

Dear Dr. Harris,

Below we have included our point-by-point responses to the reviewers and a copy of the latexdiff showing the revisions we have made to our ACPD manuscript.

Sincerely,  
Eric Sofen

**We thank Ian for his detailed and thoughtful comments on our manuscript. His comments are below and our replies are highlighted in bold.**

General comments.

This paper addresses a topic important to the development of atmospheric chemistry, how to improve the global coverage of surface ozone observations which are needed to more effectively constrain global atmospheric chemistry models. The paper is overall well-presented and appears to be without any significant scientific flaws and is appropriate for publication in ACP. A previous study addressed the more difficult issue of detecting trends in tropospheric ozone from ozonesondes and surface stations but with less sophisticated methods (Prinn 1988).

Specific comments. There is one aspect of the logic of the paper that could do with a more explicit recognition, at least in my opinion. The purpose of the analyses is to determine where more surface ozone observations should be made including both for improved direct observations and improved testing of chemical transport models. The analyses the authors perform are based on the deseasonalized output of a chemical transport model. The level of skill of chemical transport models in determining the residual variations in surface ozone after the seasonal variations are removed is probably not well quantified. So this must have some influence on the results presented. I do not think any variation in method is required, just a specific acknowledgement of the circular nature of the process. A few lines would suffice.

The information in this paper is contained in one Table and 11 Figures. The information in the Figures should be of a quality that a reader can determine the plotted quantity from the figures for any area of the world of interest to them. As indicated in Technical corrections, I do not think this standard is met in 4 of the Figures.

Technical corrections/suggestions.

Page 21026. Line 2, perhaps replace “almost 50 years” with “more than 40 years”. The DASIBI UV photometer for surface ozone measurements in the global networks first appeared in the early 1970’s. **Done**

Page 21026. Line 14 include coverage for the continent of Australia. **Done**

Page 21027. Line 16/17. The purpose of the GAW network is not primarily scientific, but rather to address key environmental issues (See WMO IGACO Plan). **Done**

Page 21030. Line 5/6 Is it “covariance” rather than “similar variability” that is being used?

**“Covariance” and “similar variability” are equivalent. We prefer to begin with a common-language description and then provide a technical description of the covariance calculation.**

Page 21032 Line 4. As the authors acknowledge, the large footprints may be erroneous due to the missing initial ozone destruction in springtime and photochemical production in summertime.

**We agree with the reviewer that the footprints may be smaller than indicated here and have highlighted the lack of ozone destruction during the spring as one issues.**

Page 21033 Line 14-19. A more physically based explanation (a few lines) of what the k-mean cluster analysis is would be useful for most readers. **Done.**

Page 21043 lines 20-26. I find the conclusion of this discussion unsatisfactory for a scientific paper. The authors statement that the data is critical is right. They need to state that they know observations are being made in these areas, and come to the conclusion they do, or advocate increased observations in these areas. We have updated the text here based on the reviewer’s comments.

**Our approach did not identify additional sites in China or India due to the small area (short lifetime) of any observational site. However, observations are being made in these countries. For example, China has an extensive air quality monitoring network where current data is publicly available online as Air Quality Index (AQI) values (<http://aqicn.org/>), but there is no available archive of historical data or direct reporting of concentrations. Until these observations are generally available for scientific evaluation it is difficult to know where further observations capabilities are needed.**

Page 21051 Figure 1. Include latitude and longitude, make the coastline bolder, mark Cape Verde Observatory more clearly.

**We have included the latitude and longitude on the figure and the location of CVO in the figure caption and have attempted to improve the aesthetic of the plot.**

Page 21054 Figure 4. I am puzzled that there is no desert in Australia in the Figure.

**The classification is determined based on an automated clustering algorithm. We have named the cluster that is illustrated in grey “desert” but it is clearly not a perfect classification, as portions of India and Central Asia fall into that classification. The lack of grey “desert” region in Australia may point to deficiencies in the Australian emissions inventories. Also note that the Atacama and Kalahari are not classified as desert, so there may be less distinction in the atmospheric chemistry between Southern Hemisphere desert and other Southern Hemisphere land areas that leads to them being merged into a single cluster.**

Page 21055 Figure 5. This figure is difficult to read with inadequate colour contrast.

**We have attempted to improve the aesthetics of the plot by changing the colormap from continuous to discrete and darkening the footprint shading.**

Page 21056 Figure 6 (a) and (b). These figures are difficult to read with inadequate colour contrast.

**We have attempted to improve the aesthetics of the plot by using discrete colormaps. We have chosen to stick with the basic colormap as it is one of the few multi-color perceptually uniform sequential colormaps available and it does not pose a serious hindrance to those with common forms of color-blindness (none of these properties are satisfied by the “Jet” or “rainbow” colormaps).**

Prinn, R.G., Toward an improved global network for determination of tropospheric ozone climatology and trends, *J. Atmos. Chem.*, 6, 281-298, 1988.

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**We thank the reviewer for their detailed and thoughtful comments on our manuscript. His comments are below and our replies are highlighted in bold.**

My overall impression of this study is that it provides a very unique,

thoughtful and valuable analysis of the representativeness of the present-day and (hopefully) future ozone monitoring network. As described below the paper needs a revision but I think my recommendations can be more or less easily addressed. My only major concern is the handling of the mountain top sites when determining the footprint of each site.

Major comments:

Regarding the method of determining the region represented by each station, how is terrain taken into account? For a marine boundary layer site the method seems straight forward and reasonable. But what about a high elevation site like Mauna Loa? GEOS-Chem at 2x2.5 degrees cannot resolve the terrain of Mauna Loa. As far as the model is concerned Mauna Loa Observatory is floating in the free troposphere at 3.4 km. This site is clearly representative of the lower free troposphere and not the marine boundary layer. If you use the model to compare ozone at the surface at that latitude and longitude you will be looking at the marine boundary layer, not the lower free troposphere. The same is true for land-locked high elevation sites like Zugspitze. The authors need to address this problem.

**The reviewer makes a good point. However, all of our footprints are calculated for the surface. Hawaii has an EPA measurement site near sea level so the footprint observed is representative of that site. Similarly for the high Swiss sites which have local lower sites close by. The only site which is at high-elevation without nearby sites in the dataset is located at Mt. Kenya**

All sites are sampled from the lowest vertical level in the model. Of course, the vertical resolution of models presents a constant challenge in comparing models and observations. GEOS-Chem uses a hybrid sigma (terrain-following) - pressure grid. Thus, grid boxes in the Alps are elevated above sea level, although the details of the orography are not resolved and heights of mountaintop sites will be underestimated. The general underestimate of mountaintop sites will likely lead to an underestimate in the size of their footprints, as ozone in the free troposphere will have a longer lifetime and will be transported a longer distance before chemistry and transport processes lead to a decorrelation with the observing site.

Computing all of the correlations is already a substantial computational load ( $\sim 12000^2$  calculations taking several hours over 64 processors). Adding vertical levels beyond the surface would increase the conceptual complexity as well: what do overlaps of a



**surface footprint and a footprint from the second vertical level mean?**

The writing style of this paper is too “chatty” and is better suited for the lecture hall rather than a research publication. A good example is on page 21028 where the change in the composition of the atmosphere is referred to as a global scale experiment. I understand the analogy and this is fine to say in a lecture to grab the attention of your audience but of course humans clearly are not conducting an organized experiment on the atmosphere, the change is a by-product of human activity.

**We have reworded the text on page 21028 and have attempted to remove the chatty text in other parts of the paper while trying to ensure readability and conciseness.**

P21027 line 4 Here you say that expanding the ozone monitoring network will benefit the development of policy. This is a science paper and not a policy paper and without explicit considerations as to how the expanded network would benefit policy development the paper should steer clear of making such a statement. A better statement would be “would provide a significant long term benefit to our understanding of the composition of the atmosphere, information which will also be available for consideration by air quality control managers and policy makers”.

**We have updated the text as requested.**

Page 21032 The biome analysis is nice but from the figure it’s difficult to tell the degree to which the various biomes are monitored for ozone. Please provide a table that lists the biomes and provides the percent area covered by the ozone monitors.

**We acknowledge that this would provide some additional information however, the biomes data that we have is a vector description and it would be a non-trivial activity for us to calculate this for small enhancement in the value of the paper. If pushed we would have to remove this section rather than be able to comply with the reviewer.**

Minor Comments: if no explanation is given for a comment please insert the suggested text into the appropriate place in the manuscript.

P21026 line 24 the models **Done**

p21027 line 6 Tropospheric ozone is an air pollutant that impairs human respiratory function and damages both crops and natural vegetation. **Done**

P21027 line 10 What do you mean by prediction, are you talking about model estimates? If so you should say: “the accurate model estimation of

the concentration of observed tropospheric ozone is...” **Done**

P21027 line 21 ozone is inadequately measured **Done**

p21027 line 26 Scientists make conclusions based on data but data don’t make conclusions by themselves. “...instrumentation provides an essential validation/verification of these remotely sensed observations.” **Done**

Page 21028 line 1 coverage of surface ozone observations in problematic. **Done**

Page 21028 line 21 No site, urban or rural, can be representative of global ozone, a site can’t even be representative of hemispheric ozone. So here just say that urban sites are not representative of regional conditions. **Done**

Page 21028 line 24 ...data are held by many individual PIs Line 24 ...do not provide long term observations **Done**

Page 21029 lines 3-8 This paragraph needs a lot of work: “...from the perspective of surface coverage, biosphere/atmosphere interactions, chemical regimes and chemical transport model evaluation. We then assess the best locations for new sites to improve our understanding of surface ozone and we conclude with a list of locations that we argue would best expand our observing capabilities.” **Done**

Page 21029 line 11 The first sentence is not necessary, just begin with: “An idealized surface ozone network would provide...” **Done**

Page 21030 line 1 The sentence beginning with “A similar approach” is just repeating what you said in the previous sentence. Delete this sentence and simply reference Henne et al in the previous sentence.

**There is a distinction between the two. In the first case, a set of back trajectories, such as from the Hysplit model, are often used to characterize the area that a site represents, particularly in the case of seasonal- or shorter-term field campaigns. The Henne 2010 approach is more general, accounting for both forward and backward trajectories and is likely calculated in a somewhat different manner from the individual trajectories.**

Page 21031 line 14 Would sound better as: How useful are these sites for observing the... **Done**

Page 21031 line 22 Drop “so” **Done**

Page 21032 line 1 Drop one of the onlys **Done**

Page 21032 Replace so with therefore **Done**

Page 20132 line 15 Not sure what you mean by “an uncertain chemistry”. Please re-phrase.

Changed to “Biogenic emissions also play an important role in tropospheric oxidant chemistry, and the chemistry between tropospheric oxidants and biogenic volatile organic compounds is highly complex and uncertain.”

Page 21036 The last sentence has structural problems

Revised to “Secondly, we look at the projected future trends in ozone, in these same models, to identify regions with large projected changes in ozone concentrations which should be monitored.”

Page 21044 line 14 South America **Done**

Page 20144 line 20 straightforward **Done**

# How to most effectively expand the global surface ozone observing network

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## Abstract

Surface ozone observations with modern instrumentation have been made around the world for ~~almost 50~~more than 40 years. Some of these observations have been made as one-off activities with short term, specific science objectives and some have been made as part of wider networks which have provided a foundational infrastructure of data collection, calibration, quality control and dissemination. These observations provide a fundamental underpinning to our understanding of tropospheric chemistry, air quality policy, atmosphere-biosphere interactions, etc. Sofen et al. (2015) brought together 8 of these networks to provide a single dataset of surface ozone observations. We investigate how representative this combined dataset is of global surface ozone using the output from a global atmospheric chemistry model. We estimate that on an area basis, 25 % of the globe is observed (34 % land, 21 % ocean). Whereas Europe and North America have almost complete coverage, other continents~~such as~~, Africa, South America, Australia, and Asia (12–17 %) show significant gaps. Antarctica is surprisingly well observed (78 %). Little monitoring occurs over the oceans with the tropical and southern oceans particularly poorly represented. The surface ozone over key biomes such as tropical forests and savanna is almost completely unmonitored. A chemical cluster analysis suggests that a significant number of observations are made of polluted air masses, but cleaner air masses whether over the land or ocean (especially again in the tropics) are significantly under observed. The current network is unlikely to see the impact of ENSO but may be capable of detecting other planetary scale signals. Model assessment and validation activities are hampered by a lack of observations in regions where ~~they the~~ models differ substantially, as is the ability to monitor likely changes in surface ozone over the next century.

Using our methodology we are able to suggest new sites which would help to close the gap in our ability to measure global surface ozone. An additional 20 surface ozone monitoring sites (a 20 % increase in the WMO GAW ozone sites or a 1 % increase in the total background network) located on 10 islands and in 10 continental regions would almost double the area observed. The cost of this addition to the network is small compared to

other expenditure on atmospheric composition research infrastructure and would provide a significant long term benefit to our understanding of the composition of the atmosphere ~~and in the development of policy~~, information which will also be available for consideration by air quality control managers and policy makers.

## 1 Introduction

Tropospheric ozone is an air pollutant that ~~causes human respiratory problems~~ (McDonnell et al., 1993; Bell et al., 2004; The Royal Society, 2008). ~~It also impairs human respiratory function~~ (McDonnell et al., 1993; Bell et al., 2004; The Royal Society, 2008) and damages both crops and natural ~~plant life~~ vegetation (Bell and Treshow, 2002). It is a greenhouse gas (Myhre et al., 2013) and plays a central role in tropospheric oxidant chemistry (Finlayson-Pitts and Pitts, 1997). Given that it is relatively easy to measure, the accurate ~~prediction~~ the accurate model estimation of the concentration of observed tropospheric ozone is often used (rightly or wrongly) as a central assessment for our ability to understand tropospheric chemistry. Over the past forty years, a number of publicly available surface ozone networks have been created. Some have been created in response to air quality legislation (e.g. US (CASTNET), Canada (CAPMON), Europe (EMEP)). Others are global in their scope (e.g. World Meteorological Organization Global Atmosphere Watch (GAW)) and are primarily ~~scientific in their outlook~~ for global atmospheric monitoring in of the composition of the atmosphere to study global environmental questions. The size, scope and locations of the measurement sites within these networks has been determined by a combination of scientific questions, cost, political expediency, serendipity, political necessity and convenience rather than by a systematic attempt to provide globally coverage. It seems likely that the global distribution of surface ozone is ~~unmeasured~~ inadequately measured even by the sum of these networks.

Satellites are able to fill in some of these gaps but they are typically less responsive to changes in the surface concentration of ozone (where human and vegetative impacts occur) than to changes higher in the column, and some form of ground-truthing from relatively sim-

ple instrumentation provides an essential validation/verification of ~~their conclusion~~these remotely sensed observations.

This lack of global coverage of surface ozone observations is problematic. The composition of the atmosphere is changing due to changes in emission, land use, climate, etc. This is part of an ongoing change since the the pre-industrial era. Industrialisation has led to increasing concentrations of ozone (Volz and Kley, 1988; Marenco et al., 1994; Staehelein et al., 1994; Cooper et al., 2014) but our record of this change is poor both temporally (systematic measurements typically are available from the 1980s onwards) and spatially (observations are sparse). ~~There has essentially been a planetary scale experiment to change the composition~~As emissions have been altered by human activity, these changes, their implications for the chemistry of the atmosphere~~without a planetary scale capability to observe the results. This experiment with atmospheric, and the further impacts of that chemistry have not been fully observed.~~ Atmospheric composition is continuing to change with emissions of ozone precursors in some regions (North America and Europe) dropping, whereas emissions in other regions (Asia, Africa, South America) forecast to increase (The Royal Society, 2008). ~~If this future experiment is to be appropriately monitored~~In order to better observe and quantify these ongoing and future changes in a global manner that can enhance our scientific understanding and guide environmental policy, the observational network needs to be fit for this purpose.

Here, we investigate how well the present day (1971–2013), publicly available surface ozone observations networks cover the globe and how that surface ozone network may be expanded to improve coverage. We base our assessment on the compilation of surface ozone data made by Sofen et al. (2015). This dataset includes sites from WMO GAW, US EPA AQS, CASTNET, EU AirBase, EU EMEP, Canadian NAPS, CAPMON, and EANET. Details of the networks and the location of the sites can be found in Sofen et al. (2015). The dataset is quality-controlled to remove urban sites (which are not representative of ~~global or~~ regional conditions) and sites with poor quality data, leaving 2389 sites. We assume that every site in the dataset is currently active. We know that there are measurements from many other locations, but the data ~~is either are~~ not easily accessible (data ~~is held by~~

~~a raft of~~ are held by many individual PIs in a range of different file formats), ~~does do~~ not provide a long term observation (measurements are for short periods of time  $< 5$  years), ~~and or~~ may not be sufficiently quality controlled. The Tropospheric Ozone Assessment Report (TOAR) (<http://www.igacproject.org/TOAR>) is attempting to provide a data framework to collect these datasets, provide some post-processing quality control and allow for dissemination. In the future TOAR may provide a framework for the inclusion of a much wider set of observations than those used here.

Here, we consider the representativeness of the Sofen et al. (2015) data from the perspective of ~~four science areas related to~~ surface coverage, biosphere/atmosphere ~~interaction~~ interactions, chemical regimes, and chemical transport model evaluation. ~~From this, we assess where it is most useful to add~~ We then assess the best locations for new sites to improve our understanding of surface ozone and ~~present we conclude with~~ a list of locations that we argue would best expand our observing capabilities.

## 2 Current spatial coverage

### 2.1 Representativeness

~~It is impossible to measure the ozone concentration everywhere at infinite spatial resolution but a consistent representation of the surface coverage across the Earth's surface would be useful for global model studies, trend analysis, monitoring, and impact assessment.~~ An idealised network of surface observations would provide measurements at a fine enough spatial resolution to enable the reconstruction of the surface ozone field globally, taking into account the varying lifetime of ozone and local meteorology, at a scale that would be useful for global model studies, trend analysis, monitoring, and impact assessment.

Each ozone measurement site represents not just the mixing ratio of ozone at that point in space, but is also representative of a “footprint” both upwind and downwind of that location. The ozone lifetime and transport patterns determine the extent of this footprint. If the local ozone lifetime is short at that location, a measurement will be representative of a small foot-



print; if the transport is rapid, a measurement will be representative of a larger footprint. A number of methods can be used to evaluate this footprint. One common approach to determining a site's footprint is to use back trajectories, but this fails to take into account either the spatial and temporal variability in the ozone lifetime or that an ozone measurement is also representative of a region downwind of the site. A similar approach uses a fixed time-frame catchment area in a Lagrangian tracer model (e.g. Henne et al., 2010) [to define footprints for many European air quality sites](#), but this [also](#) fails to account for how the lifetime of ozone varies across the Earth. Instead, we use an Eulerian forward atmospheric chemical transport model (GEOS-Chem v9-01-03; [geos-chem.org](http://geos-chem.org)) (Bey et al., 2001; Parrella et al., 2012) to determine footprints based on areas of similar variability in ozone. We use monthly mean surface ozone concentration calculated using the  $2^\circ \times 2.5^\circ$  version of the model, run from 1 January 2005 to 31 December 2011. We de-seasonalise this output by removing the mean annual cycle from each model grid-box. Then for each observing site, we calculate the correlation coefficient ( $R$ ) between the ozone in that grid box and the ozone in all other model surface grid boxes. The footprint associated with a site is then determined by the model grid boxes with a  $R \geq 0.707$ . This threshold is chosen such that  $R^2 \geq 0.5$ , or at least half the non-seasonal variance in ozone in any grid box in the footprint may be explained by the ozone observation at the observing site. To ensure that the footprint only includes grid boxes contiguous with the observing site, we use a random walk process that goes “down-hill” from the observing site until  $R < 0.707$ . The random walk process is allowed to wrap across the International Date Line ( $180^\circ$ ). We thus assume an area is “observed” from the perspective of ozone if it falls within the footprint of a monitoring site. This footprint approach is similar to that taken by Messié and Chavez (2011) to determine the spatial coherence in sea surface temperature anomalies.

Figure 1 illustrates how the size and shape of the footprint for the Cape Verde Observatory varies as the threshold is changed from 0.1 to 0.9. There will be some differences in the footprint determination between different model simulation and between different models. However, based on testing with results from the GFDL AM3 model results used for ACCMIP

(Young et al., 2013; Donner et al., 2011) (not shown) we do not believe that this provides a significant uncertainty.

Individual footprints are approximately symmetric upwind and downwind of the observing site. The footprints do not represent back-trajectories, but the areas of similar ozone both backward and forward from the observation site. Footprints are typically larger over the ocean where ozone surface deposition is slow and ozone concentrations are more homogeneous due to the lack of emissions. They are larger towards the poles than the tropics as ozone lifetimes are longer. Figure 2 shows the coverage map made up from the composite of all of the individual site footprints. Given the present day network of ozone observations and our evaluation of their global footprint we are now able to evaluate the representativeness of this network. The “representativeness” however depends on the question that is being addressed. Here we evaluate a range of questions.

1. What fraction of the planet is covered? How does this split between ocean and land? How well observed are the different continents?
2. Which biomes are being monitored?
3. Which atmospheric chemical regimes are being sampled?
4. How useful are these observations for constraining uncertainty in global models?
5. How useful are these **observations** sites for observing the predicted future changes in atmospheric composition?

## 2.2 Area

It is obvious from Fig. 2 that the coverage of observations is not global. We find that 25 % of the Earth’s surface is covered, 21 % over the oceans and 34 % over the land. Only twice as much of the northern hemisphere is covered compared to the southern hemisphere (33 % vs. 18 %) despite 99 % of the observations being in the North. This is due to the larger footprints in the Southern Hemisphere (mainly ocean with low dry deposition rates and

so-long ozone lifetimes) and overlapping footprints in the Northern Hemisphere. While the entirety of Europe and 80 % of North America are covered, the other continents are much less well represented with 17 % of South America, 14 % of Oceania and Australia, and 12 % each of Africa and Asia covered. 78 % of Antarctica is represented despite ~~only~~ having only 7 sites. This is due to the very large footprints associated with these Antarctic sites because (at least in the model) there are no local precursor emissions, no photochemistry during the polar night, ~~and~~ uniform deposition, and inadequate springtime ozone destruction, all leading to a very long ozone lifetime and little spatial variability on monthly timescales.

It is evident that the air quality networks in Europe and North America do a good job in representing the regional and background concentrations of ozone. The story for the rest of the world (other than Antarctica) is much more mixed with 12–17 % of the land area of these continents being monitored. Ocean coverage is generally poor, with the tropical and southern hemisphere oceans being particularly poorly observed.

## 2.3 Biomes

The impact of ozone on the biosphere is a critical feedback in the Earth system (Ainsworth et al., 2012). Ozone can diminish plant function and ~~so~~ therefore slow carbon dioxide uptake. This may have a significant impact of climate by impacting the terrestrial carbon budget and atmospheric CO<sub>2</sub> concentrations (Sitch et al., 2007). Biogenic emissions also play an important role in tropospheric oxidant chemistry ~~with an uncertain chemistry~~, and the chemistry between tropospheric oxidants and biogenic volatile organic compounds is highly complex and uncertain (Lelieveld et al., 2008). Thus, understanding the ozone exposure of different biomes is important when considering the atmosphere-biosphere feedback.

In Fig. 3 we show the global coverage of the current ozone observing capability on a map of terrestrial biomes (Olson and Dinerstein, 2002; The Nature Conservancy, 2012). With North America and Europe completely covered, the temperate grasslands, temperate forests, and North American boreal forests are probably well represented within the ozone observing networks. However, the map reveals that the network lacks coverage over trop-

ical forests, tropical/subtropical grasslands, Eurasian temperate and montane grasslands (e.g. steppe), and the large area of boreal forest across Asia.

Some of these unobserved biomes are critical for the budget of ozone, for the uptake of  $\text{CO}_2$  (which may be impacted by  $\text{O}_3$  uptake to plants), and are subject to significant land use change. Given the lack of ongoing monitoring of these biomes, the impact of land use change on composition and vice versa, is not being monitored. The lack of available long term observation of ozone in the major tropical forested regions of South America, Africa and Asia appears to be a critical failing of the current network.

## 2.4 Chemical regimes

The previous assessments have focused on the geographical distribution of measurements (latitude, longitude). However, the atmosphere could also be split in terms of chemical coordinates. We use a cluster analysis approach applied to chemical transport model output to define chemical regimes and explore how well the different chemical regimes are being observed. From a one year GEOS-Chem full chemistry ( $\text{NO}_x$ – $\text{O}_x$ – $\text{BrO}_x$ – $\text{HC}$ –aerosol) simulation (v9-01-03;  $2^\circ \times 2.5^\circ$ ), we extract monthly mean concentrations of all chemical tracers as well as the photolysis rate of ozone and concentration of OH. We exclude sea salt and carbonaceous aerosol species as they occur at very low concentration (model precision) in some gridboxes. Logs of the concentrations are taken of the remaining 52 species ( $x$ ) and they are then normalized (so that that  $\overline{\log_{10} x} = 0$  and  $\sigma_{\log_{10} x} = 1$ ). A k-mean cluster analysis (Pedregosa et al., 2011) on the normalised logged model concentrations determines areas of similar chemistry. [K-mean clustering groups data in to  \$k\$  clusters based on the Euclidian distance, in this case in normalized chemical concentration space, between each data point and the mean of each cluster.](#) Using ten to fifteen clusters produces qualitatively understandable chemical regimes. The chemical regimes using twelve clusters are illustrated in Fig. 4a. In Fig. 4b, a bar chart shows the total global surface area and the observed area of each cluster.

The regimes are spatially coherent, while also reflecting similar chemistry across multiple regions. For example, the Northeastern US, Western Europe, India and Eastern China are

classified in the same cluster which we described as “polluted”. While there are difference in the chemistry of these regions they are (from the perspective of the model, at least) small compared to the difference with other clusters. We identify clusters associated with polluted regions, biogenic emissions, biomass burning, polar regions, deserts, and several oceanic regions. The oceanic classifications exhibit a zonal banding due to the strong latitudinal and seasonally-driven inter-hemispheric dependence in some of the input tracers.

At least 50 % of boreal, polluted and Antarctic grid box are covered. For the polluted case this reflects the total coverage of polluted grid boxes over Europe, North America and some of East Asia but very little coverage from anywhere else in the world. Much of the Boreal coverage again comes from European and North American air quality networks. The large fractional coverage of Antarctica is due to the long lifetime of ozone in this region.

Antarctica’s coverage contrasts to that of the Arctic (36 %). Here a similar number of observations fail to characterise the region due to its proximity to Europe and North America emissions which leads to higher levels of variability and thus smaller footprints. Northern extratropical oceans (42 %) are reasonably observed due to measurements made from the European, North American and East Asian networks that extend beyond the continental regions and from islands in the Atlantic.

All of the other classifications show low coverage. Lowest is the Southern tropical ocean cluster where only 9 % of the air within that cluster could be considered measured which is achieved by the GAW site in Samoa. Chemically critical environments such as the tropical forest where fluxes of VOCs are high and our understanding of the chemistry is poor are again barely observed.

Similar to the analysis by area and biome, we conclude that we probably have good observations of the composition of Antarctic, polluted, and boreal environments. Virtually all other types of air mass are poorly observed.

## 2.5 Climate-chemistry modes of variability

The composition of the atmosphere responds to changes in emissions, solar radiation, deposition, transport etc. The leading global mode of inter-annual variability in the transport

is the El Niño-Southern Oscillation (ENSO) pattern (Zhang et al., 1997). The impact of ENSO on tropical and extratropical tropospheric ozone has been detected from satellite observations (Ziemke et al., 2010, 2015) and is captured by global models (Oman et al., 2011; Zhang et al., 2015), but has not been observed at the surface. However, the ability of models of atmospheric composition to correctly respond to this large scale forcing may be a critical test of their performance.

Spectral analysis of the surface ozone from a 30-year chemistry-climate simulation using the ACCMIP GFDL AM3 model simulation (Young et al., 2013; Donner et al., 2011) reveals a peak in the power spectrum at the ENSO timescale of 3.8 years. The spatial pattern of surface ozone anomalies associated with that timescale is shown in Fig. 5. The modelled surface ozone ENSO signal is small (at most 1.1 ppbv peak-to-trough) with opposite phases between the eastern and western Pacific. The small amplitude may be in part due to the quasi-periodic nature of ENSO. This broadens the peak in the power spectrum and means that the variability associated with the particular frequency shown in Figure 5 may be a low estimate of total ENSO variability. There are no sites located directly in the area of modelled maximal ENSO variability that could assess its magnitude observationally which probably explains why an ENSO signal has not been observed in surface observations. The existing WMO GAW site at Samoa, may be able to observe some of the variability but this is outside the region of the largest ENSO signal seen in the model. Making long-term ozone observations from sites such as the Galapagos Islands or the Marquesas Islands in the central Pacific would allow the impact of ENSO on ozone concentrations to be observed. Observations in the western Pacific from islands such as Guam or Palau would also be valuable for this purpose, although the footprints associated with this area are smaller than those in the eastern Pacific.

The second-order patterns of climate variability such as the Arctic Oscillation (Thompson and Wallace, 1998), Atlantic Multidecadal Oscillation, and Pacific Decadal Oscillation (Deser et al., 2010; Messié and Chavez, 2011) are located in the northern Atlantic and Pacific basins with their climate impacts primarily felt in North America and Europe which are

well covered by ozone observations. It should be feasible to investigate the surface ozone response to these signals.

Other large scale transport flows are the global monsoonal flows. The impact of monsoonal flows on ozone in locations such as India, West Africa, and Southeast Asia (Trenberth et al., 2000; He et al., 2008) are unlikely to be observed in the current observational networks but those in North America will most likely be observed.

## 2.6 Model evaluation

One valuable use for ozone observations is to evaluate the ability of atmospheric chemistry transport models to capture the distribution and variability of ozone. This provides an evaluation of models' ability to aid our assessment of air quality, climate, and fundamental atmospheric chemistry. Tropospheric column evaluations based on satellites or ozone-sondes may be most important for climate purposes, as the column total and vertical distribution determines ozone's potency as a greenhouse gas. However, near-surface observations are necessary for evaluating the capabilities of chemical transport models with respect to air quality and food security questions. When existing observational datasets are used to evaluate surface ozone, results and the conclusions drawn from them may be biased heavily towards the conditions in Europe and North America due to the overwhelming number of observations in these regions. Careful statistical weighting may help to alleviate this bias, but is made challenging by the varying size of site footprints described above and a corresponding variation in the spatial autocorrelation of observations.

Using the Atmospheric Chemistry and Climate Model Inter-comparison Project (ACCMIP) ensemble of model results (Lamarque et al., 2013), we compare the usefulness of the surface ozone network in two ways. Firstly, we investigate the spatial distribution of the inter-model spread in present day annual mean ozone, which provides an indication of where uncertainty in models is highest, and so where observations may be useful to differentiate between model. Secondly, we look at the projected future trends in ozone, in the these same models, the to identify regions there will be a change with large projected changes in ozone concentrations which should be monitored.

### 2.6.1 Inter-model variability

Following the work of Young et al. (2013), we calculate the standard deviation of annual mean surface ozone between eight ACCMIP models (Lamarque et al., 2013) for the present day (2005–2010). Figure 6 shows the ensemble uncertainty (standard deviation between annual means) in both (a) absolute and (b) fractional terms.

Figure 6a illustrates that the greatest absolute variability between models occurs over industrialised areas of the Eastern US, Eastern Asia and Europe as well as Greenland and through a general belt across the northern extra-tropics. Given that the ACCMIP project specified emissions, the differences over polluted industrialised regions probably reflects differences in model chemistry scheme and the treatment of VOC emissions speciation. These uncertainties appear to be transported through the Northern Hemisphere leading to a wide band of difference between models. The high uncertainty over Greenland is spatially coherent and may reflect issues with the representation of orography between models. Our ability to measure ozone (and so provide a constraint on models) is very good over North America and Europe. There is some capability in East Asia but China is missing. Differences in the northern extra-tropical Pacific will be unmeasured with the potential for some measurement over the northern-extra tropical Atlantic. Measurements made over Greenland allow the large variability here to be analysed.

Different locations become important on a fractional basis. As well as the polluted regions described earlier, large fractional uncertainty occurs over the Amazon and tropical oceans, notably the tropical western Pacific, which are essentially unobserved. Again similar to the comments in the previous sections the addition of a relatively small number of observations in key regions would help to constrain global model uncertainty.

### 2.6.2 Trends

Ozone concentrations are thought to have increased significantly over the twentieth century (Marenco et al., 1994; Staehelin et al., 1994; Cooper et al., 2014), but the magnitude and spatial distribution of this change is uncertain due to a lack of observations over this period.



Simulations of future ozone suggest similar changes over the next 50–100 years. However, this time we have the opportunity to monitor this change if observations are made in the correct places.

Figure 7 illustrates the mean changes in surface ozone between 2005–2010 and 2095–2100 in the RCP2.6 and RCP8.5 scenarios in the ACCMIP models (Lamarque et al., 2013). Trends are nearly identical to those calculated by Young et al. (2013). These RCP2.6 and RCP8.5 scenarios suggest opposite trends in surface ozone at 2100, due to the differing assumptions inherent in the emissions scenarios. Surface ozone in RCP2.6 decreases by 10 ppbv over large swaths of the Northern Hemisphere midlatitudes and up to 15–20 ppbv over the United States. The only areas of increasing ozone in RCP2.6 are over West and Central Africa. While the general drop in surface ozone would be observable with the current network, the increase in West Africa would go unmeasured by the the current observational network.

In contrast, the RCP8.5 scenario suggests the potential for 5–7 ppbv increases over polar regions and Northern Hemispheric oceans and increases of 7–15 ppbv over portions of India, the Middle East, and sub-Saharan Africa together with an increase in the hemispheric background. Again the current observational network would not be capable of measuring the changes in these locations.

Significant changes are forecast in surface ozone over the next decades. In some regions this change should be observed by the current network (North America and Europe). However, much of this change will occur in regions with no observational capability.

### **3 Where to add observations**

From the previous analysis, the current surface ozone monitoring network fails to make measurements in key regions (China, India, Amazon, Africa, tropical oceans, Southern Ocean). This has implications for our ability to understand the processes going on in the atmosphere and provide robust policy advice. There are ongoing efforts to improve the global atmospheric observational capability through the establishment or enhancement of surface

sites. There may be a range of reasons for establishing a new ozone monitoring site based on the local political requirements, practicality, finance, and scientific goals, but if they are to be established, basing the site choice on maximising the global benefit would be a sensible criterion. To aid in this, we evaluate each model grid box which does not currently have an observational site for its potential ability to improve the global measurement network.

Our primary metric for evaluation is the additional surface area that a surface ozone measurement in that location would bring. The additional area is the area of that site's footprint, minus any area already covered by the footprints of existing observing sites. Figure 8 illustrates the amount of additional area that each grid box would add to the global coverage if an ozone monitor were installed there. Areas in grey show no additional benefit from siting an observation there. The lighter the colour the greater the area covered. There is significant variation between sites due to the lifetime of ozone (longer over the ocean than over the land, longer towards the poles than towards the tropics), transport pathways, and the positioning of existing ozone sites.

The areas that would provide the greatest additional contribution to the area coverage of ozone observations come from the Atlantic and Indian sectors of the Southern Ocean. This is due to the large footprint size in those regions, combined with few existing Southern Hemisphere measurements outside of Antarctica. The terrestrial areas that will provide the greatest additional area coverage are northeastern Russia and the southern Amazon basin. Many of these recommended areas also align well with other characteristics related to ozone where we wish to expand coverage (e.g. tropical forest biomes or areas of high inter-model uncertainty).

Figures 9 and 10, shows the footprints from the top ten distinct locations for oceanic and terrestrial areas, respectively. Table 1 summarizes how each of the sites relates to the characteristics described in Sects. 2.2–2.6.2 above.

### 3.1 Oceanic sites

The oceanic sites that could add the most additional area are located over the Southern Ocean, and the tropical and southern subtropical Pacific and Atlantic Oceans (Fig. 9). The

challenge to expanding the observing capacity over oceanic regions is finding nearby islands that provide suitable environments and infrastructure for long-term continuous measurements. For ozone measurements, these requirements include a steady electricity supply, a temperature-controlled room for the instrument, an internet connection for remote data access, and regular (approximately monthly) or as-needed visits by trained staff.

There are a range of options in the Southern Ocean. In the Atlantic sector, Bouvet Island, South Georgia and the South Sandwich Islands provide potential site locations and all presently house research stations (British and Norwegian). Peter I Island and Scott Island are the only two options in the Pacific sector. Unfortunately, both are rugged and uninhabited and the only scientific infrastructure are automatic weather stations, making them less suitable options. Near Australia and New Zealand, the Macquarie Island Station is a potential site that would cover portions of the Pacific and Indian sectors of the Southern Ocean. In the Indian Ocean sector, the Crozet Islands have a permanent research station run by the French, the Prince Edward Islands host a South African research station, and the French Kerguelen Islands are populated and host multiple research stations.

In the eastern Pacific, potential islands to host ozone sites include the Pitcairn Islands, Marquesas Islands, and Galapagos Islands. One notable contribution from these locations is that they could potentially capture an ENSO signal in surface ozone. The tropical Pacific also represents the tropical oceanic chemical regime (Fig. 4) that has the least coverage of any of the chemical regimes. The tropical Pacific is also an area of high fractional inter-model spread (Fig. 6b). We are aware of measurements currently being made on the Galapagos Islands (Wang et al., 2014, A Saiz-Lopez, personal communication, 2015) and the long-term reporting of this data to the GAW network or TOAR dataset would provide a significant global benefit.

In the southern and tropical Atlantic Ocean, there are four islands that are good candidates for new ozone sites. St. Helena and Ascension Islands both already host WMO GAW sites. Flask and sonde measurements are already made on a regular basis at the GAW site on Ascension, so there is already technical staff available on the island for the infrequent maintenance of an ozone instrument. The island of Tristan da Cunha is the most remote

human settlement on Earth. While it does not host a GAW station, Tristan da Cunha hosts a Comprehensive Test Ban Treaty scientific station that is accompanied by some other instruments such as a Danish Meteorological Institute/Danish National Space Institute magnetometer. Closer to South America is the Brazilian island of Fernando de Noronha, which is a UNESCO World Heritage environmental preservation site 354 km off the coast of Brazil, but it is populated and has an airport.

The one oceanic area in the Northern Hemisphere that is notably lacking in observations and could provide a large incremental increase in the coverage of the Earth's surface based on Fig. 8 is the North Pacific near the Aleutian Islands. A site in the Aleutian Islands would have a footprint covering the Bering Sea and Kamchatka Peninsula. It could also potentially provide constraints on the large inter-model spread in Northern Hemisphere background marine environments and observe long-range transport of pollution events from Asia.

It appears that a relatively small number (10) of ozone instruments distributed on inhabited islands, many with pre-existing scientific research infrastructure, would provide a significant enhancement of the area of the world covered by ozone observations.

### 3.2 Continental sites

The ten terrestrial sites that provide the greatest additional area coverage are located in the Amazon basin, central Africa, northern Asia, and Australia (Fig. 10).

The Amazon and central African sites (Fig. 10a, c, e, h) are tropical forest regions with high biodiversity. These regions are also characterised by very high biogenic and biomass burning emissions and intensive photochemistry (Guenther et al., 2012; Giglio et al., 2013). They are also areas of high inter-model spread (Fig. 6) and high uncertainty in their future trends (Fig. 7) due to uncertainties in both emissions of ozone precursors and chemistry. All of these characteristics point to these regions as important locations for additional atmospheric monitoring. The footprint of Fig. 10e is near the WMO GAW global site of Arembe as well as the long-term ozonesonde site at Natal, Brazil. Near Fig. 10a is the Manaus, Brazil GAW contributing site.

The North African location (Fig. 10g) is very remote. The nearest WMO GAW global station at Assekrem, Algeria has been measuring ozone since the year 1997 but measures in a different meteorological regime. Setting up a measurement site in this region would be challenging.

Regions of northern Asia including Mongolia (Fig. 10d) and the Krasnoyarsk (Fig. 10f) and Kamchatka (Fig. 10b) territories of Russia would also represent large additions to the area covered by the ozone observing network. The regions are covered by the steppe and boreal forest biomes. These regions are broadly speaking rugged and remote, but there are existing WMO GAW regional stations in the region. On the Kamchatka Peninsula (Fig. 10b), the Petropavlovsk-Kamchatskiy World Ozone and Ultraviolet Radiation Data Centre (WOUDC) station measures total column ozone and solar radiation, as does the Tomsk site in Krasnoyarsk (Fig. 10f). In Mongolia near the location marked in Fig. 10d, there is a regional WMO GAW station at Ulaan Uul that presently collects greenhouse gas flask samples. Figure 10i is further west in Kazakhstan, near the Black Sea. There are several WOUDC stations, but none have reported ozone column data to the WMO in several years.

Finally, we find one location (Fig. 10j) in west Australia. Australia already has a network of ozone sites for air quality monitoring, but data is collected at the state level and is not readily accessible. That said, air quality networks focus on urban air pollution issues, and additional background sites in Australia beyond the WMO GAW site at Cape Grim, Tasmania are warranted and would bring global benefits. The nearest existing WMO GAW site is located at Darwin, but this is a coastal site that will capture more of the maritime conditions of the Timor Sea.

Unlike the marine sites, establishing ozone measurements at the terrestrial sites may be more problematic. Political and security difficulties will make countries like Zimbabwe, Niger, or the Democratic Republic of the Congo challenging locations to establish long term ozone monitoring sites. Remote locations in the Sahara or the Eurasian steppe would be logistically challenging. However, the addition of ozone measurements to sites with pre-

existing infrastructure, or the inclusion of measurements already being made into international databases would provide significant benefits for relatively little cost.

### 3.3 Impact on global coverage

The global impact of making surface ozone measurements at these additional twenty surface zone monitoring sites is shown in red in Fig. 11. The global area coverage would improve from 25 to 44 %, with 48 % of the land and 43 % of the ocean covered. Southern Hemisphere coverage is improved to 49 % surpassing that in the Northern Hemisphere (39 %). Coverage of South America would increase to 40 %, African coverage to 30 %, Asian coverage to 26 %, and Oceania to 30 %. Changes over North America and Antarctica are at the < 1 % level.

A limitation of our approach is that China and India would remain notably unobserved. The high concentrations of pollution (notably  $\text{NO}_x$ ) in these regions gives ozone a short lifetime and so any additional site in these regions adds little to the global coverage. However, given the variability in the modelled ozone for these regions and their probable trend over the next decades additional measurement and or reporting of existing measurements is critical. The challenge here is probably to get existing data into [publicly accessible](#) databases where they can be further used. [For example, China has an extensive air quality monitoring network where current data is publicly available online as Air Quality Index \(AQI\) values \(<http://aqicn.org/>, but there is no available archive of historical data or direct reporting of concentrations. Until these observations are generally available for scientific evaluation it is difficult to know where further observations capabilities are needed.](#)

## 4 Conclusions

We have investigated a dataset of readily available, long term surface network ozone data (Sofen et al., 2015) for its global coverage. This coverage can be interpreted in different ways depending upon the science/policy goal and we investigate the coverage from a range of perspectives.

Large countries such as China, India and Brazil are essentially un-represented in the Sofen et al. (2015) dataset. However, there are increasing efforts to make appropriate measurements in these regions. If these measurements were reported through to a national or international network and the data made available, they would fill a significant hole in the representation of global surface ozone. The Tropospheric Ozone Assessment Report project (<http://www.igacproject.org/TOAR>) is attempting improve access to existing measurements in these regions.

Our analysis shows that the most scientific benefit is probably accrued by making measurements in the forested tropical regions ~~of Southern~~ of South America, Africa, and Southeast Asia/Oceania. From a spatial perspective, and probably from a logistical perspective, large benefits would accrue from measurements on islands in the tropics and southern ocean with pre-existing scientific infrastructure. Adding the top ten maritime and terrestrial sites that contribute the largest additional area to the global coverage increases the fraction of the Earth covered by observing site footprints from 25 to 44 %.

We have provided a rather ~~straightforwards~~ straightforward analysis of the data that begins to provide a structure for the systematic expansion of the network of global surface ozone observations. However, more complex methodologies exist. Just as the scientific utility of new satellites observations are simulated before their launch (Zoogman et al., 2011) more complex methodologies could be applied to finding the optimal location for new surface ozone observations. However, it seems unlikely that there is much benefit to be gained from making these observations over North America, Europe or Antarctica and there is probably significant benefits in making these relatively cheap and easy observations virtually anywhere else in the world.

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**Table 1.** Summary of potential ozone sites.

Fig.	Lat.	Lon.	Area (10 <sup>6</sup> km <sup>2</sup> )	Nearby Islands or Biomes <sup>a</sup>	Country	Chem. Regimes <sup>b</sup>	ENSO	Inter-model Uncertainty <sup>c</sup>	Trends	
									RCP2.6 <sup>d</sup>	RCP8.5 <sup>e</sup>
9A	-47	1.25	17.3	Bouvet Is., S. Georgia & S. Sandwich Is.	Norway, UK	S. Ocean		4.8 (22.2%)	-2.7	4.3
9B	-45	76.25	16.8	Kerguelen Is.	France	S. Ocean		4.7 (20.7%)	-2.6	4.4
9C	-55	-136.25	11.6	Peter I Is., Scott Is.	Norway, N.Z.	S. Ocean		4.7 (21.8%)	-2.7	3.6
9D	-59	173.75	11.0	Macquarie Is.	Australia	S. Ocean, S. Subtrop. Oc.		4.6 (21.8%)	-2.6	3.9
9E	-11	-121.25	8.3	Pitcairn Is.	UK	Trop. Oc.	Yes	3.8 (20.1%)	-3.2	<b>0.5</b>
9F	-5	-151.25	6.6	Marquesas Is.	France	Trop. Oc.	Yes	4.2 (28.8%)	-3.7	<b>-0.6</b>
9G	-21	-86.25	6.5	Galapagos Is.	Ecuador	S. Subtrop. Oc.	Yes	4.1 (17.1%)	-3.4	3.5
9H	-27	-11.25	5.8	St. Helena, Tristan da Cunha	UK	S. Subtrop. Oc.		4.1 (16.9%)	-3.8	3.9
9I	51	168.75	4.8	Aleutian Is.	USA, Russia	N. Extratrop. Oc., Arctic		<b>8.4</b> (21.8%)	<b>-13.1</b>	<b>6.1</b>
9J	-3	-28.75	4.6	Ascension Is., Fernando de Noronha	UK, Brazil	S. Subtrop. Oc.		5.3 (22.1%)	-4.0	<b>0.6</b>
10A	-9	-58.75	4.4	TrMF	Brazil	Trop. Forest		4.7 (24.5%)	-4.6	<b>0.7</b>
10B	57	161.25	3.0	Tund	Russia	Boreal, Arctic		<b>7.7</b> (22.4%)	-10.3	4.2
10C	-17	28.75	2.9	TrG	Zimbabwe	Trop. Land		4.5 (14.9%)	-2.7	<b>7.0</b>
10D	43	108.75	2.4	Des, TeG, BoF, MnG	Mongolia	Boreal		6.6 (15.5%)	<b>-12.5</b>	4.3
10E	-7	-38.75	2.4	Des, TrG	Brazil	Trop. Land		3.8 (16.1%)	-4.9	1.4
10F	61	88.75	2.4	BoF, Tund	Russia	Boreal, Polluted		<b>7.7</b> (23.9%)	-9.2	3.5
10G	19	13.75	2.4	Des, TrG	Niger	Desert		4.3 (12.0%)	-9.6	3.8
10H	-3	26.25	2.3	TrMF, TrG	DR Congo	Trop. Forest		4.0 (15.2%)	<b>0.9</b>	4.9
10I	49	61.25	1.9	Des, TeG, BoF	Kazakhstan	Desert, Polluted		5.5 (15.3%)	-11.2	3.1
10J	-21	131.25	1.9	Des, TrG	Australia	Tropical land		4.8 (19.1%)	-4.8	2.2

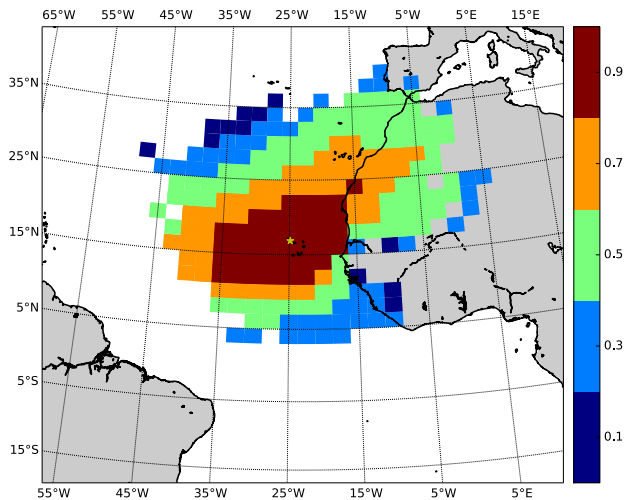
<sup>a</sup> Biomes are not specified for oceanic sites, as islands stations will primarily see marine air. The biomes listed include all the biomes covered in each footprint. Biome abbreviations refer to: Wat (Water), RoIc (Rock and Ice), Mang (Mangroves), Des (Deserts and xeric shrublands), Med (Mediterranean forests, woodlands and scrubs), Tund (Tundra), MnG (Montane grasslands and shrublands), FIG (Flooded grasslands and savannas), TeG (Temperate grasslands, savannas, and shrub-lands), TrG (Tropical and subtropical grasslands, savannas, and shrublands), BoF (Boreal forests/taiga), TeCF (Temperate conifer forests), TeBF (Temperate broadleaf and mixed forests), TrCF (Tropical and subtropical coniferous forests), TrBF (Tropical and subtropical dry broadleaf forests), TrMF (Tropical and subtropical moist broadleaf forests).

<sup>b</sup> The chemical regimes listed include all the chemical regimes covered in each footprint

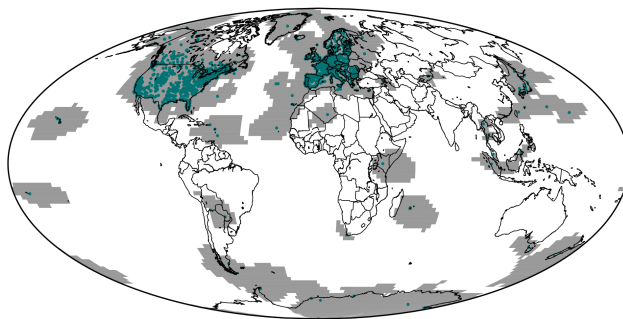
<sup>c</sup> The global area-weighted mean inter-model uncertainty is  $5.4 \pm 1.7$  ppb (20 %  $\pm$  15 %). Bolded values indicate sites where the inter-model variability is more than one s.d. above the global mean.

<sup>d</sup> The global area-weighted mean trend for RCP2.6 is  $-7.0 \pm 4.5$  ppb. Bolded values indicate sites where the local trend is more than one s.d. away from the global mean.

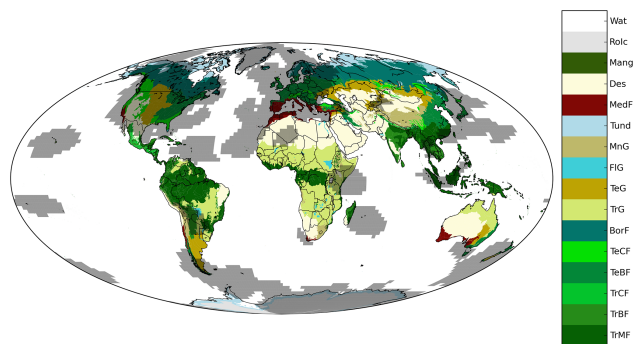
<sup>e</sup> The global area-weighted mean trend for RCP8.5 is  $3.2 \pm 2.1$  ppb. Bolded values indicate sites where the local trend is more than one s.d. away from the global mean.



**Figure 1.** Example of the site footprint for the Cape Verde Observatory (~~blue marker~~yellow star; 16° 51' 49" N, 24° 52' 2" W) derived using thresholds of  $R = 0.1$  to  $0.9$  in the spatial correlation of monthly anomalies in surface ozone from the GEOS-Chem model.

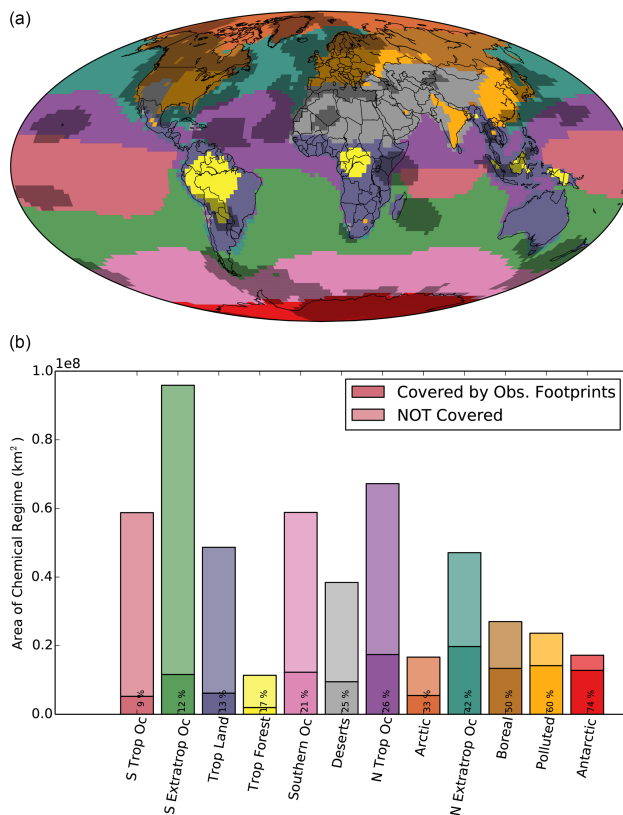


**Figure 2.** Map of surface ozone observational coverage based on the composite of the footprints for existing background ozone observing sites. Blue markers indicate site locations, and the grey areas their footprints.

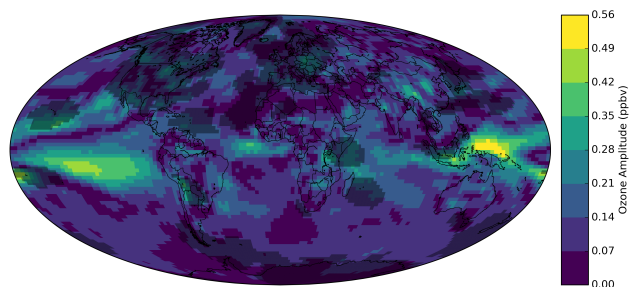


**Figure 3.** Map of terrestrial biomes (Olson and Dinerstein, 2002; The Nature Conservancy, 2012) shaded by ozone observational coverage. Dark regions of the biomes are areas that are covered by the footprints of ozone sites; light regions do not have ozone observations. Biome abbreviations refer to: Wat (Water), Rolc (Rock and Ice), Mang (Mangroves), Des (Deserts and xeric shrublands), Med (Mediterranean forests, woodlands and scrubs), Tund (Tundra), MnG (Montane grasslands and shrublands), FIG (Flooded grasslands and savannas), TeG (Temperate grasslands, savannas, and shrublands), TrG (Tropical and subtropical grasslands, savannas, and shrublands), BorF (Boreal forests/taiga), TeCF (Temperate conifer forests), TeBF (Temperate broadleaf and mixed forests), TrCF (Tropical and subtropical coniferous forests), TrBF (Tropical and subtropical dry broadleaf forests), TrMF (Tropical and subtropical moist broadleaf forests).

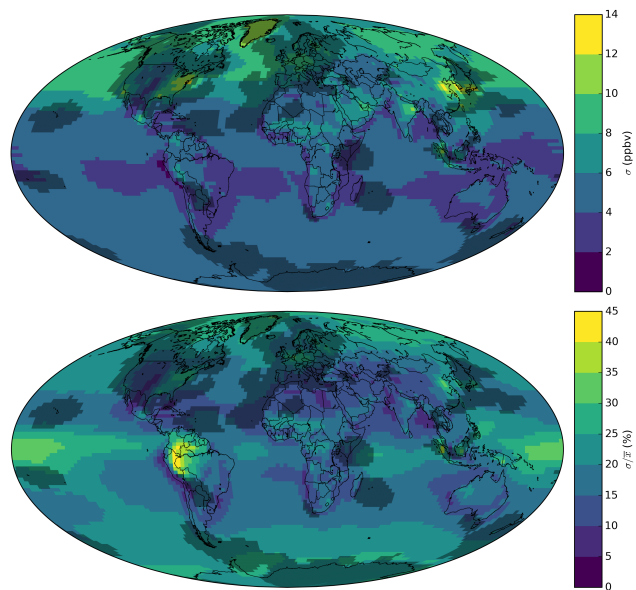




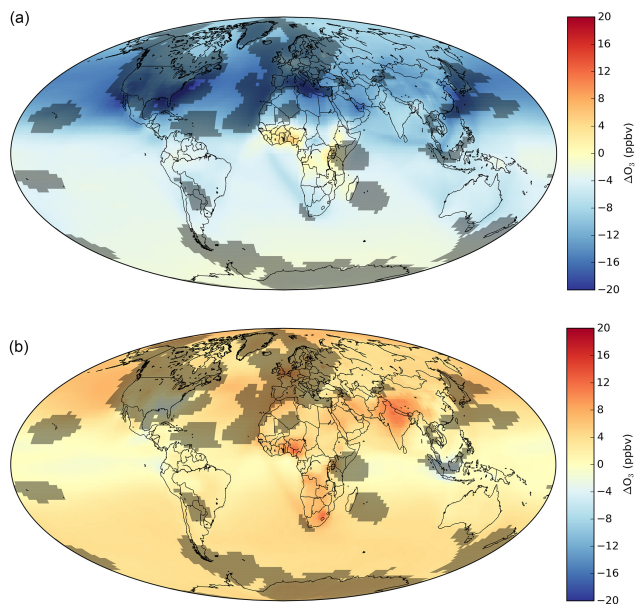
**Figure 4.** Chemical regimes defined by a cluster analysis of GEOS-Chem tracers. **(a)** shows a map of the chemical regimes, with shading indicating areas covered by the footprints of existing ozone sites. In **(b)**, the area of each chemical regime from **(a)** that are covered (dark) or are not covered (light) by the “footprints” of ozone observing sites. The bars are ordered by increasing fractional coverage shown above regime label.



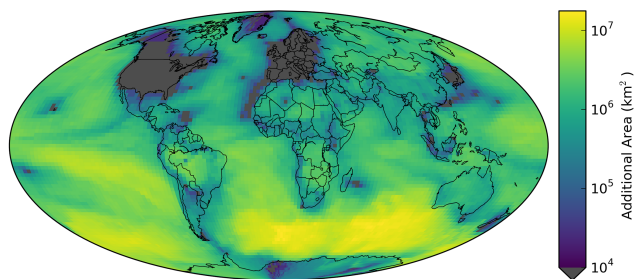
**Figure 5.** Amplitude of variability (from a Spectral/Fourier analysis) in surface ozone at the periodicity of ENSO (1403 days) in a free-running simulation using the GFDL chemistry-climate model.



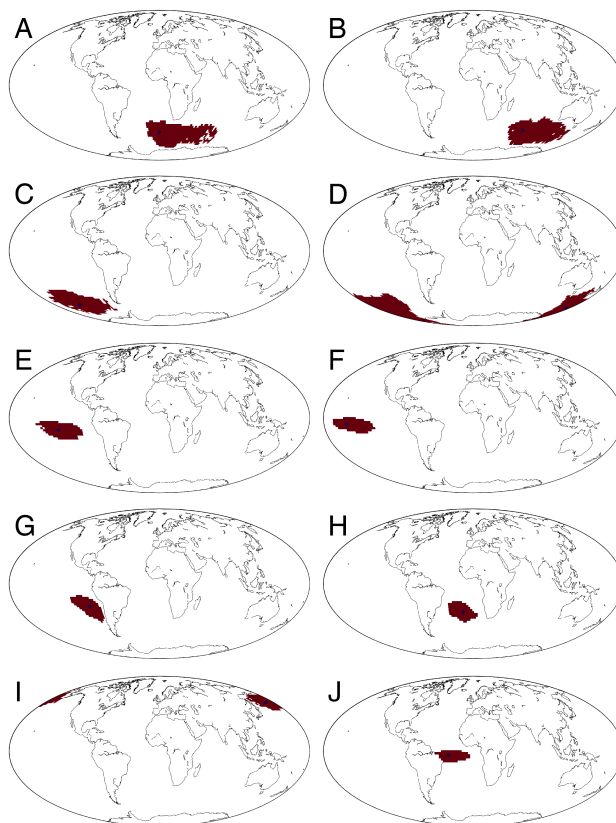
**Figure 6.** Map of the (a) absolute and (b) fractional standard deviation in annual mean surface ozone between ACCMIP models, following Young et al. (2013). Shaded regions are areas that are covered by the footprints of ozone sites; light regions do not have ozone observations.



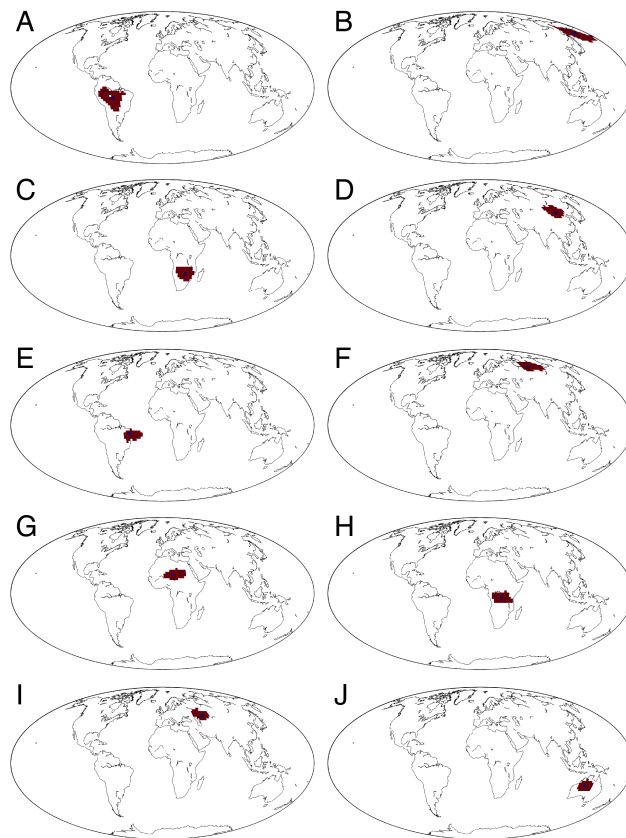
**Figure 7.** Maps showing the mean trend across ACCMIP models in annual mean surface ozone from 2005–2010 to 2095–2110 for scenarios **(a)** RCP2.6 and **(b)** RCP8.5, following Young et al. (2013). Shaded regions are areas that are covered by the footprints of ozone sites; light regions do not have ozone observations.



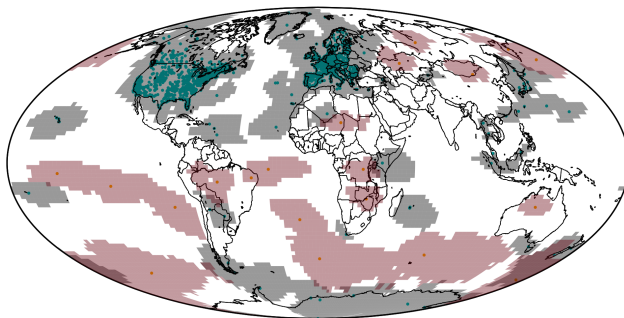
**Figure 8.** Additional area that a site in each grid box would contribute to the global ozone data coverage calculated based on footprints from GEOS-Chem.



**Figure 9.** Footprints for the top 10 grid boxes from distinct oceanic areas that will provide the greatest increase in the global coverage of ozone.



**Figure 10.** Footprints for the top 10 grid boxes from distinct terrestrial areas that will provide the greatest increase in the global coverage of ozone.



**Figure 11.** Map of surface ozone observational coverage as in Fig. 2 (blue dots indicate current measurement sites and grey areas their footprints) with the twenty additional sites and footprints from Figs. 9 and 10 added as red dots and pink areas to illustrate the global coverage if ozone measurements were made at these sites.