In the shortwave wavelength range, spectrally resolved observations from space are increasingly used for trace gas retrievals but have only rarely been exploited for cloud and aerosol applications. This is surprising, since spectrally resolved irradiance and radiance measurements taken from the ground and from aircraft suggest that cloud-aerosol research could benefit significantly from using spectral information. In fact, it can be argued that the key to some of the outstanding questions regarding the radiative effects of clouds and aerosols lies in the spectral dimension. To support this statement, some recent measurement and modeling results of solar spectral radiation will be discussed. The first example is related to cloud heterogeneities, which may have led to a systematic overestimation of the true cloud absorption in past experiments. Irradiance measurements in tropical ice clouds have shown that the main contributor to this bias, net horizontal photon transport, can be detected using the spectral shape of cloud albedo or apparent absorption (vertical component of flux divergence), and that the true (unbiased) absorption can be spectrally attributed to individual absorbers (clouds drops, ice crystals, gas absorbers, and aerosol particles). In a second example, a broken cloud field that is immersed in an aerosol layer will be considered. Due to the complexity of this problem, the irradiance measurements can only be reproduced by 3D radiative transfer calculations. Various spectral ranges exhibit different sensitivity to aerosol and cloud optical properties, 3D cloud effects, molecular scattering and absorption, and the surface; they thus contain information about both cloud and aerosol properties. As a third example, a new retrieval technique for cloud optical thickness and effective radius will be presented and developed for transmitted spectral radiance observations that relies on spectral slopes of normalized transmittance rather than absolute transmittance values. The low sensitivity of transmitted radiance to effective radius has until recently caused an unacceptably large retrieval error of this quantity. The new technique reduces the retrieval uncertainty, particularly the dependence on radiometric calibration. This is illustrated with data from the recent CalNex experiment. Currently, how to optimally propagate various errors into the retrieved product(s) is explored: radiometric uncertainties (largely bias-driven, or spectrally correlated), noise (dark currents, spectrally uncorrelated), and forward model errors (e.g., cloud heterogeneity). Finally, the term "cloud-aerosol spectroscopy" for a new type of retrievals is proposed.