

THE ROLE OF TROPICAL ATMOSPHERIC CHEMISTRY IN GLOBAL CHANGE RESEARCH: THE NEED FOR RESEARCH IN THE TROPICS AND SUBTROPICS

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The main permanent components of the atmosphere N_2 , O_2 and Ar together make up more than 99.9 volume % of the atmosphere. Nevertheless, the Earth's climate and the chemistry of the atmosphere are mainly determined by the remaining minor constituents, which because of their relatively low abundance are significantly affected by human activities, in particular by fossil fuel and biomass burning, chemical manufacturing, agriculture and land use changes. Most abundant among these gases is carbon dioxide, which plays essential roles as the carbon feedstock for the photosynthesis of plant matter and for the Earth's climate. CO_2 , however, does not play any significant role in the chemistry of the atmosphere. Among the chemically active gases, methane (CH_4) is most abundant with a volume-mixing ratio of about 1.7 ppmv, compared to a pre-industrial value of only about 0.7 ppmv. Methane plays important roles in the photochemistry of both the troposphere and the stratosphere. The next most abundant gas of chemical importance is nitrous oxide (N_2O). Chemically almost inert in the troposphere, N_2O is removed from the atmosphere by photochemical destruction in the stratosphere. A fraction of the nitrous oxide is thereby oxidized to nitric oxide (NO), which, together with NO_2 , acts as a catalyst in an ozone-destroying cycle of reactions. In the natural stratosphere, the production of ozone (O_3) by the photodissociation of O_2 is largely balanced by its catalytic destruction by NO_x ($= NO + NO_2$). Because the abundance of N_2O is increasing by 0.2 – 0.3%/year, partially due to the increased production of N_2O in soils as a consequence of the rapidly growing application of N-fertilizer, there is an anthropogenic effect on

stratospheric ozone, although at a relatively slow rate. The most important anthropogenic impact on stratospheric ozone is due to the emissions of a series of entirely manmade chlorine – (and bromine) containing compounds, in particular CFCl_3 , CF_2Cl_2 , and CCl_4 . As with N_2O , these gases are only removed from the atmosphere by photodissociation in the stratosphere, thereby producing Cl and ClO radicals, which, even more efficiently than NO_x , diminish ozone, by catalytic reactions. Most surprisingly, the strongest depletions in stratospheric ozone have been found to occur over Antarctica during the springtime months of September and October. Exactly in the height region (14-21 km) where naturally, and until about 2 decades ago, a maximum in O_3 concentrations was found, O_3 has now totally vanished, resulting in major depletions in the total ozone abundance in the atmosphere and major increases in the fluxes of biologically damaging ultraviolet, so-called UV-B, radiation at the Earth surface.

The functions of ozone in the atmosphere are manifold. It acts as a filter against solar ultraviolet radiation, thereby protecting the biosphere from a large fraction of the biologically active radiation of wavelengths less than about 310 nm. About 90% of all ozone is located in the stratosphere and 10% in the troposphere. Both are substantially affected by human activities. Contrary to what has happened in the stratosphere, ozone concentrations in the troposphere have increased, as is clearly noticed during photochemical smog episodes, but also more generally in regions that are affected by anthropogenic emissions of methane and other hydrocarbon, carbon monoxide (CO) and nitric oxide (NO), such as the mid-latitude zone of the northern hemisphere and also the continental tropics and subtropics as a consequence of biomass burning during the dry season. Ozone is deleterious to the biosphere, affecting human health and plant growth, especially agricultural productivity.

The role of ozone in the troposphere is, however, not only negative. In fact, it fulfils a very important function in the removal of almost all gases that are emitted into the atmosphere by nature and human activities. The latter occurs mainly via reactions with hydroxyl (OH) radicals which are largely formed by the absorption of solar UV-B radiation by ozone, leading to the production of electronically excited O atoms which have enough energy to react with water vapour to produce hydroxyl radicals. Despite very low tropospheric concentrations, globally averaging about 4×10^{-14} by volume, it is hydroxyl, and not abundant molecular oxygen (O_2), which is responsible for cleaning the atmosphere. Because of maximum abundance of UV-B radiation and water vapour, the concentrations of hydroxyl radi-

cal are largest in the tropics and subtropics. A quantitative understanding of the chemistry of the atmosphere requires, therefore, good knowledge of the chemistry of the tropics and subtropics.

Specifically in the tropics and subtropics there exist major gaps in knowledge and observations of many key species in tropospheric chemistry, in the first place of ozone, but also of those species which determine the oxidizing efficiency (that is OH concentrations) of the atmosphere, such as CO, hydrocarbons and NO_x. The continental tropics and subtropics are already substantially affected by mostly human-caused biomass burning. In future, agricultural and industrial activities will particularly grow in these regions on the globe. The study of the influence of these on atmospheric chemistry (e. g. ozone and hydroxyl concentrations) and climate is an important task for the atmospheric chemistry community. This requires much enhanced research activities in the tropical world which should also involve researchers from the developing world.

The present state of quantitative knowledge about particulate matter in the troposphere is even in worse shape than that of the gas phase. The role of aerosol is manifold:

1. Particulate matter can influence the chemistry of the atmosphere by providing surfaces and liquid media for chemical reactions, which often can not take place in the gas phase.
2. By the scattering and absorption of solar radiation, particulate matter plays a substantial role in the radiative properties of the atmosphere and, therefore, in the Earth's climate.
3. This influence is emphasized by the fact that atmospheric particles can serve as condensation and ice-forming nuclei. Recent studies have indicated the possibility that climate warming due to increasing levels of greenhouse gases has been substantially counteracted by the backscattering to space of solar radiation, both directly from the aerosol under cloudfree conditions or indirectly by increased albedo of clouds. Although it appears that calculated and observed temperature trends agree much better with each other when optical aerosol effects are included in global climate models, thus providing some evidence for the significance of the aerosol-climate feedback, those conclusions are still based on rather weak grounds, again largely because of lack of knowledge about the physiochemical properties and distributions of atmospheric aerosol. In particular, the above mentioned model runs were performed only considering sulfate aerosols, which are strongly derived from coal and oil burning. However, several addition-

al types of aerosol, which can likewise be influenced by human activities, are emitted into the atmosphere, such as

- * smoke sunlight-absorbing aerosol, mostly from tropical and subtropical biomass burning;
- * soil dust;
- * organic aerosol, resulting from gaseous organic precursor emissions from vegetation;
- * seasalt particles.

Clouds can provide major pathways for the chemical processing of natural and anthropogenic emissions. While this chemical cloud effect has been studied for a few major components such as SO_2 , there are many more soluble and reactive atmospheric constituents whose cloud processing is largely unknown.

Neither the quantities of emissions nor the global distribution of these aerosol are even approximately known, but one thing is clear: they all play important roles in the climate and the chemistry of the atmosphere. And again, also here the main gaps in knowledge may well be in the tropics and subtropics.

The lack of knowledge also concerns the interactions of gas phase species with the aerosols. As an example, in all climate simulations the calculated distributions of sulfate aerosols have been conducted, neglecting potential interactions of anthropogenic SO_2 with the other types of aerosol. This may well mean that much of the sulfur, which is emitted into the atmosphere, may be deposited on other aerosol, such as soil dust and seasalt particles. If that is so, then no additional sulfate particle formation could take place in regions with high emissions of such particles, implying that the sulfate cooling effect may have been substantially overestimated. What is needed most now are measurements of the emissions and global distributions of the various kinds of aerosol, especially in the tropics and subtropics.

Closely connected to what has been said above about the atmospheric chemistry aspects of global change, are biosphere/atmosphere interactions, as many of the chemically and climatologically important trace gases are likewise to a substantial degree emitted into the atmosphere by the biosphere. Besides CO_2 and N_2O , we mention especially NO , CH_4 , and reactive hydrocarbons which together have a substantial impact on O_3 and OH concentrations and which are increasingly impacted by human activities.

I propose, therefore, that in future "global change" research substantial attention is given to the tropics and subtropics. This also requires the

involvement and training of local scientists who participate in joint field programmes: a strong scientific basis in this part of the world will in future not only benefit progress in science, but will also lead to greatly improved scientific inputs in political decision making.