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The climate penalty for clean fossil fuel combustion

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The climate penalty for clean fossil fuel combustion

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Abstract

To cope with the world's growing demand for energy, a large number of coal-fired power plants are currently in operation or under construction. To prevent environmental damage from acidic sulphur and particulate emissions, many such installations are equipped with flue gas cleaning technology that reduces the emitted amounts of sulphur dioxide (SO₂) and nitrogen dioxide (NO₂). However, the consequences of this technology for aerosol emissions, and in particular the regional scale impact on cloud microphysics, have not been studied until now. We performed airborne investigations to measure aerosol size distributions in the air masses downwind of coal-fired power installations. We show how the current generation of clean technology reduces the emission of sulphur and fine particulate matter, but leads to an unanticipated increase in the direct emission of ultrafine particles (1–10 nm median diameter) which are highly effective precursors of cloud condensation nuclei (CCN). Our analysis shows how these additional ultrafine particles modify cloud microphysics, as well as precipitation intensity and distribution on a regional scale downwind of emission sources. Effectively, the number of small water droplets is increased, thus reducing the water available for large droplets and rain formation. The corresponding changes in the precipitation budget with a shift from more frequent steady rain to occasionally more vigorous rain events, or even a significant regional reduction of annual precipitation, introduce an unanticipated risk for regional climate and agricultural production, especially in semi-arid climate zones.

1 Introduction

Most global climate model based studies conclude that anthropogenic sulphur aerosols are beneficial for cooling the earth and thus counteracting the global warming effect of greenhouse gases (Lelieveld and Heinzenberg, 1992). A reduction of sulphur dioxide emissions, such as by cleaning of stack flue gases from fossil fuel burning, is thus

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claimed to accelerate global warming (Arneth et al., 2009; Shindell and Faluvegi, 2009). The basis of this beneficial climate effect of sulphur emissions is the assumption that power plants emit sulphur as SO_2 which then slowly reacts to form sulphate aerosol within several days with the sulphur accumulating primarily on already existing particle surfaces, increasing the scattering of solar radiation by aerosol (Arneth et al., 2009). This process takes several days, distributing sulphur compounds in the atmosphere from regional-scale emissions to a continental to global scale (Manteklow et al., 2009).

Although direct gas-to-particle conversion and a new production of ultrafine particles from SO_2 emissions is considered to be of minor importance to the production of sulphate aerosols in current models (Lohmann et al., 1999, 2000; Liepert et al., 2004; Kazil et al., 2010) and emission estimates (Stier et al., 2006) which form the basis of the model calculations (Kazil et al., 2010), there is evidence of a pronounced relation between new particle production (NPP) by nucleation and sulphuric acid concentrations (Paasonen et al., 2010; Birmili et al., 2003). This indicates the importance of sulphuric acid to the aerosol number budget. New particle production is the source of the ultrafine (Aitken mode) particles which enhance cloud albedo via the number of CCN. At first sight, ultrafine particles from NPP are therefore considered to be beneficial for the Earth's radiation budget by increasing cloud albedo, especially when they appear over the ocean Kazil et al., 2010, Jones et al., 2009). However, the magnitude of this effect is still uncertain (Wang et al., 2011).

Models are not yet able to quantify precisely this radiative impact (Feingold and Siebert, 2009). Furthermore, and adding to the uncertainty, cloud process models (Khain, 2009) also anticipate other concurrent climate effects like a modification of precipitation from warm clouds, which leads to a suppression of low-intensity steady rain and a shift to more vigorous downpours from clouds involving an ice phase (Konwar et al., 2010). Thus, cloud modification by anthropogenic CCN not only has radiative effects, but may also result in secondary climate impacts via the three-dimensional redistribution of water vapour amount and precipitation intensity on larger scales, because modern, “clean”, power plants emit a reduced mass of SO_2 , but higher numbers

of extremely small, nm-sized, ultrafine sulphate aerosols (Junkermann et al., 2011). These tiny aerosols (~ 1 nm diameter) are produced inside the power plant stack in the cleaning chain, with up to 3 % of the initial SO_2 converted into SO_3^- or H_2SO_4 (Murphy, 2007). In the atmosphere, these ultrafine particles then grow rapidly (Junkermann et al., 2011). Sulphuric acid particles become active as CCN already at a size of ~ 50 nm (Hamed et al., 2010; Adams and Seinfeld, 2003). Over continental areas they can be expected to have a less “beneficial” climate side effect than SO_2 emissions, by affecting the large-scale precipitation distribution (Junkermann et al., 2009; Flossmann and Wobrock, 2010). This is illustrated in Fig. 1, where the ultrafine aerosols alter the number of CCN and create brighter clouds (Twomey, 1974).

2 Experimental

Airborne measurements of ultrafine particles in the Karlsruhe (Germany) and Xilinhot (Inner Mongolia) power plant plumes, as well as over the boreal forest in Finland (O’Dowd et al., 2009) were performed using an ultralight aircraft (Junkermann, 2001) as a platform. Measurements in Australia (Junkermann et al., 2009) were made with a motorised glider (DIMONA, HK36) provided and operated by Airborne Research Australia (<http://www.airborneresearch.org.au>). Instrument installations for the determination of aerosol size distributions on both aircraft are summarised in the following table.

For wind and turbulence, the DIMONA installation includes a BAT (Best Aircraft Turbulence) probe (Crawford et al. 1992), while the ultra-light aircraft carries a smaller version of a five-hole probe (Metzger et al. 2011), both combined with a fast, 100 Hz, Inertial Navigation system RT3102, Oxford Technical Solutions, UK. For measurements over the boreal forest in Finland, the SMPS was not yet available. Instead, the production of ultrafine particles was detected as the difference of two counters for particles larger than 3 nm (TSI, model 3025) provided by the University of Helsinki and 10 nm (TSI, model 3010).

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Flight patterns were specifically designed for budget studies. On-board displays of the real time data during the flight were essential to adjust the flight patterns according to the actual weather and plume conditions and position.

3 Results and discussion

We analyzed airborne measurements of ultrafine particle size distributions in different regions of the world in order to investigate source strength, growth rates, and production rates of CCN from different emission sources. Within the COPS/TRACKS experiment we investigated the pollution plume of Karlsruhe, a medium-sized city in Germany with a population of 290 000. Here, two major sources of sulphur emissions are a 660 MW coal-fired power plant and a large petrochemical refinery (PRTR, 2009). Our measurements revealed two distinct plumes of ultrafine particles with concentrations of up to 50 000 particles cm^{-3} merging into a single plume a few km downwind. However, neither an enhancement of the fine fraction larger than $D = 300$ nm, nor increased scattering were observed (Junkermann et al., 2011). After about 2 h, these particles grew to a mean particle size of about $D = 20$ nm (Fig. 2). Based on this Lagrangian study, we estimated a growth rate of ~ 8 nm h^{-1} , sufficient to generate thousands of mainly sulphuric acid based CCN cm^{-3} in a size range larger than 50 nm within a few more hours and a particle source strength of $\sim 2 \times 10^{18}$ particles s^{-1} . For this particle flux calculation we used the particles in the plume cross section multiplied by the measured wind speed (Table 2).

Unexpectedly, compared with previous studies on new particle production (O'Dowd et al., 2009), fresh particles were emitted even in the late afternoon. This primary emission of ultrafine particles is independent of ambient conditions, because the particles are produced inside the stack already (Schulz and Smyrniotis, 2005) and do not require any radiation-dependent atmospheric initial nucleation reactions. However, particle growth after emission still depends on photochemical reactions, as is the case for natural biogenic new particle formation (Kulmala et al., 2004). During growth, these

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particles are then distributed regionally downwind, where they become effective as CCN. The atmospheric lifetime of these tiny particles is several days, enough for transport over more than 1000 km.

The process is not limited to the studied power plant in Germany. We found similar particle plumes with even higher numbers of ultrafine particles during recent airborne experiments in 2009 over the pristine grasslands of Inner Mongolia (Fig. 3). While background number counts of ultrafine particles over the steppe were $\sim 800\text{--}1200\text{ cm}^{-3}$, far lower than those found in Germany and close to the natural continental background level. We observed up to 80 000 ultrafine particles cm^{-3} on more than 50 % of the flying days of the experiments in Inner Mongolia, always downwind of a “modern” coal-fired power plant about 60 km northwest of our research area. Again, the number of fine, visible particles, $D > 300\text{ nm}$, remained unaffected on a very low level ($< 15\text{ particles cm}^{-3}$). The particle production rate from this source was even higher than that in Karlsruhe (Table 2). Overall, these data demonstrate a substantial emission of ultrafine particles from “clean” large fossil fuel combustion plants. Although this emission amounts to less than 3 % of the total power plant sulphur emission, it is increasing the number of sulphur-related cloud condensation nuclei by up to an order of magnitude depending on the regional CCN background.

Similarly, in a third plume experiment in South Australia we were able to detect a particle plume originating from the Port Augusta coal fired power plant while travelling on the road with the instrumentation described in Table 1. The source strength of this small (250 MW) plant was estimated to 0.8×10^{18} ($D = 20\text{ nm}$) particles s^{-1} . As the particle emission from power plants happens at about 200 m above ground and is subject to an additional altitude gain due to the high temperatures of the flue gas, the injection height into the atmosphere is typically about 350 m. Even under convective conditions in the vicinity of the source these emissions are not detectable on the ground. It takes about 8 to 10 km of transport before such a plume becomes visible with ground based instrumentation. In the Australian experiment we crossed the plume about 40 km from the source. This is different from the “old” plume investigations where a visible plume

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can be seen and followed only under stable conditions. The measurements presented here were all done under highly convective conditions with rapid mixing throughout the whole planetary boundary layer.

The importance of this local anthropogenic particle production can be assessed by comparing the particle numbers produced in the “clean” power plant plumes with those found during other investigations where ultrafine particles were of either anthropogenic or natural origin. Typical examples given in the following are plume studies from coal combustion before flue gas cleaning procedures were implemented (Pueschel and Van Valin, 1978), motorway traffic emissions (Rosenbohm et al., 2005; Imhoff et al., 2005), and biogenic nucleation events as observed over boreal forests (O’Dowd et al., 2009) (Table 2).

Pueschel and Van Valin (1978) performed studies during several campaigns between 1973 and 1976. The “dirty” plants studied at that time emitted large quantities of sulphur dioxide and visible fine and coarse aerosols. Although the total sulphur emission was more than an order of magnitude larger in 1976 compared with that of the 2007 Karlsruhe plume, the total number of cloud condensation nuclei, measured with a CCN counter, was significantly lower. For a power plant much larger than that at Karlsruhe (Four Corners, TX, 2200 MW), 10^{15} – 10^{16} particles per second in CCN size ($D > 50$ nm for sulphuric acid particles) were estimated. At that time, this value was considered to be comparable to the natural production of CCN from that power plant’s entire $\sim 10\,000$ km² plume area. The total production rate of 20 nm aerosols from the two “clean” point sources in our Karlsruhe plume, by contrast, was three orders of magnitude higher (Table 2).

Traffic emissions are another major anthropogenic source of ultrafine particles. Cars emit particles in the $D = 15$ – 20 nm range. From measurements alongside a busy motorway north of Karlsruhe (Rosenbohm et al., 2005; Imhoff et al., 2005), the source strength during the day can be estimated to be up to $\sim 4 \times 10^{14}$ s⁻¹ km⁻¹ at a car frequency of 60 000 cars / day, much lower at night. Here we used the data published and calculated the source strength of the line emission source based on the concentrations,

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vertical distributions and wind speeds given in these papers. Compared to the motorway line source, one “clean” power plant produces a similar amount of ultrafine particles as $\sim 1/3$ of the German motorway system (12 000 km) (Table 2).

Ultrafine particles acting as sources for CCN are produced naturally as well from DMS emissions over the oceans as from biogenic emissions over the continents. This biogenic particle production is also the source of the typical blue haze over continental forest ranges (Rasmussen and Went, 1965). The continental particles originate from regionally distributed sources over forests during new particle formation which is frequently occurring in short time nucleation bursts (Kulmala et al., 2004). A single “typical” nucleation event over the boreal forest in Hyytiälä, Finland in Spring 2003 produced ~ 8000 new particles cm^{-3} with a mean size of 20 nm, well distributed within the 800 m planetary boundary layer. These measurements were performed by continuous vertical profiles flown between about 5 m above the ground to the free troposphere during the built up of the mixed layer using the microlight aircraft in the vicinity of the Hyytiälä SMEAR station (Fig. 4). The flights also covered the vicinity of the station with a mixture of forests and lakes up to about 30 km in diameter. At that time of the year the lakes were still covered with a thick ice layer. Maximum number densities of $D = 20$ nm particles were reached about three hours after the initiation of nucleation. Hence, the total number of biogenic ultrafine particles injected into the planetary boundary layer was in the order of $\sim 10^{15}$ particles $\text{km}^{-2} \text{s}^{-1}$. However, nucleation requires dry sunny and preferably cold conditions that prevail at that location on about 90 days per year only (Manninen et al., 2010), while a nucleation event typically lasts only 2–3 h. The annual average particle source strength over boreal forest is thus about $2 \times 10^{13} \text{s}^{-1} \text{km}^{-2}$, comparable to Pueschel and Van Valin’s (1978) old power plant plume area estimates (Table 2). As new particle production from biogenic precursors is facilitated in the presence of anthropogenic sulphuric acid (Zhang et al., 2009; Paasonen et al., 2010) this possibly is an upper estimate for biogenic production in the area.

Ultrafine particle production from “clean” fossil fuel thus turns out to be one of the main anthropogenic sources of cloud condensation nuclei. One “clean” power

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plant (~600 MW electricity production) produces as many ultrafine particles as about 4000 km of motorway, or as half of the boreal forest area in Finland. This is due to the change from “dirty” power generation to “clean” operation, which reduces the total sulphur emissions, while the production of CCN precursors is considerably increased.

5 Power plant particle productions are even more important as they emit effectively into mid-elevations of the planetary boundary layer, where the particles are much less likely to be rapidly deposited than those produced at ground level and have a high probability to end up as CCN.

10 These results also explain why concurrent with the significant reduction of SO₂ emissions in Europe in the last decades, the few available data show that the number of CCN at ground level decreased only marginally (Hamed et al., 2010).

Enhanced CCN number concentrations can increase the cloud albedo (Twomey, 1974), partially compensating the loss of additional scattering by sulphate coatings on pre-existing aerosols (Arneth et al., 2009). However, it is also important to consider 15 the precipitation effects of the increased CCN numbers. Experiments are scarce, as long-term, large-scale monitoring of aerosols and precipitation is required. We performed additional airborne experiments on the interaction of particles and CCN with clouds and precipitation over one of the very few regions suitable for such experiments in the world, the South West of Western Australia. Here, two adjacent homogeneous 20 land use areas divided merely by a fence and both spanning several thousands of km² allow for direct comparisons of the effect of land use on CCN and related precipitation patterns. The fence separates an agricultural area suffering from a reduction of precipitation from a natural forest showing a slight increase in precipitation in the last decades. Long-term, statistically significant precipitation data are available for this 25 area. We were able to relate the reduction in precipitation to the large-scale land use change associated with a doubling of the CCN number concentration (Junkermann et al., 2009).

Model calculations of the particle growth in the Karlsruhe power plant plume confirm the experimental results observed as shown by our application of the regional model

COSMO-ART (Vogel et al., 2009). Applied to the aerosol production in the Karlsruhe power plant plume case our model reproduces well both, the growth of the ultrafine particles in the plume and the dependence on the emission altitude. Changing from pure SO₂ to 97 % SO₂ and 3 % H₂SO₄ (gas phase) emission (Schulz and Smyrniotis, 2005), doubles the number of particles and increases their size by 40 %. This tendency would be even stronger for a direct emission of 1 nm droplets instead of H₂SO₄ molecules. Emission into altitudes of 300 to 500 m agrees well with the observations and was confirmed to increase the efficiency of CCN production, as particle deposition on the ground does not play a major role any more (Junkermann et al., 2011).

Due to the long-range transport of the ultrafine particles for several days and the 6–8 h required to grow to CCN, local effects are sparse, but anthropogenic CCN effects can be expected on regional or even larger scales. Lower growth rates would only shift the onset of the cloud impact slightly further downwind from the source region. We found that a doubling of CCN in pristine Western Australia led to a regional reduction in precipitation by ~20 %. Also and irrespective of the recent floods, a regional decline of precipitation by 40 % since 1970 is reported for Northeastern Australia, which is claimed to be linked to regional aerosol air pollution (Bigg, 2008), although concurrent aerosol measurements are lacking. Within the last 40 years, however, more than 10 large coal-fired power plants were taken into operation.

To indicate the magnitude of the ultrafine particle emissions by single power plants we can also compare the plant emissions with requirements for particle production in global solar radiation management (SRM) proposals. The climate impact of increasing numbers of CCN is currently under intense discussion in regard to geo-engineering activities to modify the albedo of marine stratocumulus clouds in order to counteract global warming. Recent global models propose a possible continental-to-global shift of precipitation as a result of such artificial regional increases of maritime CCN (Bala et al., 2008; Keith et al., 2010). In these simulations, the earth's surface is divided into 6596 cells of $7.7 \times 10^4 \text{ km}^2$ each. SRM-experiments would require to triple the number of CCN in about 15 % of the marine cells, generating additionally $1.2 \times 10^{17} \text{ s}^{-1}$

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particles per cell, proposed to be produced by spraying seawater into tiny droplets of ~800 nm size (Salter et al., 2008). This size depends on the technical problems to spray seawater in tiny droplets. Smaller droplets would be as effective.

One power plant of the Karlsruhe size would be sufficient per cell even under the slightly higher pre-existing CCN numbers to double the 100–250 cm⁻³ CCN over pristine continental regions compared to 40–60 cm⁻³ over the oceans assuming that a small sulphuric acid CCN of 50 nm is as efficient as a fine seawater droplet.

When extrapolating from the conditions measured at single power plants in Europe, Inner Mongolia and Australia, the large number of already operational or planned “clean” coal-fired power plants in China, which is covering the size of 130 cells (Inner Mongolia ~15 cells) would be sufficient for a regional continental solar radiation management project (Latham et al., 2008; Salter et al., 2008). Although they may not yet be statistically significant, the shifts in precipitation intensity and distribution adversely affect agricultural production already now (Zhao and Running, 2010; Piao et al., 2010).

4 Summary and conclusions

We carried out airborne studies to quantify the budget of ultrafine particles as cloud condensation nuclei (CCN) precursors. We found that modern implementations of fossil fuel (coal) burning power plants are one of the major sources of ultrafine particles in the planetary boundary layer on a regional scale exceeding other known major sources. We considered 20 nm particles as a proxy for CCN precursors to be able to compare with other sources. Sulphuric acid particles would need to grow further to approximately 50 nm before they can be activated as CCN, possibly slightly reducing the numbers due to coagulation processes. However the total numbers of particles produced by “clean” power plants are so large that our comparison analysis would not be affected. “Clean” technology coal fired power plants turned out to be the major source of ultrafine particles in mid elevations of the planetary boundary layer in pristine and moderately polluted conditions. Only in very highly polluted regions other industrial

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sources could possibly compete. Here a loss to pre-existing accumulation mode particles during plume dispersion can also be significant.

Although cleaning of flue gas to reduce SO₂, sulphate and nitrogen dioxide oxide emissions is mandatory to protect the environment from acidification of rain, our results show that the technique currently used has some less beneficial side effects. The enhanced number of CCN might partially compensate the loss in aerosol scattering by an increase in the brightness of clouds. Probably, the most important unanticipated implication of cleaning fossil fuel power plants on the regional scale is the reduction of steady, low-intensity precipitation. This does not necessarily change the total precipitation amount, but the precipitation's spatial and intensity distribution. The water trapped temporarily in the atmosphere either in the gas phase or in tiny droplets too small to produce rain can be effectively transported over large distances. Finally under conditions with additional forcing of precipitation production, for example by orography, it favours more intense rainfall or even vigorous flooding which can take place thousands of km away. These effects, especially their localisation, are still rather uncertain among the few available global models (Jones et al., 2009) and were not covered by our regional scale model calculations.

Modern “clean” plants in the near future will replace the remaining old “dirty” facilities in industrialised countries, while developing countries will directly install “clean” new plants. As they are still considered to be clean, new coal fired power plants are now also being constructed in extremely sensitive semi-arid agricultural regions close to the coal mines (China Post, 2009, <http://www.power-technology.com/projects/kogan/>), where additional ultrafine particulate emissions might have severe negative consequences on the regional water budget.

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The analysis we report here is thus important in highlighting an unexpected effect of clean technology. While reducing sulphur emissions is well known to be associated with a climate penalty through light scattering effects (Arneth et al., 2009), our results show that flue cleaning technology actually introduces a new problem, in the form of increased emissions of ultrafine particles. By creating an oversupply of cloud condensation nuclei, these provide an additional mechanism to alter climate directly, by altering precipitation patterns.

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Table 1. Aircraft instrumentation.

Parameter	Instrument	Time resolution
Aerosol size distribution $D = 5\text{--}350\text{ nm}$	SMPS, GRIMM 5403	120 s
Aerosol size distribution $D = 300\text{ nm--}20\text{ }\mu\text{m}$	OPC, GRIMM 1.108	6 s
Aerosol number concentration $D > 10\text{ nm}$	CPC, TSI3010	1 s
Extinction coefficient	Nephelometer HSS AVMIII	1 s
Wind speed and direction	Turbulence probe	0.1 s
Temperature, dew point	Chilled mirror, MeteoLab TP3	1 s
Radiation up- and down-welling	Pyranometer, Licor LI-200	1 s
Photolysis rates JO^1D and JNO_2	actinic flux filterradiometers	1 s
Cloud droplets (DIMONA only)	FSSP-100, DMT, Boulder, CO	1 s

s: D, median particle diameter; SMPS, Scanning Mobility Particle Sizer; OPC, Optical Particle Counter; CPC, Condensation Particle Counter; FSSP, Forward Scattering Spectrometer Probe; HVMIII Airborne Visibility Meter.

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Table 2. Particle production rate ($D = 20$ nm, CCN precursors), power plants operating 24 h per day (hd^{-1}), motorway 15hd^{-1} , boreal forest nucleation 3hd^{-1} on 25 % of all days calculated as annual average production s^{-1} . “Dirty” power plant study, ($D > 50$ nm, CCN).

Study	site/area Year	average annual particle production	Source type
Karlsruhe “clean” power plant	2007	$2 \times 10^{18} \text{s}^{-1}$	point
Xilinhot “clean” power plant	2009	$>7 \times 10^{18} \text{s}^{-1}$	point
Port Augusta “clean” power plant	2011	$0.8 \times 10^{18} \text{s}^{-1}$	point
Four Corners “dirty” power plant	1976	$10^{15} - 10^{16} \text{s}^{-1}$	point
Motorway	2004	$4 \times 10^{14} \text{s}^{-1} \text{km}^{-1}$	line
Boreal forest Hyytiälä, Finland	2002	$2 \times 10^{13} \text{s}^{-1} \text{km}^{-2}$	area

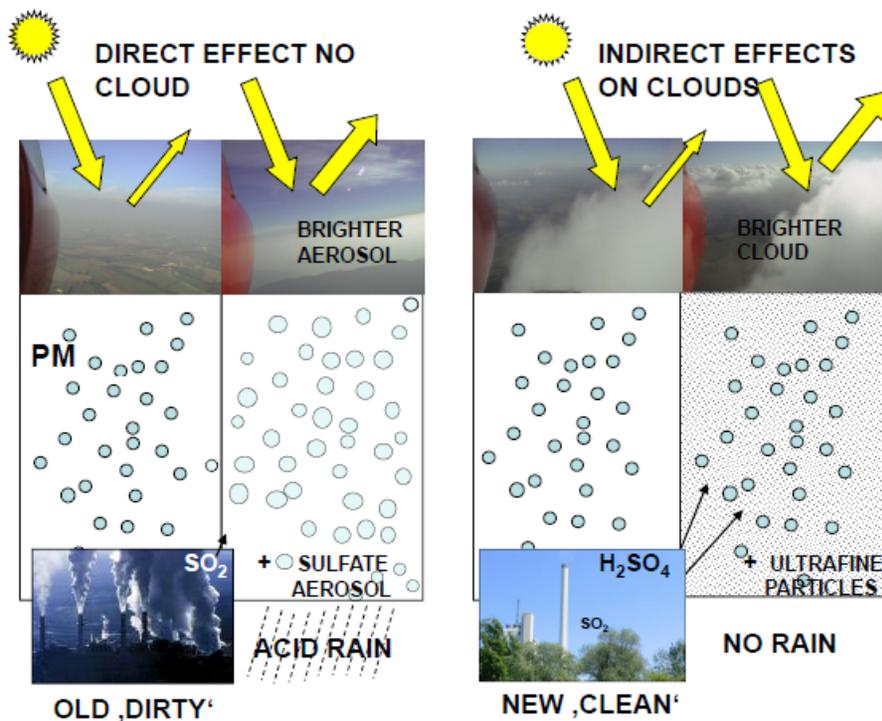


Fig. 1. Old versus new “clean” power plant emission: left (old power plant) – sulphur dioxide increases the size and scattering coefficient of particulate matter (PM), but not the number of existing or co-emitted particles; right (new power plant) – directly emitted H₂SO₄ increases the CCN number density and affects cloud microphysics by brighter clouds and precipitation suppression.

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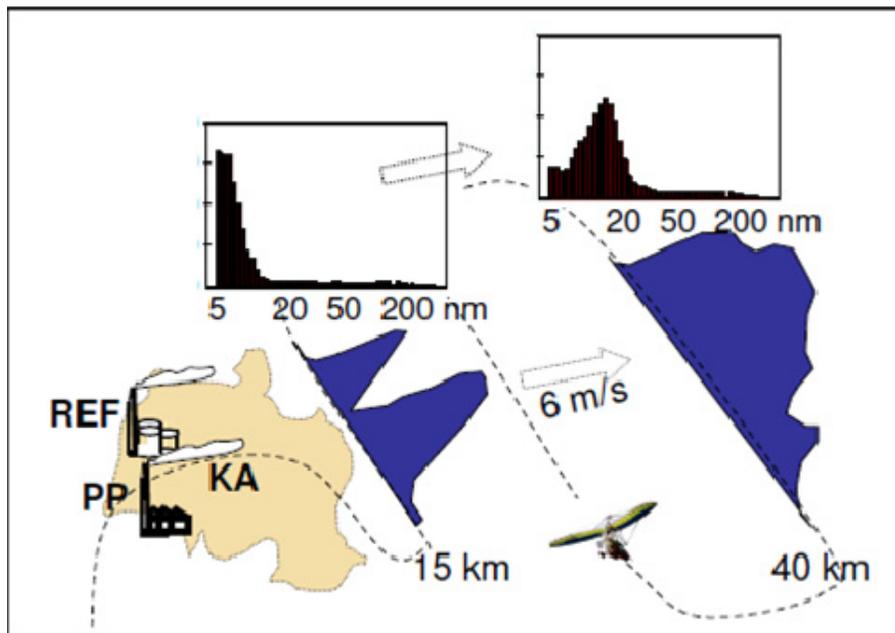


Fig. 2. Two point sources, one plume. Development of a particle plume at 500 m above ground from two elevated sources, the Karlsruhe (KA) power plant (PP) and the MIRO refinery (REF) after 15 km and 40 km of transport cross-sections (blue) and size distributions (black inserts). Maximum total numbers of ultrafine particles ($D > 10$ nm) reach $\sim 50\,000\text{ cm}^{-3}$. These particles are expected to grow into CCN sizes after transport for another ~ 100 to 150 km.

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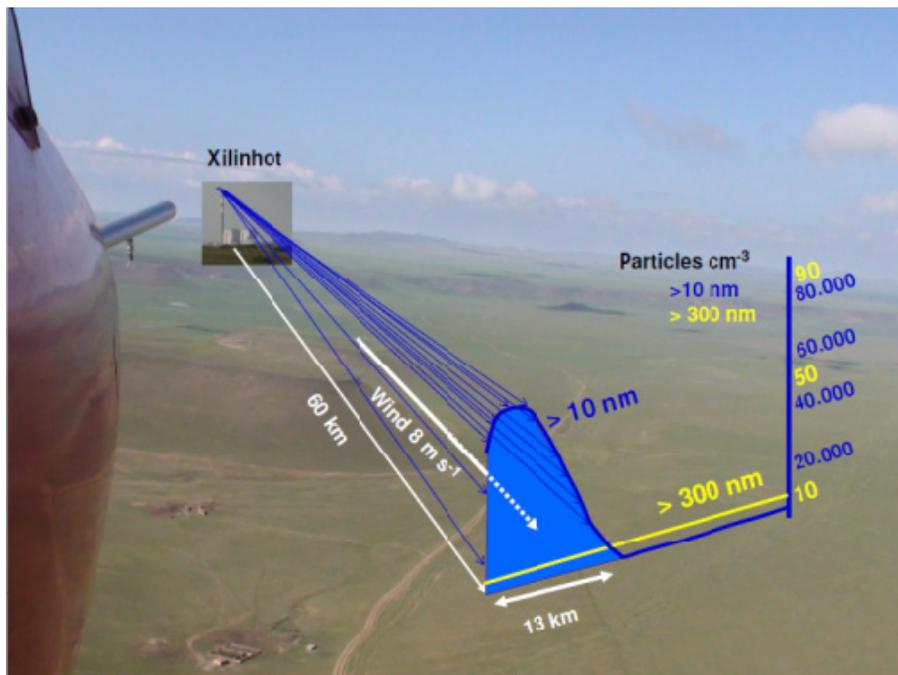


Fig. 3. Sketch of the ultrafine particle plume over the Inner Mongolian steppe downwind of Xilinhot (6 July 2009). Particle number density outside plume area $<1200\text{ cm}^{-3}$. Horizontal flight track 50 m above ground 40 km. Blue: ultrafine particles, yellow: fine particles $D>300\text{ nm}$.

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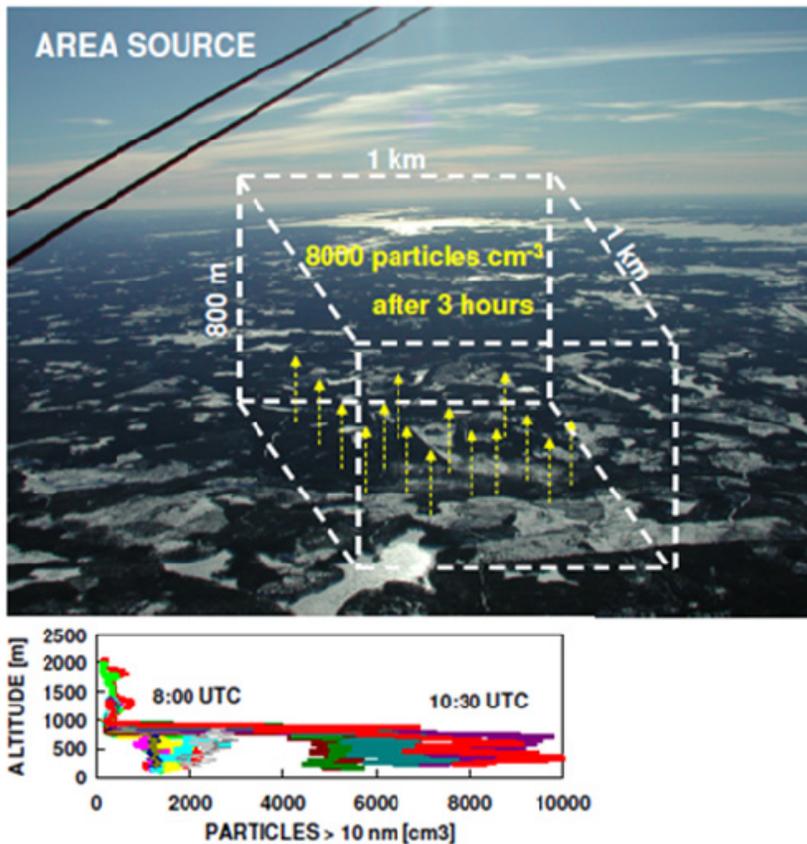


Fig. 4. Particle production over the boreal forest of Finland. 20 consecutive vertical profiles were flown over and in the vicinity of the ground based station of Hyytiälä between the free troposphere and about 5 m above ground to follow the development of the particle number concentrations and the growth of the mixed layer in the morning. Panel b shows the individual profiles of particles in the $D > 10$ nm range.

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