

**Table S1:** Information on the individual AOD, Sulphate, PM<sub>2.5</sub>, OC, N<sub>3</sub> and N<sub>50</sub> measurement data sets (campaigns and ground station networks) used in the constraint analysis in this study.

Observed Quantity	Name	Type	Period taken from	Region covered	Links and references
AOD (440 nm)	AERONET (Aerosol Robotic Network)	ground station	2005 – 2015	Worldwide	<a href="https://aeronet.gsfc.nasa.gov/">https://aeronet.gsfc.nasa.gov/</a>
Sulphate	IMPROVE (Interagency Monitoring of Protected Visual Environments)	ground station (202 stations)	2005-2009	North America (USA)	<a href="http://vista.cira.colostate.edu/Improve/">http://vista.cira.colostate.edu/Improve/</a>
Sulphate	EMEP (The European Monitoring and Evaluation Programme)	ground station (63 stations)	2005-2009	Europe	Tørseth et al. (2012); <a href="http://www.emep.int/">http://www.emep.int/</a> ; <a href="http://ebas.nilu.no/">http://ebas.nilu.no/</a>
Sulphate	EANET (Acid Deposition Monitoring Network in East Asia)	ground station (25 stations)	2005-2009	East Asia	<a href="https://www.eanet.asia/">https://www.eanet.asia/</a>
Sulphate	SORPES (Station for Observing Regional Processes of the Earth System)	ground station (1 station)	9 May 2014 – 30 April 2015	Nanjing, East China	Ding et al. (2016) Data obtained directly from the data contact: Prof. Aijun Ding, Nanjing University, China.
PM <sub>2.5</sub>	IMPROVE (Interagency Monitoring of Protected Visual Environments)	ground station (169 stations)	2008	North America (USA)	<a href="http://vista.cira.colostate.edu/Improve/">http://vista.cira.colostate.edu/Improve/</a>
PM <sub>2.5</sub>	WMO GAW–WDCA (World Meteorological Organisation’s Global Atmosphere Watch – World Data Centre for Aerosols) data for Europe	ground station (41 stations)	2007-2010	Europe	<a href="https://www.gaw-wdca.org">https://www.gaw-wdca.org</a> <a href="http://ebas.nilu.no/">http://ebas.nilu.no/</a> ; Tørseth et al. (2012);
PM <sub>2.5</sub>	A-PAD (Asia-Pacific Aerosol Database)	ground station (18 stations)	2008	Asia	Atanacio et al. (2016)
PM <sub>2.5</sub>	NAPS (The Canadian National Air Pollution Surveillance Program)	ground station (113 stations)	2008	Canada	Galarneau et al. (2016); <a href="http://maps-cartes.ec.gc.ca/rnsps-naps/data.aspx?lang=en">http://maps-cartes.ec.gc.ca/rnsps-naps/data.aspx?lang=en</a>
PM <sub>2.5</sub>	ANSTO (Australian Nuclear Science and Technology Organisation) stations	ground station (8 stations)	2008	Australia (NE coast)	Cohen and Atanacio (2015)
PM <sub>2.5</sub>	South American measurement station data.	ground station (4 sites)	Alta Floresta: 2002-2005 Manaus: 2008-2011 Porto Vehlo: 2009-2012 Santarem: 2000-2007	South America	Artaxo et al. (2013); data as used in Reddington et al. (2018). Data obtained directly from the data contact: Paulo Artaxo, University of São Paulo, Brazil, as part of the GASSP project (Reddington et al., 2017).

PM <sub>2.5</sub>	Taiwan measurement station data.	ground station (7 stations)	2008	Taiwan	Data obtained from the scientific literature: Fang and Chang (2010).
PM <sub>2.5</sub>	South Africa measurement station data	ground station (2 sites; temporary measurement trailer)	2007 – 2008	South Africa	Vakkari et al. (2013)  The data was provided courtesy of the project principal investigator: V. Vakkari, Univ. of Helsinki, Finland.
PM <sub>2.5</sub>	SORPES (Station for Observing Regional Processes of the Earth System)	ground station (1 station)	2012	Nanjing, East China	Ding et al. (2016)  Data obtained directly from the data contact: Prof. Aijun Ding, Nanjing University, China.
OC	VOCALS (The VAMOS Ocean-Cloud-Atmosphere-Land Study)	ship and aircraft campaign	October – December 2008	South East Pacific	Allen et al. (2011); Hawkins et al. (2010); Wood et al. (2011)
OC	CalNex (The 2010 California Research at the Nexus of Air Quality and Climate Change)	ship campaign	May – June 2010	California	Ryerson et al. (2013)
OC	WACS (The Western Atlantic Climate Study)	ship campaign	August 2012 May 2014	Western Atlantic	Quinn et al. (2014)
OC	ICEALOT (The International Chemistry Experiment in the Arctic Lower Troposphere)	ship campaign	19 March – 24 April 2008	Sub-Arctic Greenland Sea (region between Iceland and Svalbard)	Zhang et al. (2014); Frossard et al. (2011)
OC	DYNAMO (Dynamics of the Madden-Julian Oscillation)	ship campaign	October – December 2011	Indian Ocean (nr Thailand)	DeWitt et al. (2013)
OC	NEAQS-2004 (The New England Air Quality Study of 2004)	ship campaign	June – August 2004	Western Atlantic (from Boston, USA)	Quinn et al. (2006); Wang et al. (2007)
OC	TEXAQS06 (The Texas Air Quality study)	ship campaign	27 July – 11 September 2006	Gulf of Mexico (off Texas coastline)	Bates et al. (2008)
OC	RHaMBLe (Reactive Halogens in the Marine Boundary Layer)	ship campaign	May – June 2007	North Atlantic	Allan et al. (2009)  Data obtained directly from the data contact:

					James Allan, University of Manchester, UK.
OC	ACCACIA (Aerosol-Cloud Coupling And Climate Interactions in the Arctic)	ship campaign	March – April and July 2013	Arctic	Allan et al. (2015) Data obtained directly from the data contact: James Allan, University of Manchester, UK.
OC	AMS Global Database	ground station (41 stations)	2001 - 2010	Worldwide	Zhang et al. References for the individual sites used are provided in the acknowledgement in Appendix B of the paper.
OC	ACTRIS (Aerosols, Clouds, and Trace Gases Research Infrastructure)	ground station (10 stations)	2006 – 2009	Europe	<a href="https://www.actris.eu/">https://www.actris.eu/</a> ; <a href="http://ebas.nilu.no/">http://ebas.nilu.no/</a>
OC	SORPES (Station for Observing Regional Processes of the Earth System)	ground station (1 station)	9 May 2014 – 30 April 2015	Nanjing, East China	Ding et al. (2016) Data obtained directly from the data contact: Prof. Aijun Ding, Nanjing University, China.
OC	AMF Stations	ground station (3 stations)	2011 – 2015	Eastern North Atlantic, Southern Great Plains (USA), Cape Cod (USA)	Atmospheric Radiation Measurement (ARM) user facility (2014a, 2014b)
OC	Chilbolton	ground station (1 station)	7 January – 19 March 2009	England	Crippa et al. (2014) Data obtained directly from the data contact: Gerard Capes, University of Manchester, UK.
OC	COPS	ground station (1 station)	23 June – 27 July 2007	Hornisgrinde, Germany	Irwin et al. (2010); Jones et al. (2011) Data obtained directly from the data contact: Jonathan Crosier, University of Manchester, UK.
OC	Holme Moss	ground station (1 station)	7 November – 4 December 2006	England	Liu et al. (2011) Data obtained directly from the data contact: Dantong Liu, University of Manchester, UK.
OC	OP3	ground station (1 station)	24 June – 24 July 2008	South-east Asia	Hewitt et al. (2010) Data obtained directly from the data contact: James Allan, University of Manchester, UK.

OC	MC4	ground station (1 station)	June – July 2011	Weybourne, England	Liu et al. (2013) Data obtained directly from the data contact: Dantong Liu, University of Manchester, UK.
N3	ACTRIS (Aerosols, Clouds, and Trace Gases Research Infrastructure)	ground station (15 stations)	1997 – 2015	12 European sites; 1 Arctic, 1 Antarctic and 1 Indian site	Asmi et al. (2013) <a href="https://www.actris.eu/">https://www.actris.eu/</a> ; <a href="http://ebas.nilu.no/">http://ebas.nilu.no/</a>
N50	ACE 1(The First Aerosol Characterization experiment)	Ship and aircraft campaign	October – November 1995 (ship) November – December 1995 (aircraft)	Southern and Pacific Oceans	Bates et al. (1998); Clarke et al. (1998)
N50	VOCALS (The VAMOS Ocean-Cloud- Atmosphere-Land Study)	ship and aircraft campaign	October – December 2008	South East Pacific	Allen et al. (2011); Hawkins et al. (2010); Wood et al. (2011)
N50	DOE ARM MAGIC (Marine ARM GPCI Investigation of Clouds)	ship campaign	September 2012 – October 2013 (excluding January – April 2013)	North Pacific	Lewis and Teixeira (2015) <a href="https://www.bnl.gov/envsci/cloud/campaigns/MAGIC/">https://www.bnl.gov/envsci/cloud/campaigns/MAGIC/</a>
N50	CalNex (The 2010 California Research at the Nexus of Air Quality and Climate Change)	ship campaign	May – June 2010	California	Ryerson et al. (2013)
N50	WACS (The Western Atlantic Climate Study)	ship campaign	August 2012	Western Atlantic	Quinn et al. (2014)
N50	NEAQS – 2002 (The New England Air Quality Study of 2002)	ship campaign	July – August 2002	Western Atlantic (from Boston, USA)	Bates et al. (2005); Quinn and Bates (2005) <a href="https://www.esrl.noaa.gov/csd/projects/neaqs/">https://www.esrl.noaa.gov/csd/projects/neaqs/</a>
N50	ARCTAS (The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites campaign)	aircraft campaign	April 2008 June – July 2008	North America; Canadian / Alaskan Arctic	McNaughton et al. (2011)
N50	ASCOS (The Arctic Summer Cloud Ocean Study)	ship campaign	August – September 2008	Arctic 87°N	Tjernström et al. (2014); Heintzenberg and Leck (2012); Chang et al. (2011)

N50	ICEALOT (The International Chemistry Experiment in the Arctic Lower Troposphere)	ship campaign	19 March – 24 April 2008	Sub-Arctic Greenland Sea (region between Iceland and Svalbard)	Frossard et al. (2011)
N50	Aerosols99	ship campaign	January – February 1999	North and South Atlantic	Bates et al. (2001) This campaign and the INDOEX campaign (below) together were a transitional cruise through the North and South Atlantic (Aerosols99) and an intensive scientific campaign in the Indian ocean (INDOEX).
N50	DYNAMO (Dynamics of the Madden-Julian Oscillation)	ship campaign	October – December 2011	Indian Ocean	DeWitt et al. (2013)
N50	INDOEX (Indian Ocean Experiment)	ship campaign	February – March 1999	Indian Ocean	Quinn and Bates (2005); Ramanathan et al. (2001)
N50	PEM-Tropics (A)	aircraft campaign	August – September 1996	Remote tropical Pacific	Fenn et al. (1999)
N50	PEM-Tropics (B)	aircraft campaign	March – April 1999	Remote tropical Pacific	Raper et al. (2001)
N50	PASE (The Pacific Atmospheric Sulphur Experiment)	aircraft campaign	August 2007	Pacific (East of Christmas Islands)	Hudson and Noble (2009)
N50	NAURU99	ship campaign	23 June – 17 July 1999	Indonesia (tropical western Pacific)	<a href="https://www.esrl.noaa.gov/psd/psd3/air-sea/nauru99/">https://www.esrl.noaa.gov/psd/psd3/air-sea/nauru99/</a> Long and McFarlane (2012)
N50	ACE-ASIA (The Asian Pacific Regional Aerosol Characterization Experiment)	ship campaign	March – April 2001	North-eastern Pacific near Japan	Bates et al. (2004); Huebert et al. (2003)
N50	NEAQS-2004 (The New England Air Quality Study of 2004)	ship campaign	June – August 2004	Western Atlantic (from Boston, USA)	Quinn et al. (2006); Wang et al. (2007)
N50	TEXAQS06 (The Texas Air Quality study)	ship campaign	27 July – 11 September 2006	Gulf of Mexico (off Texas coastline)	Bates et al. (2008)

N50	Alert measurement station	ground station (1 station)	2013	Canada	Leaitch et al. (2013)
N50	Whistler Peak measurement station	ground station (1 station)	2011	Canada	Takahama et al. (2011)
N50	Egbert measurement station	ground station (1 station)	2008	Canada	Jeong et al. (2010); Pierce et al. (2014)
N50	South Africa measurement station data	ground station (temporary measurement trailer)	2007 – 2008	South Africa	Vakkari et al. (2013) The data was provided courtesy of the project principal investigator: V. Vakkari, Univ. of Helsinki, Finland.
N50	Tiksi Polar measurement station	ground station (1 station)	2013	Russian Arctic	Asmi et al. (2016) Obtained via the online data portal: IASOA (International Arctic Systems for Observing the Atmosphere).
N50	Gual Pahari measurement station	ground station (1 station)	2008	Northern India	Hyvärinen et al. (2010) Obtained via the online data portal: WDCA (World Data Centre for Aerosol).
N50	Troll measurement station	ground station (1 station)	2008	Antarctic	Fiebig et al. (2009) Obtained via the online data portal: WDCA (World Data Centre for Aerosol).
N50	European measurement stations	ground station (26 stations)	2008	Europe	Asmi et al. (2011) Measurements were obtained via the EBAS data portal: <a href="http://ebas.nilu.no/">http://ebas.nilu.no/</a> and processed by Asmi et al. (2011) to N50 concentration.

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