

Springer Theses

Recognizing Outstanding Ph.D. Research

Catherine E. Scott

The Biogeochemical Impacts of Forests and the Implications for Climate Change Mitigation

 Springer

Springer Theses

Recognizing Outstanding Ph.D. Research

Aims and Scope

The series “Springer Theses” brings together a selection of the very best Ph.D. theses from around the world and across the physical sciences. Nominated and endorsed by two recognized specialists, each published volume has been selected for its scientific excellence and the high impact of its contents for the pertinent field of research. For greater accessibility to non-specialists, the published versions include an extended introduction, as well as a foreword by the student’s supervisor explaining the special relevance of the work for the field. As a whole, the series will provide a valuable resource both for newcomers to the research fields described, and for other scientists seeking detailed background information on special questions. Finally, it provides an accredited documentation of the valuable contributions made by today’s younger generation of scientists.

Theses are accepted into the series by invited nomination only and must fulfill all of the following criteria

- They must be written in good English.
- The topic should fall within the confines of Chemistry, Physics, Earth Sciences, Engineering and related interdisciplinary fields such as Materials, Nanoscience, Chemical Engineering, Complex Systems and Biophysics.
- The work reported in the thesis must represent a significant scientific advance.
- If the thesis includes previously published material, permission to reproduce this must be gained from the respective copyright holder.
- They must have been examined and passed during the 12 months prior to nomination.
- Each thesis should include a foreword by the supervisor outlining the significance of its content.
- The theses should have a clearly defined structure including an introduction accessible to scientists not expert in that particular field.

More information about this series at <http://www.springer.com/series/8790>

Catherine E. Scott

The Biogeochemical Impacts of Forests and the Implications for Climate Change Mitigation

Doctoral Thesis accepted by
the University of Leeds, UK

Author

Dr. Catherine E. Scott
School of Earth and Environment
University of Leeds
Leeds
UK

Supervisors

Dr. Dominick Spracklen
Prof. Piers Forster
Prof. Kenneth Carslaw
School of Earth and Environment
University of Leeds
Leeds
UK

ISSN 2190-5053

ISBN 978-3-319-07850-2

DOI 10.1007/978-3-319-07851-9

ISSN 2190-5061 (electronic)

ISBN 978-3-319-07851-9 (eBook)

Library of Congress Control Number: 2014943244

Springer Cham Heidelberg New York Dordrecht London

© Springer International Publishing Switzerland 2014

This work is subject to copyright. All rights are reserved by the Publisher, whether the whole or part of the material is concerned, specifically the rights of translation, reprinting, reuse of illustrations, recitation, broadcasting, reproduction on microfilms or in any other physical way, and transmission or information storage and retrieval, electronic adaptation, computer software, or by similar or dissimilar methodology now known or hereafter developed. Exempted from this legal reservation are brief excerpts in connection with reviews or scholarly analysis or material supplied specifically for the purpose of being entered and executed on a computer system, for exclusive use by the purchaser of the work. Duplication of this publication or parts thereof is permitted only under the provisions of the Copyright Law of the Publisher's location, in its current version, and permission for use must always be obtained from Springer. Permissions for use may be obtained through RightsLink at the Copyright Clearance Center. Violations are liable to prosecution under the respective Copyright Law. The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.

While the advice and information in this book are believed to be true and accurate at the date of publication, neither the authors nor the editors nor the publisher can accept any legal responsibility for any errors or omissions that may be made. The publisher makes no warranty, express or implied, with respect to the material contained herein.

Printed on acid-free paper

Springer is part of Springer Science+Business Media (www.springer.com)

Parts of this thesis have been published in the following journal articles:

C. E. Scott, A. Rap, D. V. Spracklen, P. M. Forster, K. S. Carslaw, G. W. Mann, K. J. Pringle, N. Kivekäs, M. Kulmala, H. Lihavainen and P. Tunved (2014) “The Direct and Indirect Radiative Effects of Biogenic Secondary Organic Aerosol”, *Atmospheric Chemistry and Physics*, **14**, 447–470.

F. Riccobono, S. Schobesberger, C. E. Scott *et al.*, (2014) “Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles”, *Science*, **344** (6185), 717–721.

C. E. Scott, D. V. Spracklen, J. R. Pierce, I. Riipinen, S. D. D’Andrea, A. Rap, K. S. Carslaw, P. M. Forster, M. Kulmala, G. W. Mann, K. Pringle (2014) “Impact of partitioning approach on the indirect radiative effect of biogenic secondary organic aerosol”, (in prep).

Supervisor's Foreword

Forests and vegetation emit biogenic volatile organic compounds (BVOCs) into the atmosphere which, once oxidised, can partition into the particle phase, forming secondary organic aerosol (SOA). This thesis reports a unique and comprehensive analysis of the impact of BVOC emissions on atmospheric aerosol and climate. A state-of-the-art global aerosol microphysics model is used to make the first detailed assessment of the impact of BVOC emissions on aerosol microphysical properties, improving our understanding of the role of these emissions in controlling the Earth's climate.

Laboratory experiments, ambient atmospheric observations and model simulations are combined to provide new evidence that the oxidation products of BVOCs participate in the first steps of particle formation. This finding means that BVOCs have a larger impact on climate than previously thought, with the global radiative effect of biogenic SOA estimated to be half the net anthropogenic radiative forcing of climate. The thesis also reports on the implications for the climate impact of forests. Accounting for the climate impacts of SOA, alongside the carbon cycle and surface albedo effects that have been studied in previous work, increases the total warming effect of global deforestation by 21 %. The thesis suggests that deforestation warms climate more than previously thought.

Leeds, UK, June 2014

Dr. Dominick Spracklen

Acknowledgments

Firstly I would like to thank my supervisors: Piers Forster, Dom Spracklen and Ken Carslaw. Without their guidance, advice and support over the past four years, this work, and the production of this thesis would not have been possible. I am extremely grateful for the opportunities they have given me, and the enthusiasm with which they have provided supervision.

Secondly, I would like to thank Alexandru Rap for calculating the direct radiative forcings presented in Chap. 4, Jeff Pierce for conducting the TOMAS simulations discussed in Chap. 5 and Steve Arnold for running the Community Land Model used in Chap. 6. I would also like to thank Francesco Riccobono and the “CLOUD” team for providing the mechanism for new particle formation described in Chap. 3. For providing the measurement data used in Chap. 3, I would like to thank: Markku Kulmala, Pasi Aalto, Peter Tunved, Niku Kivekas and Heikki Lihavainen. I would also like to thank Ilona Riipinen and Steve D’Andrea with whom I have collaborated. Also thanks to Richard Rigby, without whose tireless IT support, this work would definitely not have been possible!

During the course of my Ph.D. I have very much valued being part of three great research groups: The Physical Climate Change group, the Aerosol Modelling group and the Biosphere-Atmosphere Group, and would like to thank all members for making me feel very welcome. Deserving of a particular mention for their help and advice over the years are: Anja, Carly, Annabel, Eimear, Alex, Matt, Helen, Tom, Kirsty, Graham, Lindsay, Jo, Steve P., Lawrence, Jayne, Julia, Neil, Susi, Sarah L., Ros, Mike and Sarah M.

For supporting me over the past four years, I would like to thank my Mum, Dad and Sally; Chris, Adèle and Wayne (and Myla!); Erin, Zarashpe, Ed, Tim, Rosey, Susie, Louisa and Al; Vicky, Amy, my fellow 2009–2013 Ph.D. gang: Amber, Jenn, Dave, Bradley and James; and last but definitely not least, thank you to Ross for everything!

Contents

1	Introduction	1
1.1	The Climate System	1
1.2	Forests and Their Impact on the Climate	2
1.2.1	Historical Changes to Forest Distribution	3
1.2.2	Role of Forests in the Carbon Cycle	5
1.2.3	Biogeophysical Impacts of Forests	5
1.2.4	An Additional Biogeochemical Impact of Forests?	7
1.3	Anthropogenic Climate Change	18
1.3.1	Drivers of Climate Change	18
1.3.2	Climate Change Mitigation	20
1.4	Aims of Thesis	22
	References	23
2	Model Description	35
2.1	Introduction to GLOMAP	35
2.1.1	Representation of the Aerosol Size Distribution	35
2.1.2	Gas-Phase Emissions and Processes	38
2.1.3	Primary Particulate Emissions	40
2.1.4	Microphysical Processes	41
2.1.5	Meteorological Conditions	46
2.2	Calculation of Cloud Condensation Nuclei Concentrations	46
2.3	Suitability of GLOMAP-Mode for This Work	47
2.4	Comparison to Observations	47
	References	48
3	The Impact of Biogenic SOA on Particle and Cloud Condensation Nuclei Concentration	53
3.1	Introduction	53
3.2	Experimental Design	53
3.2.1	Yield	53
3.2.2	New Particle Formation	55

3.2.3	Characteristics of Primary Carbonaceous Emissions	57
3.2.4	Presence of Anthropogenic Emissions	58
3.3	Results	58
3.3.1	Changes to Total Particle Concentration	58
3.3.2	Changes to Cloud Condensation Nuclei Concentrations	59
3.3.3	Sensitivity to New Particle Formation	62
3.4	Comparison to Observations	63
3.4.1	Seasonal Cycle at Forested Sites	63
3.4.2	Seasonal Cycle in Total Particle Concentration	65
3.4.3	Cloud Condensation Nuclei Concentrations	69
3.5	Summary and Conclusions	70
	References	71
4	The Radiative Impact of Biogenic SOA	75
4.1	Introduction	75
4.2	Experimental Setup	75
4.2.1	The Edwards–Slingo Radiative Transfer Model	75
4.2.2	Direct Radiative Effect	76
4.2.3	Aerosol Indirect Effect	76
4.3	Results	78
4.3.1	Direct Radiative Effect	78
4.3.2	First Aerosol Indirect Radiative Effect	79
4.4	Sensitivity to Anthropogenic Emissions	85
4.5	Summary and Conclusions	88
	References	90
5	The Impact of Volatility Treatment on the Radiative Effect of Biogenic SOA	93
5.1	Introduction	93
5.1.1	The Volatility Treatment of SOA	93
5.2	Experimental Setup	94
5.3	Results	96
5.3.1	Changes to Cloud Droplet Number Concentration	97
5.3.2	First Aerosol Indirect Effect	100
5.4	Summary and Conclusions	103
	References	105
6	The Radiative Effects of Deforestation	107
6.1	Introduction	107
6.2	Experimental Setup	107
6.2.1	The Community Land Model and MEGAN	107
6.2.2	Changes to GLOMAP-Mode Model Setup	113
6.2.3	Deforestation Experiments	114

6.3	Results	116
6.3.1	First Aerosol Indirect Effect	116
6.3.2	Other Radiative Effects of Forests	118
6.4	Summary and Conclusions	121
	References	123
7	Conclusions, Implications and Further Work	125
7.1	Summary of Results	125
7.1.1	Implications for Understanding and Mitigating Future Climate Change	128
7.2	Further Work	129
7.2.1	Secondary Organic Aerosol in GLOMAP	130
7.2.2	Dry Deposition in GLOMAP	130
7.2.3	Tropospheric Chemistry	130
	References	131

Abbreviations and Acronyms

AIE	Aerosol Indirect Effect
AOGCM	Atmosphere–Ocean General Circulation Model
BHN	Binary Homogeneous Nucleation
BLN	Boundary Layer Nucleation
BVOC	Biogenic Volatile Organic Compound
CCN	Cloud Condensation Nuclei
CDNC	Cloud Droplet Number Concentration
CLM	Community Land Model
CO _{2e}	Carbon Dioxide Equivalent
DRE	Direct Radiative Effect
ECS	Equilibrium Climate Sensitivity
E–S	Edwards–Slingo Radiative Transfer Model
ESM	Earth System Model
FAO	Food and Agriculture Organisation of the United Nations
GCM	General Circulation Model
GFED	Global Fire Emissions Database
GHG	Greenhouse Gas
GLOMAP	Global Model of Aerosol Processes
IPCC	Intergovernmental Panel on Climate Change
LW	Longwave
MEGAN	Model of Emissions of Gases and Aerosol from Nature
ppbv	Parts per billion by volume
pptv	Parts per trillion by volume
RCP	Representative Concentration Pathway
RE	Radiative Effect
RF	Radiative Forcing
SMPS	Scanning Mobility Particle Sizer
SOA	Secondary Organic Aerosol
SRES	Special Report on Emission Scenarios
SS	Supersaturation
SS _{max}	Maximum Supersaturation

SW	Shortwave
TOA	Top of the Atmosphere
UNFCCC	United Nations Framework Convention on Climate Change

Chapter 1

Introduction

Vegetation emits biogenic volatile organic compounds (BVOCs) into the atmosphere which, once oxidised, may partition into the particle-phase forming secondary organic aerosol (SOA). In this thesis, the climatic impacts of biogenic SOA are quantified, using a detailed global aerosol microphysics model, and the sensitivity of these radiative effects to the representation of various atmospheric processes is examined.

The radiative effects of biogenic SOA have implications for the climatic impact of forests and any changes to their distribution. In this thesis, simple deforestation scenarios are used to quantify the radiative effects of potential changes to the magnitude and distribution of biogenic SOA production.

1.1 The Climate System

The energy balance of the Earth system is governed by fluxes of incoming and outgoing radiation. Shortwave (SW) solar radiation reaching the Earth system is either reflected or absorbed, as dictated by the nature of the surface and composition of the atmosphere. Figure 1.1 depicts the estimated global average energy flows within the Earth system and indicates that the atmosphere exerts considerable control over how much incoming radiation reaches the surface.

Almost half of the incoming solar radiation is absorbed at the Earth's surface and re-emitted at longer wavelengths (LW); the Earth system behaves almost as a black body with a total emissive power (σT^4) proportional to the fourth power of its temperature, T , where σ is the Stefan-Boltzmann constant. In its equilibrium state, the amount of energy entering the Earth system is equal to that which leaves. If LW radiation is prevented from leaving, the system shifts in order to maintain the required outgoing flux, with an increase in emissive temperature. As such, changes to atmospheric composition, levels of incoming SW radiation, and

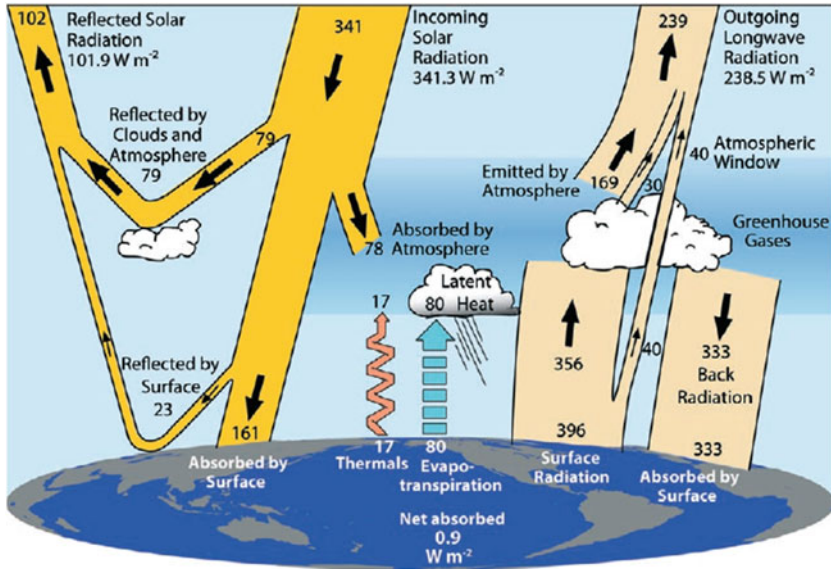


Fig. 1.1 Schematic of the estimated global average energy budget (values in W m^{-2}) for March 2000 to May 2004; reproduced from Trenberth et al. [225]

properties of the Earth's surface that initiate a radiative imbalance, can alter the temperature.

A radiative forcing (RF) may be assigned to physical factors on the basis of their ability to perturb the Earth's radiative balance, over a given time period. The RF is most commonly defined as the net change in radiative flux (i.e. downwards minus upwards) at the tropopause, after allowing for stratospheric temperatures to readjust to radiative equilibrium but with surface and tropospheric temperatures and state held fixed at their unperturbed values [58, 82, 189]. As the surface and troposphere are strongly coupled by convective heat transfer, there is a direct relationship between the RF across the tropopause and the global mean equilibrium temperature change at the Earth's surface (ΔT_s), such that $\Delta T_s = \lambda \text{ RF}$ [189], where λ represents the *climate sensitivity parameter* (in $\text{K W}^{-1} \text{m}^2$). However, the relationship between RF and transient climate change is not straightforward, and consideration must be given to the temporal and spatial variability of the forcing agent.

1.2 Forests and Their Impact on the Climate

Approximately 31 % of the Earth's land area (4.03 billion hectares) is presently covered by forest (Fig. 1.2); one third of this being undisturbed primary forest [55]. More than half of the world's current forest cover lies within just five countries: The Russian Federation, Brazil, Canada, the United States of America and China [55].