Speeding up the Atmosphere

Experimental oxidation studies of ambient and laboratory aerosols using a flow reactor

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Abstract

Apart from gases, the air we breath consist of tiny, so called, aerosol particles. A cubic metre of air in a relatively clean environment can consist of several billion aerosol particles. The impact of these particles on human health and on climate is significant. According to WHO, particles affect more humans than any other pollutant, and is closely related to mortality and morbidity. Further, it has been estimated that anthropogenic particles have cooled the climate, masking up to 50% of the warming caused by greenhouse gases. A large portion of the smallest particles consists of organic compounds, much of which is formed by atmospheric oxidation reactions. This material is called secondary organic aerosol (SOA). In this thesis, submicron secondary aerosol particles have been investigated using an oxidation flow reactor (OFR). Inside the reactor, large concentrations of oxidants produce secondary particulate material in a matter of minutes, thereby speeding up the naturally occuring atmospheric processes.

In two laboratory studies, we have investigated the effect of mixing anthropogenic primary and secondary particles with biogenic SOA. In line with expectations, the anthropogenic and biogenic organic precursors mixed, and, in a non-linear way, formed more particulate mass than would otherwise be the case. Further, the effect of wet anthropogenic salt particles on SOA formation was investigated. The produced SOA mass in the presence of wet particles was significantly higher than if the particles were dry. This effect is believed to be very important in the atmosphere since water is always present. In both these studies it was shown that the SOA mass formed in OFRs at low particle mass concentrations, is underestimated due to the limited time for condensation of vapours in the reactor.

In a separate study, the SOA formation from biomass burning was investigated. Biogenic SOA dominates on a global scale, but very high concentrations are only formed in the proximity of anthropogenic sources. It was shown that SOA formation from the emissions of a modern wood stove can be large enough to dominate over the primary particle emissions. To estimate the secondary particle formation potential of ambient air, two field studies were performed. In both studies, the simulated atmospheric processing of the background air did not produce much secondary particle mass. From this, it can be concluded that the chosen measurement sites were relatively clean, but the results also point to the efficiency of atmospheric processing. However, in one of the studies, targeting ship emissions at a coastal site, plumes of secondary material, of the same magnitude as the background aerosol particle concentration, was formed. This demonstrates the importance of considering atmospheric processing and the advantage of using OFRs in field studies.

The relative importance of SOA in the atmospheric aerosol is believed to increase in the future. Due to complex feedbacks and the many variables affecting SOA formation, it is difficult to parameterize in a simple enough manner fit for global models. Both detailed and large-scale processes needs further investigation to improve estimations of SOA radiative forcing and the anthropogenic effect on biogenic SOA. The popularity of oxidation flow reactors in SOA research is likely to increase. The five original research manuscripts included in this thesis contributes specifically to the OFR research community, but also to improved understanding of SOA formation in the the anthropocene in general.

Key words: Secondary organic aerosols, oxidation flow reactor, volatile organic compounds, climate

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Experimental oxidation studies of ambient and laboratory aerosols using a flow reactor

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Coverphoto shows the thin line of Earth's atmosphere and the settling sun, taken by the crew of the International Space Station. Image credit: NASA

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TO THE MOST SACRED MAJESTY, THE KING

Sir,

One day, while I was walking in your Majesty's palace, where I sometimes come to enjoy the sight of your magnificent presence, I saw a ghastly billow of smoke coming from one or two tunnels between Northumberland House and Scotland Yard.

(…)

In this short text I'd like to suggest a means of removing this nauseating smoke. This will make your palace and this city one of the sweetest and most splendid places to live in the world and can be done with almost no cost to you. If the plantations around the town are improved, they will produce greenery which will lend its wonderful scent to your palaces and make them seem like they have been magically transported to the happy parts of the Arab world which lie amongst shrubbery and precious spices.

(…)

I shall go into greater detail about this later in these pages if God permits me to finish them.

-John Evelyn (Fumifugium or The inconveniencie of the aer and smoak of London dissipated together with some remedies, 1661)

Contents

Papers included in this thesis

I. **Secondary organic aerosol from VOC mixtures in an oxidation flow reactor.** Ahlberg, E., Falk, J., Eriksson, A. C., Holst, T., Brune, W. H., Kristensson, A., Roldin, P. and Svenningsson, B.

Atmospheric Environment, 161, 210, 2017. (published under an open access license)

II. **Effect of salt seed particle surface area, composition and phase on secondary organic aerosol mass yields in oxidation flow reactors.** Ahlberg, E., Eriksson, A. C., Brune, W. H., Roldin, P. and Svenningsson, B.

Manuscript

III. **Impacts of combustion conditions and photochemical processing on the light absorption of biomass combustion.** Martinsson, J., Eriksson, A. C., Nielsen, I. E., Malmborg, V. B., Ahlberg, E., Andersen, C., Lindgren, R., Nystrom, R., Nordin, E. Z., Brune, W. H., Svenningsson, B., Swietlicki, E., Boman, C. and Pagels, J. H.

Environmental Science & Technology, 49, 14663, 2015. (published under an open access license)

IV. **No particle mass enhancement from induced atmospheric ageing at a rural site in northern Europe.** Ahlberg, E., Ausmeel, S., Eriksson, A. C., Holst, T., Karlsson, T., Brune, W. H., Roldin, P., Kristensson, A. and Svenningsson, B.

Manuscript

V. **Coastal measurements of ship plume aerosol downwind a major shipping lane in the Baltic Sea Sulphur Emission Control Area: Physicochemical characterization, simulated atmospheric ageing and impact on air quality.** Ausmeel, S., Ahlberg, E, Eriksson, A. C., Spanne, M. and Kristensson, A.

Manuscript

Author's contribution

I. I came up with the idea for the paper. I was responsible for planning and designing the experiments. I conducted most of the experiments. I analyzed all data and wrote the manuscript with comments from the coauthors.

II. I came up with the idea for the paper. I was responsible for planning and designing the experiments. I conducted the experiments. I analyzed all data and wrote the manuscript with comments from the coauthors.

III. I performed initial tests and calibrations of the oxidation flow reactor. I took part in planning and designing the experiments. With C.A., I analyzed OH exposure, reactivity and suppression data. I wrote minor parts of the manuscript dealing with the reactor.

IV. I was responsible for the coordination of the campaign. I was responsible for planning and designing the experiments. I conducted the measurements together with the coauthors. I analyzed all particle data and wrote the manuscript with comments from the coauthors.

V. I took part in planning and designing the measurements. I took part in conducting the measurements. I had the overall responsibility for the oxidation flow reactor measurements. I analyzed data related to the reactor measurements of the summer campaign and wrote the parts of the manuscript dealing with the reactor.

Related publications

Peer-reviewed papers

Time-resolved characterization of primary particle emissions and secondary particle formation from a modern gasoline passenger car. Karjalainen, P., Timonen, H., Saukko, E., Kuuluvainen, H., Saarikoski, S., Aakko-Saksa, P., Murtonen, T., Bloss, M., Dal Maso, M., Simonen, P., **Ahlberg, E.**, Svenningsson, B., Brune, W. H., Hillamo, R., Keskinen, J. and Ronkko, T. *Atmospheric Chemistry and Physics*, 16, 8559, 2016.

Influence of fuel ethanol content on primary emissions and secondary aerosol formation potential for a modern flex-fuel gasoline vehicle. Timonen, H., Karjalainen, P., Saukko, E., Saarikoski, S., Aakko-Saksa, P., Simonen, P., Murtonen, T., Dal Maso, M., Kuuluvainen, H., Bloss, M., **Ahlberg, E.**, Svenningsson, B., Pagels, J., Brune, W. H., Keskinen, J., Worsnop, D. R., Hillamo, R. and Ronkko, T. *Atmospheric Chemistry and Physics*, 17, 5311, 2017.

Carbonaceous aerosol source apportionment using the Aethalometer model - evaluation by radiocarbon and levoglucosan analysis at a rural background site in southern Sweden. Martinsson, J., Azeem, H. A., Sporre, M. K., Bergstrom, R., **Ahlberg, E.**, Ostrom, E., Kristensson, A., Swietlicki, E. and Stenstrom, K. E.: *Atmospheric Chemistry and Physics*, 17, 4265, 2017.

Lead author abstracts

Characterization and first field deployment of the Lund University PAM chamber. **Ahlberg, E.**, Frosch, M., Brune, W. H., Svenningsson, B. NOSA, Helsingør 2012

Simulated atmospheric ageing of wintertime ambient aerosol at a rural background site in southern Sweden. **Ahlberg, E.**, Eriksson, A. C., Brune, W.H., Kristensson, A., Sporre, M., and Svenningsson, B. Faraday Discussions 165, Leeds 2013.

Characterization and first field deployment of the Lund University PAM chamber for biogenic VOC ageing studies. **Ahlberg, E.**, Frosch, M., Brune, W. H., Svenningsson, B. LUCCI annual report 2013.

Investigations into enhanced biogenic secondary organic aerosol from ageing experiments of VOC-mixtures. **Ahlberg, E.**, Falk, J., Eriksson, A: C., Roldin, P., Swietlicki, E., Brune, W. H., Kristensson, A., Svenningsson, B. Biogenic hydrocarbons in the atmosphere, Girona 2014.

Simulated atmospheric ageing of wintertime ambient aerosol at a rural background site in southern Sweden **Ahlberg, E.**, Eriksson, A: C., Brune, W. H., Kristensson, A., Sporre, M. and Svenningsson, B. LUCCI annual report 2014.

Low Particle Formation from Induced Ageing of Ambient Air at a Salix Plantation. **Ahlberg, E.**, Eriksson, A: C., Ausmeel, S., Karlsson, T., Holst, T, Kristensson, A., Svennigsson, B. ICOAAMS 13, Lund 2016.

Effect of seed particle composition, phase and concentration on secondary organic aerosol yields and chemistry in an oxidation flow reactor. **Ahlberg, E.**, Eriksson, A. C., Kristensson, A., Svenningsson, B. NOSA, Lund 2017.

Populärvetenskaplig sammanfattning

Forskningen i denna avhandling handlar i hög grad om att försöka härma kemin i jordens atmosfär. Den sista mätningen till denna avhandling gjordes för bara några veckor sedan, under ett fruktmellanmål tillsammans med mina barn. Jag bestämde mig för att testa ett otroligt påstående som säger att om jorden krymptes ihop till ett äpple är atmosfären lika tunn som äpplets skal. D.v.s.:

$$\frac{tjocklek_{äppleskal}}{diameter_{äpple}} = \frac{höjd_{atmosfären}}{diameter_{jorden}}$$

Med parametrar från ett äpple (Royal Gala tror jag det var) och jordens diameter, får vi att atmosfärens höjd är 74 km. Åker man så långt upp ligger mer än 99,9 % av atmosfärens massa under en, så påståendet stämmer rätt bra (med denna typ av äpple). Men allt väder, nästan alla utsläpp och den kemin vi försökt simulera, sker i troposfären vilket är de lägsta ca 10 km av atmosfären. Tar man bara hänsyn till troposfären skulle äpplet behöva vara nära en halvmeter i diameter, men behålla samma skal, för att påståendet skulle gälla. Exemplet med äpplet ger en bra bild av hur tunn vår atmosfär är, och att vad vi släpper ut spelar roll.

Utan vår atmosfär skulle inget levande klara sig. Den ger oss ett gynnsamt klimat, skyddar oss från farlig strålning och bidrar med vatten och syre. Syret, som kommer från fotosyntesen, gör att atmosfären är väldigt reaktiv och kan rena sig själv från en mängd skadliga ämnen. Men det finns en gräns för hur mycket atmosfären kan hantera. Somliga föroreningar stannar bara några timmar eller dagar i atmosfären innan de reagerar eller faller ut, medan andra kan stanna i mer än ett århundrade. Ofta omvandlas en typ av förorening till en annan, till exempel kan gaser klumpa ihop sig till större partiklar. Dessutom kan reaktioner med en förorening påverka livstiden hos andra. Alla dessa återkopplingar, tillsammans med meteorologisk dynamik, gör att föroreningars påverkan på klimatet kan vara svår att förutspå.

Luftföroreningar är inget nytt fenomen. När John Evelyn skrev Fumifugium (1661), ett tidigt arbete om luftföroreningar, var han mycket bekymrad över kungens hälsa och de oestetiska effekterna av partiklar och gaser från stadens koleldning. Fortfarande har många städer i världen liknande synliga problem, men även till synes ren luft innehåller små partiklar som påverkar både hälsa och klimatet. I en normalren miljö i Sverige andas vi in ett par miljoner sådana partiklar per andetag och upp emot 100 miljarder per dag. Det är faktiskt ganska lite, om man jämför med de gasmolekyler vi andas in (ca 10^{22} stycken per andetag, och ungefär 10-250 miljoner gånger mer massa), men på grund av partiklarnas egenskaper kan de skapa stora hälsoproblem och påverka klimatet på ett helt annat sätt än gaser. Partiklarna sprider ljus på ett sätt som motverkar växthuseffekten och påverkar molnen så till den grad att varenda molndroppe bildas runt en partikel. Genom dessa effekter maskerar partiklar en stor andel av de klimatförändringar som mänskliga utsläpp av växthusgaser borde gett upphov till. Tyvärr är partiklarnas klimatpåverkan mycket osäker, vilket gör att mycket forskning krävs.

Partiklarna i atmosfären är både naturliga och från mänskliga aktiviteter, typiskt förbränning av något slag (ved, olja, kol, bensin). Globalt sett dominerar naturliga partiklar, medan de från mänskliga aktiviteter ofta dominerar i städer. De största partiklarna kan vara damm, små sandkorn eller pollen. De minsta partiklarna, som är i fokus i denna avhandling, består ofta av organiska ämnen och salter. De senaste decennierna har det blivit allt tydligare att en stor andel av de minsta partiklarna bildas genom reaktioner mellan gasmolekyler och så kallade oxidanter (väldigt reaktiva molekyler) i atmosfären. Dessa kallas sekundära partiklar, i kontrast till primära partiklar som släpps ut direkt från källan, såsom en skorsten. Ofta kan man själv ana när det finns mycket partikelbildande gaser i närheten. Det luktar t.ex. skarpt av aromater när du tankar bilen. Den mer angenäma doften av skog och träd, kommer även den från gaser som bildar partiklar i atmosfären, och dessa naturliga utsläpp är faktiskt mycket större än de som vi människor släpper ut. Genom att skapa höga koncentrationer av samma oxidanter som finns i atmosfären, och blanda med dessa gaser, kan man simulera den sekundära partikelbildning som sker i atmosfären. Oxidation i atmosfären kallas också åldring.

I denna avhandling har en kammare med höga oxidantkoncentrationer använts för att studera sekundära partiklar, både i laboratorie- och utomhusmiljö. Grundforskning rörande den experimentella metoden har varit en viktig del av arbetet, men den bakomliggande frågan har varit miljövetenskaplig: hur påverkar människan? Resultaten visar att mänskliga utsläpp, från t.ex. vedeldning eller transport, kan skapa stora mängder sekundära partiklar, ibland flera gånger mer än de primära partikelutsläppen. Men, eftersom växternas utsläpp av partikelbildande gaser är större, handlar människans påverkan också mycket om förändrade förutsättningar för de naturliga sekundära partiklarna. Partikelbildning är oftast inte en linjär process, vilket gör att ett plus ett ofta blir större än två. När mänskliga och naturliga utsläpp blandas i atmosfären kan partiklar bildas både snabbare och i större mängd, än om de isolerats från varandra. De naturliga sekundära partiklarna är därför inte alltid helt naturliga. Alltså, för att kunna redogöra för hur människan påverkar klimatet genom atmosfäriska partiklar krävs en stor mängd kunskap om detaljerade och invecklade processer som dessutom ofta är återkopplade. Denna avhandling är ett litet bidrag till denna kunskap.

1. Introduction

1.1 Background and aim

Humans affect all aspects of the biosphere. Although not all geologists agree (Monastersky, 2015), we have affected the earth to such a degree that it has been proposed that a geological epoch should be named after us, the Anthropocene (Crutzen, 2002; Zalasiewicz *et al.*, 2010). Not least the atmosphere have changed since the emergence of humans. Burning of biomass and fossil fuels emit large amounts of trace gases and particles, but the emergence of agriculture is also believed to have changed the atmosphere during the last couple of thousand years (Ruddiman *et al.*, 2011). Although relatively pristine areas and untouched forests exist, today it is hard to imagine a carbon atom which does not see any influence of humans during its cycle from the atmosphere to the biota, and back again.

Thanks to the evolution of life on earth, which spawned oxygen emissions, our atmosphere is highly reactive. Oxidation reactions makes the atmosphere somewhat self-cleansing. Atmospheric oxidation of several trace gases form so called secondary particles which can be both organic and inorganic. Particles in the atmosphere (commonly referred to as an aerosol, see chapter 2 for definitions) is considered an air pollutant. Particles affect more people than any other pollutant and air pollution is regarded by WHO as "the world's largest single environmental health risk" (WHO, 2014) But, in opposite to greenhouse gases, particles have a negative radiative forcing (Figure 1, IPCC, 2013), which can be viewed as a positive effect. Biogenic (produced by life processes) organic trace gas emissions, which are much larger than the human emissions, are believed to increase in a warmer climate (Penuelas and Staudt, 2010). This in turn can increase particle concentrations that cool the climate, forming a feedback loop (Kulmala *et al.*, 2004). Although a simplification, and a small piece of a much larger puzzle, this loop fits well into the Gaia hypothesis. This hypothesis was proposed by James Lovelock during the 1960´s as he was thinking about ways to remotely detect life on other planets. It proposes that the biosphere, acting as a single organism, maintains favourable conditions for itself by regulating atmospheric oxygen levels and temperature, among several other things. More recently, Lovelock have warned that anthropogenic emissions of pollutants have offset the regulating feedbacks of Gaia (poetically shown in the titles of the books *The revenge of Gaia* [2006] and *The vanishing face of Gaia: A final warning*, [2009]).

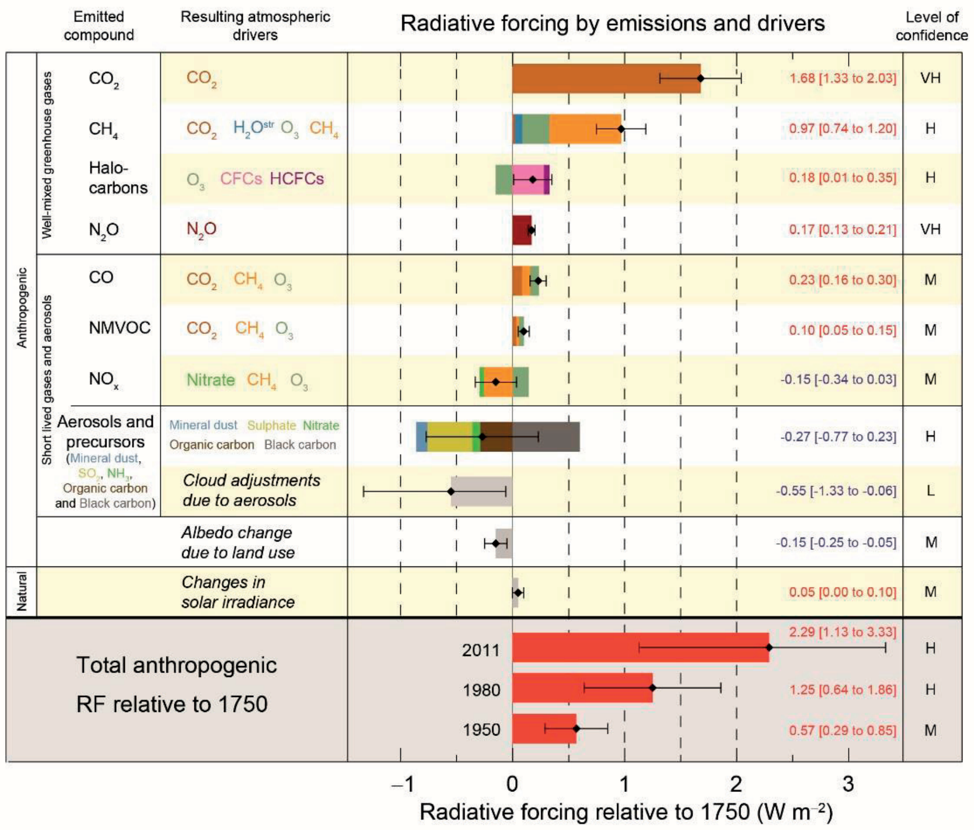

Figure 1. Originally published in the IPCC AR5 (IPCC, 2013). Original caption: Radiative forcing estimates in 2011 relative to 1750 and aggregated uncertainties for the main drivers of climate change. Values are global average radiative forcing (RF14), partitioned according to the emitted compounds or processes that result in a combination of drivers. The best estimates of the net radiative forcing are shown as black diamonds with corresponding uncertainty intervals; the numerical values are provided on the right of the figure, together with the confidence level in the net forcing (VH – very high, H – high, M – medium, L – low, VL – very low). Albedo forcing due to black carbon on snow and ice is included in the black carbon aerosol bar. Small forcings due to contrails (0.05 W m–2, including contrail induced cirrus), and HFCs, PFCs and SF6 (total 0.03 W m–2) are not shown. Concentration-based RFs for gases can be obtained by summing the like-coloured bars. Volcanic forcing is not included as its episodic nature makes is difficult to compare to other forcing mechanisms. Total anthropogenic radiative forcing is provided for three different years relative to 1750.

Research on atmospheric particles in general is motivated by the effects on health and climate. The uncertainty of the radiative forcing of particles remains to be very high in comparison with that of greenhouse gases (Figure 1). Secondary organic species are ubiquitous and are now recognized as a very important part of the atmospheric aerosol, but their dynamic nature and the complexity of gas to particle partitioning makes their influence on climate highly uncertain. Although secondary organic aerosols are gaining interest and are now included, to varying degrees of complexity, in many global atmospheric models (Tsigaridis *et al.*, 2014), their radiative forcing was not assessed in the latest report from IPCC "since the formation depends on a variety of factors not currently sufficiently quantified" (Stocker *et al.*, 2013).

The research presented on the following pages deals with experimentally induced oxidation of trace gases emitted from both vegetation and human activities, and the subsequent particle formation of the reaction products. I have studied fundamental properties of the particles, such as how much is formed from a certain amount of gas phase, in laboratory experiments, but also performed oxidation experiments in ambient air. All measurements were made using an oxidation flow reactor.

The overall aim of this research has been to quantify some of the anthropogenic influences on atmospheric secondary organic particles. This thesis contributes to the research field of secondary organic aerosols in general, and is of particular interest to the growing and vivid group, within the aerosol science community, that makes use of oxidation flow reactors and the results they provide.

1.2 The atmospheric aerosol and its impacts

An aerosol, from aero-solution, is a collection of particles suspended in a gas. Therefore, the atmosphere can be regarded as an aerosol. For particles to stay suspended in the gas, they have to be small. The largest ones fall to the ground due to gravity, but there's no defined upper limit. Atmospheric aerosol particles are mostly below 10 μm in diameter (a human hair is about 75 μm in diameter). The lower limit of an aerosol particle is on the order of a few nm. Smaller particles move more easily, and are affected by the surrounding air molecules bombarding them from every angle at a breakneck speed of 500 m/s (at room temperature). The smallest particles move around so much that they are very likely to hit something, be it another particle or a wall, and get stuck. The largest particles are phlegmatic, opposing every shift in direction that the surrounding air makes. They just continue in a straight line and are therefore also lost to surfaces easily. Particles in the "just right" size range (around 100 nm) can stay suspended for a long time, and under the right circumstances, be transported over hemispheric scales. Commonly quoted atmospheric aerosol particle lifetimes are of the order of hours to weeks (Boucher, 2015). Particles that can avoid obstacles and follow the air stream, are also the ones that can be transported deep into the human respiratory tract.

Particles are always present in the atmosphere, but at highly varying concentrations. Natural sources include sea spray, dust, volcanic eruptions and primary and secondary organics. The most common anthropogenic sources are all connected to the burning of fuels, be it for cooking, heating or transportation. Regular high concentrations of particles are found exclusively close to anthropogenic sources, but on a global scale natural sources dominate.

The negative health effects of particles are well documented (Pope and Dockery, 2006). A recent synthesis report concluded that pollution is the cause of 16% of all deaths worldwide, which is "three times more deaths than from AIDS, tuberculosis, and malaria combined and 15 times more than from all wars and other forms of violence" (Landrigan *et al.*, 2017). Pollution problems are worst in low- and middle-income countries, but there is little evidence for a lower limit on health effects from particles (WHO, 2006). It has been estimated that almost 90% of humans live in areas where the WHO air quality guidelines for particles are exceeded (Brauer *et al.*, 2016). Lelieveld *et al.* (2017) argued that Clean Air should be added to the United Nations sustainable development goals.

Atmospheric aerosol particles play an important but complex role in the climate system. Excluding black carbon aerosol particles, which absorb light, aerosol particles cool the climate by directly scattering light back to space and by acting as cloud condensation nuclei. Particle properties such as size and hygroscopicity govern the ability for water droplet growth. The influence of particles on clouds is multilayered and far from fully understood (Fan *et al.*, 2016; Rosenfeld *et al.*, 2014; Seinfeld *et al.*, 2016), but in general, aerosol-cloud interactions are believed to lead to a fairly strong negative radiative forcing (Boucher *et al.*, 2013). The fact that aerosol particles act on climate in the opposite way as greenhouse gases, leads to a potential masking effect, which can be on the order of 50% of the global warming caused by greenhouse gases (Unger *et al.*, 2017).

The complexity of aerosol-climate interaction is reflected in the high uncertainty of total aerosol radiative forcing (Figure1, IPCC, 2013). In fact, the relative uncertainty of the aerosol radiative forcing have increased in each report since 1993 (Myhre *et al.*, 2015). Carslaw *et al.* (2013) showed that a lot of the variance in models of cloud radiative forcing is attributable to uncertainties in natural emissions of aerosols and aerosol precursors, pointing to the importance of understanding pre-industrial aerosol radiative forcing.

1.3 Secondary organic aerosols

The scientific picture of organic aerosols have changed significantly during the last decades (for a short history see Donahue *et al.*, 2012). Organics in submicron particles are found everywhere and consist to a large degree of secondary material (so called secondary organic aerosol, SOA) formed from volatile organic compounds (VOCs) that undergo oxidation processes in the atmosphere (Hallquist *et al.*, 2009; Spracklen *et al.*, 2011; Zhang *et al.*, 2007). Organic mass concentrations are comparable to or larger than submicron particulate sulphate throughout the northern hemisphere (Jimenez *et al.*, 2009). It is now well established that the organic portion is exposed to atmospheric ageing processes by oxidation throughout its lifetime, and that the molecules partition between the gas and particle phases (Donahue *et al.*, 2013; Jimenez *et al.*, 2009;

Robinson *et al.*, 2007). However, there are still large uncertainties in the total SOA budget (Spracklen *et al.*, 2011).

VOCs are emitted from anthropogenic and biogenic sources. That emissions from vegetation can create atmospheric particles has been known ever since Went speculated in the origin of blue hazes over distant mountains (Went, 1960). Many plants release 1-5% of the carbon fixed as non-methane VOCs (Chapin *et al.*, 2002; Llusià and Peñuelas, 2000; Sharkey, 1996), with a total global emission estimated at over 1000 Tg C y^{-1} (Guenther *et al.*, 1995), widely exceeding that from anthropogenic activities (IPCC, 2001). Not accounting for these emissions in the carbon cycle may overestimate the carbon sink of plants. The emissions, which increase with temperature and photosynthetically active radiation (PAR) (Guenther *et al.*, 1993), have been shown to be a means of communication and a way to cope with biotic and abiotic stresses (Laothawornkitkul *et al.*, 2009; Monson *et al.*, 2013; Peņuelas and Llusiā, 2003; Sharkey *et al.*, 2008). Most non-methane biogenic volatile organic compounds (BVOCs), emitted from terrestrial and marine plants (Kesselmeier and Staudt, 1999; O'Dowd *et al.*, 2004), are highly reactive and are readily oxidized in the atmosphere, with corresponding short lifetimes (Guenther *et al.*, 1995). Anthropogenic VOCs are mainly emitted from combustion, with the largest sources being motor vehicles and biomass burning (IPCC, 2001). Common anthropogenic SOA precursors are aromatics.

There has been a dramatic increase in SOA research since the 1990s, which can be seen in the search result from scientific journal search engines (Figure 2). Published articles per year containing the phrase "secondary organic aerosol" in either the title, abstract or keywords have gone up from around 20 per year in the early 2000´s to over 400 in 2016. As can be expected, the scientific understanding of SOA has significantly increased with increased research and new instrumentation, but new discoveries also comes with new questions and uncertainties (e.g. Glasius and Goldstein, 2016).

Naturally, the uncertainties in VOC emissions and SOA formation processes are reflected in estimates of climate and health effects. Much of the SOA research deals with either fundamental or atmospheric processes, with fewer studies on the health effects. Tuet *et al.* (2017) investigated the chemical oxidative potential of several samples and found that SOA, especially from anthropogenic sources, is important to consider in health effect studies. Estimates of the radiative forcing of SOA are hampered by the necessary simplifications made when SOA is included in global models (Shrivastava *et al.*, 2017). Spracklen *et al.* (2011) estimated a direct radiative forcing of anthropogenically controlled SOA of -0.26 W m^{-2} and radiative forcing of -0.6 W m^{-2} from the indirect cloud effect. Stressing the uncertainties, Scott *et al.* (2014) calculated present-day SOA radiative effects (as opposed to forcings) of up to -0.78 and -0.77 W m^{-2} for the direct and indirect aerosol effects, respectively.

SOA is often underestimated in models (Heald *et al.*, 2005; Tsigaridis *et al.*, 2014). Simple models use experimentally determined SOA mass yields. These are calculated by the ratio of formed SOA mass to the mass of the reacted gas. Recent research have

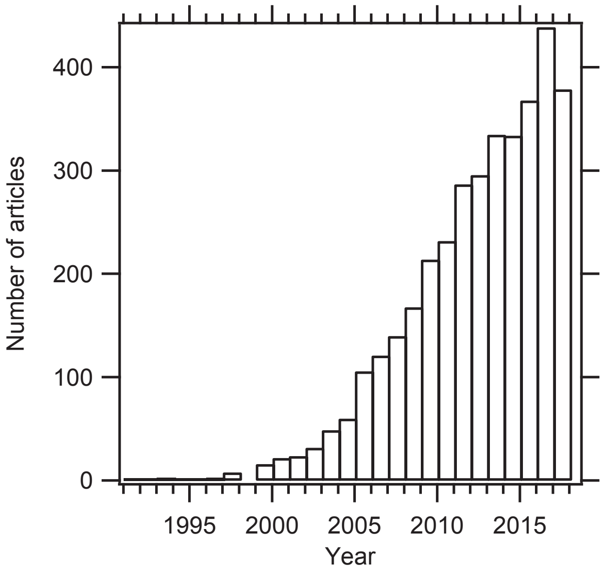

Figure 2. Number of articles published per year containing the phrase "secondary organic aerosol" in either title, abstract or keywords according to Web of Science.

pointed to the importance of accounting for gas phase losses in these experiments (Zhang *et al.*, 2014) which potentially can decrease model-measurement discrepancy (Cappa *et al.*, 2016). Hodzic *et al.* (2016) suggest that a stronger production and faster removal of SOA can lead to a better agreement between models and measurements.

1.4 Biogenic secondary organic aerosols in the Anthropocene

Humans affect all aspects of the biosphere and biogenic SOA is not an exception. Since biogenic VOC emissions, and hence biogenic SOA source strength, are much larger than anthropogenic emissions, it is of great importance to understand the impacts from humans on both biogenic emissions and subsequent particle formation. Carlton *et al.* (2010), investigating eastern U.S.A., found that more than 50% of the biogenic SOA was formed due to anthropogenic influence, and concluded that the classification of SOA based on origin only is not sufficient. Spracklen *et al.* (2011) compared a model with a global dataset of organic aerosol and found the best model-measurement agreement when a large source of SOA from VOCs that correlated with anthropogenic CO emissions was added, but the best chemical agreement was found when a large source of biogenic SOA was added.

Humans affect biogenic SOA formation by altering the sources, atmospheric chemistry and physical processes of SOA formation (for an overview see Hoyle *et al.*, 2011). The perhaps most direct and simple effect is that of land-use change. Cutting down a forest, to plant crops significantly changes the emissions of the area. A staggering 83% of the land surface is today directly influenced by humans (Sanderson *et al.*, 2002). Unger *et al.* (2014) found that the reduction in VOC emissions by land use changes, from forest to agriculture, since preindustrial times have cooled the climate. This is due to the fact that the radiative forcing from decreases in methane (by less competition for oxidants) and tropospheric ozone (formed in VOC photochemistry), which are greenhouse gases, is larger than that from decreases in biogenic SOA (which cools the climate). However, aerosol cloud interactions were not considered in this study.

Water is always present in aerosol particles, often in a substantial amount, affecting both health and climate effects, but is sometimes overlooked and rarely measured (Nguyen *et al.*, 2016). The term anthropogenic water was used by Carlton and Turpin (2013) referring to the fact that some anthropogenic aerosol sources are hygroscopic (such as ammonium sulphate), attracting more water to the particles. This facilitates partitioning of water soluble organic molecules to the particles, an effect which was studied in paper II. In a similar way, an organic particle phase can increase the partitioning of organic gases to particles, provided that they mix (Odum *et al.*, 1996; Pankow, 1994). Mixing of anthropogenic and biogenic SOA was investigated in paper I.

The atmospheric composition and chemistry is altered by human emissions and affect SOA formation in a number of ways. SOA mass yields differ, with different concentrations of NO_X.(Kroll *et al.*, 2005; Ng *et al.*, 2007). Further, NO_X concentrations alter the photochemical production of ozone and hydroxyl radicals, but also determine the nighttime production of nitrate radicals, which all react with biogenic VOCs. In the particle phase, anthropogenic emissions often increase the acidity which may catalyze further reactions of biogenic species and increase SOA mass yields (Gao *et al.*, 2004; Surratt *et al.*, 2007).

1.5 Simulating atmospheric ageing

Atmospheric SOA formation is experimentally simulated in laboratories using various types of chambers (an overview is given in Schwantes *et al.*, 2017). Large (several m^3) environmental chambers have traditionally been used. These may require several hours of flushing prior to experiments to clean the chambers, and the experiments themselves are also time consuming. During the last decade oxidation flow reactors (OFR) have been increasingly used. These are smaller, and have a higher surface-to-volume ratio which is generally bad due to increased wall interactions. However, the short residence time (minutes) in flow reactors compensate for this and parameters such as mass yield and chemical composition have been shown to be similar in OFRs and smog chambers (Bruns *et al.*, 2015; Lambe *et al.*, 2015).

The reactor used in the research included in this thesis is a potential aerosol mass (PAM) OFR designed at Pennsylvania State University (Figure 3). It is a simple design; a 13 l metal cylinder with two UV lamps mounted inside. The UV lamps produce atmospheric oxidants from oxygen and water, at very high concentrations. Translated using ambient oxidant concentrations, the reactor can simulate up to two weeks of atmospheric ageing. It was first introduced in 2007 (Kang *et al.*, 2007). Between 2007 and 2014 15 papers using the PAM reactor were published. Since then, the reactor has been commercialized and in 2017 around 20 papers will be published. The reactor has been thoroughly characterized during the years since its introduction and several important issues and potential pitfalls have been investigated. Important advances include studies of unwanted chemistry (Li *et al.*, 2015; Ortega *et al.*, 2013; Peng *et al.*, 2016; Peng *et al.*, 2015) and gas phase losses (Palm *et al.*, 2016), among other things. Due to the commercialization and ease of use, research using OFRs is likely to increase in the coming years.

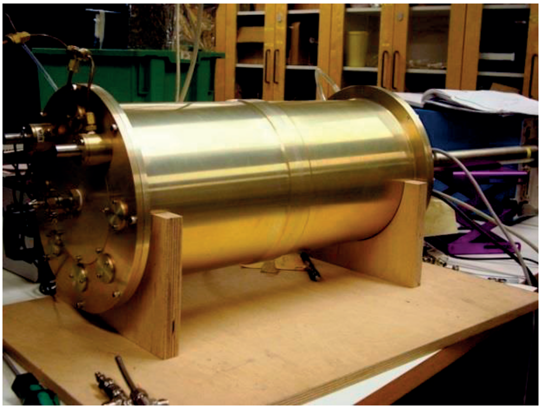

Figure 3. The PAM oxidation flow reactor used in all appended papers of this thesis, provided by Penn State University. The reactor is 42 cm long and has two UV lamps mounted inside that produce extreme amounts of oxidants, several orders of magnitude higher than ambient concentrations. The residence time and oxidant concentration can be varied.

2. Summary of appended papers

2.1 Organic aerosol mixtures (I)

In paper I we investigated the SOA mass yields of VOC mixtures. Partitioning theory predicts increased SOA mass yield with increasing organic particulate mass (Pankow, 1994). This assumption is valid if all organics mix ideally, and is one way in which anthropogenic emissions can increase biogenic SOA (and vice versa, dashed lines in Figure 4). Research have shown that this is not always the case, e.g. freshly emitted organics may not mix with SOA (Cappa and Wilson, 2011; Robinson *et al.*, 2013; Song *et al.*, 2007; Vaden *et al.*, 2010). Models traditioinally treat organics as an ideal mixture (Kanakidou *et al.*, 2005), but experimental evidence of complex SOA mixtures have been lacking (Hoyle *et al.*, 2011).

We performed experiments with an OFR and VOC mixtures containing 2-4 common biogenic and anthropogenic SOA precursors. The results were analyzed comparing data with a simple model constructed by assuming ideal mixing. We saw very good fits between model and measurements in mixtures not containing the VOC myrcene, and concluded that aged SOA mixes ideally regardless of the source. Mixtures containing myrcene had higher yields than the model predicted. This was explained by the fact that myrcene produced a lot more small particles than other VOC, providing a larger surface area for condensation. This result is important in assessing particle growth and transportation, but specifically, it showed that the time scales of the OFR was insufficient for complete condensation of particle forming gases.

2.2 Effect of seed particle composition and phase on SOA formation (II)

In paper II the effect of liquid water on SOA mass yields was investigated. Atmospheric aerosol water is ubiquitous but rarely measured. The amount of water depends on the hygroscopicity of the particle compounds and may increase the uptake of gases. Salt particles are generally more hygroscopic than organics. Anthropogenic particles con-

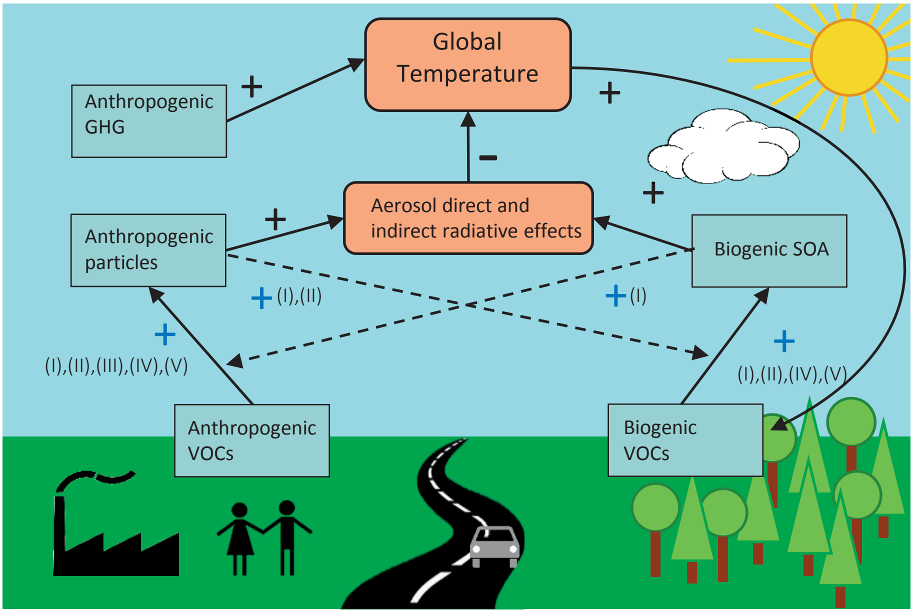

Figure 4. Simplified schematic of the effects of volatile organic compounds (VOCs) and SOA feedbacks with climate. Dashed lines show effects on VOC to particle processes. Blue signs indicate processes investigated in this thesis and corresponding numerals indicate the paper in which these were studied. This picture is merely a selected subset of a much larger system and several processes which are not discussed in this thesis are omitted for clarity. Examples of important processes excluded in the picture include the effect of aerosols on diffuse radiation and biogenic VOC emissions, and the effect of increased GHG lifetimes from VOCs by competition for oxidants.

taining salt can therefore increase the SOA mass yields of biogenic precursors (dashed line in Figure 4). Laboratory studies on this effect have been inconsistent.

Dry or liquid ammonium sulphate and ammonium nitrate seed particles were injected into the OFR together with a mixture of VOCs. To quantify the condensation sink effect, discovered in paper I, a range of seed particle concentrations was used. We found that the SOA mass yield increased by a factor ~2 for dry particles, and on average a factor 3.2 with wet seed particles compared to experiments without seed particles. This suggests that the effect of particulate water is substantial, with a 60% higher mass yield when the seed particles were wet.

2.3 Induced ageing of ambient and anthropogenic aerosols (III-V)

The focus of papers III-V is on anthropogenic and ambient SOA sources.

Paper III is a laboratory study of SOA forming potential and optical properties at different stages of the burn cycle in a conventional wood stove. This was investigated with the OFR by sampling aerosol from a steel chamber enclosing diluted emission samples from a modern wood stove. The effect on the optical properties of the aerosol from reactor ageing was small. SOA formation was highly dependent on the burn conditions. At high burn-rate SOA was up to a factor 10 higher than the primary organic emissions and more than a factor 2 higher than black carbon emissions. At other burn rates, the SOA to primary particle mass concentration ratio was around a factor 2. In this study, the OH exposure was likely higher than needed to produce maximum amounts of SOA. Further, there was no correction for particle losses in the reactor or vapor losses in the steel chamber or reactor, indicating the reported enhancements are a lower limit. In terms of SOA formation, this variability highlights the uncertainties and difficulties in comparing experiments and parameterize emissions for use in models, but also the importance of sound recommendations for wood stove use.

In paper IV we used the OFR to measure the particle formation potential of ambient air in a rural area. The OFR was placed together with particle and VOC monitoring instruments in a Salix plantation. Salix is a bioenergy crop that can be burned directly or be refined to produce ethanol. Bioenergy crops are likely to increase in the future as they are regarded as a sustainable alternative to fossil fuel use. If these crops increase substantially, they will change the VOC emissions from arable land, and consequently also change SOA formation. However, the field site of these measurements was too small to be expected to give a response in the particle measurements. During the campaign, the simulated atmospheric ageing in the OFR resulted in increased O:C ratios, commonly used as a metric for volatility and aerosol age, but also for hygroscopicity. The reactor processing also led to net particle mass losses. This was likely due to a combination of evaporation and oxidation reactions leading to fragmentation of SOA precursors. The VOC measurements, together with relatively low ambient particle concentrations showed that the campaign averages was likely too low to result in significant SOA formation. Furthermore, the fact that extended atmospheric processing lead to decreased SOA concentrations show that the contribution from the nearest sources is low and that natural atmospheric processing acts fast on these emissions. To our knowledge, this is the first reported measurement using an OFR on ambient air in Europe.

Paper V is a summary of two field measurement campaigns, during winter and summer, targeting ship particle emissions. The field site, located in southern Sweden, is approximately 30 minutes downwind a major ship lane. The daily contribution from ship plumes to particle concentration was low in terms of mass, but up to 23 % in terms of number concentrations. The heterogeneity of the ship plume characteristics (particle volume, number and chemical composition) was striking. OFR processing was evaluated during parts of the summer campaign. In contrast to paper IV, reactor processing of the regional background air did not lead to net losses, despite a similar set-up. This can be due to similar magnitudes of loss and formation, but the ambient aerosols were likely also different between the two studies. On some occasions reactor processing led to significant secondary organic and sulphate particle formation, with relative volume concentration increases of a factor 1.5-2.5, and absolute mass concentration increases of about 3-15 µg m^{-3}. Four of these periods were identified during five days of measurements. The results point to the advantages of ambient OFR measurements and the importance of secondary particle formation in the characterization of source emissions.

3. Concluding remarks

The research presented on the following pages have been performed using an oxidation flow reactor. The overall aim has been to study the anthropogenic influence on secondary aerosol formation from both anthropogenic and biogenic precursors. Since flow reactors have not been used extensively for a very long time, the experimental method is under development, and has been given a lot of attention in the papers of this thesis as well as in most papers using OFRs. The advantages of using OFRs have been shown during the last decade.

Incorporating an OFR in field and laboratory measurements is relatively easy. But, more work on the characterization and best practices of the reactor needs to be done. For example, the short residence time is problematic when estimating SOA formation potential at low particle mass concentrations, due to a low condensation sink. This is especially problematic in rural or background measurements, where the condensation sink is generally low. A solution to this problem is to artificially increase the concentration of particles, but such experiments need careful consideration, so that other parameters affecting the SOA formation are not perturbed. I am certain that oxidation flow reactor design and best practices will change in the future, to minimize this type of problem, but intercomparisons and repetition experiments could also benefit the community.

The appended papers in this thesis are both laboratory and ambient studies. We have shown that anthropogenic emissions can form large amount of SOA on itself, but also that they interact with biogenic SOA in ways that increase the overall particle mass concentration. Yet, when processing ambient air, the SOA mass formed in an OFR is not always that great. This is partly because of dilution of the emissions, and inefficient partitioning in the reactor, but also because the atmosphere itself is an efficient low temperature reactor (Rohrer *et al.*, 2014). Many experiments in the past have not taken into account gas phase losses and other variables affecting SOA formation, and thus have not been able to produce the maximum possible SOA mass. For example, many experiments are performed under dry conditions, which is never the case in the atmosphere. It is likely that a large portion of global SOA comes from partitioning of gases to a water containing particle, be it cloud droplets or hygroscopic salt particles. The behavior of SOA in a dynamic water phase, similar to the atmospheric aerosol, is to my knowledge not yet systematically investigated. Further, the temperature has a huge effect on the volatility of gases. The temperatures at which atmospheric SOA formation takes place are naturally much lower than in most laboratories, and with saturation vapour pressures and transformation enthalpies not accurately known (Bilde *et al.*, 2015), it is difficult to translate the room temperature results to atmospheric conditions.

Developments in SOA research during the last decades have to a large degree focused on detailed mechanisms. Although increasing knowledge, this have complicated the picture, rather than decreased the uncertainties. Several recent reviews point to the importance of continued broad-based research covering both detailed chemical processes and large-scale field measurements (Fan *et al.*, 2016; Glasius and Goldstein, 2016; McNeill, 2017; Seinfeld *et al.*, 2016; Shrivastava *et al.*, 2017). Knowledge gaps pointed out in these reviews include deposition, vertical profiling and cloud interactions, among other things.

With a title inspired by Charles Dickens, Pandis *et al.* (2016) used five case studies in Europe, U.S.A and Mexico, to show that improvements in air quality can be made with increasing awareness and regulation, but at the same time the importance of non-traditional sources is increasing. In all cities, secondary material dominated fine particular matter. Including SOA in global models may thus become more important in the future. Summing up, the increase in SOA research during the last decade is likely to continue. In my view, the future of aerosol research looks big, with a wide range of research needed to be done. SOA and OFR experiments in both laboratory and ambient settings will be important.

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