions of short-lived forcers such as air pollutants in hopes that a reduction in warming could be achieved in the short-term (10-30 years). However, owing to the inexorable link between air pollution and climate change, it is critical that existing and proposed mitigation policies take into account the impacts they would have on **both** air pollution and on climate. In order for this to occur, policy makers and scientist need to establish a dialogue that allows policy makers to design mitigation strategies based on the best science available and for scientists to be able to examine the impact of different policy strategies on air pollution and climate.

Therefore, the IGBP Air Pollution & Climate initiative seeks to open a science-policy dialogue to examine the multiple implications (climate, human health, ecosystems, and food and water security) of existing and proposed mitigation policies that address air pollution and climate change in the near and long-term.

Key objectives

- Synthesis for policy makers on current state of knowledge on the role and interactions between air pollutants and climate change, including an assessment of uncertainties and identification of gaps.
- Explore and quantify possible mitigation options within socio-economic and scientific context.
- In partnership between policy makers and scientists, assess and develop new metrics to quantify co-benefits/trade-offs of past and future pollutant reduction strategies from different emission sources on air quality, human health, climate, ecosystems, and food and water security (within the context of natural changes in the Earth system).
- Build a new multi-disciplinary research programme to tackle cross cutting issues across traditional science-policy boundaries.

Current Activities

- Submitted proposal for a session at the Planet Under Pressure 2012 Conference
- June 2011 - IGBP Workshop entitled "Tackling the Air Pollution & Climate Change Challenge: A Science-Policy Dialogue" in Arona, Italy (Invitation Only)
- Fall 2011 – IGBP Workshop in Taiwan

For More Information

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Atmospheric Chemistry and Health Initiative

By **Tong Zhu**

Understanding the health effects of air pollution is essential for setting up air quality standards, assessing the health risk of exposure to air pollutants, and evaluating cost-benefits of air pollution control policies. Extensive studies on the health effects of various air pollutants have been conducted in the past, with the efforts of air quality scientists, environmental epidemiologist, and environmental toxicologists. In recent years, exposure science has emerged as a new discipline to better quantify human exposure to environment pollutants at different scales, from personal to regional.

Given the complexity of the exposures of human population to multi-pollutants in the atmosphere and their associations with various health end points, researches on the health effects of air pollution are facing many new challenges, such as: how to identify the health effects of various aerosol particle components, *e.g.* black carbon, sulfate, and secondary organic aerosol? How to quantify the exposure to gaseous and particulate pollutants more precisely with advanced technology? How to evaluate the "co-benefits" of climate change and air pollution reduction policies?

To deal with these challenges, a deep understanding of the complicated chemical and physical processes that form atmospheric pollution, as well as advanced measurement and modeling technologies, are essential. Interdisciplinary and integrated studies involving more atmospheric chemists to work with environmental epidemiologists, toxicologists, and exposure scientists could advance our knowledge in this field greatly.

In recent years, an increasing number of atmospheric chemists have become interested in the health effects of air pollution. During the processes of exchanging ideas between the atmospheric chemists and the health scientists, gaps of scientific knowledge and methodologies have been gradually recognized. We believe that initiatives organized by IGAC could play an important role to stimulate collaborative research on the health effects of air pollution between the atmospheric

chemists and health scientists, with the hope that atmospheric chemists are able to bring new insights to the understanding of the health effects of atmospheric pollutants, by providing comprehensive understanding of the atmospheric chemistry and cutting edgy methods.

At the IGAC 2007 SSC meeting, Tong Zhu suggested an IGAC initiative on human health. At the IGAC 2010 SSC meeting in Halifax, Gufran Beig and Candice Lung formally proposed an Atmospheric Chemistry and Health initiative, which was approved by the IGAC SSC. It is an important step that IGAC moved into the field of the health effects of atmospheric pollutants. The Atmospheric Chemistry & Health Initiative will be further discussed at the IGAC 2011 SSC meeting.

Also at the IGAC 2010 SSC meeting in Halifax, iCACGP secretary Frank Dentener and IGAC co-chair Tong Zhu jointly proposed a workshop on Atmospheric Chemistry and Health, with the support of Aaron Cohen of the Health Effects Institute and Bert Brunekreef of Utrecht University. This workshop will bring leading researchers from the atmospheric chemistry and health communities together to explore the various and multi-dimensional interactions between atmospheric chemistry and human health. The overall objectives of the workshop are to:

1. Identify the key scientific questions at the interface of atmospheric chemistry and air pollution health effects:

2. Identify key areas in which integrated research is needed;

3. Discuss the benefits for scientific research and environmental and health policy that would accrue from dealing with air pollution and atmospheric chemistry in a more integrated way.

The workshop on Atmospheric Chemistry and Health will be held on Oct. 12-13, 2011 in Boston. We hope the outcome of this workshop will lay a solid ground for the Atmospheric Chemistry and Health Initiative.

IGAC China Working Group

By **Tong Zhu**

Established over 20 years ago, IGAC has played a leadership role in identifying important research fields in atmospheric chemistry, formulating scientific plans, promoting the collaboration of international scientific communities in carrying out research in atmospheric chemistry, and enhancing interdisciplinary research with atmospheric chemistry. IGAC has made important contributions to understanding a variety of atmospheric environmental problems, such as urban and regional air pollution, inter-continental transport of pollution, atmospheric chemistry/climate interactions, global climate change, biosphere/atmosphere interactions, and changing emissions from both natural and anthropogenic sources.

With the growing concerns about environmental problems at local to global scales, IGAC is expected to play an even more active role in stimulating research, fostering collaboration across traditional disciplinary boundaries, and educating young scientists in the field of atmospheric chemistry. National/Regional-scale IGAC Working Groups could help to strengthen the links between IGAC and the atmospheric scientists in different countries/regions, and it could facilitate implementation of the IGAC agenda by organizing and acting as a liaison between scientists and policy makers at the local and regional levels.

With nearly 40 years of history in atmospheric chemistry research, Chinese atmospheric chemists have been conducting frontier research in areas such as urban and regional air pollution, the climate effects and health impacts of air pollution, and are playing an increasingly important role in the international community of atmospheric chemistry. The sheer magnitude of the country of China coupled with its large, growing and economically advancing population makes it critical that we understand its role in air quality and climate at a regional to global scale. This will be best done through an organized effort within the country and through close coordination of national and international efforts. In addition, the research to date within China on air quality and health leaves them well-poised to help IGAC advance its recently established initiatives on air pollution-climate interactions and on air quality and human health. Thus, the benefits of establishing a Chinese IGAC Working Group would be two-way.

On 24 October 2010, dozens of active Chinese atmospheric chemists held a preliminary IGAC China Working Group meeting in Beijing, organized by Tong Zhu. At this meeting, the organizational structure of the IGAC China Working Group was discussed, the articles of the IGAC China Working Group were finalized, members of the IGAC China Working Group were nominated, and Tong Zhu was nominated as the group leader. A short-term work plan, including the organization of the 2012 IGAC Open Science Conference in Beijing, was drafted.

An application for Establishing the IGAC China Working Group, together the articles of the IGAC China Working Group and a list of the nominated members, was submitted to the IGAC Scientific Steering Committee. After suitable consideration, the IGAC Scientific Steering Committee approved the formation of the IGAC China Working Group in April 2011.

The IGAC China Working Group comprises twenty members with expertise in gas-phase chemistry, aerosol chemistry, meteorology, aerosol-cloud interaction, air quality, and health effects. The working group is going to hold its first meeting in August 2011.

The establishment of the IGAC China Working Group will strengthen closer contact between the Chinese atmospheric scientists with IGAC, will foster international cooperation in atmospheric chemistry and the training of young scientists, and will stimulate the development of atmospheric chemistry in China.

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