1. Introduction: clean and polluted air

1.1 What is air?

Air and atmosphere are synonymous terms, with the term *atmosphere* used more in sense of being a reservoir as part of the geosphere (another reservoir is the hydrosphere for example), while *air* used in a more narrow sense for the chemical substrate of the atmosphere[1]. Based on physico-chemical characteristics, air is a gas mixture with particles suspended therein. Note that the natural ingredients of air should be dominating; otherwise the term *exhaust* air is used. Waste gases and/or exhaust air from technical systems lead to polluted air. To avoid any misunderstanding, we will use the term air only for *atmospheric* air. Exhaust we call either gaseous or particulate *emission*.

Beside solid particulate substances, we observe (but not continuously) water droplets in air. After the main constituents of air, nitrogen (N_2) and oxygen (O_2), water (H_2O) is the chemical species ranking on the third place (however, highly variable in its concentration). Most of the water exists as vapour (i.e. gaseous) and only occasionally liquid, forming drops (which in turn form clouds, fog, precipitation)[2]. Liquid water droplets (the aqueous phase in air) contain always soluted and unsoluted substances, however, in small concentrations only[3]. Water particles (among drops also ice and snow crystals, grains and other forms) are called *hydrometeors*.

The solid non-aqueous phase, also called particulate matter (PM), may also contain water, however, as a minor constituent only. This particulate matter in air is called *atmospheric aerosol*[4]. Note that atmospheric aerosol is not identical with PM - aerosol is defined as (solid) particles suspended in air; the particles are also called *aerosol particles*. Consequently, aerosol will not be emitted but the particles forming aerosol in air. With increasing humidity (i.e. water vapor in air), those particles, which are hygroscopic, can produce so called *cloud condensation nuclei* (CCN). In the case of some supersaturation, water vapour will condense on (activated) CCN and form cloud (or fog) drops within a few seconds; this process is called *heterogeneous nucleation*. We see that water droplets only exist under very limited conditions: the existence of CCN and the occurrence of supersaturation. When water vapour is going below saturation, droplets evaporate and an aerosol particle (changing in size and chemical composition – it becomes larger and more water soluble) as well as gases remain in air. Many chemical reactions, changing the atmospheric composition, occur simultaneously in gas and aqueous phase. Moreover, transport of clouds will distribute trace species over larger areas. This complex process – nucleation, chemical reactions, evaporation and transportation – is called *cloud processing*.

Only a few cloud events (about 10%) lead to precipitation. Precipitation, also called *wet deposition*, is part of the water cycle and removes trace species (natural and pollutants) from the atmosphere. Wet deposition of trace species is part of the global matter cycles and an input for the ecosystems. The particulate and liquid phase occupy only a very small part of air volume (about 10^{-6}), but have a huge significance for chemical and physical processes in the atmosphere. Air must be described as a multi-component and multiphase system.

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1 However, air (being the region above the ground) also is used in link with *land* and *sea* for the region. The atmosphere is defined as the mass of air surrounding the Earth.

2 Liquid water exists only in form of drops (between 1 µm for smallest cloud droplets and 10 mm for largest rain drops) in air. A collected sample of drops is called *bulk*-water.

3 High concentration occurs only during droplet formation (heterogeneous nucleation) and droplet evaporation as transient state.

4 Aerosol is general defined being a gas (or gas mixture) with suspended particles, where the particles may be solid or liquid. Because of very different physical and chemical properties of PM and droplets we will not include hydrometeors into atmospheric aerosol.

For a long time air has been regarded as a unique substance. The main constituents, O_2 and N_2 have been discovered at the end of the 18^{th} century. Today we know that air consists of thousands of different chemicals from natural and man-made origin. However, many years before, people have seen that in air "non-natural" substances can be observed. With the use of coal since the 13^{th} century soot and smoke must have resulted in an air quality in cities, which was much worse than today. *John Evelyn*[5] wrote in 17^{th} century:

> *It is this horrid Smoake which obscure our Church and makes our Palace look old, which fouls our Cloth and corrupts the Waters, so as the very Rain, and refreshing Dews which fall in the several Seasons, precipitate to impure vapour, which, with its black and tenacious quality, spots and contaminants whatever is exposed to it.*

Nowadays, in a situation like this we speak of *air pollution* or *polluted air*. Different man-made processes (sources of emissions) produce waste products, which (at least partly) escape into the atmosphere as *pollutants*[6]. The term "pollutant" should be used only for man-made (anthropogenic) emitted substances. The polluted air as a result of man-made emissions may be lower in its trace species concentration than remote air, far from anthropogenic sources at special locations (e.g. in a tropical forest). The chemical mixture (which varies over time and space) of the natural atmosphere is called *remote air*. We may also use the term *clean air*, describing the natural state. Note, however, that in nature situations may occur, such as volcanic eruptions, sand storms, biomass burning, where the air is being *polluted*, that is (natural) trace concentrations are increased. Therefore, the *reference* state of remote or clean air is a climatological figure[7].

The term *clean air* is also used in air pollution control[8] as a target, i.e. to make our air cleaner[8] in sense of pollutant abatement[9].

Another difficulty in defining the difference between clean and polluted air results from the point of view of the mankind in nature. Since the use of fire, a long time before any agricultural activities, people have been changing the atmosphere. A change in the chemical composition of the air is not only given by direct emitted substances, but is also due to all influences on the biosphere, esp. a change in land use (conversion from forest into agricultural or even devastated areas), which happened on a regional scale already 2000 years ago. Assessment of the resulting biosphere-atmosphere interaction on air chemical composition is still open. Before 1850 (non-industrialized era) we can regard the air being clean out of cities. Until the mid of the 20^{th} century (man-made) pollution was limited to cities and industrialized areas (in Europe e.g. Ruhr area, Upper Silesia, Saxonia-Bohemia etc.). In the 2^{nd} half of the last century with the huge industrialization in Northern America and Europe we detected new phenomena: acidification ("acid rain"), UV-B increase ("ozone hole"), forest decline, summer smog and climate change. The scale of impact rose from local to regional and global level. At present time we will not be able to find a location on earth with clean air in the sense discussed above, unaffected by man. However, one has to consider the very small concentration of man-made constituents in air (in %):

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5 John Evelyn: FUMIFUGIUM or The Inconvenience of the AER and SMOAKE of LONDON dissipated. Cited in Finlayson-Pitts and Pitts (1986).

6 In German often "Schadstoff“ (noxe) is used in translation of "pollutant". I emphasize the only use of "Spurenstoff" (trace species), because a chemical being a noxe depends only from its concentration in relation to the object of impact (see Chapter 3.1).

7 Averaged concentration over longer time (e.g. 30 years). The problem in determining such a reference level is that today we will not find any location in the world with clean air in this strong "natural" sense.

8 Air pollution control is not fully identical with the German "Luftreinhaltung" (to keep the air clean); mostly it is used in sense of emission abatement.

9 See the programme CAFE (Clean Air For Europe).

0,01 CO_2
0,0001 CH_4
0,0001 sum of all other pollutants.

This means, (dry) air is up to 99,99% "clean" and even 99,9999% when we consider all pollutants beside carbon dioxide and methane. It was already mentioned that the "natural" air varies in its chemical composition due to different biogenic and geophysical source processes. It is still unknown whether there is a natural trend for some species. When we understand the Man as part of nature[10] and the evolution of a man-made changed Earth's system, we also have to accept that we are unable to remove the present system of a pre-industrial or even pre-human state because this means to disestablish humans. The key question remains: which parameters of the system allow the existence of humans under specific conditions?

Air should be regarded as the chemical substrate of the atmosphere where natural and man-made sources contribute to its chemical composition. Thus, it is important to ask

a) what is the ratio between natural and man-made emission
b) what are concentration variations on different time scales
c) what are really trends given by man-made origin and
d) what are concentration thresholds for effects we may not tolerate?

1.2 What is air pollution management?

Management of air pollution means to change an unwanted condition into a wanted state of air by use of all appropriate measures to *manage* (negotiate, contrive, accomplish, direct, create, administrate, service). Air pollution is an expression used for the man-made change of the chemical composition of air (the atmosphere), see Fig. 1. This state must be recorded and assessed. Recording is made by air analysis using different chemical and physical methods based on different platforms (ground-based stations and networks, air-borne measurements by planes, remote sensing by satellites). Despite of problems in spatial resolution and missing species in measurement programmes it is not more a principal problem to create huge data sets on the chemical composition of the atmosphere. The assessment, however, is still a problem because we have to include quantification of effects of changing composition or – much simpler – of single pollutants. In this context we have to consider the complex interactions and feedbacks between the pollutants as well as between the atmosphere,

[10] This statement should never be forgotten. On the other hand, humans developed in contrast to all animals strong changes in nature over thousands of years. The Russian geochemist Wladimir Iwanonowitsch Vernadski (1863-1945) was first to propose the idea of biogeochemical cycling (what is the impact of life on geology and chemistry of the Earth?). Pierre Teilhard De Chardin (1881-1955) was a Jesuit who introduced 1925 the concept of *Noosphere*, the sphere of mind superimposed on the biosphere or the sphere of life. Not until after the appearance of Man on the Earth did the *Noogenesis* begin. The term *Noogenesis* comes from the Greek word *noos = mind*. Teilhard wrote: "Man discovers that he is nothing else than evolution become conscious of itself. The consciousness of each of us is evolution looking at itself and reflecting upon itself." This concept was also used by Edouard Le Roy (1870-1954) a french mathematician and philosopher (L'exigence idealiste et le fail d'evolution. Paris, Alcan, 1927). Vernadski met both in Paris and took the term "noosphere" first in 1931 being a new dimension of the biosphere under the evolutionary influence of mankind (. . . . ., 1944, 18, pp.113-120). Today the term "anthroposphere" also is used. The idea of close interrelation between Man and biosphere is topical in understanding the "Earth System", i.e. the climate change and used by Schellnhuber (1999) with the terminology „global mind" (H.-J. Schellnhuber: Earth System Analysis and the Second Copernican Revolution. *Nature*, Suppl., 402, C19-C23, 1999) and by Crutzen (2002) with "Anthropozän" (Paul Crutzen: A Critical Analysis of the Gaia Hypothesis as a Model for Climate/Biosphere Interactions. *GAIA* 11, 96-103, 2002).

biosphere and geosphere. A further question is that of concerning the problems resulting from subsequent effects. For example, climate change may be characterized by increasing mean surface temperature due to an impact of “greenhouse gases” (see Chapter 6.2) on the radiative budget of the atmosphere. A subsequent effect (2^{nd} climate effect) may be an increase of storm number and decrease of precipitation. The *problem*, however, is given (in this example) by damages of houses and loss of agricultural crops. This *last* effect will not be accepted by the society and, consequently, result into diverse measures of *air pollution control* (here, for example, the reduction of greenhouse gas emission) which (hopefully) will control the effect. You may guess that it is extremely difficult to forecast the results of a 50% reduction of emission due to the complexity which could make the relationship (in mathematical sense) positive or negative, linear or non-linear. Fig. 1 illustrates the close interaction of the atmosphere with other reservoirs as well as the link between the physical and chemical climate.

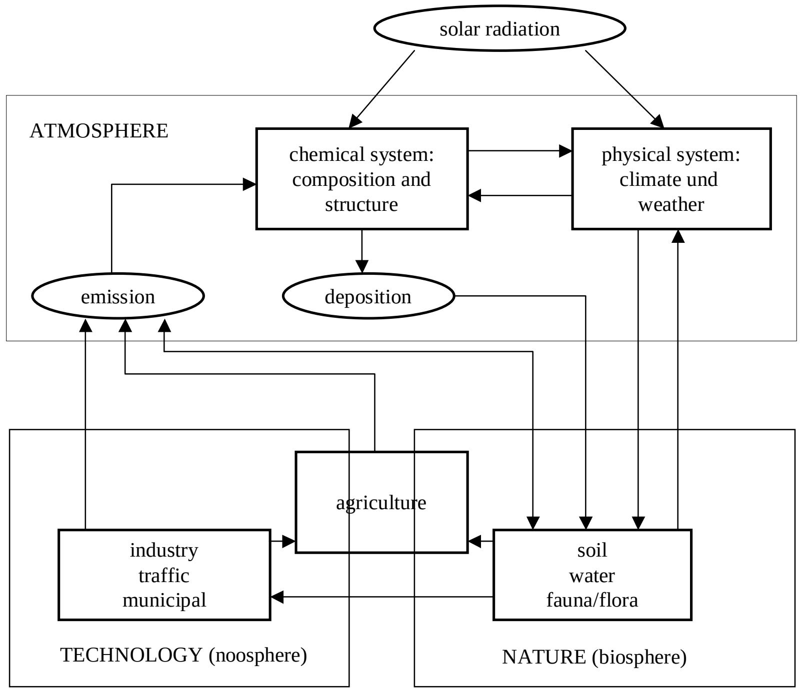

Fig. 1. System atmosphere: interaction of natural and anthropogenic processes (Earth system)

As mentioned before, the chemical composition of air has been changing since the settlement of humans. Beside the scale problem (from local to global) we have to regard the time scale. Natural climate variations (e.g. due to ice ages) had a minimum time scale of 10000 years. The man-made changes in our atmosphere over the last 2000 years were quite small until the 1850s. In the last one hundred years, however, the chemical composition was changed drastically. For many pollutants anthropogenic emissions are in the same or even larger order of magnitude than natural ones. Due to the huge population density, need (or consumption)[11] of materials and energy we totally separated our matter fluxes from natural cycles. Adaptation and restoration of natural systems is impossible within our time scale. We should not forget that “nature” is unable to assess its own condition. With other words, the

[11] It is quite an interesting question: Do we need all this consumption? What consumption do we need to realize a cultural life? Of course we move to sociological and political dimensions (life sciences) in answering these questions. But I see huge potential to economize and save resources in answering these questions and its implementation. Karl Marx wrote: “The philosophers have only managed to interpret the world in various ways. The point is to change it".

biosphere will accept all chemical and physical conditions, even worse (catastrophic) ones. Only humans evaluate the situation with the statement to accept it or not.

Thus, air pollution management comprises a set of measures, beginning with air quality assessment, evaluating the effects and disadvantages and finally installing actions to reduce emissions (Fig. 2). Because of the already described problems (how to define the reference state? How to separate between natural and anthropogenic “pollution”? What is the deviation in sense of “pollution” from a mean state?) in a scientific definition of air pollution we will use this term in the further lecture with caution. Air pollution is a common expression for the problem of a changing atmosphere. In the next subchapter we will consider, what the science to study air pollution is.

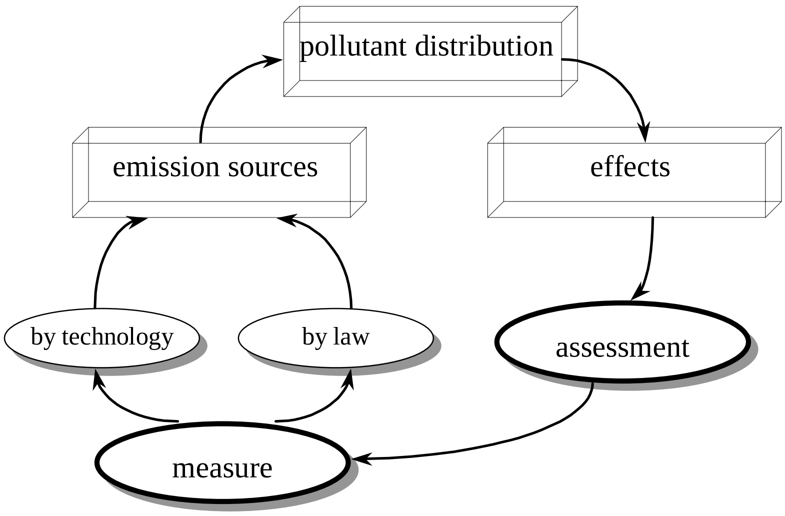

Fig. 2. The cycle of air pollution controlling

1.3 Air chemistry – the science to study air pollution

The chemical composition of air depends on the natural and man-made sources of trace species (their distribution and source strength) as well the physical (e.g. radiation, temperature, humidity, wind) and chemical conditions (other trace species) which determine transportation and transformation. Physics and chemistry can not be separated in atmosphere for understanding the fate of pollutants.

Here we will consider what the task of air (or atmospheric) chemistry in studying of air pollution is. The “physical part” of air pollution will be dealt with by (environmental)[12] meteorology. Both disciplines are essential for describing the fate of air pollutants and to provide data for assessment of atmospheric changes with the background of impacts and effects.

We define air (or atmospheric) chemistry as the science of origin and fate of trace components in air. The fate includes distribution (which is the main task of meteorology), chemical conversion, phase transfers (reservoir distribution) and deposition of species. Deposition is going on via different mechanisms from gas, particulate and droplet phases to Earth’s ground including plants animals and humans. It is the removal from atmosphere but

[12] The chair of Professor Schaller is called „Environmental Meteorology“. Within the last 10 to 20 years many things have been added to the word *environment*. But what is environment? It is the surroundings of Man. It is the world in which we live. Using the word “environmental” we emphasize that we consider a relationship between nature and society.

input to other spheres. Thus, atmospheric chemistry is not a pure chemistry and also includes other disciplines which are important for describing the interaction between atmosphere and other surrounding reservoirs (biosphere, hydrosphere, etc.). Measurements of chemical and physical parameters in air will always contain a "geographical" compartment, i.e. the particularities of the locality. General conclusions or transfer of results to other sites should be made very carefully. It is a base task in atmospheric chemistry to not only present local results of chemical composition and its variation in time, but also to find general relationships between pollutants and their behaviour under different conditions. "Pure" air chemistry can only be realized in laboratory under controlled conditions. This is also the only way to study the kinetics and mechanisms of chemical reactions (in a variety of so-called smog chambers). We need these data to model conversion processes in atmosphere. Due to transport processes, we can only simulate concentration patterns in air with Chemistry-Transport-Models (CTM). However, it is possible to use so-called box models, disregarding transport (like in a well stirred reaction vessel) to study reaction chains and mechanisms theoretically. Those model results may validate with smog chamber studies. CTM can only validate with 3-dimensional measurements in the atmosphere. Atmospheric chemical measurements always have to be accompanied by physical (meteorological) measurements.

Subdivision of air chemistry is done in many different approaches, using "classical" chemical branches like organic and analytical chemistry but more from different "subregions" of the air, see Fig. 3.

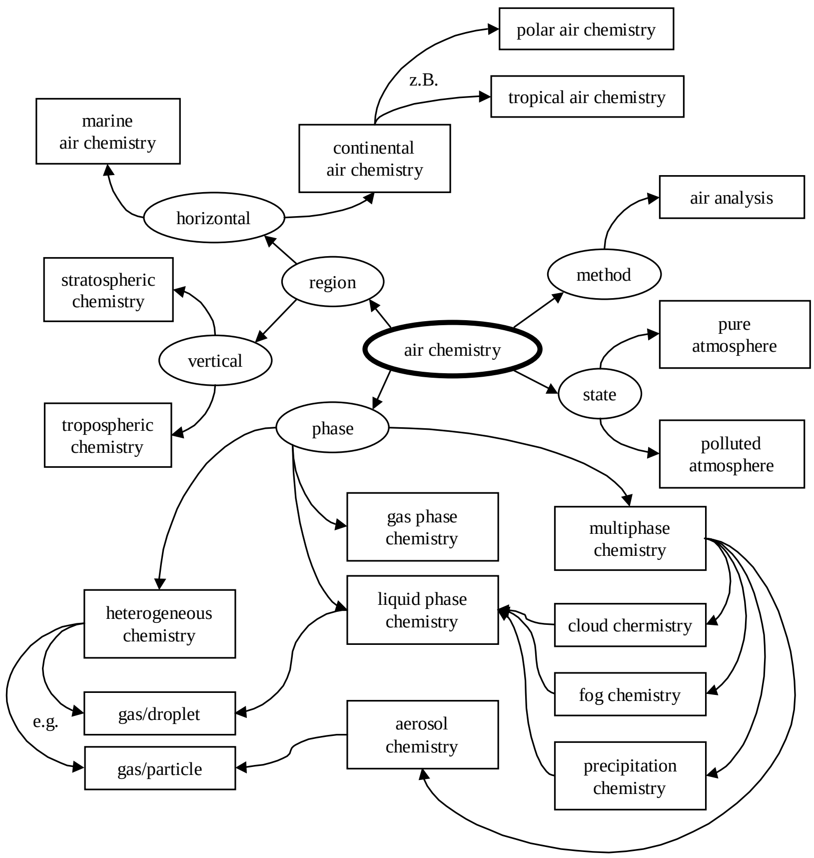

Fig. 3. Approaches to atmospheric chemistry

On the other hand, it makes sense to make "pure" physical measurements in atmosphere to study dynamical processes, radiation transfer, cloud dynamic etc. Physics of the atmosphere is the basic science of the atmosphere to describe its structure without going on a molecular level (chemistry). But very soon you will see again that "chemistry" at least in sense of the material structure of the atmosphere is needed to explain physical processes, e.g. radiation and heat budget. Meteorology is not simply physics of the atmosphere. There are tendencies to include atmospheric chemistry into meteorology. The term atmospheric chemistry was not introduced to public before the 1960s. Before terms like chemical climatology and meteorology were used. Thus, meteorology could be regarded as the science[13] of physical and chemical conditions of the atmosphere. It is not the aim of this short introduction to treatise differences between disciplines[14] but to make it clear that air pollution management demands an interdisciplinary approach.

Going back to the antique statement that the world consists of four elements (air, water, soil and fire) we can nowadays state out that the air again must be regarded as some type of micro-cosmos containing the "elements air, soil, water and fire". Expressed in modern words, air consists of gases, particles and hydrometeors "hold together" by thermodynamic (chemical and physical) forces.

1.4 The problem of air pollution

An understanding to assess polluted air, or in other terms, the air quality, is based on the problems resulting from changing chemical air composition. However, as already mentioned, not the given air concentration (burden or load) of a pollutant is the *problem* but the (possible) resulting effects (Fig. 4). In this context we will consider the load-carrying ability (in German: *Belastbarkeit*). There are definitions, for example the capability of a system to tolerate a certain degree of loading (in sense of a buffer capacity) without prejudicial changing immanently parameters responsible for the structure and function of the system (Guderian, 1977). But, how define "prejudicial changes" and "immanently parameters" to quantify air pollution impacts? The complexity of the system atmosphere makes it so tricky to identify man-made effects and to assess subsequent modifications of the system. This does not implicate that polluted air at a given location is harmful in sense of an impact.

It was already mentioned that *air pollution control* is considered as air pollution *management*. The term air pollution control (in the German sense "Luftreinhaltung") is mostly used in a sense of emission reduction due to technological measures and by law (see Fig. 2), focussing on the source of emission. By far more effective is an approach to avoid emissions (and waste) as a primary goal of the *waste-free technology*. This idea, already suggested in the 1970s, is based on a sustainable strategy using mostly closed matter cycles and reduced consumption of resources[15]. Such strategy may hardly be realized by single enterprises and will lead to a conflict between market economy and interests of the society. We hope that this conflict is temporary only and even the market economy will have the capability in analogy to self-organizing and optimizing ecological systems to follow this approach of sustainability. A sustainable system, however, must show a harmonization between production and consumption. Consumption by individuals and in sum by society

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13 In German the word „Luftkunde" would summarize this approach.

14 The term „discipline" is based on discipline, correct, fine and consequently isolate. In this way different scientific branches can be definend and delimit esp. because of practical reasons. In the last decades in the field of environmental sciences came into being many new disciplines: environmental chemistry, ecotoxicology, geoecology, ecological chemistry, environmental physics etc. On the other hand most of new findings have been found interdisciplinary and we observe a tendency to disintegrate disciplins. The best example is creating the term Earth´s sciences (the science of the Earth´s system) where biology, chemistry, geology etc. have to be involved.

15 This will be realized in the so-called solar era when energy is exclusively taken from solar flux, carbon (and materials based on C) from atmospheric CO_2 and all other elements taken from biogeochemical cycles and put (partly transformed) again into natural cycles without not tolerable changing rates of accumulation.

should have a primate (unfortunately, today production is the primary goal). The answer on the question what and how much will be consumed was until now firstly given by the difference of a dictatorship and a democratic society. So simple, however, we will not longer answer this question in future. Optimization and limitation are essential elements of air pollution control (in a wider sense of environmental protection); how far a democratic society will go this way depends strongly on the cultural level, esp. education of individuals and society at all[16]. This needs newly developed social norms but also governmental control.

High consumption lead to production using inappropriate technologies rising to levels, that *problems* have been detected at all scales (Table 1).

change (origin)	property (state)	effects of impact
acid-base budget	acidity	corrosion, forest decline
increase trop. O_3	oxidation potential	forest decline, health
increase of CO_2, CH_4 et al.	greenhouse effect	heating
increased aerosol contents	aerosol forcing	cooling
particulate composition	toxicity	health
decrease strat. O_3	UV(B) radiation	health

Tab. 1. Atmospheric environmental problems

As given in 1.2 in discussing the term "problem", based on the example of climate change, I will further explain the "levels of problem", in the following on the example of acidification. An increase of atmospheric acidity (acidification) due to changing budget of emissions forming acids and/or bases in air is not a problem for the atmosphere itself. The forester for example will observe reduced wood growth, the geologist increased erosion rates and the ecologist a changing land use structure. Therefore it is important to separate the effects (to real *problems*) on different levels (Fig. 4)

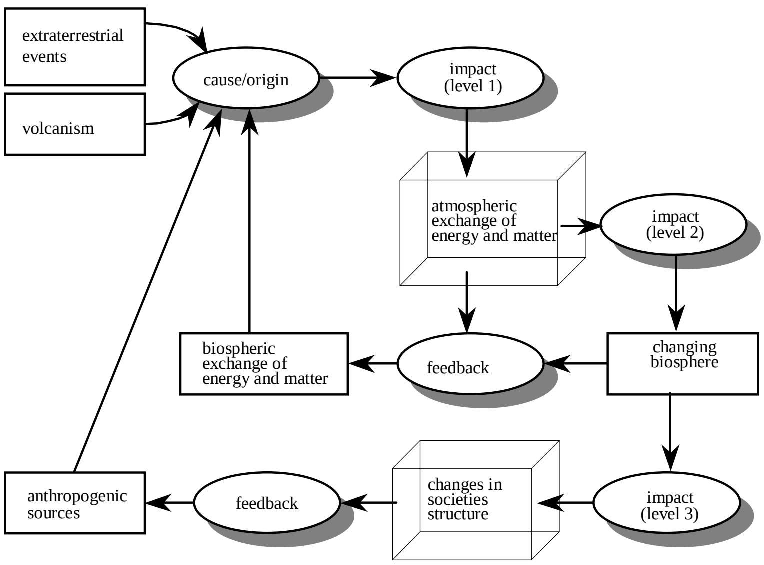

Fig. 4. Impact levels in air pollution management

We will see that man-made changes of the chemical composition of air are linked with the whole development of human society. Thus, it is not our aim in air pollution management to

[16] *Hegel* defined freedom as subjective insight into objective demands, what is the point of view in this connection.

create and maintain a natural[17], but an acceptable state of the man-environment relationship. The difficulty lies in determination of the parameters describing this acceptable state. The story of mankind shows that it has always been movement between extremes. It is the task of natural sciences to determine the threshold, where crossing the line leads to catastrophic developments of the Earth's system when concerning human existence. In connection with the climate discussion we will see that this border is very uncertain. In a society decisions on limitations in technique and air pollution control will be made by the government; their decisions also may be wrong[18]. Detailed questions (emission figure limitations, measurements methods, health effects) will be treated by national and international expert groups.

Nowadays the most important atmospheric problems (see also Table 1) are listed below (this is not a rank list – to my opinion it is impossible to state a general rank list):

- *Tropospheric ozone*, i.e. the change (increase) of atmospheric oxidation capacity. We understand the ozone formation very well; some open questions concern the multiphase chemistry. Ozone is a key substance also indicating many other (more) harmful species in air. Air pollution control in the last decade led to a decrease of ozone exceedance but not the mean concentration figure. In contrast, it seems that annual mean figures are further rising due to CH_4 and CO as the main global precursors.

- *Atmospheric acidity* (acid deposition); it was the phenomena „acid rain" detected around 30 years ago due to an increase in concentration of the main precursors SO_2 and NO, which form acids in air. Nowadays, SO_2 is not longer a problem in Europe; we see a shift to more alkaline rain (relative NH_3 increase). The acidity problem is shifted to Asia, esp. China.

- *Greenhouse gases* (CO_2, CH_4, N_2O, O_3 and others) are still with increasing rates emitted or secondary produced in air (CO_2, O_3) and lead doubtless to atmospheric heating. The problem is that formation of CO_2 formation during combustion of fossil fuels is not avoidable. Other important greenhouse gases (CH_4 and N_2O) are by-products in agriculture and linked with food production. There is no successful idea to reduce these emissions.

- *Aerosols*, esp. sulfates lead to atmospheric cooling and mask the greenhouse effect (Charlson und Heintzenberg, 1995). SO_2 as the sulfate precursor is very limited in Europe (and partly Northern America), where the net greenhouse effect will rise. In Asia SO2 still plays an important role. A main issue is the short residence time of aerosol particles (1 week) in contrast to that of greenhouse gases (years).

- *Aerosols (particulate matter)* becomes now a human health problem. It is not yet clear what the impact function is, but many studies show serious health effects on humans living under influence of high PM pollution.

- Decrease of *stratospheric ozone* due to long living tropospheric substances (first of all chlorfluorhydrocarbon and N_2O) lead to an increase of UV-B, harmful to biosphere and human skin. Other effects within the atmosphere (changing photochemistry, temperature distribution) are not well described, but may have negative feedbacks).

[17] It seems that this is exactly the aim of some organisations, but it is not only unrealistic, it is even anti-human because it may be realized only with negotiation of mankind.

[18] In my opinion the German decisions to close nuclear power station ("irreversible exit") is wrong from environmental and energetic point of view. Controlled fission is the only alternative to burning of fossil fuels with huge CO_2 emission before beginning the solar era (in about 100 years).

At this place it is pointed out, that it is not dealt with the protection of nature, but the protection of mankind. Nature itself – without man – adapts to whatever circumstances imagineable. There is not anyone to disagree that the atmosphere of Venus is facinationg, although it does not allow for any forms of life to exist. Sometime, the Earth's atmosphere will be affected by cosmic events, leaving conditions which do not allow for man to exist any further. Not only with this background man should see himself as part of nature[19].

It has become a fact, that mankind may not survive the 21st century, if consumption and production continues to grow the way it has. The limitation of resources demands for a steady development, and also results in the necessity to integrate emissions (and waste) in usable matter cycles and to use energy from the solar flux. Mankind is still far away,[20] and it seems as if current social matters have pushed all other problems into the background. Also, there seems to be an unsolvable gap between the conditions of life in "poor" (developing) and "wealthy" (developed) nations. Without adapting to specific cultural and economic circumstances problems can not be solved on a global level. However, global problems can not be solved by transferring the conditions of life of the *wealthier* to the *poorer* countries. A way must be found that does not affect the quality of life of the people in the wealthy countries in a negative way, but which is connected with a (significant) decrease in the use of resources and the production of waste materials.

19 The vision of a *perfect* man remains a dream of philosophers.

20 Still, it is not just utopia: generally all technologies are known and experience has shown that science and technology have been able to solve almost any kind of problem – *under pressure*.