AEROSOLS IN A CHANGING WORLD

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FOREWORD

Aerosols recently came into the forefront of the major global climate change topics: the greenhouse disturbance and the ozone hole. Aerosols have been somewhat forgotten although they are omnipresent. In fact, an analysis of any system comprising air (or even space) has to take into account the presence of the tiny particles which were extensively investigated more than a century ago. Aerosols are difficult chapters in many handbooks on health, warfare, nuclear safety, and global climate change. The author, who has been involved in aerosol research for more than 20 years, describes the change of focus in aerosol research resulting from changes in society. The reverse is dealt with by treating changes and trends in climate globally (greenhouse, ozone hole, and nuclear winter), regionally (nuclear safety), and locally (indoor) where aerosols play an important role. Special attention is given to the climate effects of volcanic eruptions and the conclusions which can be drawn from that for greenhouse disturbance and shorter term consequences due to extreme climatic events.

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SUMMARY

The changing scene of aerosol research is discussed. There is an apparent shift from basic to applied research. Attention is given to indoor aerosols, nuclear aerosols, and to the role of aerosols in climate, especially volcanic aerosols and those from nuclear warfare, and in view of the ozone layer destruction.

1. INTRODUCTION

Aerosols are systems of particles suspended in a gas. They are inherently unstable: even when there are no interactions with walls of enclosures, with force fields or with condensing molecules, yet changes occur due to coagulation. Scientific insight in coagulation goes back as far as 1906 (1) when Smoluchowski Ritter von Smolan's first publication on coagulation appeared (2). Apart from the inherent processes, changes in aerosol are indicators of the presence of actors; this applies to the aerosols themselves as well as to aerosol research. We shall draw some conclusions from a brief analysis of aerosol literature, showing that the world is changing for aerosol scientists. Furthermore, we shall deal with some selected topics in the field of worldwide changes where aerosols play a role.

2. AEROSOL RESEARCH

Bibliographical analysis of the period 1983-1987/88 was made of three periodicals in view of the trends in aerosol and air pollution research: "Aerosol Science and Technology," "Journal of Aerosol Science," and "Atmospheric Environment." As shown in Fig. 1,

- the aerosol-pollution oriented fraction of papers in "Aerosol Science and Technology" decreased from 41% in 1983 to 29% in 1987. Typically, the remainder comprises technologically motivated research;
- this share of aerosol-pollution papers increased in "Journal of Aerosol Science" from 26% in 1983 to 44% in 1988. The non-pollution oriented research concerns mainly basic and general instrumental aerosol research;
- the percentage of aerosol research contributions in "Atmospheric Environment" remained remarkably constant in the same period at a level of $44 \pm 2\%$.

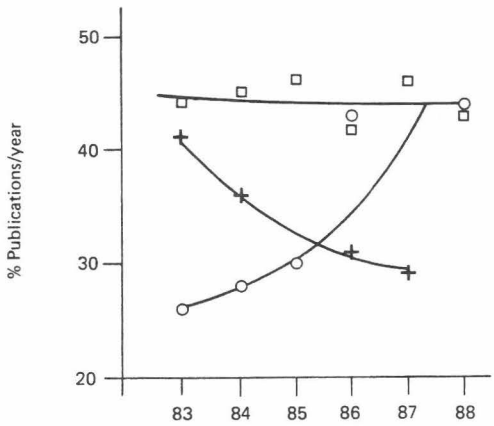

Figure 1: Percentages of aerosol-pollution oriented papers in "Journal of Aerosol Science" (○) and "Aerosol Science and Technology" (+), and the percentages of aerosol papers in "Atmospheric Environment" (□).

These data show a fast shift (within 5 years!) of the focus of aerosol research from rather basic/generic to air pollution ("Journal of Aerosol Science") and also from air-pollution to technological applications ("Aerosol Science and Technology"). The constant level of aerosol papers in the air-pollution periodical "Atmospheric Environment" likely is the result of a dynamic equilibrium: input from basic/generic and drain to industrial technology of aerosol expertise. This is in accordance with observations elsewhere in the modern western society showing a tendency of increasing technocratically motivated attention for applications of science and undervaluation of basic (i.e. generic) research. The driving forces of this movement to applied research are the governmental deficits and the improved economics of the western society. In this respect we must welcome the recommendation by EPA's Science Advisory Board that EPA plan and implement a coherent long-term research program (3). Let us hope this is the first step in a good direction.

The fact that less than half of the articles in "Atmospheric Environment" are aerosol-related, needs further consideration. Air-borne contaminants are either gaseous or particulate. Beside the two parameters, concentration and composition, common to both aerosols and gases, aerosols have one parameter additionally, viz. its size distribution. One would expect therefore aerosol-related papers to have a larger share in journals about atmospheric environmental issues. This, not being so, however, can be understood from a characteristic of almost any environmental issues, viz. it usually originates from observations with an advanced measuring technique. Aerosols are much more difficult to measure as specific compounds than gases are.

3. THE INDOOR CLIMATE

The environment at home is the most important in man's life: on the average 70-80% of his or her time is spent there. Exposure can be much higher there than elsewhere (outdoors or at workplace, for the environmental quality of which ultimately not the individual is responsible). The indoor climate has at least three important aerosol aspects: the aeroallergens, radon and thoron daughter products, and tobacco smoke. In all three completely different cases we are dealing with decades long exposures. However, the changed attitude of

the western society versus energy conservation, resulting in lower ventilation rates, may easily lead to exposures exceeding acceptable limits. Moreover, various insulation measures not only influence ventilation behaviour but also appear to result in increased ventilation through spaces below the floor. Besides, the lower ventilation rates increase relative humidity, often reinforced by decreased space heating. A good review is given in (5). Under these circumstances, increased formation of aeroallergens is likely to result. These allergens, of which the ones generated by the house dust mite are the most powerful, seriously affect the sensitive part of our population (several percent suffer from "chronic aspecific respiratory disease"). The faecal balls ($22 \pm 6\mu$m s.d. diameter) of the mite cause the inflammatory responses of most of the bronchial-asthmatic persons (6). There are two interesting aspects to this health problem. Firstly, how can the faecal particles of this size reach the human bronchi? Their sizes are generally too large to have a significant chance to reach the bronchi. However, because of the extremely high antigen content of the faecal particles, it would be possible that only one or a few of them already cause the inflammatory response. A second more mysterious aspect of the mite's faeces is their airborne state. It is known (7) and shown theoretically (8) that entrainment into the air of particles of sizes similar to these faecal balls is extremely difficult, though easier than smaller or larger ones. High wind velocities (several $m.s^{-1}$) are required to entrain them. Typically, the faecal balls are found in room corners where in the proper environment of house-dust and high humidity the house mite lives, each mite producing some 20 faeces per day. These locations are places with almost stagnant air and a stagnant boundary layer with a thickness several orders of magnitude larger than the allergenic particles. It is puzzling how such particles can become airborne; yet they have been observed (9) to be present in significant numbers (expressed in antigen levels: circa 5 $\mu g.m^{-3}$) when there is human activity in the room. Platts-Mills c.s. (10) guess that normal exposure is "in the form of a few ($\approx$200) particles/day." Assuming source and sinks to be roughly in balance, one may conclude that the source strength in an average 100 m^3 room is about 5000 faecal aerosol particles per hour (an average settling velocity of 10 $m.h^{-1}$ – $10\mu m$ diameter – was assumed). This is a surprisingly high value. From the connection with (human) activity, however, one might infer that some local powerful (triboelectricity-coupled?) mechanism is active. It is recommended to investigate this problem in view of the recent conclusion that 15% of the Dutch homes have conditions of potentially high aeroallergen levels (11).

The radon problem is of a completely different nature. Recent analyses of the risks due to radon in U.S. homes show that this radioactive gas contributes on the average about 10% of the total risk of lung cancer and in a significant number of houses the total risk exceeds that due to smoking (12). The review by Nero (5) treats the radon problem extensively. In fact one has to do with two radioactive gases, radon and thoron, emanating from the soil under the house floor (also other sources can be important: tapwater from local wells, or emanation from building materials like gypsum or fly-ash containing bricks or concrete). Both these gases themselves as well as their α-radioactive daughter products, which are solid, can pose health problems. The latter solid radioactive matter attaches immediately to other particles. The particle diameter of indoor aerosol being around 0.3 μm, especially that of tobacco smoke (side stream), means that once attached to these particles, highly radiotoxic daughter products of radon and thoron can penetrate deeply into the vulnerable lungs. The fraction of Dutch dwellings with radon levels exceeding criteria has been estimated (11) to be 80 $\pm$ 10%. This reflects the potentially large influence of energy conservation habits in combination with construction principles since this figure concerns only radon from the soil underlying houses.

Smoking, either directly or indirectly, affects human health substantially; this is already obvious from the fact that any epidemiological study on other agents experiences serious problems in distinguishing effects of other origin from those of smoking. There is a wealth of literature on the effects of aerosols from this peculiar human habit. As far as Dutch houses

are concerned, valuable information is given in Lebret's thesis (13). It is concluded that U.S. standards for "respirable suspended particles" are frequently being exceeded in houses of smokers. A recent overview of Dutch environmental quality (11) reports that respirable suspended particle standards are exceeded indoors in the Netherlands in more than 60% of the houses. Sidestream smoke is regarded to be the most important (14). It is of utmost importance to have quantitative knowledge of the composition of these sidestream aerosols and of their physico-chemical fate prior to inhalation because there does not exist something like the effect of "total suspended particles" or "respirable suspended particles" (15). The specific constituents of particles cause these effects. Guerin (14) gives the relevant information on the composition of tobacco smoke. One may look after a relevant measuring method in epidemiology by which tobacco smoke can be monitored, simply and continuously. An existing technique, the "black smoke," probably could serve that purpose. It is worthwhile investigating therefore whether

- there is a constant relation between the black smoke index and the level of specific toxic smoke components in smoke-loaden rooms,
- other indoor sources of elemental carbon containing aerosol like woodburning in fireplaces or candle-light could significantly interfere.

4. NUCLEAR SAFETY

Expertise should remain available also in view of sudden changes in the world. A good example is the nuclear reactor accident that took place in Chernobyl, USSR, in April 1986. Clearly, all radioactive material transported worldwide was in the form of aerosol (80% in the size range < 1 μm) apart from the noble gases and part of the iodine. Of the I-131 at least 50% was particulate (< 1 μm) (16). This, in fact, is basic knowledge needed for a warning and control system operating after a nuclear reactor accident. Dispersion and deposition of the most hazardous radionuclides emitted was strongly dependent on their physicochemical form. This is illustrated by the observed, diverging deposition velocities of the particulate and the gaseous radioactivities being 0.07 and 0.7 cm.s^{-1}, respectively (16). The particle size (< 1 μm) and patchy pattern of radioactivity deposition are correlated. Submicron particles have a unique deposition route, viz. by (in-cloud) rain-out followed by wet deposition. One of the conclusions from this is that a high risk exists in case of short distance in-cloud uptake followed by immediate precipitation of a release from a nuclear reactor accident.

From this brief survey of a major nuclear accident it may be evident that a living aerosol expertise is of utmost importance. To have a slow die-back of aerosol expertise is dangerous: we have to expect the unexpected. It is unrealistic to assume gases to be more important than aerosols in hygiene; the opposite may be true.

Another interesting branch of nuclear safety oriented aerosol research is that in the field of transport of radioactive particles inside the safety containment after melt-down of a nuclear reactor. This kind of aerosol research has the typical features of safety research. Safety assessment may be based on a simplified picture of the accident provided the simplifications are on the conservative (safe) side. For this purpose then rather basic research can be performed which still has direct application. This field of nuclear aerosol research created an international research community which generated extensive and important progress in aerosol science (e.g. on coagulation, aerodynamics, phoretic processes, and condensation) which expertise has been used and remains to be used in other fields. The author's thesis is an example of such spin-off of nuclear aerosol expertise, viz. to the field of air pollution (17). No general aerosol conference is really general without one or more contributions on nuclear aerosols (see e.g. (2)).

The changing world concerning nuclear power, e.g. (18), with its obvious anti-nuclear content, could easily lead to a disaster for an outstanding piece of aerosol expertise if not other fields of aerosol research appear. Funding of nuclear aerosol research is steadily decreasing.

5. AEROSOLS AND CLIMATE CHANGES

As stated above, an aerosol is an inherently unstable system, even an aerosol in free space. An enclosed aerosol is subject to processes additional to coagulation. Aerosol mass will be lost by interaction with spatial boundaries due to which also the particle size distribution may coarsen, become finer or even remain unchanged. Any situation where the aerosol remains persistent while spatial borders are present, implies that there is a continuous source of aerosol. This is the case in the Earth's atmosphere which is loaded with several Mtons of aerosol from the dynamic equilibrium between the many, natural and anthropogenic, sources and the (wet and dry deposition) sinks of aerosols (4). When, in the world's atmosphere, aerosols are changing, this generally should be due to changes, suddenly or slowly, in the generation rate of aerosol; the atmospheric sinks are less likely to be subject to large-scale changes. This paper, dealing with aerosols in a changing world, will review briefly the climatic impacts of aerosols, like from "nuclear war," volcanic eruptions, the role of aerosols in the ozone layer destruction and its relation to the greenhouse effect.

The global atmosphere is a relatively thin layer (Earth's radius = ca. 6500 km; atmospheric height = ca. 50 km) and has a density which decreases quickly (exponentially) with increasing height. The inner shell of about 10 km thickness, the troposphere, has only limited exchange with the rest of the atmosphere, especially the stratosphere, the next shell. Therefore, residence times of aerosol particles in the troposphere are much shorter (a few days, maximum two weeks) than in the stratosphere (more than one year), where no walls remove particles and where gravity forces hamper particles to diffuse into space. Reck (19) and Jäger (20) have reviewed the effects of particles on climate. Aerosols play an important role in the climate system through their influence on the radiation balance as well as by providing condensation nuclei for cloud formation and by gas/particle interactions. Coakley c.s. (21), using a simple two-layered atmosphere model, shows that the existing background aerosol causes a global surface cooling of 2-3 K. From large volcanic eruptions it is also evident that aerosols significantly influence the global climate; this will be dealt with below, together with other global effects from volcanic aerosols, e.g. on the stratospheric ozone.

Though earlier trials of mathematically modelling the global climate have been valuable, the development of climate simulation models really started in the early eighties: the El Chichón eruption, the Nuclear War and also paleoclimatic changes were large-scale events to try to simulate (22). An extensive, 100 pages, overview of the relation between volcanic eruptions and the global climate since the last Ice Age has been given by Lamb (23). He concludes that a relation exists between major volcanic eruptions and the below-average temperatures of the subsequent two years. One of the most striking illustrations of this relation is the course of the temperature after the Agung eruption in 1963 (Indonesia), (Fig. 2). Stratospheric heating was about 5 K and at sea level the average temperature was 0.4 K below normal (24). It is generally accepted that the sulfur compounds (SO_2, H_2S, COS) in volcanic eruptions are the climatologically most important part of these emissions (25). Already in the early seventies, Castleman Jr. et al. proved that the so-called Junge aerosol layer in the stratosphere originates from volcanic emissions and consists predominantly of sulphate (26). From an 11-year stratospheric sulphate aerosol measuring program, Sedlacek (27) deduced that the 1/e folding time of a volcanic sulphate injection is 11 months and that there is an annual increase of stratospheric sulphate of 6-9%. This increase could be due to COS (28). However, Berresheim and Jäschke (29) argue that the yearly non-eruptive S-emission of volcanoes

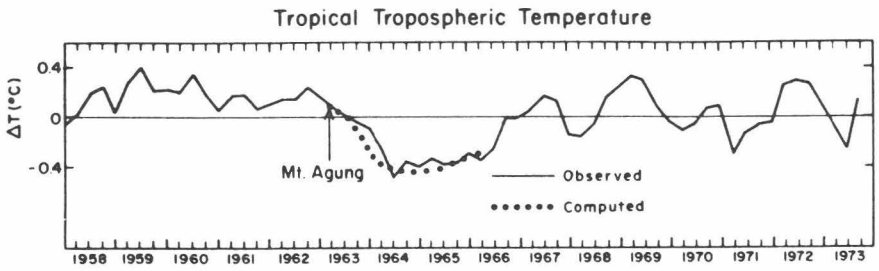

Figure 2: Observed tropospheric temperatures between 30° N and 30° S and computer temperatures after the eruption of Mount Agung, assuming that the added stratospheric aerosols are sulfuric acid and the average depth of the mixed layer of the ocean is 70 m (24).

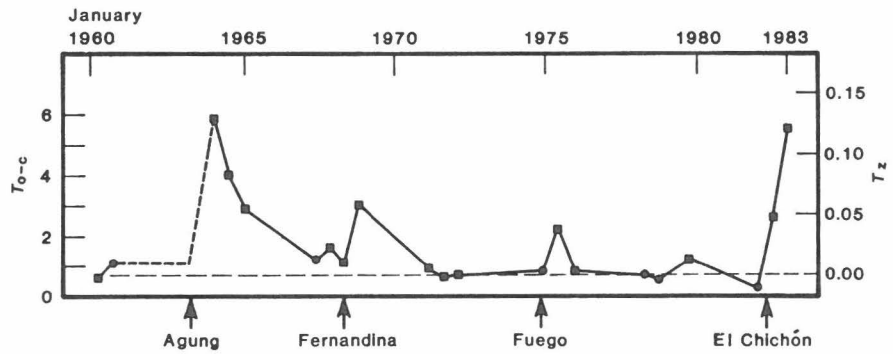

Figure 3: Volcanic aerosol loading estimates obtained from observations of 21 lunar eclipses between 1960 and 1982. Global average optical depths (right-hand scale) are derived from observed minus calculated eclipse brightness (left-hand scale) (34).

(mainly sulphate) is one order of magnitude larger than the eruptive emission rate. Hence, the troposphere/stratosphere exchange of this sulphate could explain the general increase of 6-9%. Baldwin et al (30) regard volcanic emissions to have been the main influence on the global temperature up till 1940. Tropospheric warming due to greenhouse gases may be compensated temporarily and partially by aerosols formed in the stratosphere.

The major eruption of El Chiçón in April 1982 (almost exactly four years before "Chernobyl"), triggered a number of research programs, the results of some of them still being evaluated. Also preliminary results have been presented. The temperature at 30-mbar increased significantly above its values during the previous 18 years (31). By this eruption about 20 Mtons of sulphate appeared in the stratosphere; one year later this was about 8 Mtons (25). Brightness measurements of moon eclipses indicate that the global aerosol loading from El Chichón is of a magnitude similar to that of Agung (32), figure 3. The absence of a significant tropospheric cooling by the El Chicón eruption in contrast to the Agung eruption, and the resulting disastrous El Niño of 1982/83, brought Angell (33) to the hypothesis that there is energy transfer between the El Chicón eruption and El Niño: global cooling or heating can never be considered without taking into account a large-scale coupling with oceanic processes. According to Handler (34), stratospheric aerosol influences El Niño events through changes in equator-to-pole temperatures.

Altogether, the geo/atmosphere system is extremely complicated, though intriguing: many coupling mechanisms are active, the most complicating ones are perhaps those involving phase transitions (e.g. cloud formation and its enormous influence on the Earth's heat balance). Stratospheric aerosols appear to play a role by their interaction with light quanta. Insight in the impact of volcanic aerosols on climate has been utilized also in the problem of the Nuclear Winter as is obvious from the names of authors writing on both subjects.

The Nuclear Winter situation is a hypothetical situation of large smoke injections into the atmosphere by fires following a nuclear war. The analyses of the climatic effects of such smoke emissions were not free from political choices as may be clear from the widely diverging magnitudes of smoke injection and climatic models used. Rampino discussed the appropriateness of using volcanic eruptions as a basis for estimating the effects of a nuclear war (36). From a brief survey of major volcanic eruptions Rampino comes to the conclusion that maximum aerosol optical depths of 40 (ca. 10^{16}g) were probably correlated with low global temperatures some 17-14 Myr ago. Such optical depths are similar to those assumed in the most severe nuclear war scenarios. SCOPE-ICSU has estimated the amount of smoke that would remain in the atmosphere for some time after mass fires from a nuclear war: 30 Mtons which consist mainly of blackish elemental carbon. Calculations show that regional temperature drops of 15-35 K are possible in the weeks after an attack (37). The main uncertainties stem from effects of early scavenging of the smoke: enormous amounts of water vapour (10-100 Gtons) are also produced by the fires. Ghan et al. (38) has identified several feedback processes which are not present or incorrectly treated in models, like boundary layer stability under Nuclear Winter conditions, interactive ground hydrology and aerosol coagulation. The patchy pattern of smoke is also insufficiently dealt with. Input data on initial mass and physical characteristics of the smoke particles have to be better defined too. The importance of the latter uncertainty is also stressed by Penner (39) and Small (40). Mannis (41) has considered the possibility of smoke penetration into the stratosphere (with its inherently long residence time); he shows that only simultaneous burning of large city-centers may lead to injection into the lower stratosphere. It is assumed that solar heating of the smoke with its strongly absorbing soot component is an important vertical driving force, requiring a non-existing sophisticated global circulation model. Large-scale forest fires may be regarded as examples; they have had some influence on climate on a meso-scale, like the ones in Kalimantan, Indonesia, and most likely have led to atmospheric disturbances in Singapore and Malaysia (19), though not to the extent shown by the above quoted calculations. A Nuclear Winter due to a nuclear war is becoming less likely due to the successful negotiations on nuclear disarmament between the superpowers. Nevertheless, there is still more than sufficient nuclear arsenal to have fires to the extent as assumed in most of the model calculations. Fortunately, the world has changed in such a manner that any intentional nuclear attack is much more remote than ever before. Meanwhile, it is obvious that spin-off of aerosol knowledge to and from the Nuclear Winter assessments has taken place. Possibly, even these aerosol studies eased the nuclear disarmament.

Another worldwide change which has caused (and increasingly causes) concern, is that of ozone-layer depletion. Also here aerosols appear to play an essential role (aerosols not in the sense of spray-cans), either in the complicated physicochemistry of the ozone depletion or by influencing the ozone measurements. Stratospheric ozone depletion is well-known to the public as leading to a potentially large direct threat to health. The stratospheric ozone layer being a protective shield against damaging UV radiation, less ozone in the atmospheric column above him or her increases the risk of skin cancer. Excellent reviews of the subject can be found in (42-44). In fact, two phenomena are included in the ozone depletion: the Antarctic so-called Ozone Hole which appears in spring, and higher latitude ozone depletion. These phenomena differ not only in spatial and temporal aspects but also in the success they can be modelled. For the Ozone Hole the model of ozone scavenging by derivatives of anthropogenic

chloro-fluorocarbons fits the observations reasonably well. However, there is considerable uncertainty about the northern mid-latitude depletion in wintertime, being greater (30-50%) than calculated (45-47). There is sufficient scientific evidence of the overwhelming role played by manmade chlorine compounds in the stratospheric ozone depletion. The 1987 Montreal "Protocol on Substances that Deplete the Ozone Layer" is a sign of the willingness of those involved to lower the chloro-fluorocarbon emissions considerably. In order to have the depletion to disappear this is, however, not sufficient mainly because of the long atmospheric lifetimes ($\approx$ 100 years) of the chloro-fluorocarbons. The phenomenon is rather complicated with a number of feedbacks and indirect physical processes (e.g. CO_2, chloro-fluorocarbons, and aerosols, and also ozone itself may change stratospheric thermohydraulic conditions which are of importance for ozone destruction rates and stratospheric circulation). The photochemistry of ozone, its chemical destruction by chlorine compounds formed from man-made halogen compounds is dealt with extensively by Cicerone (48) and by Stolarski (46).

The surprise of the recent years is the role played by heterogeneous chemical reactions at the surface of aerosol particles in the stratospheric ozone depletion. Aerosol particles (mainly sulphate) act as nuclei in the formation of stratospheric ice clouds. There is evidence of significant heterogeneous activity by ice surfaces (185 K) in the essential N_2O_5/HCl system producing Cl_2 and HOCl, and fixing HNO_3 at the ice surface (49). The formation of HOCl and Cl_2 is extremely important because after wintertime (the ice surfaces are present as "polar stratospheric clouds" in wintertime) the sun starts fotolysing Cl_2 and HOCl resulting in the formation of chlorine atoms or chlorine monoxide, components in the ozone destructing Cl-cycle. The role of the aerosol particles, therefore, is the increased formation of active chlorine compounds (Cl_2 and HOCl) from low active ones like HCl and $ClONO_2$. The polar stratospheric clouds consist of particles of ice and HNO_3 grown on nuclei which probably were composed of sulphuric acid and which are of volcanic origin; see above (50-51). The presence of HNO_3 on the larger ice particles (several microns) and their probable disappearance from stratosphere by settling of these particles is another important role of the (polar) aerosol in ozone depletion because of HNO_3 being a sink for the ozone destructing chlorine atoms. Recently, Kerr (52) reported sticking probabilities of nitrogenous molecules of 0.01-0.1 to sulfuric acid which is a value sufficiently high to make the Junge layer a possible actor in the mid-latitude ozone layer destruction.

6. CONCLUSIONS AND SUMMARY

1. There is a significant fast shift in aerosol research from basic to applied technology, probably due to the changing origin of research funding. This could harm a well-balanced continuous broadening and deepening of scientific insight in aerosols. Investments are needed with return in the long term which are of only limited interest to industry. Profits of air pollution oriented aerosol research are often economically hidden or indirect. In order to survive in this struggle for aerosol expertise life it is necessary to have controlled application of this expertise in these areas of new attention (industry, climatology, indoor air pollution, etc.) in such a manner that a certain minimum of basic research can be carried out and that the sophisticated aerosol equipment can be maintained at a modern level.

2. Information on indoor aerosols has to be improved to a level similar to that of outdoor aerosols. There are a number of important and interesting problems, e.g. concerning the entrainment of settled particles in the indoor air, and concerning the influence of space heating on aerosol behaviour. The indoor environment will become more and more a health problem with increasing energy conservation.

3. Nuclear aerosol research has greatly improved aerosol knowledge. Safety research allows performing rather basic research on experimental systems by conservatively simulating

accident conditions.

4. To a large extent, knowledge of volcanic aerosols has been used in Nuclear Winter studies. The insight gained from these activities in effects of large-scale clouds on visible, IR and near-UV radiation is very useful in lowering the uncertainties of the "greenhouse" problem.

5. Though experimentally complicated, the heterogeneous chemistry of stratospheric cloud simulating aerosols has to be investigated under well-defined laboratory conditions. Still the question has to be answered of the validity of simulating the gas/particle system by a system of a gas and a single large solid surface.

6. Worldwide changing anthropogenic influences raise new subjects to be studied by aerosol research groups. Research management has to be aware of the important knowledge present in aerosol research groups which knowledge has to be used sooner or later.

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