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Formation and growth rates of ultrafine atmospheric particles: a review of observations

M. Kulmala[a,*], H. Vehkamäki[a], T. Petäjä[a], M. Dal Maso[a], A. Lauri[a], V.-M. Kerminen[b], W. Birmili[c], P.H. McMurry[d]

[a] *Department of Physical Sciences, Division of Atmospheric Sciences P.O. Box 64, Gustaf Hällströminkatu 2, FIN-00014 University of Helsinki, Finland*
[b] *Finnish Meteorological Institute, Air Quality Research, Sahaajankatu 20E, FIN-00880 Helsinki, Finland*
[c] *Division of Environmental Health and Risk Management, University of Birmingham, Edgbaston B15 2TT, UK*
[d] *Department of Mechanical Engineering, 111 Church Street SE, Minneapolis, MN 35455, USA*

Abstract

Over the past decade, the formation and growth of nanometer-size atmospheric aerosol particles have been observed at a number of sites around the world. Measurements of particle formation have been performed on different platforms (ground, ships, aircraft) and over different time periods (campaign or continuous-type measurements). The development during the 1990s of new instruments to measure nanoparticle size distributions and several gases that participate in nucleation have enabled these new discoveries. Measurements during nucleation episodes of evolving size distributions down to 3 nm can be used to calculate the apparent source rate of 3-nm particles and the particle growth rate. We have collected existing data from the literature and data banks (campaigns and continuous measurements), representing more than 100 individual investigations. We conclude that the formation rate of 3-nm particles is often in the range 0.01–10 cm^{-3} s^{-1} in the boundary layer. However, in urban areas formation rates are often higher than this (up to 100 cm^{-3} s^{-1}), and rates as high as 10^4–10^5 cm^{-3} s^{-1} have been observed in coastal areas and industrial plumes. Typical particle growth rates are in the range 1–20 nm h^{-1} in mid-latitudes depending on the temperature and the availability of condensable vapours. Over polar areas the growth rate can be as low as 0.1 nm h^{-1}. Because nucleation can lead to a significant increase in the number concentration of cloud condensation nuclei, global climate models will require reliable models for nucleation.

Keywords: Atmospheric aerosols; Nucleation; Condensation growth

* Corresponding author. Tel.: +358-9-19150756; fax: +358-9-19150860.
E-mail address: markku.kulmala@helsinki.fi (M. Kulmala).

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1. Introduction

Aerosol particles are ubiquitous in the Earth's atmosphere and influence our quality of life in many different ways. In urban environments, aerosol particles can affect human health through their inhalation (Wichmann & Peters, 2000; Stieb, Judek, & Burnett, 2002). In a global troposphere, and particularly downwind from major pollution sources, aerosol particles are thought to contribute to climate change patterns (Stott et al., 2000; Ramanathan, Crutzen, Kiehl, & Rosenfeld, 2001; Yu, Saxena, & Zhao, 2001; Menon, Del Genio, Koch, & Tselioudis, 2002). Understanding these effects requires detailed information on how aerosol particles enter the atmosphere and how they are transformed there before being removed by dry or wet deposition. Key processes in this respect are the formation of new atmospheric particles and their subsequent growth to larger sizes.

Aitken (1897) was the first to report evidence for new particle formation in the atmosphere. However, quantitative measurements of aerosol formation and growth rates have required the recent developments in instrumentation for measuring size distributions down to sizes as small as 3 nm in diameter (McMurry, 2000a). We refer to the 3–20 nm particles as the "nucleation mode" (called sometimes also the ultrafine mode), since nucleation and growth from gaseous precursors leads to the formation of such very small particles. Other particle modes that have been previously documented are the Aitken nuclei (20–90 nm), accumulation (90–1000 nm) and coarse (particles $>$ 1000 nm in diameter) modes.

Many studies conducted in the free troposphere, and especially near clouds and close to the tropopause, have detected large numbers of very small, 3–15 nm diameter aerosol particles (e.g. Hoffman, 1993; Perry & Hobbs, 1994; Hoppel, Frick, Fitzgerald, & Larson, 1994; Clarke et al., 1998b; Clarke, Kapustin, Eisele, Weber, & McMurry, 1999a; Clarke et al., 1999b; Nyeki et al., 1999; Keil & Wendisch, 2001; Weber et al., 2001b; Twohy et al., 2002). In the continental boundary layer, there are frequent observations of recent nucleation events, i.e. the formation of ultrafine particles detected at a few nm, accompanied by the subsequent growth of these particles to $\sim$ 100 nm within the next 1–2 days. Such observations span from the northernmost sub-arctic Lapland to the remote boreal forest (Kulmala, Toivonen, Mäkelä, & Laaksonen, 1998; Mäkelä et al., 1997) to suburban Helsinki (Väkevä et al., 2000), to urban Atlanta, Pittsburgh and St. Louis (Woo, Chen, Pui, & McMurry, 2001; Stanier, Khlystov, & Pandis, 2002; Shi and Qian, 2003), to industrialised agricultural regions in Germany (Birmili & Wiedensohler, 2000a; Birmili et al., 2003) and also to coastal environments around Europe (O'Dowd et al., 1999). Nucleation has been observed with monitors on mountains (Weber, McMurry, Eisele, & Tanner, 1995; Weber et al., 1996, 1997), and evidence for the role of biogenic emissions in aerosol formation has also been reported (Kavouras, Mihalopoulos, & Stephanou, 1998; Weber et al., 1998). A limitation of most observations is that measurements were either made at a fixed point (ground), or on platforms not necessarily moving along with the same air parcel. Observations of new particle formation may therefore be biased by spatial variations of constituents in different air parcels.

A variety of different nucleation mechanisms have been proposed for the atmosphere. The most widely studied ones are the binary water–sulphuric acid nucleation (e.g. Kulmala & Laaksonen, 1990), ternary water–sulphuric acid–ammonia nucleation (Kulmala et al., 2000c) and ion-induced nucleation (Yu & Turco, 2000). A technique is available for measuring sulphuric acid vapour, and such measurements have been reported for a few nucleation studies. Techniques for measuring

ammonia with high time resolution at ppt levels are now becoming available, but measurements of ammonia during nucleation events are rare (e.g. Berresheim et al., 2002). Organic vapours could, in principle, participate in nucleation, but nucleation mechanisms that involve organics have not yet been identified. It appears very likely, however, that organics contribute to growth of nucleated particles (O'Dowd, Aalto, Hämeri, Kulmala, & Hoffmann, 2002b). In practise it is very important to investigate nucleation and growth processes separately, since different species can participate in these processes.

In this review we summarise recent observations of particle formation and growth. Altogether these measurements span a broad range of both geographical locations and ambient conditions. Where possible, we report the formation rate of 3 nm particles, because 3 nm is the current minimum detectable size. Some studies involved the use of instruments with a minimum detectable size that is larger than 3 nm. In such cases we estimated particle formation rates at the minimum detectable size. Growth rates can also be determined from measured nucleation mode size distributions.

There are several studies in which there is clear evidence on aerosol formation but no quantitative estimation of particle production rates is possible (e.g. Aitken, 1897). An ideal situation in this regard is when continuous size distribution measurements of particles > 3 nm are available. This is the case at the SMEAR II station in Finland (Kulmala et al., 2001) and at several U.S.E.P.A supersites, including those in Atlanta (Woo et al., 2001), Pittsburgh (Stanier et al., 2002) and St. Louis (Shi and Qian, 2003). Such data enable the determination of both particle formation and growth rates.

2. On observations

In this study we review more than 100 publications that report observations of ultrafine particles in the atmosphere. The studies included are presented in Table 1, from which one can see the number of each paper (to be used later), the authors, and the location (latitude, longitude, name of the place) and the measurement time period. A global map showing the measurement locations is presented in Fig. 1. As can be seen, measurements have been performed all over the world, even though Europe and North America are much better represented than other regions of the world.

The investigations are based on either long-term monitoring or intensive short-term measurements. Only a few continuous long-term studies have been carried out. Given the benefits of data analysis on a climatological basis, more measurements of this type are clearly desirable. The measurement platforms can be divided into three different types: ground-based, ship-based and airborne. In airborne measurements often only particle number concentrations have been measured, with no information on particle formation and growth rates. In some cases this kind of information is available, as was the case for aircraft measurements conducted in the plume from a penguin colony over the Macquarie Island (Weber et al., 1998) or for more recent measurements in the polluted continental boundary layer by Brock et al. (2002, 2003).

Observations can also be categorized by altitude, latitude, degree of pollution influence, etc. In this respect, a distinction can made between different altitudes (the boundary layer and the lower, middle and upper free troposphere), latitudes (tropics, mid-latitudes, high latitudes, polar regions) and the overall degree of pollution (remote marine, polluted marine, remote continental, rural, urban). Table 1 summarises also the different measurement platforms used, along with the air mass type.

Table 1

Ref	Authors (year)	Place	Latitude	Longitude	Date	Air mass		Air mass type	Platform
1	Koponen (2003)	Antarctica	S 73 03	W13 25	January 2000, 2001	CBL	Arctic	Coastal	Land
2	Jaenicke et al. (1992)	Atka Bay, Antarctic	S70 37	W008 22	82–90	CBL	Arctic	Coastal	Land
3	Gras (1993)	Mawson, Antarctica	S67 36	E062 53	85–91	CBL	Arctic	Coastal	Land
4	Park, Sakurai, Vollmers, and McMurry (2003)	South Pole	S89.997	W102.0	12/98 & 12/00	CBL	Arctic	Remote	Land
5	Ito (1993)	Syowa, Antarctica	S69 00	E039 35	8/78–12/78	CBL	Arctic	Coastal	Land
6	Aalto, Kulmala, and Nilsson (1995)	Värriö	N67 46	E029 35	6/93–8/94	CBL	Sub-Arctic	Remote rural	Land
7	Kulmala et al. (1996)	Värriö	N67 46	E029 35		CBL	Sub-Arctic	Remote rural	Land
8	Pirjola, Laaksonen, Aalto, and Kulmala (1998)	Värriö	N67 46	E029 35	6/93–8/94	CBL	Sub-Arctic	Remote rural	Land
9	Dal Maso (2002)	Värriö	N67 46	E029 35	12/97–7/01	CBL	Sub-Arctic	Remote rural	Land
10	McMurry et al. (2003)	Atlanta, GA	N33.777	W84.414	8/1/98–8/30/99	CBL	Mid-latitudes	Urban	Land
11	Shi, Evans, Khan, and Harrison (2001)	Birmingham	N52 30	W001 52	10/98–2/99	CBL	Mid-latitudes	Urban	Land
12	Weber et al. (1997)	Colorado Rockies	N39 59	W105 35	93	CBL	Mid-latitudes	Rural	Land
13	Keil and Wendisch (2001)	Dresden area	N51 [40] N52 20	E013 [2] E014 5	96–98	CBL	Mid-latitudes	Rural	Aircraft
14	Neusüss et al. (2002)	Falkenberg/ Germany	N52	E14	7/98–8/98	CBL	Mid-latitudes	Rural	Land
15	Wiedensohler et al. (1997a)	Great Dun Fell	N54 41	W002 27	4/93–5/93	CBL	Mid-latitudes	Rural, cloud interaction	Land
16	Birmili et al. (1999b)	Great Dun Fell/United Kingdom	N55	W-3	3/95–4/95	CBL	Mid-latitudes	Rural, cloud interaction	Land

17	Verheggen and Mozurkewich (2002)	Hastings, Canada			25.8.1993	CBL	Mid-latitudes	Rural	Land
18	Birmili et al. (2000b)	Hohenpeissenberg	N47 48	E011 1	1998–2000	CBL	Mid-latitudes	Rural	Land
19	Birmili et al. (2003)	Hohenpeissenberg	N47 48	E011 1	1998–2000	CBL	Mid-latitudes	Rural	Land
20	Birmili et al. (2001a)	Holme Moss/United Kingdom	N54	E-1	3/2001	CBL	Mid-latitudes	Rural	Land
21	Mäkelä et al. (1997)	Hyytiälä	N61 51	E024 17	2/96–10/96	CBL	Mid-latitudes	Rural	Land
22	Kulmala et al. (1998)	Hyytiälä	N61 51	E024 17	2/96–1/97	CBL	Mid-latitudes	Rural	Land
23	Mäkelä, Koponen, Aalto, and Kulmala (1999)	Hyytiälä	N61 51	E024 17	2/96–1/97	CBL	Mid-latitudes	Rural	Land
24	Dal Maso et al. (2000)	Hyytiälä	N61 51	E024 17	96–99	CBL	Mid-latitudes	Rural	Land
25	Kulmala et al. (2000a)	Hyytiälä	N61 51	E024 17	3/99–4/99	CBL	Mid-latitudes	Rural	Land
26	Mäkelä et al. (2000)	Hyytiälä	N61 51	E024 17	1/96–9/99	CBL	Mid-latitudes	Rural	Land
27	Aalto et al. (2001)	Hyytiälä	N61 51	E024 17	4/98–5/98, 7/98–8/98, 3/99–4/99	CBL	Mid-latitudes	Rural	Land
28	Hämeri et al. (2001)	Hyytiälä	N61 51	E024 17	3/99–4/99	CBL	Mid-latitudes	Rural	Land
29	Kulmala et al. (2001)	Hyytiälä	N61 51	E024 17	04/99	CBL	Mid-latitudes	Rural	Land
30	Kulmala et al. (2003)	Hyytiälä	N61 51	E024 17		CBL	Mid-latitudes	Rural	
31	Salm and Tamm (2000)	Hyytiälä/ Tahkuse	N61 51	E024 17	3/99–4/99	CBL	Mid-latitudes	Rural	Land
32	Marti et al. (1997)	Idaho Hill, Rocky Mountains (Boulder, Colorado)	N40 00	W105 16	09/93	CBL	Mid-latitudes	Rural	Land
33	Leaitch et al. (1999)	Kejimkujik	N44 26	W065 12	07/96	CBL	Mid-latitudes	Rural	Land

Table 1 (*continued*)

Ref	Authors (year)	Place	Latitude	Longitude	Date	Air mass		Air mass type	Platform
34	Wehner and Wiedensohler (2002)	Leipzig, Germany	N52	E13	2/97–3/00	CBL	Mid-latitudes	Urban	Land
35	Hämeri et al. (1996)	Luukki	N60 18	E024 41	4/93–5/93	CBL	Mid-latitudes	Urban	Land
36	Väkevä et al. (2000)	Luukki	N60 18	E024 41	2/97–3/07	CBL	Mid-latitudes	Urban	Land
37	O'Connor and McGovern (1991)	Mace Head	N53 19	W009 54		CBL	Mid-latitudes	Coastal	Land
38	McGovern, Jennings, Connor, and Simmonds (1996)	Mace Head	N53 19, N58	W009 54	4/91	CBL	Mid-latitudes	Coastal	Land
39	O'Dowd, Hill, Smith, Geever, and Jennings (1998b)	Mace Head	N53 19	W009 54	6/96?	CBL	Mid-latitudes	Coastal	Land
40	McGovern (1999)	Mace Head	N53 19	W009 54	90–92	CBL	Mid-latitudes	Coastal	Land
41	Grenfell et al. (1999)	Mace Head	N53 19	W009 54	96–97	CBL	Mid-latitudes	Coastal	Land
42	O'Dowd et al. (1999)	Mace Head	N53 19	W009 54	05/97	CBL	Mid-latitudes	Coastal	
43	O'Dowd et al. (2000a)	Mace Head	N53 19	W009 54	9/98,6/99	CBL	Mid-latitudes	Coastal	Land
44	O'Dowd et al. (2000b)	Mace Head	N53.2–N54.2	W009.2–W010.2	06/99	CBL	Mid-latitudes	Coastal	Aircraft
45	Kulmala, Dal Maso, Mäkelä, and O'Dowd (2000b)	Mace Head	N53 19	W009 54		CBL	Mid-latitudes	Coastal	
46	O'Dowd (2001)	Mace Head	N53 19	W009 54	05/97	CBL	Mid-latitudes	Coastal	Land
47	O'Dowd et al. (2002a)	Mace Head	N53 19	W009 54		CBL	Mid-latitudes	Coastal	Land
48	Dal Maso et al. (2002)	Mace Head	N53 19	W009 54		CBL	Mid-latitudes	Coastal	Land

49	O'Dowd, Geever, Hill, Smith, and Jennings (1998a)	Mace Head/Outer Hebrides	N53 19, N58	W009 54 W7	96/94	CBL	Mid-latitudes	Coastal	Land
50	Williams et al. (1998)	Manchester	N53 30	W002 15	11/97–12/97	CBL	Mid-latitudes	Urban	Land
51	Williams et al. (2000)	Manchester	N53 30	W002 15	12/97	CBL	Mid-latitudes	Urban	Land
52	Birmili et al. (2000b)	Melpitz	N51 32	E012 56	3/96–8/97	CBL	Mid-latitudes	Rural	Land
53	Birmili, Wiedensohler, Heintzenberg, and Lehmann (2001b)	Melpitz	N51 32	E012 56	3/96–8/97	CBL	Mid-latitudes	Rural	Land
54	Mihalopoulos et al. (1992)	Penmarch (Brittany)	N47 47	W006 40	06/89	CBL	Mid-latitudes	Coastal	Land
55	Stanier et al. (2002)	Pittsburgh, USA				CBL	Mid-latitudes	Urban	Land
56	Zeromskiene, Ulevicius, and Mordas (2000)	Preila	N55 21	E021 04	6/97–8/97	CBL	Mid-latitudes	Coastal	Land
57	Ulevicius, Mordas, and Plauskaite (2002)	Preila	N55 21	E021 04	7/97–8/97	CBL	Mid-latitudes	Coastal	Land
58	Koutsenogii and Jaenicke (1994)	Siberia:Lake Baikal/ Novosibirsk	N53, N55	E104, E83	7/91–8/91, 6/92–7/92	CBL	Mid-latitudes	Rural	Land
59	Horrak, Salm, and Tammet (1998)	Tahkuse	N58 31	E024 56	9/93–10/94	CBL	Mid-latitudes	Rural	Land
60	Horrak, Salm, and Tammet (2000)	Tahkuse	N58 31	E024 56	9/93–10/94	CBL	Mid-latitudes	Rural	Land
61	Winklmayer (1987)	Vienna	N48 15	E016 25	85–86	CBL	Mid-latitudes	Urban	Land
62	Harrison et al. (2000)	Weyborne	N52 57	E001 7	06/95	CBL	Mid-latitudes	Rural	Land
63	Coe et al. (2000)	Weyborne	N52 57	E001 7	06/98	CBL	Mid-latitudes	Rural	Land
64	McMurry, Woo, Weber, Chen, and Pui (2000)					CBL	Mid-latitudes	Urban	Modelling type

Table 1 (*continued*)

Ref	Authors (year)	Place	Latitude	Longitude	Date	Air mass		Air mass type	Platform
65	Woo et al. (2001)	Atlanta	N33 45	W084 23	8/98–8/99	CBL	Subtropic	Urban	Land
66	Kavouras, Mihalopoulos, and Stephanou (1999b)	Pertouli, Greece	N39 32	E021 20	7/97–8/97	CBL	Subtropic	Rural	Land
67	Weise, Birmili, Wiedensohler, and Covert (1998)	Sagres/ Portugal	N37	W9	6/97–7/97	CBL	Subtropic	Coastal	Land
68	Misaki (1964)	Socorro	N34 02	W106 54	63	CBL	Subtropic	Rural, desert	Land
69	Kavouras et al. (1998)	Tabua, Portugal	N40 21	W008 02	08/96	CBL	Subtropic	Rural	Land
70	Kavouras et al. (1999a)	Tabua, Portugal	N40 21	W008 02	08/96	CBL	Subtropic	Rural	Land
71	Andreae et al. (1992)	Congo	N01 27	E018 04	02/88	CBL	Tropic	Rural	Aircraft/ balloon
72	Baumgartner et al. (2000)	Mexico City	N19 15	W099 11	11/97	CBL	Tropic	Urban plume	Land
73	Zhou, Swietlicki, Hansson, and Artaxo (2002)	Balbina, Brasil	S1	W59	3–4/98	CBL	Tropic	Rural, rain forest	Land
74	Baltensperger et al. (1997)	Jungfraujoch	N46 33	W007 59	4/92–5/92	CBL & FT	Mid-latitudes	Remote rural	Land
75	Weingartner, Nyeki, and Baltensperger (1999)	Jungfraujoch	N46 33	W007 59	3/97–5/98	CBL & FT	Mid-latitudes	Remote rural	Land
76	Brock et al. (2002, 2003)	Eastern USA	N31–40	W86–94	7/99	FT	Mid-latitudes	Exhaust plume/ industrial	Aircraft
77	Brock et al. (2003)	Houston, Texas	N29 –N30.5	W94.5 –W96	8/2000 –9/2000	FT	Mid-latitudes	Exhaust plume/ industrial	Aircraft
78	Nyeki et al. (1999)	Jungfraujoch	N46 33	W007 59	07/97	FT	Mid-latitudes		Aircraft
79	Weber et al. (2001b)	South Pacific	S40–S60	E145 –E159	11/95	FT	Mid-latitudes	Cloud interaction	Aircraft

80	de Reus et al. (2000)	Tenerife	N29	W018	07/97	FT	Mid-latitudes		Aircraft
81	Schröder and Ström (1997)	Western Europe	N51–N55	W004 –E011	07/94	FT	Mid-latitudes		Aircraft
82	Twohy et al. (2002)	Wisconsin	N42–N47	W88–W91	5/96	FT	Mid-latitudes		Aircraft
83	Clarke et al. (1999b)	Pacific	N20–S20	W100 –W160	8/96–10/96	FT	Tropic		Aircraft
84	Clarke (1993)	Pacific Ocean	N75–S60	W120 –E130	5/90–6/90	FT	Tropic & Mid-latitudes		Aircraft
85	Petzold, Döpelheuer, Brock, and Schröder (1999)	Germany				FT		Exhaust plume/ aircraft	Aircraft
86	Brock et al. (2000)					FT		Aircraft exhaust plume	Aircraft
87	de Reus et al. (1998)	Ireland	N50 –N58	W000 –W015	5/96–6/96	FT & Lower S	Mid-latitudes		Aircraft
88	Wiedensohler et al. (1996)	Arctic ocean	N70 –N90		8/91–10/91	MBL	Arctic		Ship
89	Covert et al. (1996b)	Arctic ocean	N70 –N90		8/91–10/91	MBL	Arctic		Ship
90	Leck and Bigg (1999)	Arctic ocean	N85 –N87		7/96–8/96	MBL	Arctic		Ship
91	Aalto and Nilsson (2003)	Arctic ocean			19,911,996	MBL	Arctic		
92	Weber et al. (1998)	Macquarie Island	S54 30	E159	11/95– 12/95	MBL	Mid-latitudes	Biogenic emissions	Aircraft+ land
93	Hoppel et al. (1994)	Oregon coast	N44	W125	08/92	MBL	Mid-latitudes		Airship
94	Perry and Hobbs (1994)	Pacific Ocean (off Oregon coast)	N44	W130	11/92	MBL	Mid-latitudes	Cloud interaction	Aircraft
95	Bates et al. (1998)	South of Australia	S40–S55	E135 –E160	11/95 –12/95	MBL	Mid-latitudes		Ship
96	Hegg, Radke, and Hobbs (1991)	Washington state coast	N40 –N55	W125 –W135	6/89, 4/90	MBL	Mid-latitudes		Aircraft
97	Covert, Kapustin, Quinn, and Bates (1992)	Washington state coast	N40 –N55	W125 –W135	4/91–1/91	MBL	Mid-latitudes		Ship

Table 1 (*continued*)

Ref	Authors (year)	Place	Latitude	Longitude	Date	Air mass		Air mass type	Platform
98	Hegg, Covert, and Kapustin (1992)	Washington state coast	N40 –N55	W125– W135	04/91	MBL	Mid-latitudes		Ship/ aircraft
99	Weber et al. (1995)	Mauna Loa, HI	N19 32	W155 34	6/28/92– 7/27/92	MBL	Subtropic	Volcanic emissions	Land
100	Ito (1980)	Minamito-rishima/ Chichijima Is.	N24, N27	E154, E142	10/75– 11/75, 10/76– 11/76	MBL	Subtropic		Land
101	Van Dingenen, Raes, and Jensen (1995)	North Atlantic	N25 –N45	W010 –W065	9/92–10/92	MBL	Subtropic		Ship
102	Hoppel, Fitzgerald, Frick, Larson, and Mack (1990b)	North Atlantic	N20 –N55	W000 –W80	3/83–4/84	MBL	Subtropic & Mid-latitudes		Ship
103	Hegg, Radke, and Hobbs (1990)	Southern California coast Washington state coast	N23, N40 –N55	W112, W125 –W135	7/87 6/89	MBL	Subtropic & Mid-latitudes	Cloud interaction	Aircraft
104	Clarke et al. (1998b)	Coast of Ecuador/ Panama	N08	W085– W086	09/96	MBL	Tropic		Aircraft
105	Weber et al. (2001a)	Hawaii-Tahiti	N21–S22	W194 –W220	2/99–3/99	MBL	Tropic		Aircraft
106	Covert, Kapustin, Bates, and Quinn (1996a)	Mid-Pacific	N55–S70	W060 –W150	92–93	MBL	Tropic & Mid-latitudes		Ship
107	Hoppel and Frick (1990a)	Seattle–Hawaii–Tahiti	N20–S55	W150	2/84–5/84	MBL	Tropic & Mid-latitudes		Ship
108	Zaizen, Ikegami, Tsutsumi, Makino, and Okada (1996)	Pacific Ocean (Australia to Japan)	N38–S38	E140 –E154	01/94	MBL	Tropic & Subtropic		Aircraft
109	Clarke et al. (1999a)					MBL			Review
110	Weber et al. (1999)					MBL			Review

111	Weber et al. (1996)	Mauna Loa, Colorado Rockies	N19 32, N39 59	W155 34, W105 35	6/28/92–7/27/92; 90/5/93–9/29/93	MBL & CBL	Subtropic & Mid-latitudes	Refers to 2 other papers	Land
112	Clarke et al. (1998a)	South of Australia and the Equator	S40–S44	E140 –E147	11/95 –12/95	MBL & FT	Mid-latitudes		Aircraft
113	Raes, Van Dingenen, Cuevas, Van Velhoven, and Prospero (1997)	Tenerife	N28 18	W016 30	07/94	MBL & FT	Subtropic	Coastal	Land
114	de Reus et al. (2001)	Indian Ocean	N10–S10	E67–E80	2/99–3/99	MBL & FT	Tropic		Aircraft
115	Clarke and Kapustin (2002)					MBL & FT			Review
116	McMurry (2000a,b)								Review
117	Shi and Qian (2003)	St. Louis	N38.6	W90.2	4/01–4/02	CBL	Mid-latitude	Urban	Land
118	Brock, Hamill, Wilson, Jonsson, and Chan (1995)				87–94	UT & lowerS	Tropic, mid-latitudes & polar		Aircraft
119	Ferek, Hobbs, Radke, and Herring (1995)	Barrow/ Deadhoese, Alaska	N71.18	W156.47	6/90–5/92	MBL	Arctic	Coastal	Aircraft + land
120	Hoffman (1993)	Laramie, Wyoming	N41	W105	1971–1990	T	Mid-latitudes	Rural	Balloon
121	Radke and Hobbs (1991)	Cascade Mnts, Washington,	N45–50 N45–50	W120 W120	May 11, 1989	FT	Mid-latitudes	Rural	Aircraft
122	Thornton, Bandy, Blomquist, Bradshaw, and Blake (1997)	Western Pacific Ocean	N38–S38	E140 –E154	1991,1994	MBL & FT	Tropic & Sub-tropic		Aircraft
123	Wang et al. (2000)	North Atlantic	N20 –N70	W80–E10	10–11/97	UT	Mid-latitudes		Aircraft
124	Davison et al. (1996)	Antarctic	S68–S73	0–W20	12/92	MBL	Antarctic	Remote	Ship

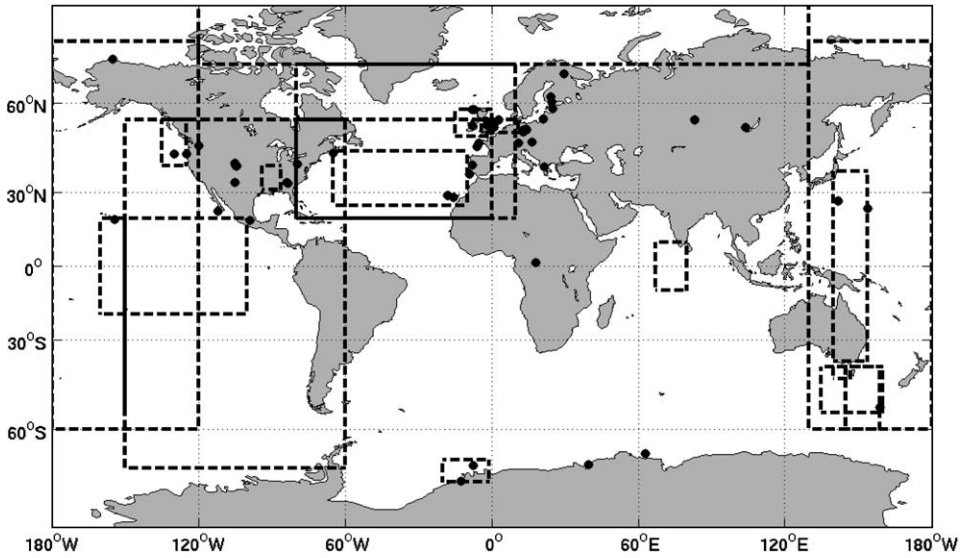

Fig. 1. Global map. The dots indicate observation sites, the dashed lines and rectangles indicate regions where airborne or ship observations have been made.

3. Instrumentation

Studies of atmospheric particle formation and growth require measurements of nucleation mode particles (< 20 nm). Simultaneous measurements of nucleating gases can provide further insights into mechanisms. Here, we give a brief summary of the relevant methods, their characteristics, and limitations. For more detailed and historical aspects of aerosol measurement technology, the reader is referred to the rich body of literature on the subject (e.g. McMurry, 2000a, b; Flagan, 1998).

Particle formation and growth rates can be inferred from measurements of nanoparticle size distributions. The following should be considered when selecting measurement strategies:

- detection of small particles (current limit is ca. 3 nm, but smaller would be better);
- time resolution ~ 10 min for ground-based or ship measurements, or between about a second and minute for aircraft measurements;
- size resolution involving multiple channels in the 3–20 nm range in order to detect a possible growth of particles after nucleation;
- the ability to measure low nucleation mode concentrations (< 500 cm^{-3}), such as are found in clean and remote atmospheres;
- the ability to measure high nucleation mode concentrations ($> 10^5$ cm^{-3}), such as are found during intense nucleation bursts occurring in coastal and continental environments.

Measurements that provide information on the concentrations of the nucleating gases (or their precursors) and the composition of freshly nucleated particles provide further insights. A technique

for the measurement of gas phase sulphuric acid at concentrations down to about 10^4 cm^{-3} is available (Eisele & Tanner, 1993), and techniques for measuring ammonia in the ppt range with high time resolution have recently been deployed. The hygroscopicity and volatility of freshly nucleated particles can be measured with the nano-TDMA (Hämeri et al., 2001); such measurements provide constraints on the composition of growing particles. Also, progress on measurements of the composition of sub-10 nm particles has recently been reported (Voisin, Smith, Sakurai, McMurry, & Eisele, 2003). Information on the charging state of nucleated particles can help to differentiate ion-induced from other nucleation mechanisms.

3.1. Condensation nucleus counter (CNC)

The laminar flow CNC is the instrument most widely used to measure atmospheric particle number concentrations. Its basic working principle is that the sampled aerosol flows over a warm reservoir of a working fluid where it becomes saturated with a condensable vapour (Agarwal & Sem, 1980). During subsequent cooling in a condensor the vapour becomes supersaturated, causing particles to grow into large liquid droplets of ca. 10 μm in size, which are individually detected by light scattering. CNCs detect the particles larger than a particular cut-off size, which is a function of the supersaturation achieved in the condensor section of the CNC. A particular improvement regarding the lowest detectable size was made by wrapping the particle sample flow in a saturated sheath flow, thus activating 50% of all 3 nm particles (UCPC Model 3025, TSI Inc., St. Paul, Minnesota; Stolzenburg & McMurry, 1991). The size dependent collection efficiencies of various commercial types of CNCs have been compared, e.g., in Wiedensohler et al. (1997a, b). As a CNC detects single particles it is able to detect low particle concentrations. Operating two CNCs in parallel, each having different lower cut-off diameters, enables the measurement of nucleation mode number concentrations in a specified size range (e.g., 3–10 nm) by subtracting their readings. The high time resolution that can be achieved with this method (1 s) makes it a preferred choice for deployment on mobile platforms, such as an aircraft. CNCs have been used in almost all studies presented in this overview.

3.2. Pulse height analysis (PHA)

Measurements have shown that within laminar flow CNCs, the final droplet size after condensation decreases with decreasing size for particles smaller than 10 nm (Saros, Weber, Marti, & McMurry, 1996). This size-dependent growth can be used to infer size distributions of sub-10 nm particles. Such measurements are carried out by measuring the "pulse height" produced by the optical detector in the CNC. Pulse heights decrease with decreasing size; size distributions are obtained by mathematically "inverting" measured pulse height distributions (Weber et al., 1998). Measurements have shown that particles larger than 10 nm all grow to the same final size, so the PHA technique can only provide information on size distributions of sub-10 nm particles. The drawback of the PHA is that the resolution of particle size distribution is not as good as obtained with SMPS or DMPS systems (Wiedensohler, Aalto, Covert, Heintzenberg, & McMurry, 1994). A recent methodological development involved a laboratory calibration of the PHA system with nanoparticles of various compositions, including pure ionic and organic compounds. It was found that the final droplet size of pure organic nanoparticles of a given size was larger than the final droplet size of ionic particles of

the same initial size. This observation was used to conclude that newly formed particles in Hyytiälä behave more like pure organic particles than like ionic particles (O'Dowd et al., 2002b).

3.3. Electromobility classification

Operating an electrical classifier upstream of a CNC enables the measurement of particle size distributions. Differential mobility analysers (DMA) segregate particles in an electrical field, and yield particles of a narrow monodisperse electrical mobility (Knutson & Whitby, 1975). A particle's electrical mobility varies in proportion to its electrical charge and inversely with its Stokes' diameter. Mobility distributions are obtained by using a CNC to measure the concentration downstream of a DMA for a range of classifying voltages. Particle size distributions are obtained from such measurements by carrying out a mathematical inversion that takes account of the size-dependent distribution of charges on particles (e.g., Alofs & Balakumar, 1982). DMAs are available in various designs, with recent developments focussing on a more efficient transmission of the smallest sizes < 10 nm (Winklmayr, Reischl, Linde, & Berner, 1991; Chen et al., 1998). A frequently used instrumental set-up of a Differential Mobility Particle Sizer (DMPS) in ground-based or ship-based experiments involves two DMAs covering a wide size range, such as 3–700 nm, and two separate CNCs to count particles (e.g., Birmili, Stratmann, & Wiedensohler, 1999a; Aalto et al., 2001). The time required to measure an atmospheric aerosol size distribution depends primarily on the time required to obtain a statistically significant number of CNC counts at each classifying voltage. A measurement period of often 10 min provides a viable compromise between size resolution, time resolution, and particle counting statistics for most atmospheric applications. DMA–CNC systems may also be operated as scanning mobility particle sizers (SMPS; Wang & Flagan, 1990), whereby particle concentrations are measured as the classifying voltage is increased at a continuous rate. SMPS scan times as short as 2 min are possible, albeit in a trade-off against sizing accuracy and particle counting statistics.

An alternative class of instruments based on electric mobility analysis are air ion mobility spectrometers (e.g., Misaki, 1964; Horrak et al., 1998, and references therein). Ion mobilities are segregated very similarly as in a DMPS, but an array of electrometers is typically used to simultaneously measure the various mobility fractions. Unlike DMPS and SMPS systems, which utilise bipolar chargers to bring the aerosol to Boltzmann equilibrium before they are classified by the DMA, ion mobility spectrometers measure naturally occurring mobility distributions. Ion mobility spectrometers can detect charged particles of any size, extending down to the range of molecular ions (ca. 0.4 nm). A limitation is that the sensitivity of electrometers limits the lowest detectable particle concentration to $\sim 50\ \mathrm{cm}^{-3}$.

3.4. Future needs

The body of available experimental studies suggests that a full understanding of atmospheric new particle formation processes depends on further instrumental improvements. It would clearly be an advantage to be able to count neutral particles smaller than 3 nm. Further needs address the determination of physico-chemical properties (e.g., solubility in different solvents), and the chemical composition of nucleation mode particles. Measurements of gas phase species that participate in nucleation and growth are also essential.

4. Formation and growth rates of atmospheric aerosol particles

4.1. Estimation of the particle formation and growth rates

Critical clusters formed by atmospheric nucleation events cannot yet be measured quantitatively due to instrumental limitations. Only one measurement of clusters during nucleation events has been reported, and it showed that clusters were present when 2.7–4 nm particles were detected (Weber et al., 1995). More work on the distribution and composition of such clusters is needed to refine our understanding of atmospheric nucleation.

Because critical clusters cannot yet be measured, we are unable to measure the true atmospheric nucleation rate but rather the formation rate of particles of some larger diameter D. The diameter D corresponds typically to the CNC detection limit, which is presently 3 nm or greater.

Mathematically, the particle formation rate, J_D, is equal to the flux of particles past the size D because of their growth:

$$J_D(t) = \left.\frac{\mathrm{d}n(D_p,t)}{\mathrm{d}D_p}\right|_D \times \left.\frac{\mathrm{d}D_p}{\mathrm{d}t}\right|_D . \tag{1}$$

Here t is the time and $n(D_p,t)$ is the particle number size distribution. In order to apply Eq. (1), both the particle number size distribution function and particle growth rate at the size D must be known. This kind of information is rarely available.

Rather than estimating an instantaneous particle formation rate $J_D(t)$, one usually averages J_D over some time interval Δt. The most frequently used selection for Δt is the duration of the particle formation event, although shorter time intervals are also sometimes used. After time averaging, we obtain

$$\left.\frac{\Delta N_{D,D_{\max}}}{\Delta t}\right|_{\text{observed}} = J_D - \left.\frac{\Delta N_{D,D_{\max}}}{\Delta t}\right|_{\text{self-coag}} - \left.\frac{\Delta N_{D,D_{\max}}}{\Delta t}\right|_{\text{coag-scav}} - \left.\frac{\Delta N_{D,D_{\max}}}{\Delta t}\right|_{\text{transport}}, \tag{2}$$

where $N_{D,D_{\max}}$ is the total particle number concentration in the size range $[D, D_{\max}]$ and $D_{\max}$ is the maximum size the critical clusters can reach because of their growth during Δt. The first term in Eq. (2) is the observed change in $N_{D,D_{\max}}$ during Δt and can be obtained from particle size distribution or number concentration measurements. The second and third terms in the right hand side represent the loss of particles in the size range $[D, D_{\max}]$ by self-coagulation and coagulational scavenging to larger pre-existing particles (particles with sizes $D_p > D_{\max}$), respectively. The last term in Eq. (2) represents the influence of air mass transport to $N_{D,D_{\max}}$. This term may become important in fixed-site measurements exposed to inhomogeneous air masses, or in systems subject to strong dilution.

When the effects of both coagulation and transport are small compared with particle production, Eq. (2) reduces to

$$J_D \approx \left.\frac{\Delta N_{D,D_{\max}}}{\Delta t}\right|_{\text{observed}} . \tag{3}$$

This is perhaps the most common formula used to estimate an atmospheric particle formation rate. Eq. (3) is often an excellent approximation to Eq. (2) in relatively clean, homogenous air masses. However, it may significantly underestimate the true particle production rate when the nuclei particle

number concentration is very high ($> 10^5$ cm^{-3}). Also if the pre-existing particle concentration is very high J_D values are underestimated.

Estimating the particle growth rate, GR, requires information on the time evolution of the mean diameter of the new particle (nucleation) mode, D_m. With this information, GR can be calculated from the formula

$$\mathrm{GR} = \frac{\Delta D_m}{\Delta t}, \tag{4}$$

where D_m belongs to the size range $[D, D_{max}]$. Eq. (4) fails in the case of high continuous nucleation producing new particles during the particle growth (see e.g. Clement, Ford, Twohy, Weinheimer, & Campos, 2002; O'Dowd et al., 1999). Frequently, suitable data on particle number size distributions to calculate GR are not available. In this case GR can be estimated if one has information on the time evolution of nucleating precursors and the total nuclei number concentration:

$$\mathrm{GR} = \frac{D - D_{crit}}{t_{prod} - t_{nuc}}. \tag{5}$$

Here D_{crit} is the critical size of nucleated particles t_{prod} is the time at which particle formation is observed to begin, and t_{nuc} is the time nucleation is assumed to have begun. The quantity D_{crit} is obtained from a nucleation theory, while t_{nuc} is typically set equal to the time at which the concentration of the assumed nucleating precursors such as sulphuric acid is observed to increase. Eq. (5) can be applied to systems having a regular diurnal cycle for both nanoparticles and nucleating precursors. Due to the many necessary assumptions, however, growth rates determined using Eq. (5) are subject to significant uncertainties.

4.2. Observations

Due to instrumental limitations we cannot observe the very smallest ultrafine particles (less than 3 nm in diameter) during nucleation bursts. Therefore, we assume that we are not discussing nucleation directly. Usually particles start to be observed at an instrumental detection limit of round 3–5 nm, which means that they have already grown for some time starting from the size of a nucleated cluster that is around 1 nm. In the case of J_{10} and J_{15} the needed growth time is much longer. Ion mobility spectrometer data can be used to infer formation rates of charged particles smaller than 3 nm (e.g. Horrak et al., 1998) when ion-induced nucleation is occurring or to detect charged fraction of growing stable neutral clusters.

It is also worth noting that the evolution towards larger particle sizes seen in the particle size spectra during the particle formation process is always interpreted as a particle growth process. Since most measurement spots are actually fixed, they record the data in an "Eulerian system". This interpretation of growth already includes the assumption that the aerosol must be quite homogeneous in a larger-scale air mass.

An example of a typical particle formation event measured by a DMPS is shown in Fig. 2 as a contour plot (Mäkelä et al., 1997; Mäkelä et al., 2000). The ordinate of Fig. 2 is particle diameter and the abscissa is time of day. The grey-scale shading indicates the value of the particle size distribution. Before midday newly formed particles with diameters between about 4 and 10 nm enter the detectable size range, after which they grow at a rate of a few nm h^{-1}, reaching sizes between about 20 and 50 nm by the evening.

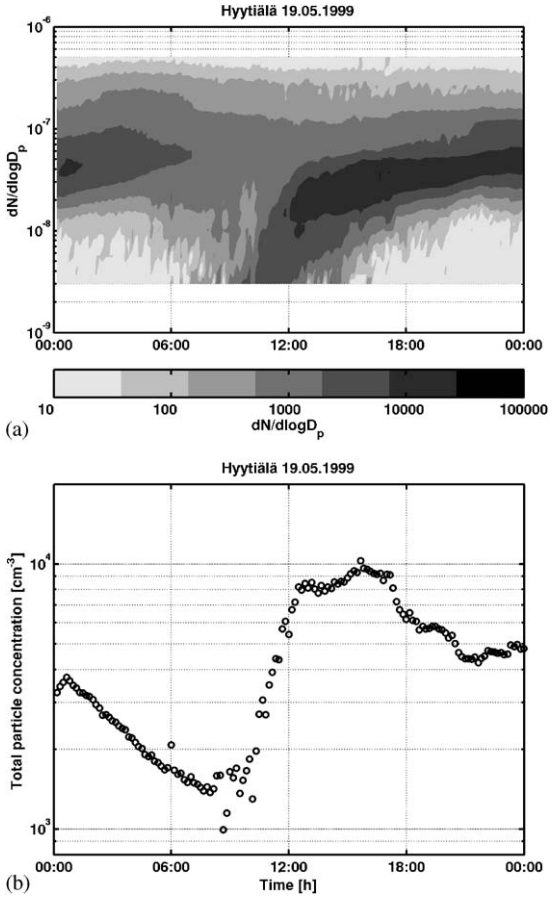

Fig. 2. Typical particle formation event in Hyytiälä boreal forest site on 13th March 1996: particle size distribution data as a surface plot (a), and total particle number concentration versus time (b).

From the data sets similar to Fig. 2 it is possible to estimate the quantities Δt, $\Delta N_{D,D_{\max}}$ and ΔD_{m}, after which both J_D and GR can obtained using Eqs. (3) and (4), respectively. The main problem in this approach is to distinguish between new and pre-existing particles. If there are plenty of background particles, or if the size distribution fluctuates much, it is rather difficult to pinpoint which size classes belong to the event mode and one has to make an educated guess. Also the start and the end of an event are often difficult to determine because of fluctuations in the number concentrations in the smallest DMPS-channels. These uncertainties could result in rather large errors in the calculated values.

Fig. 3 shows an example of mean size of nucleated particles versus time of day. The diameter growth rate is the slope of this line. The data in Fig. 3 show that particle growth rates are remarkably constant during daylight hours. Such constant growth rates appear to be typical during regional nucleation events.

Table 2 summarises observed particle formation and growth rates. Most of the data are from literature, in addition to which our own unpublished data have been used. The particle formation

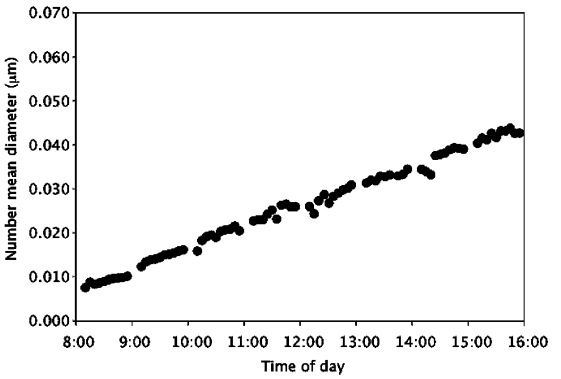

Fig. 3. Mean size versus time of day during a regional nucleation event in St. Louis on September 1, 2001 (Shi and Qian, 2003).

rates have been divided into three subgroups: J_3, J_{10} and J_{15}, which correspond to the formation rates of 3, 10 and 15 nm particles, respectively. The number of observed particle formation events is also included. This number gives information on the data reliability: the higher the number the more reliable the data can be considered. In practise we have more than 60 studies from which the formation rate can be estimated, and correspondingly more than 50 studies for the growth rate. From all other studies we can conclude that aerosol formation has been observed. However, no quantitative numbers for formation or growth rates can be estimated.

It appears that several types of atmospheric processes lead to particle formation. Fig. 2 shows an example of a "regional nucleation event." During such events the growth of nucleated particles continues throughout the day, regardless of the wind direction. Several studies using multiple measurement stations (Kulmala et al., 2001; Stanier et al., 2002) have shown that such events can occur more or less uniformly in air masses that extend over distances of hundreds of kilometers. Nucleation in the outflows of convective clouds are likely to be of global significance due to the large volumes of air near the inter-tropical convergence zone where such nucleation events are routinely observed (Clarke, 1993; Clarke, Li, & Litchy, 1996; Clarke et al., 1998a, 1999b). Similar phenomena have also been observed in outflows of mid-latitude convective storms (Twohy et al., 2002).

Intense nucleation bursts have also been observed over the exposed surface zone at Mace Head (O'Dowd, 2001; O'Dowd et al., 2002a) and in industrial plumes that contain SO_2 (Woo et al., 2001; Brock et al., 2002, 2003). During most observed nucleation bursts, the maximum in the particle size distribution occurred at sizes below 15 nm , thus indicating their recent formation. Observations from the ground of the Amazon rain forest (Zhou, Swietlicki, Hansson, & Artaxo, 2002) report the regular occurrence of newly formed particles at relatively large diameters (about 30 nm), which could be indicative of an abundant vapour reservoir in that region, being responsible for the rapid particle growth prior to observation. With only one exception (Wiedensohler et al., 1997a), observed nucleation events always occur during daytime, suggesting that photochemistry plays a central role in this process.

Typical observed formation rates of 3-nm particles, J_3, in boundary layer regional nucleation events are in the range 0.01–10 cm^{-3} s^{-1}. Rates higher than this (up to 100 cm^{-3} s^{-1}) are often observed in urban areas. Very high values of J_3 are observed over coastal zones (10^4–10^5 cm^{-3} s^{-1}) and in

Table 2

Ref.	Authors (year)	# events	J_3 (cm^{-3} s^{-1})	J_{10} (cm^{-3} s^{-1})	J_{15}(cm^{-3} s^{-1})	GR (nm h^{-1})
1	Koponen (2003)	Couple	0.5			1–2
3	Gras (1993)					0.1
4	Park et al. (2003)	1				0.13
5	Ito (1993)			3.8×10^{-4} (needed to eplain observations)		
8	Pirjola et al. (1998)	155				1.9–3.8
9	Dal Maso (2002)	141		J_7: 0.38(+/−0.07)		1–10
10	McMurry et al. (2003)	23	20–70 (regional)			2–6
11	Shi et al. (2001)	1	5–50	—	—	4
12	Weber et al. (1997)	9	0.1–1			0.5–2
13	Keil and Wendisch (2001)	5	250	110	—	—
14	Neusüss et al. (2002)	7	5.7(+/ − 1.1)			3.5(+/ − 1.0)
15	Wiedensohler et al. (1997a)		0.1			∼ 1
16	Birmili et al. (1999a)	1	20(+/ − 10)			∼ 20
17	Verheggen and Mozurkewich (2002)	1		ca 5–40		4
18	Birmili et al. (2000b)	1	3			2.1
19	Birmili et al. (2003)	117	1.11(0.01–8.8)			2.61(0.5–8.41)
20	Birmili et al. (2001a)	10	1.41(+/ − 0.3)			3.0(+/ − 1.1)
21	Mäkelä et al. (1997)	Roughly 50 (3)	∼ 3			3.3
22	Kulmala et al. (1998)	55 (10)	3–6			3–20
23	Mäkelä et al. (1999)	2	0.4–0.5			2–3
24	Dal Maso et al. (2000)	50–60	0.1–1			Median=5, range 2–8
25	Kulmala et al. (2000a)		1–2			2.2–2.8
26	Mäkelä et al. (2000)	184	0.001–1			1–17
27	Aalto et al. (2001)	30	0.002–0.6			3–4
29	Kulmala et al. (2001)	3?	1			2.2–10.5
30	Kulmala et al. (2003)		0.1–3.4			1.3–5.0
31	Salm and Tamm (2000)		J1.9: 0.05–0.1			3.5–5
32	Marti et al. (1997)		0.1			
33	Leaitch et al. (1999)			0.3		5.0–10
34	Wehner and Wiedensohler (2002)	105	13(+/ − 1.2)			
36	Väkevä et al. (2000)	3				4–6
37	O'Connor and McGovern (1991)			0.4		
38	McGovern et al. (1996)	5				
39	O'Dowd et al. (1998b)		10^3–1.5×10^4			8
40	McGovern (1999)	8	10–10000			
41	Grenfell et al. (1999)		100–10000			
42	O'Dowd et al. (1999)		1000–10000			20
43	O'Dowd et al. (2000a)	25	20000–100000			
44	O'Dowd et al. (2000b)					10
45	Kulmala et al. (2000b)		300–60000			8–10
47	O'Dowd et al. (2002a)	600	1e4–1e5			15–200
48	Dal Maso et al. (2002)		400–1e5			15–200
49	O'Dowd et al. (1998a)		1000–15000			8
50	Williams et al. (1998)		≫0.2			

Table 2 (*continued*)

Ref.	Authors (year)	# events	J_3 (cm^{-3} s^{-1})	J_{10} (cm^{-3} s^{-1})	J_{15}(cm^{-3} s^{-1})	GR (nm h^{-1})
52	Birmili and Wiedensohler (2000a)	151	11.1 (0.06–74)	—	—	< 10 nm: 4.1(1.1–6-1)
53	Birmili et al. (2001b)	151	11.1 (0.06–74)	—	—	< 10 nm: 4.1(1.1–6-1) 2.1
54	Mihalopoulos et al. (1992)	10			16	
55	Stanier et al. (2002)	42				4–5
56	Zeromskiene et al. (2000)			1–1.5		
57	Ulevicius et al. (2002)			30–40		8–12
58	Koutsenogii and Jaenicke (1994)				1.0(J_{tot})	
59	Horrak et al. (1998)	4	0.1–1.0			4–5
60	Horrak et al. (2000)	101	0.1–1.0			
61	Winklmayer (1987)	2	2.5			
62	Harrison et al. (2000)	4	0.91			1.3
63	Coe et al. (2000)	6	< 0.05			2.6–8
65	Woo et al. (2001)	23	10–15		—	—
66	Kavouras et al. (1999b)		3–5			
67	Weise et al. (1998)	8	~ 2			~ 4
68	Misaki (1964)	1			0.5	8.9
69	Kavouras et al. (1998)					~ 2
70	Kavouras et al. (1999a)			0.9		3
74	Baltensperger et al. (1997)	1–2		0.5		
75	Weingartner et al. (1999)	4		0.14		3–4.5
78	Nyeki et al. (1999)				10–100	
79	Weber et al. (2001b)					> 0.33
80	de Reus et al. (2000)	1				
83	Clarke et al. (1999b)					~ 0.4
91	Aalto and Nilsson (2003)					0.2
92	Weber et al. (1998)		0.5–10			2–5
95	Bates et al. (1998)	13				1
97	Covert et al. (1992)	1–2	0.15–0.17			
98	Hegg et al. (1992)	1	0.7			
99	Weber et al. (1995)		~ 0.5			Lower limit: 0.003–1
102	Hoppel et al. (1990b)			0.6		
105	Weber et al. (2001a)					1,5–6
107	Hoppel and Frick (1990a)			0.6		
110	Weber et al. (1999)		0.5–10			
111	Weber et al. (1996)		0.5–10			2–5
112	Clarke et al. (1998a)					To detectable sizes few sec/hrs, to aitken size in hours/days.
113	Raes et al. (1997)					2–3
117	Shi and Qian (2003)	85	1–80 (regional events)			0.5–9
124	Davison et al. (1996)	3	3.2			

SO_2-laden industrial plumes. Typical particle growth rates are between 1 and 20 nm h^{-1}, although exceptions can be found. Some estimates in coastal areas give growth rates as high as 200 nm h^{-1}. The smallest reported growth rates are around 0.1 nm h^{-1} and have been observed in clean polar areas.

5. Discussion

The annual variations in growth rates during regional nucleation events in the Hyytiälä forest (Mäkelä et al., 2000), rural Hohenpeissenberg (Birmili et al., 2003), and urban St. Louis (Shi and Qian, 2003) are shown in Fig. 4. Note that in all locations the growth rates during the summer range from 4 to 10 nm h^{-1}. The growth rates during the winter are considerably lower (0.5–2.5 nm h^{-1}). The GR data in Table 2 show qualitatively that the rates are significantly lower at the poles than at mid-latitudes. When growth rates and gaseous H_2SO_4 concentrations were both measured during nucleation events, calculations show that H_2SO_4 condensation typically accounts for only 10 to 30% of the observed growth (Weber et al., 1997; Weber et al., 1998; Birmili et al., 2003).

Fig. 5 shows the fraction of days in each month of the year during which regional nucleation events were observed in the Hyytiälä forest, Hohenpeissenberg and St. Louis. Note that in all locations events are observed throughout the year, although there appear to be seasonal patterns. In Hyytiälä, frequencies peak in the March to May period and a smaller secondary peak is observed in September. Hohenpeissenberg shows the highest frequencies in spring and winter. In St. Louis, frequencies are the lowest during the winter (December–March) with no clear seasonal pattern during the rest of the year.

Fig. 6 shows measured concentrations of gaseous H_2SO_4 and 2.7–4 nm particles during a nucleation event in the Rocky Mountains. Note that the H_2SO_4 concentration began to rise immediately after sunrise and reached its maximum shortly before the noon. Concentrations of the ultrafine particles followed a similar pattern but with approximately a 1.5-h lag. Based on such observations, we conclude that there is an apparent association between particle formation and gaseous sulphuric acid. These observations are consistent with the hypothesis that H_2SO_4 participates in nucleation. However, since the chemical composition of nucleating clusters has not been measured thus far, no direct proof on the involvement of H_2SO_4 in this process is available.

Fig. 7 shows the relative acidity of sulphuric acid versus the relative humidity measured during nucleation events. The relative acidity is defined as the measured H_2SO_4 partial pressure divided by its saturation partial pressure above a liquid H_2SO_4 surface. Also shown are theoretical predictions for the critical relative acidity in case of the binary water–sulphuric acid and ternary water–sulphuric acid–ammonia nucleation. An ammonia concentration of 1 ppt was assumed when plotting the curve representing the ternary theory. Note that the data fall into two distinct groups: measurements made aloft in cloud outflows (open circles) and those made near or at the ground level (solid circles). The former are consistent with the binary theory, whereas the latter demonstrate nucleation to occur at much lower relative acidities, although exceptions to this pattern have been observed (Weber et al., 2001a). It is possible that other gases participating in nucleation, such as ammonia, are present close to the surface, which causes the difference. Although supported by recent findings by Kulmala et al. (2002), more frequent ammonia measurements during nucleation events at various locations are needed to verify this idea.

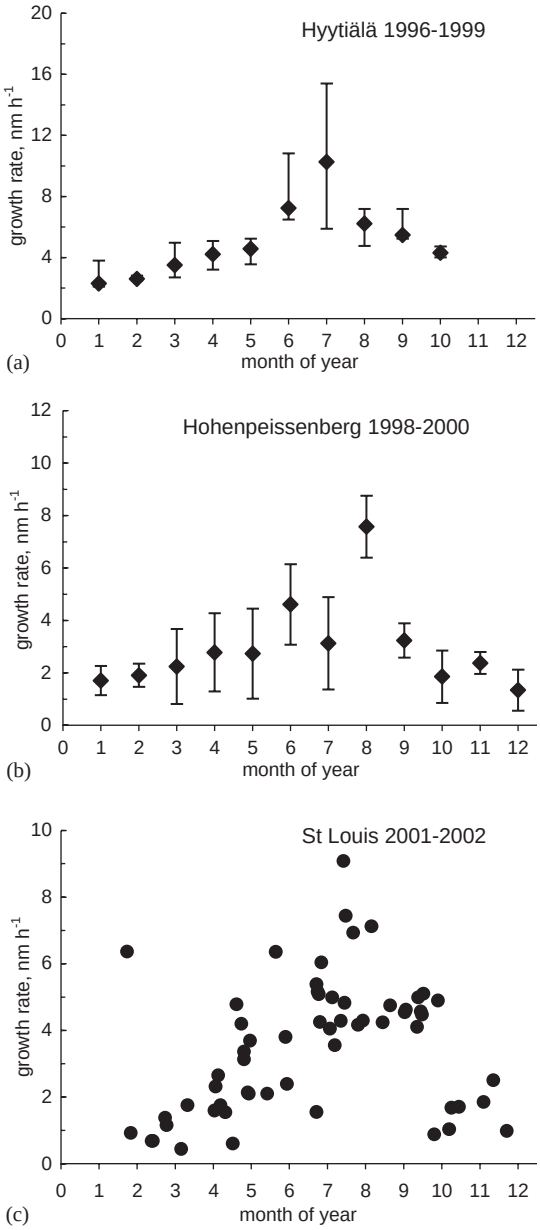

Fig. 4. Monthly variations of the growth rate during regional nucleation events in (a) Hyytiälä, (b) Hohenpeissenberg (Birmili et al., 2003), and (c) urban St. Louis (Shi and Qian, 2003).

In order to get some understanding on these observations, let us have a brief look at the primary factors affecting atmospheric nucleation events (Fig. 8). The driving force for both nucleation and particle growth is the concentration of "non-volatile" vapours. Potential candidates for such vapours are sulphuric acid and some yet unidentified organic compounds, all formed by oxidation reactions

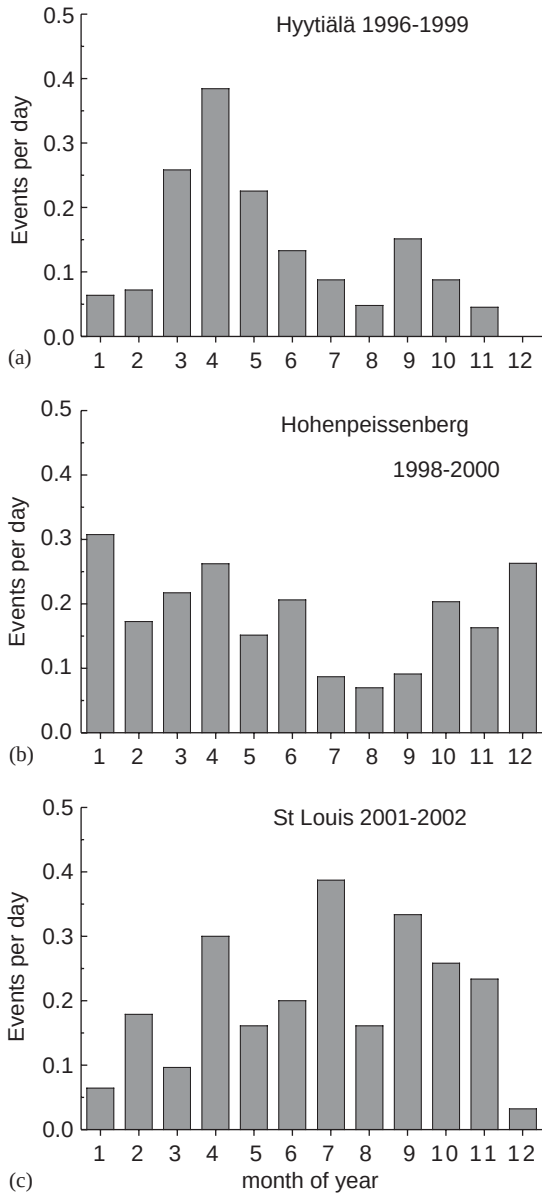

Fig. 5. Frequency of regional particle formation events versus month for (a) Hyytiälä (b) Hohenpeissenberg (Birmili et al., 2003), and (c) St. Louis (Shi and Qian, 2003).

involving suitable precursor gases (sulphur dioxide, volatile organic compounds). The "non-volatile" vapour concentrations are expected to be positively correlated with their precursor gas and oxidant (hydroxyl and nitrate radicals, ozone) concentrations, and negatively correlated with the pre-existing aerosol particle concentration. The real nucleation rate, J_{real}, as well as the particle growth rate, GR,

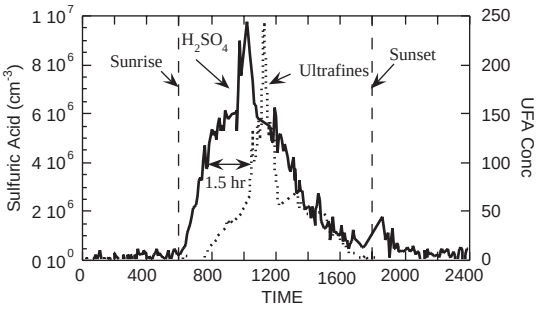

Fig. 6. Time series plot of sulphuric acid vapour and 2.7–4 nm particles during a nucleation event at Idaho Hill, CO on 21 September, 1993 (Weber et al., 1997).

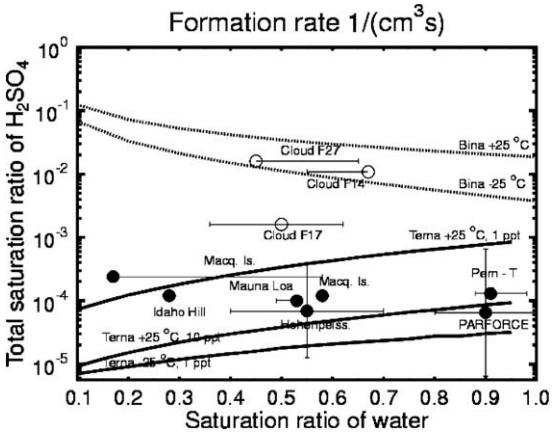

Fig. 7. Sulphuric acid relative acidity versus relative humidity during nucleation events in various locations. The data shown in open circles were measured aloft, while the solid circles were measured at or close to the surface. Both modelled ternary with 1 ppt ammonia and binary nucleation rates (1 cm^{-3} s^{-1}) are also indicated . See also Weber et al. (1999).

are both expected to be positively correlated with "non-volatile" vapour concentrations. As already discussed in Section 4, we are unable to measure J_{real} due to instrumental limitations and must rely on measurements of J_D. Under steady state conditions the value of J_D is smaller than J_{real}, since small (< 3 nm) clusters coagulate very effectively with pre-existing larger particles. Theoretical considerations indicate that J_D is expected to be positively correlated with both J_{real} and GR, and negatively correlated with the pre-existing aerosol particle concentrations (Kerminen, Pirjola & Kulmala, 2001; Kerminen & Kulmala, 2002).

From Fig. 8 we can see that J_D is not directly connected with precursor gas concentrations, nor even with "non-volatile" vapour concentrations. This explains why the association between observed particle formation rate and concentration of various trace gases, excluding sulphuric acid, has remained ambiguous. A very likely relation to be observed is an anti-correlation between J_D and the concentration of pre-existing aerosol particles. This is because larger pre-existing particle

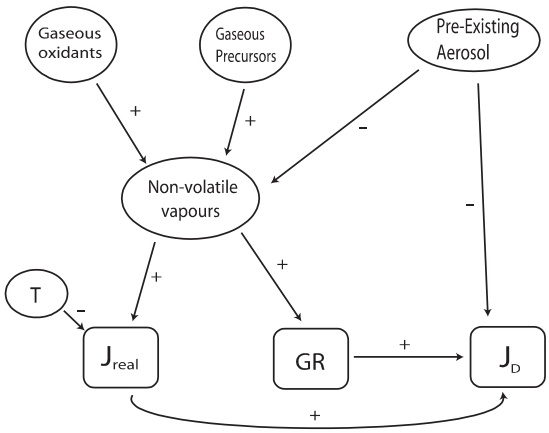

Fig. 8. Schematic picture on relations between the new-particle formation rate (J_{real} and J_D) and the concentrations of precursor gases, their oxidants and pre-existing aerosol particles. Plusses (minuses) indicate a positive (negative) correlation between the two quantities. Meteorological factors other than temperature (T) have not been included.

concentrations decrease J_D by accelerating the coagulational scavenging of small nuclei and by decreasing "non-volatile" vapour concentrations (which reduces both J_{real} and GR). However, if pre-existing particle and precursor gas concentrations are strongly correlated to each other, as might be the case in plumes, also a positive correlation between the particle formation rate and the concentration of pre-existing aerosol particles can be observed.

Since concentrations of "non-volatile" vapours are proportional to the concentration ratio between their precursor gases and pre-existing aerosol particles, the nucleation itself can take place in both clean and polluted environments. However, due to the strong coupling of J_D to both pre-existing particle concentrations and GR, we never observe these events unless the nuclei grow fast enough to avoid being scavenged by coagulation before reaching detectable sizes. For this reason, low particle growth rates of the order $0.1\ \mathrm{nm\ h^{-1}}$ can be observed only in very clean air (which have low pre-existing particle concentrations), such as the air encountered over polar areas.

Altogether, most observations made so far support the idea that nucleation and subsequent particle growth are uncoupled under atmospheric conditions (Kulmala, Pirjola, & Mäkelä, 2000c). The associations between J_D and H_2SO_4 vapour concentration are suggestive of the involvement of H_2SO_4 in atmospheric nucleation, yet the actual nucleation mechanism remains to be definitively identified. The few observations made in the free troposphere are consistent with the binary water–sulphuric acid nucleation, whereas in the boundary layer a third nucleating component (such as ammonia) or a totally different nucleation mechanism is clearly needed. We are not able to rule out the possibility that ion-induced nucleation occurs in the boundary layer. Observed growth rates of nucleated particles cannot usually be explained by the condensation of sulphuric and associated inorganic compounds (water and ammonia). While organic compounds having a very low saturation vapour pressure would appear to be the most likely candidates for the growth of nucleated particles, the identity of these compounds remains to be revealed.

6. Summary and conclusions

The formation and growth of new aerosol particles is of growing interest due to their climatic and health effects. The question "How and under what conditions does new particle formation occur?" has exercised the minds of meteorologists and physicists since the time of John Aitken, who in the late 1880s built the first apparatus to measure the number of dust and fog particles. However, only during the last 10 years has the measurement technology developed to such a level that size distributions nanometer-size particles can be measured in the atmosphere.

In recent years the formation and growth of nanometer-size atmospheric aerosol particles have been observed at many different sites. These measurements have been performed on ships, aircraft and fixed sampling sites during both intensive campaigns and continuous ground-based measurements. From measured size distributions one can infer the particle growth rate, whereas from measured number concentrations an apparent source rates of 3 nm (or 10 or 15 nm) particles is obtained. We have collected existing data from more than 100 studies reported in the literature and existing data banks (campaigns and continuous measurements). Aerosol formation has been observed in many different locations and environmental conditions, including the free troposphere, urban plumes, clean arctic air, coastal environments, etc. Aerosol formation is often related to atmospheric mixing processes, such as the evolution of a continental boundary layer or the mixing of stratospheric and tropospheric air near the tropopause.

Based on this review we conclude that typical particle growth rates are 1–20 nm h^{-1} in mid-latitudes, depending on the temperature and concentration of condensable vapours. However, growth rates as low as 0.1 nm h^{-1} can be seen over in Antarctic and Arctic regions. The formation rate of 3 nm particles during regional nucleation events lies typically in the range 0.01–10 particles cm^{-3} s^{-1} in the boundary layer. In coastal environments and industrial plumes, however, formation rates as high as 10^4–10^5 particles cm^{-3} s^{-1} have been reported. Particle formation and growth events can be observed throughout the year, but particle growth rates are clearly bigger during the summer than in winter.

From the present study it is not possible to decide what is the most relevant nucleation mechanism in the atmosphere. It may be that more than one nucleation process is operating in the atmosphere. The most realistic candidate mechanisms include (1) homogeneous binary water–sulphuric acid nucleation when ammonia concentrations are very low, (2) homogeneous water–sulphuric acid–ammonia nucleation, (3) ion-induced nucleation of binary (water–sulphuric acid) or ternary vapours or of organic vapours, and (4) barrierless (kinetically controlled) homogeneous nucleation. Recent studies (Napari, Noppel, Vehkamäki, & Kulmala, 2002a; Napari, Kulmala, & Vehkamäki, 2002b) have shown that some other inorganic systems are not able to make particles under atmospheric conditions. However, the data presented in this paper generally supports the hypothesis that sulphuric acid, while participating in nucleation, accounts usually for only a portion of the particle growth. This supports the proposed uncoupling between the nucleation and growth of atmospheric aerosols, particularly in lower troposphere.

Globally, the formation of new particles and their subsequent growth seem to occur almost everywhere. The new particle can, depending on the location, increase the concentration of cloud condensation nuclei by a factor more than two over the course of 1 day. We can therefore conclude that atmospheric new-particle production is an important process that must be understood and include when developing global climate models. Future work should include continuous observa-

tions of aerosol particle size distributions in diverse locations, measurements of gaseous compounds participating in nucleation and growth, and determination of the chemical composition and other properties of nucleated particles. Measurements concerning the distribution and composition of nucleating clusters would significantly add to our understanding of the nucleation process itself. Finally, the possibility that ion-induced nucleation is occurring cannot be overlooked. Future work should include measurements of ion mobility distributions and ion compositions.

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