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Microphysical Interactions between Cosmic Galactic Rays and Clouds: „Missing Link“ in the Climate Discussion? Hypotheses, Indications and the Difficulties of Enquiry. Part I: The IPCC 2007 perspective

I. Why atmospheric phase transitions are that important?

The understanding of atmospheric phase transitions such as the formation of aerosols, hydrosols and hydrometeors is of crucial importance for the epignosis, diagnosis and prognosis of the spatiotemporal evolution of the Earth's atmosphere. Phase transitions are associated with transformation of energy and matter (e.g., its state of order) in the atmosphere from one quality to another. For example, the condensation of atmospheric water vapour is associated with the formation of small, radiatively active droplets and the release of latent energy[1], which can enhance the kinetic energy in the updraft cores of deep convective cloud systems due to the work of buoyancy forces. In this way, condensation can trigger the formation of severe thunderstorms, heavy rainfalls, flash floods etc. As a consequence, thermodynamic and kinetic processes at microscopic scales can lead, on principle, to hydrothermodynamical effects at scales which are larger by many orders of magnitude, under circumstances associated with disastrous macroscopic aftermaths for population, economy etc.

II. Importance of clouds for weather and climate and their prediction

The most impressive manifestation of atmospheric phase transitions is the formation of clouds, able to be experienced both by their visibility and sometimes also by their audibility in a plethora of beautiful forms and guises. Clouds control Earth's weather and regulate its climate (vide infra) by cooling the atmosphere due to reflecting incoming visible-wavelength solar radiation and warming the surface by trapping outgoing infrared radiation (Baker and Peter, 2008). They produce rain and snow, impacting Earth's weather, landscapes and vegetation zones. Small-scale processes causing large-scale effects of clouds are: (a) Updraughts and downdraughts inside of clouds and turbulent mixing on scales of metres to kilometres, affecting the evolution of nanometre to micrometre particles; (b) Submicrometre aerosol particles of natural and anthropogenic origin and strongly varying composition serving as nuclei for water droplet and ice crystal formation in clouds[2]; (c) Growth of cloud water droplets or ice crystals by water vapour uptake; (d) Rain formation by falling and collision of cloud droplets, which have reached diameters of tens of micrometres (Baker and Peter, 2008).

Clouds constitute the largest single source of uncertainty in weather and climate prediction (Baker and Peter, 2008). While the Numerical Weather Prediction (NWP) of most weather elements has been considerably improved over the last two decades, enhancement of rain prediction achieved during this period is comparatively low. Moreover, the quantitative precipitation forecast is still of such poor quality, that it can hardly be used for many applications, e.g., hydrology (Hense et al., 2003). This situation happens despite of large progress achieved in process understanding, physical model parameterisation, data assimilation, computer capacity etc. The reasons for that are not yet known and difficult to identify. However, present deficiencies are suspected to include (a) the incomplete modelling of the components of the water cycle, (b) gaps, non-resolved structures and errors in the initial fields, (c) inadequate methods of optimally linking observations with forecast models (assimilation of data into models) and (d) basic problems in our understanding and interpretation of deterministic NWP models (Hense et al., 2003). In view of the little progress in rain prediction over nearly two decades one might ask: How far we really away from the theoretical limits of cloud and rain prediction?

Recently, Lange (2007) discussed the consequences of cross-linking and nonlinearity in complex systems, such as the Earth's atmosphere, for the predictability of future system states. The future of a complex system essentially depends on whether the sum of all destabilising or that of all stabilising feedbacks will dominate the system behaviour. Employing basic principles of system theory by means of deterministic reference models, the author instructively demonstrated, that this question cannot be answered on the base of model simulations. The reasons for that are twofold:

1. *Existence of „stochastic chaos"*: It is caused by (a) the coexistence of a multitude of repelling and attracting fix points (repellors and attractors) in high-dimensional phase spaces, (b) the existence of metastable states of attractors (stability against small perturbations), (c) the increase of the

number of attractors with increasing degree of system cross-linking, (d) the increase of unpredictability, which attractor currently controls the system evolution, with increasing number of attractors (i. e., increasing probability of random attractor changes);

2. *Existence of „deterministic chaos"*: It is a direct consequence of the existence of nonperiodic solutions of the deterministic hydrothermodynamic flow equations, e. g., the weather and climate prediction is based on (known as „butterfly effect"). Thereafter unpredictability can occur, even though the actual state point of a system in the phase space permanently stays within the attracting region of a single attractor and the trajectory of the state point is solely dynamically controlled without any perturbation. Then, the evolution of the system remains unpredictable as determinism could only be exploited, if the initial conditions are known with unendingly accuracy. In reality, (a) the accuracy of the initial condition is limited by the observation error (e.g., detection limits of measuring instruments), and (b) unpredictable subscale fluctuations of the system state near phase transitions (bifurcation points) might become very important for the system future. The mastery of the deterministic chaos is, on principle, impossible due to the uncertainty principle of quantum physics.

With respect to climate prediction the author emphasised, that the question whether the sum of all destabilising or the sum of all stabilising feedbacks will dominate the system behaviour, has not been answered so far.

III. Cloud-mediated homeostasis of Earth's climate

Cloud formation is a key process ensuring the homeostasis[3] of Earth's climate over geological times. The application of this concept to atmospheric science was promoted by James Lovelock, who at the end of the 1960's formulated the idea of homeostasis of a fictive planet Gaia represented by the conceptual „daisy world" model (Lovelock, 1991). Lovelock proposed a negative feedback mechanism to explain the relative stability of Earth's climate over geological times, which became generally known as „Gaia" hypothesis[4]. At the end of the 1980's, the Gaia theory was essentially extended by Charlson et al. (1987, CLAW hypothesis). In their postulated negative feedback mechanism, abundant atmospheric sulphate aerosols are hypothesised to play a key role in stabilising Earth's climate. Thereafter, an assumed global warming caused by radiative forcing of „Greenhouse" gases would lead to an increase of the sea surface temperature and consequently, to an increase of dimethyl sulfid emissions (DMS, CH_3SCH_3) from marine phytoplankton (e.g., macroalgae) from the ocean into the atmosphere[5]. The gas-phase oxidation of DMS produces sulphur dioxide (SO_2), which further oxidises with the hydroxyl radical (OH) to form sulphuric acid vapour (H_2SO_4). Water vapour and sulphuric acid vapour are key precursor gases for the homogeneous heteromolecular (binary) nucleation of sulphate aerosols over the ocean[6]. Due to condensation and coagulation processes these microscopic particles can grow via intermediate, not yet fully understood stages from thermodynamically stable clusters via ultrafine condensation nuclei (UCN) to non-sea-salt (nss) sulphate aerosols and finally, to cloud condensation nuclei (CCN). An increase of the CCN concentration leads to an enhancement of the number concentration of cloud droplets. A higher cloud droplet concentration causes an enhancement of the cloud reflectivity for given liquid water content. An increased reflectivity of solar radiation by clouds cools the atmosphere and counteracts the initial warming. Via this route, a thermal stabilisation of Earth's climate is accomplished[7].

Though describing climate self-regulation at the global scale in a physically sound manner, the details of this mechanism are neither fully understood and theoretically worked out nor empirically verified at the process scales, respectively. Subjects of large uncertainties are the genesis of atmospheric phase transitions itself, whereas new particle formation (NPF) by homogeneous and heterogeneous nucleation, respectively, are of special importance, since both processes are necessary conditions for CCN, cloud and rain formation.

The homeostatic climate stabilisation via aerosol formation is neither the only negative feedback mechanism in Earth’s geosystem nor it has been claimed to be the primary one.

IV. Helio- and astrophysical perspective on climate change

From a non-anthropocentric point of view on climate change a subject of ongoing interest is the heliophysical component of climate variability, i.e., the impact of solar variability and total solar irradiance (TSI) on Earth’s climate. Recently, this heliophysical aspect was supplemented by an astrophysical component. Based on previous works, Svensmark (2007) used the notion „cosmoclimatology” to denote longterm changes of Earth’s climate as a result of the alteration of the intensity of galactic cosmic rays (GCRs). The latter are suspected to impact Earth’s cloudiness via ion-induced nucleation and formation of aerosols, which serve as building blocks of CCN. The modulation of the GCR influx by the solar magnetic activity is suspected to account for climatic fluctuations on decadal, centennial and millennial timescales. Changes of the galactic environment of the solar system, associated with the revolution of the Sun around the galactic centre, are predicted to have dramatic climatic consequences, including „Snowball Earth" episodes. By means of a state-of-the-art astrophysical model to describe the position of the Sun along its orbit with respect to the spiral arm pattern of the Milky Way, the cosmoclimatological theory involving ion-induced nucleation gives a physically sound explanation for the match between the spiral-arm encounters and ice-house episodes during the Phanerozoic (Perseus spiral arm: Ordovician to Silur ice-house periods; Norma arm: Carboniferous ice-house; Scutum-Crux spiral arm: Jurassic to Early Cretaceous icehouse periods; Sagittarius-Carina spiral arm: Miocene epoch, leading almost immediately (in geological terms) to Orion spur: Pliocene to Pleistocene epochs) (Svensmark, 2007, Figs. 8 & 9, see references therein). Such a perspective extends the scales of interest from molecular to galactic dimension.

In the presented here part I of the article, the IPCC 2007 perspective on the direct and indirect solar impact on climate change will be reviewed. In the forthcoming part II, I will scrutinise recent studies supporting links between the intra- and extraterrestrial weather, especially empirical findings for the indirect solar effect on Earth's climate, supposed mechanisms and cloud chamber experiments to verify/falsify existing theories.

V. The IPCC 2007 perspective on the Sun's direct and indirect effects on Earth's climate

V. 1 Climate change in the past

Earth's climate has changed on all time scales. The primary driver of past climate variations is the change of Earth's radiation budget, which can be caused by the following three pathways (Jansen et al., 2007, IPCC-WG I, Chapter 6, p. 449–450, FAQ 6.1):

1. Variation of the incoming solar radiation (e.g., by changes in the Earth's orbit[8] or of solar processes):
    a. Ice ages, occurring in regular cycles, are linked to regular variations in the Earth's orbit around the Sun (Milankovitch cycles[9]). These cycles change the amount of solar radiation received at each latitude in each season.
    b. Climatic changes can also be caused by variations of the energy output of the Sun. The solar output varies slightly (≈0.1%) over an 11-year cycle (vide infra). Sunspot observations, going back to the 17th centu-

ry, and measurements of isotopes generated by GCR, indicate long-term changes in solar activity. According to our present level of understanding, solar variability and volcanic activity are considered as leading reasons for climate change in the pre-industrial era[10].

2. Change of global albedo (i. e., variation of the fraction of reflected solar radiation, e.g., due to changes in cloud cover, atmospheric aerosols, land surface cover).
3. Alteration of the longwave energy radiated back to space (e.g., by changes in „Greenhouse" gas concentration).

V. 2 Direct and indirect solar forcing of climate change

Solar forcing of climate change is suspected to be a combination of direct forcing by the total solar irradiance (TSI) changes and indirect effects of UV radiation on the stratosphere and GCR-mediated cloud change.

(a) Direct effect of solar variability

Satellite-based TSI observations show day-to-weak variations associated with the Sun's rotation on its axis, and decadal fluctuations arising from the 11-year solar activity. The TSI levels during the past two solar minima were comparable. The contemporary TSI variability is due to the presence of sunspots (compact, dark features, where radiation is locally depleted) and faculae (extended bright features, were radiation is locally enhanced) on the Sun's disk. Neither model based facular proxy nor measured GCR 10.7 cm flux and the so-called *aa* geomagnetic index since the 1950s exhibit a significant secular trend during activity minima. Changes of surface emissivity by magnetic sunspots and facular regions are the most effective in altering irradiance. Also changes of the solar diameter are suspected to cause secular TSI changes.

TSI reconstructions over the past 400 years assumed the existence of a long-term variability component in addition to the 11-year solar cycle, in which the 17th-century Maunder Minimum TSI was reduced in the range of 0.15% to 0.3% below contemporary solar minima. This long-term temporal evolution was assumed to follow either the smoothed amplitude of the solar activity cycle or the cycle length, respectively, and was motivated by (a) the range of variability in Sun-like stars, (b) the long-term trend in geomagnetic activity and (c) solar modulation of cosmogenic isotopes. However, according to the IPCC 2007 report these arguments, which support a significant long-term TSI trend, could not be confirmed by investigations performed in 2004 and 2005 (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 190, see references therein)[11]. Among others it has been found out, that the relationship between TSI and geomagnetic and cosmogenic indices is probably nonlinear. The „open” magnetic flux (the flux extending into the heliosphere), which modulates geomagnetic activity and cosmogenic isotopes[12], can accumulate on intercycle time scales even when the „closed” magnetic flux, such as in sunspots and faculae, does not. The IPCC 2007 report cites model studies, which suggest a 0.04% increase of the TSI from the Maunder Minimum to present-day cycle minima.

^{14}C and ^{10}Be cosmogenic isotope records in tree rings and ice cores indicate solar-related cycles near 90, 200 and 2.300 years. Some evaluation studies of cosmogenic isotopes and sunspot records suggest, that the solar activity during the 12th-century Medieval Solar Maximum was comparable to the present Modern Solar Maximum. A recent study of Solanki et al. (2004) reports on exceptionally high levels of solar activity in the past 70 years, relative to the preceding 8.000 years. Nevertheless, owing to differences among isotopes records, there is still some debate on whether current levels of solar activity can be considered as *„exceptionally high”* or only *„historically high, but not exceptionally so”* (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 190).

(b) Indirect effect of solar variability

Variations of the solar UV radiation contribute significantly to TSI changes and creates and modifies the ozone layer in the stratosphere, with an indirect dynamical and radiative influence on the troposphere. Although the UV radiation energy of the spectrum at wavelengths below about 300 nm (absorbed by the atmosphere) amounts only ≈1% of the radiation energy of the Sun, its variation is by at least an order of magnitude higher than the TSI variation (15% of the total irradiance cycle). Probably, there are also effects of TSI variations on the mesosphere and thermosphere. A further contribution to indirect forcing comes from solar wind fluctuations and solar-induced heliospheric modulation of GCR.

Solar cycle changes of the UV radiation alter middle atmospheric ozone concentration, temperatures and winds, including the Quasi-Biennial Oscillation. For example, 11-year cycle signals could be identified in zonally averaged stratospheric temperature, ozone and circulation. Furthermore, a solar-cycle induced increase in global total ozone of 2 to 3% at solar cycle maximum was found, which is accompanied by a temperature response increasing with height (≈1 K at z=50 km). Solar forcing is suggested to induce a significant lower stratosphere response, probably caused by temperature changes affecting planetary wave propagation.

During episodes with high solar activity, the magnetic field of the heliosphere reduces the GCR flux in the atmosphere. Solar-induced GCR fluctuations are hypothesised to influence Earth's climate via the following mechanisms:

1. The plasma, generated by GCR ionisation in the troposphere, is part of an electric circuit, which extents from the Earth's surface to the ionosphere. Thus, GCR may affect thunderstorm electrification.
2. By altering the CCN population and hence microphysical cloud properties (e. g., droplet number concentration), GCR may induce microphysical processes analogous to the indirect effect of tropospheric aerosols.
3. Atmospheric cluster ions, generated by GCR, are preferred sites for heterogeneous nucleation of aerosols. In the case of low H_2SO_4 vapour concentration, ion-induced nucleation[13] may dominate over binary homogeneous nucleation (BHN) of H_2O and H_2SO_4. Furthermore, increased ion-induced nucleation and increased scavenging rates of aerosols are suggested to occur in turbulent regions around clouds. Thus, the presence of GCR-generated ions may influence several microphysical processes.

Several empirical findings show a correlation between globally averaged low-level cloud cover and GCR fluxes, which is hypothesised to result from the changing ionisation of the atmosphere (and subsequent ion-induced nucleation and activation of cloud nuclei) by solar-modulated GCR fluxes (e.g., empirical relation between cloud cover variations during 1984 to 1990 and the solar cycle). The IPCC 2007 report states an ongoing controversial discussion of this issue, which originates from uncertainties concerning the reality of the decadal signal itself, the phasing or anti-phasing with solar activity, and its separate dependence for low, middle and high clouds.

The IPCC 2007 report refers to the following con's: (a) The GCR time series does not correspond to global total cloud cover after 1991 or to global low-level cloud cover after 1994 without unproven de-trending. (b) The correlation is significant with low-level cloud cover based only on infrared (not visible) detection. (c) Multi-decadal (1952 to 1997) time series of cloud cover from ship synoptic reports do not exhibit a relationship to GCR.

On the other hand the report refers to a statistically significant positive correlation between cloud cover over the UK and GCR flux during 1951 to 2000, though contrarily, cloud cover anomalies from 1900 to 1987 over the USA do have a signal at 11 years, which is anti-phased with the GCR flux.

VI. Summary of IPCC 2007 point of view and conclusions

The IPCC 2007 perspective on the direct and indirect solar forcing of climate change can be summarised as follows:

- TSI changes are stated to be not the major cause of the temperature change in the second half of the 20th century *„unless those changes can induce unknown large feedbacks in the climate system. The effects of galactic cosmic rays on the atmosphere (via cloud nucleation) and those due to shifts in the solar spectrum towards the ultraviolet (UV) range, at times of high solar activity, are largely unknown. The latter may produce changes in tropospheric circulation via changes in static stability resulting from the interaction of the increased UV radiation with stratospheric ozone. More research is needed before the magnitude of solar effects on climate can be stated with certainty"* (Le Treut et al. 2007, IPCC-WG I, Chapter 1, p. 108).
- Empirical results have strengthened the evidence for solar forcing of climate change. The most likely mechanism is considered to be some combination of direct forcing by changes in total solar irradiance, and indirect effects of ultraviolet (UV) radiation on the stratosphere. The indirect effects induced by GCR is considered as *„least certain"* and subject of ongoing research (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 188).
- Whether solar wind fluctuations or solar-induced heliospheric modulation of galactic cosmic rays also contribute indirect forcings *„remains ambiguous"* (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 192).
- Owing to the difficulty to track the influence of ions in a long chain of complex interacting processes, *„quantitative estimated of galactic cosmic-ray induced changes in aerosol and cloud formation have not been reached"* (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 192–193).
- The apparent relationship between solar variability and cloud cover may result not only from the solar-modulated GCR flux and solar-induced ozone change, but also from sea surface temperatures altered directly by TSI change and by internal variability due to El Niño-Southern Oscillation. In reality, different direct and indirect physical processes may operate simultaneously (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 193).
- While the level of scientific understanding of solar forcing due to direct TSI change has been declared as *„low"*, the one for GCR influences has been declared as *„very low"* (Forster et al., 2007, IPCC-WG I, Chapter 2, p. 193).

The IPCC 2007 point of view agrees very well with the conclusions drawn at an interdisciplinary „Workshop on Ion-Aerosol-Cloud Interactions”, which was held at CERN, 18–20 April 2001. Here, the quintessence of the „Workshop Summary Panel” discussion, given in form of question-answer dialogue, will be summarised (Wolfendale, 2001):

1. Does cosmic ray ionisation play a role in the climate? (This question did not ask whether GCRs do indeed significantly affect the climate, which was supposed to be clearly unanswerable at present.)
   a. The empirically found solar/GCR-climate correlations are sometimes present and sometimes not. This may reflect the complexity of interactions in Earth's climate system. The climate may have stable states such that a correlation may persist for some decades and then disappear for a while. Interactions with anthropogenic contributions (e.g., sulphur dioxide and its effect on cloud formation) may lead to a yet more complex climate response.
   b. Correlations do not demonstrate cause and effect. Present data are unable to separate whether the Sun-Earth coupling is via electromagnetic radiation (TSI, UV) and/or energetic GCR (galactic/solar). There is a need to understand the amplification factors, which are required to enhance the GCR impact despite their very small energy input (roughly equivalent to that of starlight) in comparison with the TSI[14].
   c. To get climate models reproducing temperature records, the solar contribution must be enhanced by a factor 3. Presently, only direct TSI changes are considered. An additional, indirect, solar contribution could either decrease or increase the projections of the anthropogenic effects[15].
   d. To explain the satellite-based solar cycle correlation with low cloud cover both electromagnetic radiation and GCRs remain candidates. The observed solar correlation is confined to low clouds, and the global correlation map of low cloud cover shows no preference for high geomagnetic latitudes. These counter-intuitive findings are deserved to be made reconcilable with the supposed solar effects.

   Vote: The distribution of votes on the question was equally divided between „?” and „Yes”, with zero votes for „No”. GCRs have the potential to affect the climate, but the question of whether they are significant is not yet answered.
2. Is the mechanism „ionisation → aerosol → cloud” microphysically understood?

a. Theoretical studies of Yu (2001) and D'Auria and Turco (2001) suggest, that ions plays an important role in the creation (ion-induced nucleation) and early growth of UCN from trace vapours such as sulphuric acid. Charges stabilise embryonic clusters. Yu (2001) proposed a physical mechanism to explain why the solar modulation is observed only in low clouds. It was demonstrated, that GCR ionisation can be the limiting factor to aerosol nucleation at low altitudes, whereas at high altitudes, where the ionisation rate is up to a factor 20 larger, other parameters such as the trace gas concentrations become the limiting factor.
b. The presence of ion-induced nucleation was confirmed by laboratory studies as well as by mass spectrometric aircraft measurements of ions up to large sizes.
c. Open questions concern (i) the effect of extra UCN, formed by GCR-induced nucleation of trace condensable vapours, on CCN, which seed cloud droplets, (ii) the influence of GCRs on the growth process of other aerosols and on the activation of CCN into droplets[16], (iii) the GCR effect on electro-freezing of super-cooled liquid droplets, (iv) the GCR effect on the global electrical circuit and electric field strength, (v) the GCR effect on the production of trace reactive chemicals (NO, OH), which could affect atmospheric chemistry at certain altitudes.

Vote: There was a 100% „Yes" vote to the question, whether the „ionisation → aerosol" mechanism is understood. The vote to the question, whether the „aerosol → cloud mechanism" is understood, was equally divided between „?" and „No", with zero votes for „Yes". Whether or not GCR-induced extra UCN have a significant effect on CCN is essentially unknown. The experimental and theoretical understanding of ionisation effects on aerosol growth and CCN activation is poor.

3. Is the scientific motivation for a cosmic ray influence on cloud cover agreed?
   a. The GCR-cloud hypothesis is considered as the „first hard clue" on the origin of observed solar-climate correlations. There is a definite hypothesis, which can be tested experimentally: „Are cosmic rays affecting cloud formation?"[17].

   Vote: There was a 100% „Yes" vote, that the scientific motivation for GCR impact on clouds is agreed.
4. Would the CLOUD[18] Atmospheric Research Facility using a particle beam, proposed by CERN, satisfy a need?

a. By means of an expansion cloud chamber, well-defined thermodynamic conditions can be produced over large volumes, and by means of a CERN particle beam, GCR conditions throughout the atmosphere can be recreated[19].
b. The CLOUD facility allows a precise simulation of the conditions inside clouds at all altitudes and latitudes (aerosol nucleation, growth, activation), and to investigate the effects of ionising particle radiation on aerosol and cloud processes (aerosol nucleation, growth, activation).
c. The following problems can be investigated by laboratory experiments: (i) the impact of ionisation on the freezing of Polar Stratospheric Clouds (PSCs), which is of crucial importance for our understanding of de-nitrification and subsequent ozone loss over the poles; (ii) GCR and Solar Cosmic Ray (SCR) impacts on atmospheric NO production by affecting lightning production; (iii) the observed correlation between the GCR intensity and the frequency of lightning; (iv) the suggested preferential activation of water droplets on negative ions, which may be responsible for charge separation in clouds, and therefore lightning; (v) the observed decrease of rainfall during Forbush decreases as well as increased rainfall during energetic SCR events; (vi) the responsibility of GCRs for fair weather ionisation throughout most of the lower atmosphere and, consequently, for the global electrical circuit and subsequent cloud microphysical effects (electro-freezing, aerosol charging, scavenging of charged aerosols by cloud droplets).

Vote: There was a 100% „Yes" vote to the question.

In view of the „very low level of understanding" of GCR impacts on climate change, as stated by the IPCC 2007 report, the microphysical interactions between cosmic galactic rays and clouds represent a subject of high relevance with respect to both basic research in atmospheric physics and climate prediction. At the same time, cloud chamber/particle beam experiments are a challenging endeavour for the science of engineering. The available indications for indirect solar effects on the Earth's climate, given by observational correlations and supported by theoretical and modelling studies, are less than „Smoking Guns", but definitively more than „Bad Shots". The extension of the scales of interest beyond Orlanski's traditional classification of characteristic time and length scales of atmospheric motions (Orlanski, 1975) toward molecular and galactic scales is a prerequisite to verify/falsify hypothesised helio- and astrophysical contributions to climate change. The elucidation of possible links between the intra- and extraterrestrial weather opens a new window for atmospheric and geophysical research.

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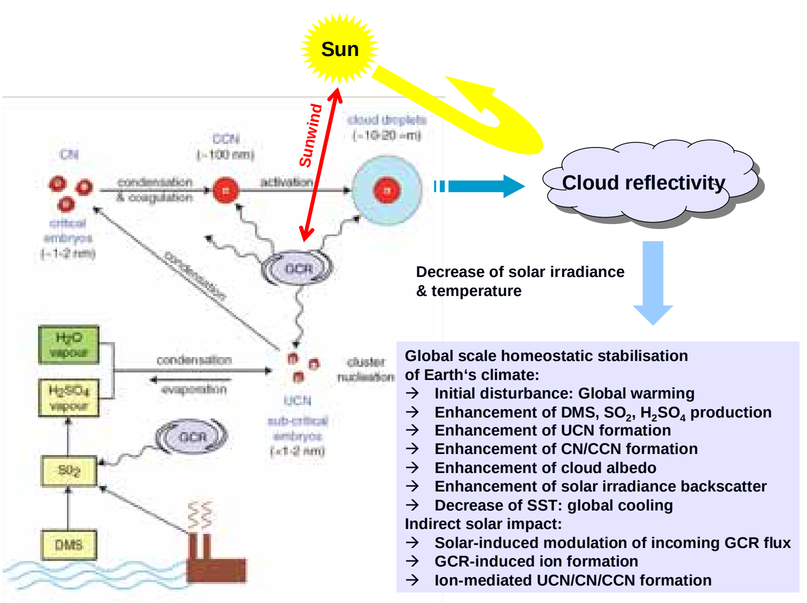

Fig. 1 Nucleation of ultrafine condensation nuclei (UCN) from water vapour (H_2O) and trace sulphuric acid vapour (H_2SO_4), followed by aerosol growth into condensation nuclei (CN) and cloud condensation nuclei (CCN), which can activate into cloud droplets. The precursor of H_2SO_4 is sulphur dioxide (SO_2), produced anthropogenically or, in the remote marine boundary layer (MBL), predominantly from dimethyl sulphide (DMS) released from plankton. The scheme shows the homeostatic principle at the global scale, which is hypothesised to stabilise Earth's climate (Gaia theory). The processes, which may be affected by cosmic galactic rays (GCRs) are indicated by wavy arrows. Charged aerosols are expected to have an enhanced growth rate and reduced evaporation relative to neutral aerosols. GCRs may also affect the activation of CCN in droplets.
(taken from Kirkby, 2001, p. 40, Fig. 35 with some modifications).

Notes

1. A phase transition is a system response to a supersaturated state. For illustration, let us consider a homo-molecular vapour with volume concentration of molecules c (number of molecules per unit volume) and mean free path λ, defined as the average distance travelled by a molecule between collisions with other molecules (intermolecular distance). The length scale λ is proportional to the mean speed of the vapour molecules ν, which increases with temperature. The maximum number of molecules, which can be maintained in the vapour phase, depends on the temperature (corresponding to the saturation vapour pressure). When the number of molecules per unit volume exceeds this maximum value (e.g., by emission or production of vapour molecules), the system tries to reduce its specific volume (i.e., the volume occupied by one molecule $\omega=1/c$) by a transition from the vapour to the liquid phase. The formation of the new liquid macro-phase performs via nucleation of molecule clusters (called embryo's) in the parturient mother phase. The decrease of ω is associated with a gain of latent energy, corresponding to the decrease of λ due to the phase transition.

This energy gain is proportional to the volume of the new embryo (volume term of formation energy). However, to separate the embryo from the mother phase an interfacial area must be formed, which requires work against the intermolecular surface forces. The specific energy to form a surface of unit area is the specific interfacial energy or surface tension. The energy consumption to build up the interface is proportional to the embryonic surface area (surface term of formation energy). The total work of cluster formation is the sum of both the volume and the surface term of formation energy. For sub-saturation, the cluster formation work is monotonously increasing with cluster size, i.e., the emission of vapour monomers from the clusters (shrinking) is energetically more favourable than monomer aggregation by the cluster (growth). For super-saturation, at first the formation work continuously increases with cluster size until a maximum, which is reached at a critical cluster size. From there, the formation energy monotonously decreases with increasing cluster size, because the volume term starts to dominate over the surface term. The formation energy of sub-critical clusters decreases with decreasing cluster size, hence cluster shrinking is energetically more favourable than cluster growth, consequently sub-critical clusters disappear. The formation energy of super-critical clusters decreases with increasing cluster size, hence cluster growth is energetically more favourable than cluster shrinking, i.e., super-critical clusters can grow without limitation.

The energy required to form a cluster of critical size is of great importance for the determination of the nucleation rate, i.e., the rate of formation of critical clusters per unit time and per unit volume of the ambient phase. Owing to thermal fluctuations, sub-critical clusters can exceed the critical size and become super-critical, whereas the probability to overcome the energy barrier decreases with the distance of the actual size from the critical size and vice versa. The probability of near-critical clusters to jump the critical size is correspondingly high. One can see, that phase transition is a clever way of nature to convert latent heat, stored in the supersaturated state (e.g., by solar-induced water vapour evaporation from the Earth’s surface), into sensible heat at the molecular scale by changing the state of order (low order of vapour, high order of droplets).

2. High concentrations of aerosols can increase the brightness of clouds and their ability to reflect solar radiation. An increased aerosol concentration alters large-scale patterns in cloud lifetimes and precipitation.
3. Homeostasis is an universal concept in physics, chemistry, biology, cybernetics, system theory, economics and social science, psychology etc. So far as known, the notion „homeostasis” was created by the physiologist Claude Bernard and published in 1865.
4. Gaia is the name of the goddess of Earth in the Hellenistic mythology. In Hesiod's Theogony (700 B.C.) the creation of Gaia was thought as the transition from chaos to cosmos. William Golding and James Lovelock used this name as a synonym for a self-regulating geophysical and biophysical mechanism on a global scale. According to this hypothesis, temperature, oxidation state, acidity as well as different physicochemical parameters of rocks and waters remain constant at each time due to homeostatic interactions maintained by massive feedback processes. These feedback processes are initiated by the „living world”, whereas the equilibrium conditions are changing dynamically with the evolution of Earth's live (not of single creatures).
5. Coccolithophorids such as *Emiliania huxleyi* and other marine microorganisms, including diatoms, produce dimethyl sulphide (DMS). Dimethyl sulphide is produced from dimethylsulfoniopropionate (DMSP) by (a) enzymatic cleavage of DMSP, (b) by viral infection of

E. huxleyi (some viral populations show dramatic CO_2 response) and (c) due to grazing of phytoplankton by microzooplankton (Wingenter et al., 2007, cf. references therein).

6. New particle formation (NPF) via nucleation over the ocean is still poorly understood. Over the last years it has been suspected, that new particles form via homogeneous nucleation of water vapour, sulphuric acid vapour and ammonia. There exists some evidence, that DMS-derived sulphuric acid is involved in NPF. However, modelling studies have shown, that while nucleation of sulphuric acid can occur, the concentration of sulphuric acid is too low to explain nucleated, thermodynamically stable clusters to grow to detectable sizes larger than 3 nm (O'Dowd et al., 2007). Field observations show a correlation of NPF with both solar flux and low tide. The identification of iodine oxides in particles and laboratory studies suggest, that UV photolysis of alkyl iodides (particularly CH_2I_2, emitted by macroalgae) in presence of ozone with subsequent homogeneous nucleation of OIO (iodide oxide) is responsible for NPF in the marine boundary layer (MBL). The following reaction mechanism is proposed to explain NPF in the MBL:

   $CH_2I_2 + h\nu \rightarrow CH_2I + I$(1)

   $I + O_3 \rightarrow IO + O_2$ (2)

   $IO + IO \rightarrow OIO + I$ (3)

   $OIO + OIO \leftrightarrow I_2O_4$ (4)

   $I_2O_4 + OIO \leftrightarrow\leftrightarrow\leftrightarrow$ „particle" (5)

   Reaction (5) represents a sequence of multiple OIO addition steps leading to the formation of a stable iodine oxide cluster (polymerisation). In this way, NPF results from single component homogeneous nucleation of OIO (Burkholder et al., 2004). It is suspected, that inhomogeneous sources of iodine oxides, i.e., „hot spots" with elevated iodine species emissions, are responsible for aerosol production in the coastal and open ocean marine boundary layer (Pechtl et al., 2006).

   Atomic iodine (I), released by photolysis of molecular iodine (I_2) and chloroiodomethane (CH_2ClI) in the MBL, can very efficiently catalytically destroy O_3 (Wingenter et al., 2007, cf. references therein):

   $I + O_3 \rightarrow IO + O_2$ (6)

   $IO + HO_2 \rightarrow HOI + O_2$ (7)

   $HOI + h\nu \rightarrow HO + I$(8)

   $O_3 + HO_2 \rightarrow HO + 2O_2$(9)

7. An increase of atmospheric CO_2 alters the seawater carbonate equilibria by shifting inorganic carbon away from carbonate (CO_3^{2-}) towards more bicarbonate (HCO_3^-) and increased ocean acidity:

   $CO_2 + H_2O \leftrightarrow HCO_3^- + H^+$ (1)

   $CO_3^{2-} + H^+ \leftrightarrow HCO_3^-$ (2)

   An increase of atmospheric CO_2 will result in a pH drop of surface seawater, an increase in hydronium (H^+) concentration and a fall in CO_3^{2-} levels. Some phytoplankton taxonomic groups favour CO_2 as their inorganic carbon source, while others consume mostly HCO_3^-. If increasing atmospheric CO_2 leads to greater DMS production, then this may contribute to the homeostasis of Earth's atmosphere (Wingenter et al., 2007, cf. references therein).

8. Over the time period from the past 800 kyr to the next 200 kyr, the obliquity (tilt) of the

Earth axis varies between 22.05° and 24.50° with a strong quasi-periodicity around 41 kyr. Changes of the obliquity impact the inter-seasonal variation of insolation as well as the annual mean insolation changes. As the obliquity impacts the annual mean insolation with opposite effects in low vs. high latitudes, there is no effect in global average insolation. The eccentricity of the Earth's orbit around the Sun has quasi-periodicities at 400 and around 100 kyr. Changes of the eccentricity modulate the Sun-Earth distance and have limited impacts on global and annual mean insolation. Nevertheless, variations of the eccentricity have an impact on the intra-annual changes of the Sun-Earth distance. In this way, a significant modulation of seasonal and latitudinal effects, induced by obliquity and climatic precession, is accomplished. The general precession of the equinoxes and the longitude of the perihelion causes periodic shifts of the position of solstices and equinoxes on the orbit relative to the perihelion. These shifts modulate the seasonal cycle of insolation with periodicities of about 19 and 23 kyr. As a result, changes in the position of the seasons on the orbit strongly modulate the latitudinal and seasonal distribution of insolation. During periods with low eccentricity (about 400 kyr and during the next 100 kyr), precession-induced changes of seasonal insolation are lower than during periods with larger eccentricity (Jansen et al., 2007, IPCC-WG I, Chapter 6, p. 445, Box 6.1 Orbital forcing).

9. According to Milankovitch's theory, ice ages are triggered by minima in summer insolation (insolation drops below a critical threshold) near 65°N, which enables winter snowfalls to persist all year and forming northern hemisphsric glacial ice sheets by snow accumulation. The onset of the last ice age about 116 kyrs corresponds to a 65°N mid-June insolation about 40 Wm^{-2} lower than today (Jansen et al., 2007, IPCC-WG I, Chapter 6, p. 445, Box 6.1 Orbital forcing). The next large reduction on northern summer insolation is expected to start in 30.000 years.
10. Between 1902 and 1957, the absolute value of the total solar irradiance (TSI) varied between 1322 and 1464 Wm^{-2}. The current estimate of the TSI is 1365 Wm^{-2}. Higher TSI values are associated with more solar faculae. Satellite missions revealed, that at the maximum of the 11-year solar activity cycle, the TSI is by about 0.1% larger than at the minimum. The TSI assumes its highest values when sunspots are at their maximum. Between 1672 and 1699 a low solar activity (sunspots) was observed, denoted as the Maunder Minimum. This time interval falls into the climate period of the Little Ice Age (from about 1350 to 1850).

    There is increasingly reliable evidence of the influence of solar cycle TSI variations on atmospheric temperatures and circulations, particularly in the higher atmosphere. From numerical simulations it was concluded, that TSI changes could cause surface temperature changes of the order of a few tenths of a degree Celsius. Satellite data in combination with historically recorded sunspot number, records of cosmogenic isotopes and characteristics of other Sun-like stars indicate quasi-periodic TSI changes of 0.24 to 0.30% on the centennial time scale (Le Treut et al. 2007, IPCC-WG I, Chapter 1, p. 107–108, see references therein).
11. One problem concerns the evaluation and interpretation of stellar data. By recent reassessment of such data it was not possible to separate low emission in non-cycling stars (Maunder-minimum type) from higher emission in cycling stars. Furthermore, the current Sun is thought to have „typical" (rather than high) activity relative to other stars. Another problem is the possibility of long-term instrumental drifts in historical indices of geomagnetic activity.
12. The open flux is well correlated with cosmogenic isotopes, which are modulated by heliospheric processes.

13. The favour of ion-induced nucleation compared to homogeneous nucleation results from two effects: Firstly, ion clusters represent nucleation sites for heterogeneous nucleation. A part of their surface serve automatically as a part of the interface area, required to separate the new embryo from the ambient mother phase. Thus, the surface formation energy is considerably decreased, if the vapour condenses on a pre-existing particle surface. Secondly, the work of Coulomb attraction forces act in the same direction as the volume term of formation energy, i.e., it decreases considerably the work required to form a cluster of critical size.
14. There are numerous examples in physics of large energy amplification factors. For example, nuclear chain reactions are released by a few initial neutrons (Wolfendale, 2001).
15. An increased solar attribution during the last century could indicate a steeper anthropogenic rise in recent decades.
16. If GCRs are able to modify the CCN number concentration in certain regions of the atmosphere, this may affect both cloud lifetime and albedo.
17. Wolfendale (2001) summarised the motivation to get deeper insight in the GCR-cloud mechanism by controlled experiments in a convincing statement: „If our only tool is correlation, we may continue another two centuries and still not be able to understand the underlying mechanism."
18. CLOUD is an acronym for Cosmic Leaving OUtdoor Droplets.
19. Theoretical studies of , e.g., Yu (2001) and D'Auria and Turco (2001) have shown, that ionisation effects are highly non-linear. Thus, experiments must reproduce ionisation rates and ionisation densities close to natural GCRs. Such conditions cannot be achieved with radioactive sources. A CERN pion beam closely duplicates natural GCRs and provides a precisely controlled and delivered particle ionisation inside the active volumes of the experiments.